Energy & Environmental Science

Rapid technological progress in white light-emitting diodes and its sources in innovation and technology spillovers

Journal:	*Energy & Environmental Science*
Manuscript ID	Draft
Article Type:	Analysis
Date Submitted by the Author:	n/a
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Supplementary Information for:
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"Rapid technological progress in white light-emitting diodes and its sources in innovation and technology spillovers" *Energy and Environmental Science* (2023)

Contents

1 Luminous Efficacy Metrics

Figure 1 in the main publication uses luminous efficacy of radiation as the primary metric to describe progress in lighting technologies. Care must be taken not to confuse this metric with luminous efficacy of source, which is used in SI11.

1.1 Luminous Efficacy of Radiation

This metric describes the match of a light-emitting diode package spectrum to the human visual system. Efficacy in lighting is dependent on the luminosity function, which describes the wavelength-dependent sensitivity of the human eye. A light source emitting very *efficiently* in the infrared yet emitting no visible light has a very low *efficacy*. The luminous efficacy of radiation K is mathematically defined as the normalized, integrated product of the spectral radiant flux of a light source with the wavelength-dependent human sensitivity to light [1]

$$K[\mathrm{lm/W}_{opt}] = \frac{\int_0^\infty K(\lambda)\phi \mathrm{d}\lambda}{\int_0^\infty \phi \mathrm{d}\lambda} \tag{SI1}$$

where

$$K \ldots \text{spectral luminous efficacy}$$
$$\phi \ldots \text{spectral radiant flux}$$
$$\lambda \ldots \text{wavelength}$$

This metric can be computed from spectral data alone and does not require additional spectral normalization. It enables straightforward comparison between the performance of different downconversion phosphors, as shown in the top panel of Fig. SI11. Light sources emitting in the far red or blue part of the spectrum have lower efficacy of radiation. Care must be taken not to confuse this efficacy metric with the *efficacy of source* described in the following subsection.

1.2 Luminous Efficacy of Source

The luminous efficacy *of a light source* η is defined as the ratio between the emitted luminous flux and the consumed electrical power [2]

$$\eta[\mathrm{lm/W}_{el}] = \frac{\phi}{P_{el}} \tag{SI2}$$

This metric is often cited in device datasheets, scientific literature and textbooks when describing the performance of light-emitting diodes. Care must be taken not to confuse this efficacy metric with the *luminous efficacy of radiation*, which depends only on the spectral characteristics of a light source. As the luminous efficacy of a light source η captures the overall device efficacy, it depends on a large number of other device properties and parameters. This makes attribution of changes in this metric to individual changes in device design or manufacturing difficult. For this reason, we do not use this metric in our study.

2 Manufacturing Cost Model

2.1 Structure of the Model

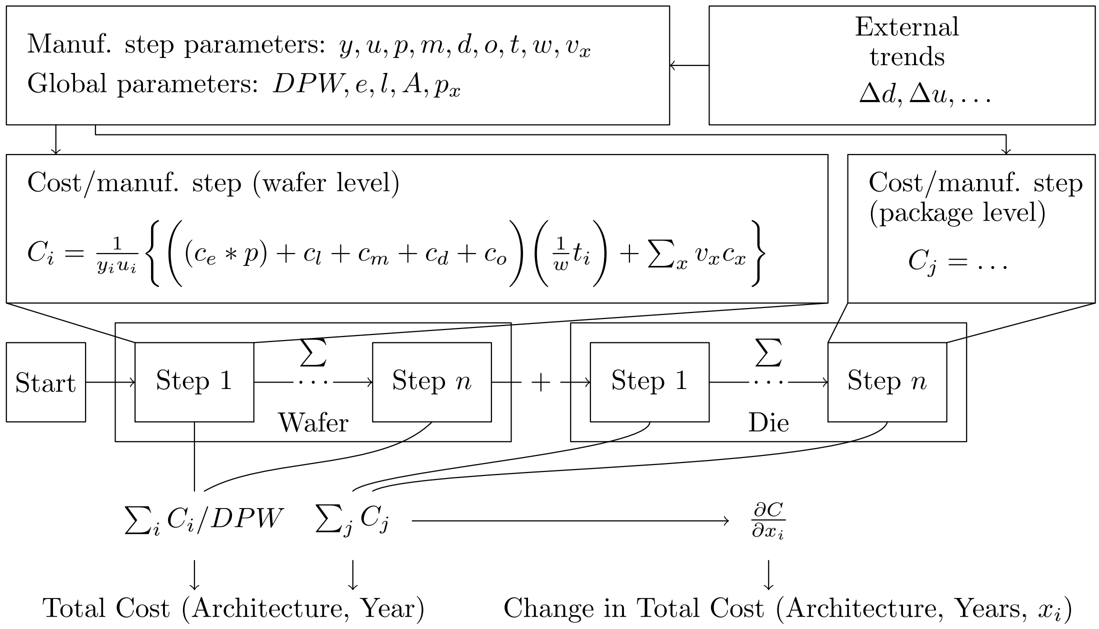

Figure SI1: Schematic diagram of the cost model showing inputs to each step and computational steps leading to the cost model outputs. For the description of cost variables, see definitions for Eq. (SI6). For the computation of total cost, see Section 2.3. For the computation of changes in total cost, see Section 3.

The cost model adapted for this publication is a microeconomic manufacturing cost model. Within the timeframe and scope laid out in the main publication, it returns the total manufacturing cost of phosphor converted warm white light-emitting diode packages. In this computation, it considers the main economic factors associated with operating and maintaining manufacturing equipment. It does not consider costs associated with research and development or those associated with the construction of manufacturing facilities. It considers market trends through their effect on manufacturing parameters.

A schematic diagram of the cost model is presented in Fig. SI1. The cost model is process step-based. It is split between the two stages of the manufacturing process: the first stage combining operations at the wafer level, followed by the LED packaging stage. The model takes as inputs parameters specific to individual manufacturing process steps (*"manufacturing step parameters"*) and parameters affecting all manufacturing steps (*"global parameters"*). The cost for each process step is then computed. The cost model returns returns the costs of individual manufacturing steps as well as the total manufacturing cost. It further considers the yield per step and returns the cumulative yield, the yielded cost per step and the yielded total manufacturing cost.

2.2 Manufacturing Process Steps by Chip Architecture

Fig. SI2-Fig. SI6 show a simplified rendering of the manufacturing process of three different chip architectures considered in the cost model.

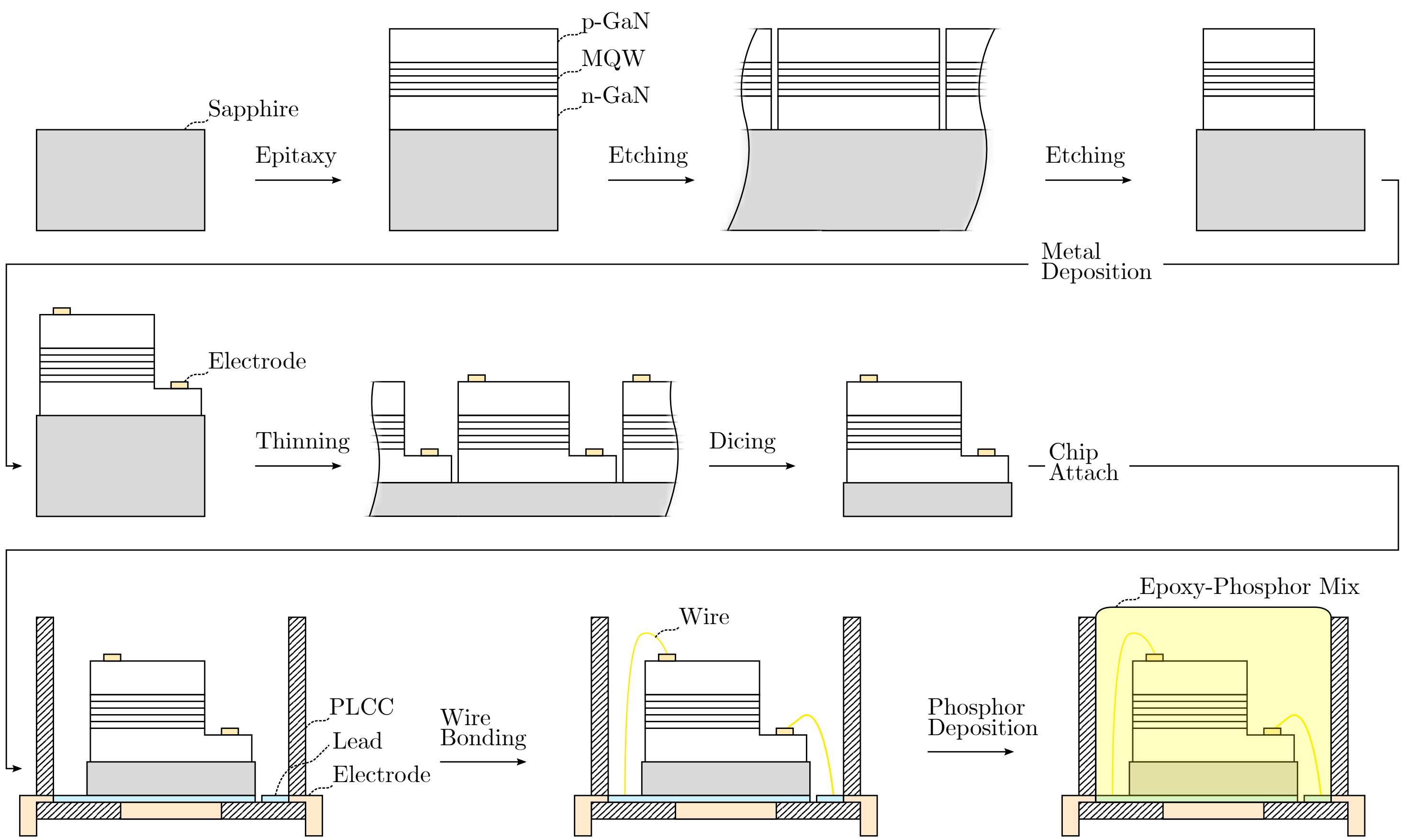

Figure SI2: Manufacturing process for a classical LED package with lateral current spreading, circa 2003. Abbreviations: MQW - multiple quantum well; PLLC - plastic leaded chip carrier.

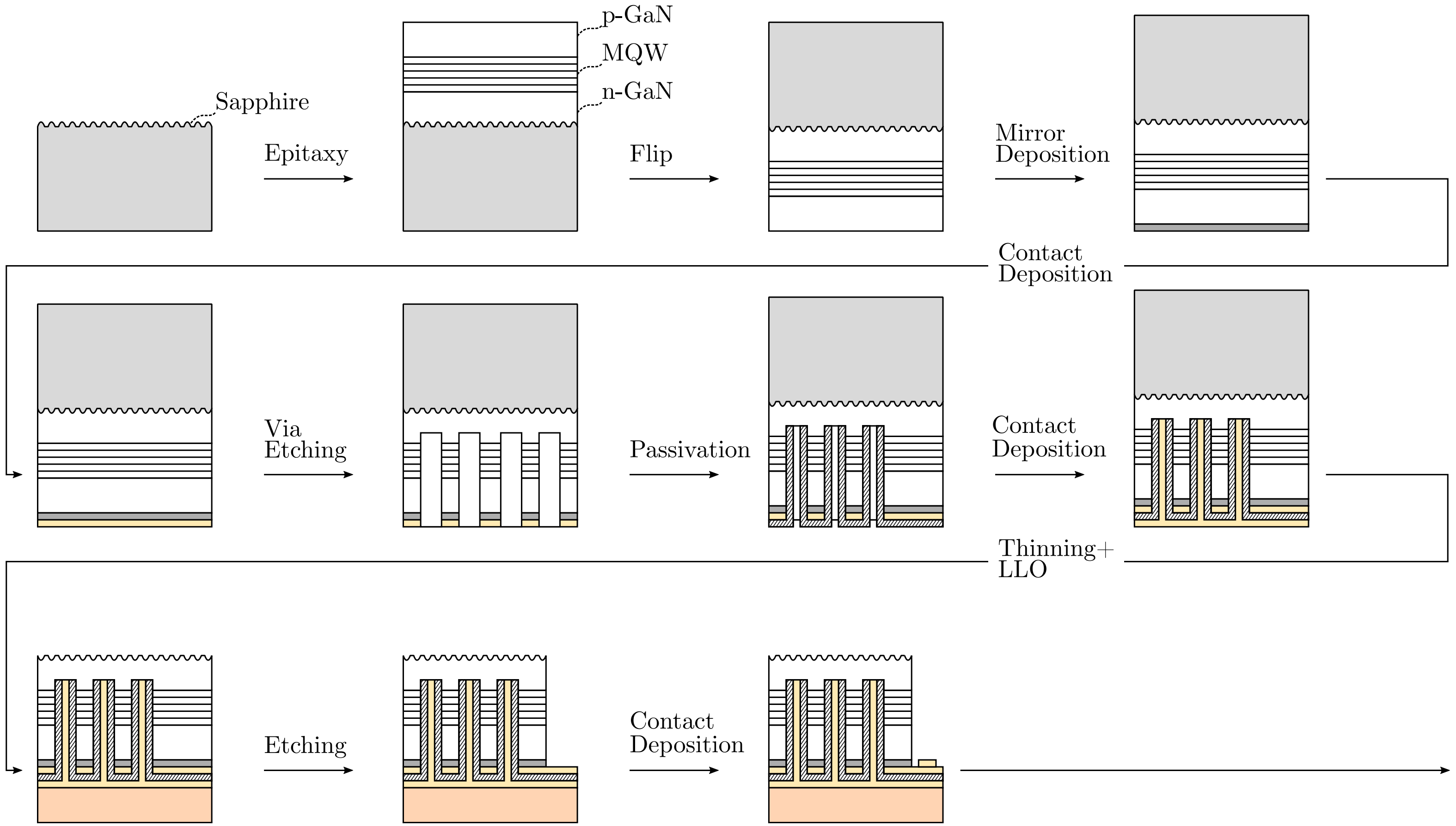

Figure SI3: (1/2) Manufacturing process for a vertical thin-film package flip-chip LED chip with vertical current spreading, circa 2012. Abbreviations: MQW - multiple quantum well; LLO - laser lift-off. Continued on next page.

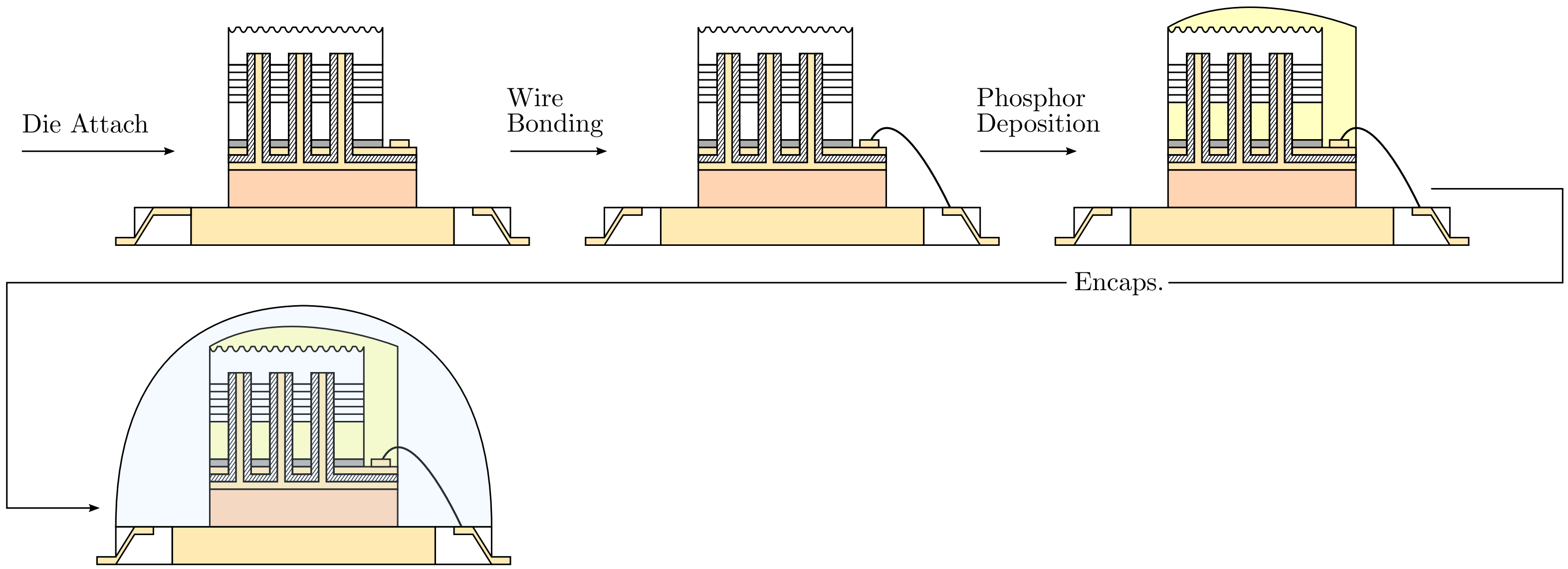

Figure SI4: (2/2) Continued from previous page. Abbreviations: Encaps. - encapsulation.

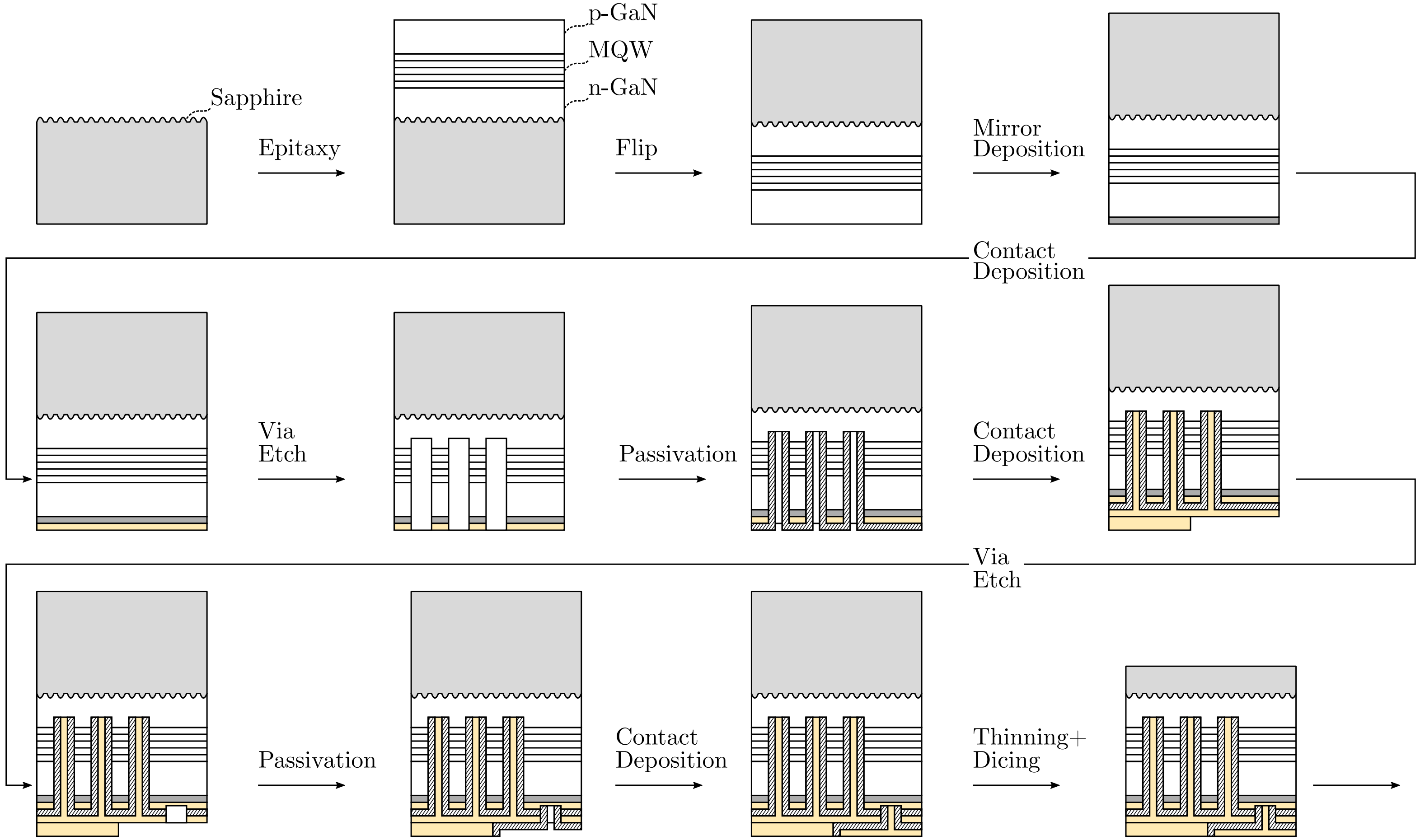

Figure SI5: (1/2) Manufacturing process for a chip scale package flip-chip LED chip with vertical current spreading, circa 2020. Abbreviations: MQW - multiple quantum well. Continued on next page.

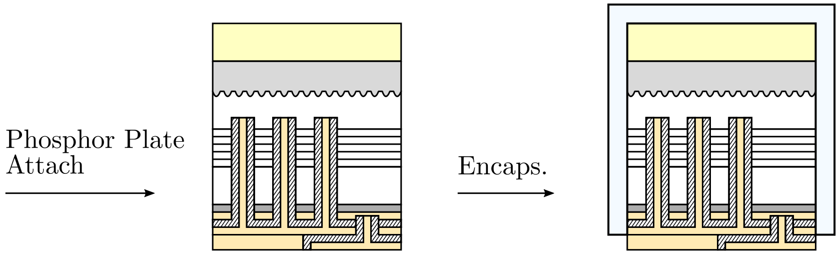

Figure SI6: (2/2) Manufacturing process for a chip scale package flip-chip LED chip with vertical current spreading, circa 2020. Continued from previous page. Abbreviations: Encaps. - encapsulation.

2.3 Computation of Manufacturing Cost

The manufacturing process of semiconductor devices can be categorized by the level at which manufacturing process steps are implemented, i.e., either at the wafer level or at the individual chip/package level. The total manufacturing cost per die is thus the sum of the total costs of all wafer processing steps and all die packaging steps.

$$C\left[\frac{\text{USD(2020)}}{\text{die}}\right] = P_S + C_w + C_p \tag{SI3}$$

where

$$\begin{aligned} P_S &\ldots \text{sapphire substrate price per die} \\ C_w &\ldots \text{wafer processing cost per die} \\ C_p &\ldots \text{die processing cost} \end{aligned}$$

The total wafer processing cost and total die packaging costs are in turn the sum of all associated process steps.

$$C_w = \sum_i C_i \tag{SI4}$$

$$C_p = \sum_j C_j \tag{SI5}$$

The cost of a single process step C_i can now be written as

$$C_i\left[\frac{\text{USD(2020)}}{\text{die}}\right] = \frac{1}{DPW}\frac{1}{y_i}\left\{\left((c_e * p) + c_l + c_m + c_d + c_o\right)_i \left(\frac{t_i}{w_i u_i}\right) + \sum_x v_x c_x\right\} \tag{SI6}$$

where the index i runs over all wafer processing steps, the index j runs over all die processing steps and the index x run over all materials.

$$\begin{aligned} DPW &\ldots \text{number of functional (i.e., successfully tested) die per wafer} \\ y &\ldots \text{process step yield} \\ u &\ldots \text{equipment utilization (relative to theoretical equipment capacity)} \\ p &\ldots \text{power consumption} \\ c_e &\ldots \text{hourly electricity cost} \\ c_m &\ldots \text{hourly maintenance cost} \\ c_d &\ldots \text{hourly depreciation cost} \\ c_l &\ldots \text{hourly labour cost} \\ c_o &\ldots \text{hourly overhead cost} \\ t_i &\ldots \text{time per run} \\ w &\ldots \text{wafers per run} \\ A &\ldots \text{wafer area} \\ v_x &\ldots \text{volume of material } x \text{ per wafer} \\ c_x &\ldots \text{cost of material } x \text{ per volume} \end{aligned}$$

The number of die per wafer D depends on the total usable wafer area. The usable area depends on the wafer diameter, the cutting street width between the chips and the exclusion zone at the rim of the wafer.

$$A_{\text{usable}} = A_{\text{wafer}} - A_{\text{cut}} - A_{\text{exclusion}} \tag{SI7}$$

Determining the usable wafer area as a function of these three parameters requires a numerical solution. However, following discussions in the literature [3], we approximate the number of functional die per wafer[1] as

$$DPW = \frac{\pi}{4}\left(\frac{d-2e}{\sqrt{a}+s/2}\right)^2 - \frac{\pi}{\sqrt{2}}\frac{d-2e}{(\sqrt{a}+s/2)^2} \tag{SI8}$$

where

$d \ldots$ wafer diameter
$e \ldots$ wafer edge exclusion zone width
$a \ldots$ die area
$s \ldots$ cutting street width

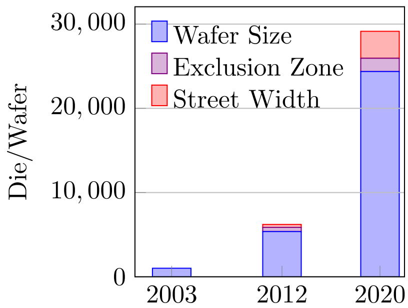

Figure SI7: Number of die per wafer for the different wafer sizes used in different years: (DPW) $d(2002) = 50\text{mm} \rightarrow 851\text{DPW}$, $d(2002) = 50\text{mm} \rightarrow 5370\text{DPW}$, $d(2020) = 100\text{mm}, \rightarrow 26,838\text{DPW}$. Source: own estimates, based on Eq. (SI8) and the dominant wafer size of the considered year, informed by expert interviews. For detailed statistics on changes in wafer size, compare also Fig. SI8.

Results are plotted in Fig. SI7, showing the increase in die per wafer (DPW) over time. Eq. (SI8) then gives us for the cost of a manufacturing step C_i in the wafer processing category

$$\begin{aligned} C_i\left[\frac{\text{USD(2020)}}{\text{die}}\right] = &\left(\frac{\pi}{4}\left(\frac{d-2e}{\sqrt{a}+s/2}\right)^2 - \frac{\pi}{\sqrt{2}}\frac{d-2e}{(\sqrt{a}+s/2)^2}\right)^{-1} \times \\ &\frac{1}{y_i}\left\{\Big((c_e * p) + c_l + c_m + c_d + c_o\Big)_i \left(\frac{t_i}{w_i u_i}\right) + \sum_x v_x c_x\right\} \end{aligned} \tag{SI9}$$

and the cost of a manufacturing step C_j in the packaging category

$$C_j\left[\frac{\text{USD(2020)}}{\text{die}}\right] = \frac{1}{y_j}\left\{\Big((c_e * p) + c_l + c_m + c_d + c_o\Big)_i \frac{c_j}{u_j} + \sum_x a v_x c_x\right\} \tag{SI10}$$

where c_j is throughput^{-1}. The total cost is thus

$$\begin{aligned} C = P_s + &\sum_i \left\{\frac{1}{DPW}\frac{1}{y_i}\left[\frac{t_i}{w_i u_i}\Big((e*p) + l + m + d + o\Big)_i + \sum_x v_x p_x\right]\right\} + \\ + &\sum_j \left\{\frac{1}{y_j}\left[\frac{c_j}{u_j}\Big((e*p) + l + m + d + o\Big)_i + \sum_x a v_x p_x\right]\right\} \end{aligned} \tag{SI11}$$

[1]often *"good die per wafer"* in the literature

Note that in keeping with the categorization introduced by the United States Department of Energy (cf. eg. [4]), certain steps from these two categories are reported separately. In the wafer processing category, the epitaxy step is reported separately due to its complexity and the large share of cost carried. In the wafer processing category, the phosphor step is reported separately.

2.4 Computation of Yielded Cost

Devices may be damaged or otherwise rendered unusable during the manufacturing process. The ratio between the number of good devices per step and the number of handled devices per step is known as the yield. Optimizing this yield is critical for reducing manufacturing cost [5]. This is because cumulative yield quickly drops as the yield from manufacturing steps with below 100% yield is multiplied. We must thus consider not only the manufacturing cost per process step, but also the cost including the yield [6][7]. While there are different mathematical approaches to including yield, we follow the definition in [6]. We write for the yielded cost C_{Y_i} of a step i with associated cost (before considering yield) C_1 and yield Y_1:

$$C_{Y_1} = \frac{C_1}{Y_1} \tag{SI12}$$

$$C_{Y_2} = \frac{C_1 + C_2}{Y_1 Y_2} - C_{Y_1} = \frac{C_1 + C_2}{Y_1 Y_2} - \frac{C_1}{Y_1} = \frac{1}{Y_1 Y_2}\Big(C_1(1 - Y_2) + C_2\Big) \tag{SI13}$$

$$C_{Y_i} = \frac{\sum_{x \le i} C_x}{\prod_{x \le i} Y_x} - \frac{\sum_{x < i} C_x}{\prod_{x < i} Y_x} \tag{SI14}$$

If a step is applied more than once, we can conveniently rewrite this in a form suited to computation within the *Excel* worksheet. Assuming step 2 is used twice, we get for the yielded cost of this step an equation of the form

$$C_{Y_2}^{(2\times)} = \left(\frac{C_1 + C_2}{Y_1 Y_2} - \frac{C_1}{Y_1}\right) + \left(\frac{C_1 + 2C_2}{Y_1 Y_2^2} - \frac{C_1 + C_2}{Y_1 Y_2}\right) \tag{SI15}$$

$$= \frac{1}{Y_1 Y_2^2}\Big(C_1(1 - Y_2^2) + 2C_2\Big) \tag{SI16}$$

which can be compared to Eq. (SI12) to find a more general form:

$$C_{Y_2}^{(n\times)} = \frac{1}{Y_1 Y_2^n}\Big(C_1(1 - Y_2^n) + nC_2\Big) \tag{SI17}$$

It can be shown by induction that the general form of a term for a step $i > 1$ repeated n times can be expressed as:

$$C_{Y_{i>1}}^{(n\times)} = \frac{1}{Y_i^{n-1} \prod_{x \le i} Y_x}\left(nC_i + \sum_{x<i} C_x(1 - Y_i^n)\right) \tag{SI18}$$

This cumulative approach to yielded cost is different from the approach taken in the original *LEDCOM* model. It uses what in the literature is described as an *"itemized approach"* to yielded cost [6]. In this approach, the yielded cost of a single process step f is described as

$$f_{\text{single}} = \frac{i + s}{y} \tag{SI19}$$

where

$$i \ldots \text{material cost of previous step}$$
$$s \ldots \text{step cost}$$

For process steps that are performed more than once, a series expression is used

$$f_{\times 2} = \frac{\frac{i+s}{y} + s}{y} \tag{SI20}$$

$$f_{\times n} = \frac{i + s(1 + y + y^2 + \cdots + y^{n-1})}{y^n} \tag{SI21}$$

This itemized approach serves as a convenient approximation, but its cumulative contributions do not equal the total yielded cost of the entire manufacturing process

$$f_{\text{total}} = \frac{\sum_i^n s_i}{\prod_i^n y_i} \neq \sum_i^n f_i \tag{SI22}$$

where n is the total number of steps. This is due to the approximation introduced through the series approximation in Eq. (SI21).

2.5 Example of Input Data Considered: Sapphire Wafers

Sapphire wafers form the substrate on which all other layers of the light-emitting diode are grown. Being transparent to radiation in the visible spectrum, it is not removed after growth in the Classical architecture or the Thin-Film Flip-Chip architecture. In the Vertical Thin-Film architecture, it is removed by means of a laser-lift-off process. Wafers can be either unpatterned or patterned, where the latter has become commonplace by 2020 due to the beneficial properties that microstructures on the surface have on layer growth [8] and light-extraction efficiency [9]. The price of sapphire substrates has decreased significantly since the year 2000, as shown in the bottom panel of Fig. SI8. This can be attributed not only to the lighting industry, but more importantly to increased supply as a result of increased demand from electronics manufacturing [10], where sapphire glass is used to protect screens and sensor interfaces from scratches [11]. Wafer sizes used in manufacturing have also increased, due to the favourable economics of large wafer processing. The market has been dominated by U.S.-based *Rubicon Technology* and Russian-based *Monocrystal*. he evolution of sapphire wafer prices and diameters used in LED manufacturing over the years is shown in top panel of Fig. SI8.

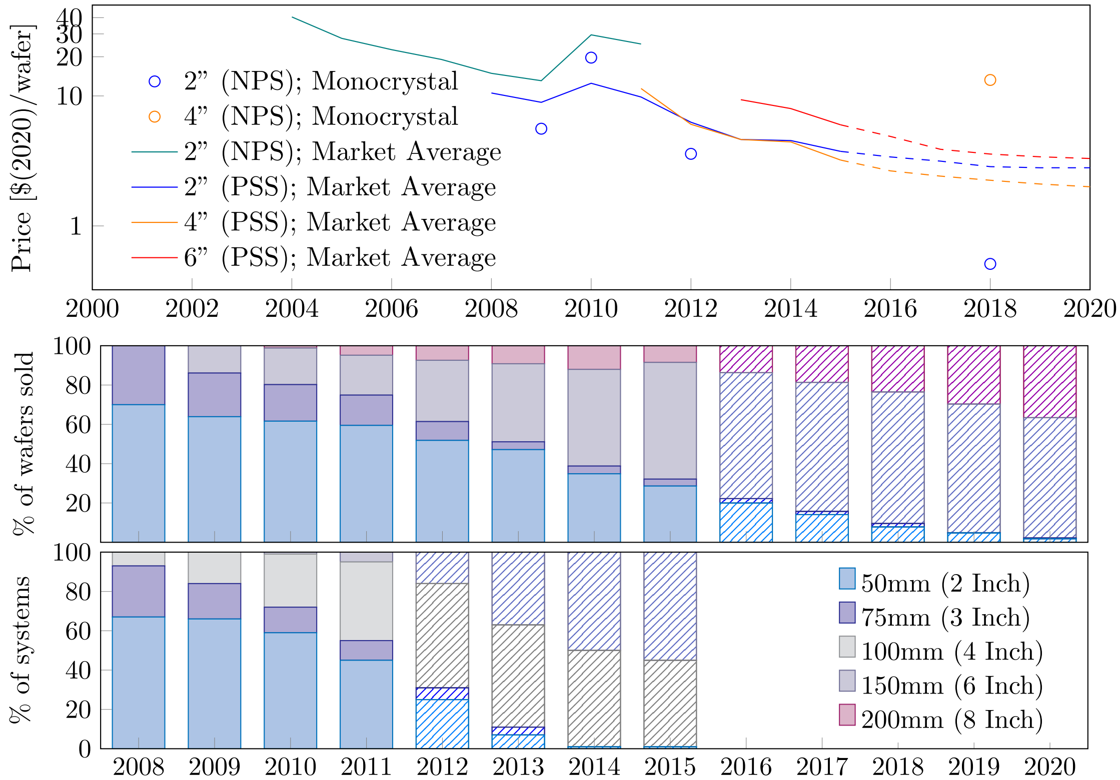

Figure SI8: Top: Historical data for sapphire substrate prices of different surface properties and diameters. Shown is data for polished surface (NPS) and patterned substrates (PSS). *Monocrystal* denotes the Russian manufacturer of the same name. Dashed lines are projections from the previous year. Sources: [12][13][10]. Bottom: Prevalence of sapphire wafer size used in the manufacturing of light-emitting diodes. Hatched bars are projections given by the sources. Sources: [14][15][10]

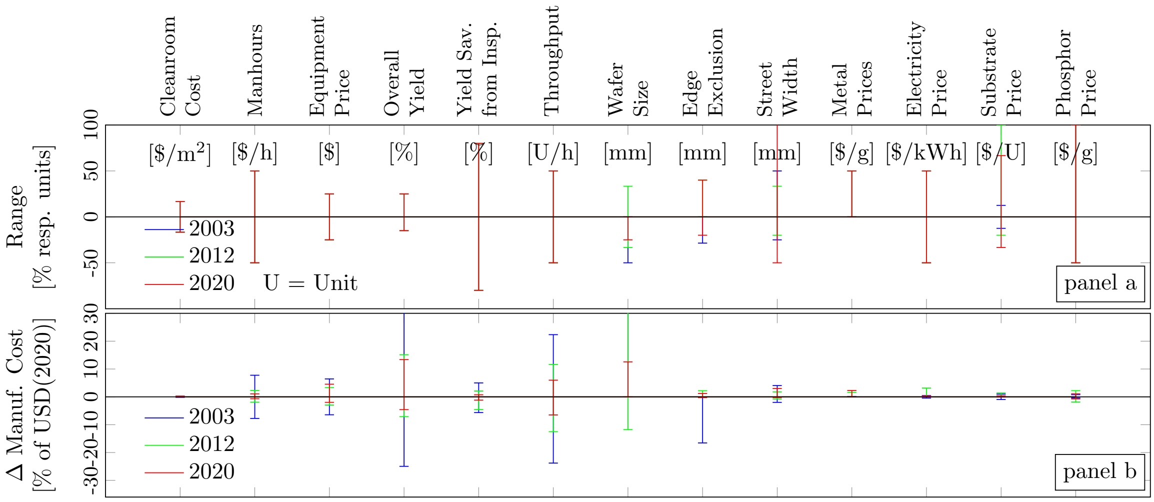

Figure SI9: Sensitivity analysis for selected cost model parameters. Panel a represents the tested range of variation of parameters indicated on horizontal axis along with their respective measurement units, as represented in Table SI1. Panel b shows the resulting range of variation in the manufacturing cost. Ranges are represented for three years considered in the cost model, where corresponding data is available, by whiskers of the following colour: 2003 – blue, 2012 – green, 2020 – red. Note the overall trend of decreasing sensitivity to cost model parameters as the number of die per wafer increases over time. Abbreviations: resp. - respective; yield sav. from insp. - yield savings from inspection.

2.6 Preliminary Sensitivity Analysis

A sensitivity analysis of the cost model has been performed using parameter variations listed in table Table SI1. The amount of parameter variation was chosen to encompass the identified range of parameter values in different manufacturing setups for each considered year. As an example, the lower range in the variation for the price of metals used in the model (+50%,-0%) was chosen because the prices quotes from the United State Geological Survey price database. Industry metals are not sold below the price of the raw material and often the markup is small compared to the price of the material. The upper range was chosen because a survey of industrial metal suppliers for semiconductor manufacturing showed that the largest markup was below 50%. The results of the analysis are shown in Fig. SI9. The cost model is generally more sensitive to the variation in parameters at smaller wafer diameters. The most sensitive parameters are global parameters, such as yield or average equipment throughput.

Table SI1: Cost model sensitivity analysis parameter list. The results of the sensitivity analysis for the parameters in this table are presented in Fig. SI9.

Parameter	*Unit*	*2003*	±[%]	*2012*	±[%]	*2020*	±[%]	Source
Cleanroom Cost	USD/m^2	3000	+16,-16	3000	+16,-16	3000	+16,-16	[16][17] [18][19]
Person-hour	FTE	100%	+50,-50	100%	+50,-50	100%	+50,-50	I
Equip. Discount	% of USD	0%	+25,-25	0%	+25,-25	0%	+25,-25	I [20]
Overall Yield	%	100%	+25,-25	100%	+25,-15	100%	+25,-25	I, [21][22] [23][17]
Inspec. Yield Savings	%/inspec.	0.5%	+80,-80	0.5%	+80,-80	0.5%	+80,-80	[24]
Overall Throughput	UPH or h^{-1}	100%	+50,-50	100%	+50,-50	100%	+50,-50	Datasheets
Wafer Diameter	mm	100	+0,-50	150	+33.3,-33.3	200	+0,-25	I and Fig. SI8
Edge Exclusion	mm	7	+0,-50	5	+40,-0	5	+40,-20	[25][26] [27][28]
Cutting Width	μm	100	+50,-25	75	+33.3,-20	20	+300,-50	[29][30] [31][32]
Metal Prices	USD/kg	100%	+50,-0	100%	+50,-0	100%	+50,-0	Datasheets
Electricity Price	USD/kWh	100%	+50,-50	100%	+50,-50	100%	+50,-50	[33][34]
Saph. Subst. Price	USD	40	+12.5,-12.5	10	+100,-20	3	+66.6,-33.3	Fig. SI8
Phosphor Prices	USD/g	100%	+50,-50	150	+0,-0	200	+0,-0	I, [35][36]

Note: The units for the values in columns *2003-2020* are indicated in the column *Units.* If values in columns *2003-2020* are instead given in %, this indicates that the parameters were varied by a set percentage from their respective model baselines. Abbreviations: FTE - full-time equivalent; UPH - units per hour; Equip. Discount - equipment discount (sales rebate for large purchases); Inspec. Yield Savings - yield savings from inspection (early detection and alleviation of issues in the manufacturing workflow).

2.7 Comparison with Reported Industry Data and DOE Projections

We conducted a comparison of the LED manufacturing cost structure produced by our cost model with previously reported US DOE calculations and projections based on the LEDCOM model and industry data provided as part of industry round table discussions (see Fig. SI10). We note some differences between the results of our model and the cost structure reported or projected by the DOE. For instance, the share of the epitaxy step is consistently larger in the DOE data. This can in part be explained by our model relying on state-of-the-art equipment at a virtual US-based manufacturing location, while industry might not run low-power and mid-power chip production on these, more expensive, reactors. In addition, the manufacturing lines of the majority of manufacturers is located in Asia. We also note that the share of the substrate price in the DOE data is much larger than in our model, which can in part be explained by the overestimation of the actual price of sapphire wafers in earlier projections for 2015-2020. Finally, the relative importance of the packaging part of the manufacturing process is very similar in our model and in the DOE results in 2012. However, in the DOE projections it significantly decreases by 2020, while in our model it retains and even increases its share of the total cost. This trend has been independently confirmed by researchers and industry reports on wafer-level packaging [37][38][39], showing better performance of our cost model compared to earlier DOE model projections

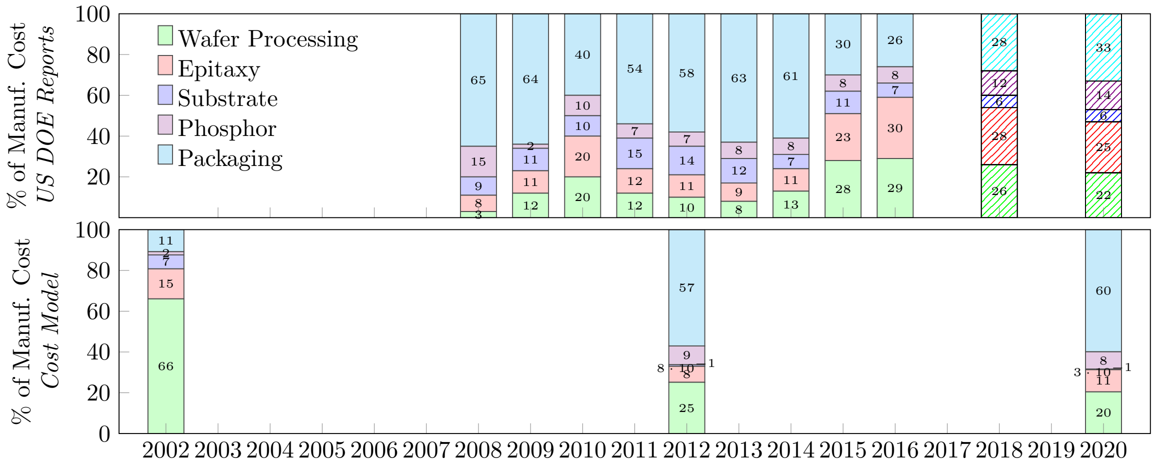

Figure SI10: Comparison of LED manufacturing cost structure between our manufacturing cost model (bottom panel) and previously published US DOE manufacturing cost calculations and projections based on industry data and LEDCOM model (top panel). Hatched bars are projections given by US DOE for 2018 and 2020. Sources for top panel: [40][41][42][43][44][45][46]. Sources for bottom panel: own elaboration based on the cost model described in Section Section 2.

3 Computation of Contribution of Individual Variables

To quantify the drivers of changes in manufacturing cost or device performance among many contributing factors, one would need to identify the magnitude of contribution to these changes made by single variables in equations Eq. (SI6) and Eq. (SI10). Mathematically, given a function C which describes the manufacturing cost or performance function of a device at time t,

$$F = ab + cd \tag{SI23}$$

and input variables a, b, c, d, we are looking for the contribution ΔF_a made by a single variable a to the total change in the function value ΔF between points t_0, t_1 such, that

$$\Delta F = F(t_1) - C(t_0) = \sum_{i=a,\dots,d} \Delta F_i \tag{SI24}$$

The infinitesimal contribution to the total funcation value by the infinitesimal change in an input variable is defined through the total differential of the function as

$$\mathrm{d}F(x_1(t), x_2(t), \dots) = \sum_i \frac{\partial F}{\partial x_i}\frac{\mathrm{d}x_i}{\mathrm{d}t} = \sum_i \frac{\partial F}{\partial x_i}\Delta x_i \tag{SI25}$$

where x_i is an input cost variable. The contribution of the change Δx_1 in variable x_1 to the total function value F over the period $t_0 < t < t_1$ is then

$$\Delta F_{x_1} = \int_{t=t_1}^{t_2} \frac{\partial F}{\partial x_1}\frac{\mathrm{d}x_1}{\mathrm{d}t}\mathrm{d}t \tag{SI26}$$

However, data on the input variables is not available in continuous time. Disaggregating the contribution of single variables to the change in the cost or performance is thus not straightforward in our model. This problem does not arise in cost models which compute cost changes directly, such as [47] [48]. In our work, we propose to address this problem by following an approach developed by Kavlak et al. [49]. For the detailed derivation, we refer to this publication.

The function F as a function of a vector of input model variables $\vec{r} = (r_1, r_2, \dots)$ is defined as

$$F(\vec{r}) = F(r_1, r_2, \dots) = \sum_i F_i \tag{SI27}$$

where

$$F_i(\vec{r}) = F_i^0 \prod_w g_{iw}(r_w) \tag{SI28}$$

Using logarithmic differentiation, the integral from Eq. (SI26) can be rewritten as

$$\Delta F_x = \int_{t=t_0}^{t_1} F(t)\frac{\partial \ln F}{\partial x}\frac{\mathrm{d}x}{\mathrm{d}t}\mathrm{d}t \tag{SI29}$$

where for $F(t)$ a constant $F(t) \approx \tilde{F}$ can be chosen such that $\Delta F_{x_i} = \Delta F$. In practice, this constant value can be approximated through the geometric and arithmetic means $\tilde{F} \approx \frac{2}{3}F_i^{\mathrm{geo}} + \frac{1}{3}\overline{F_i}$. The contribution of a single cost model variable r_z can then be written as

$$\Delta F_z(t_1, t_2) \approx \sum_i \tilde{F}_i \ln \frac{g_{iz}(t_2)}{g_{iz}(t_1)} \tag{SI30}$$

We use this approach to estimate the effect of individual innovations and spillovers on LED device performance, as discussed in section 4.3 and shown in Figure 5 in the main article. Due to time constraints, we leave the quantification of the impact of different drivers of cost reductions with this methodological approach for our future work.

4 Non-Phosphor-Related LED Innovations

Table SI2 summarizes LED innovations and technology and manufacturing process improvements identified in this study as affecting key white LED sub-efficiencies, as discussed in section 5.1 and shown in Figures 4-6 in the main article. Note that phosphor-related LED innovations affecting consumer experience metrics, along with spectral efficiency, are presented separately in Table 1 in the main publication and discussed in detail in Section 5.

Table SI2: LED innovations and technology improvements affecting key white LED device sub-efficiencies. The table includes innovations and technology improvements affecting forward voltage efficiency, light extraction efficiency, internal quantum efficiency, and light conversion efficiency. For a list of innovations affecting spectral efficiency, see Table 1 in the main article.

Forward Voltage Efficiency (VFE) η_{Vf}

Year	*Innovation*	*Area of Improvement*	*Spillover*	*Source*
1999	Indium Tin Oxide Current spreading layer	Contact resistance	Yes	[50]
1998	Digitated electrodes	Contact resistance	No	[51]
Ongoing	Epitaxy improvements and better doping	Polarization and bulk resistance	No	I
1998	Silver p-Contacts	Contact resistance	No	[52]

Light extraction efficiency (LEE) η_{LE}

Year	*Innovation*	*Area of improvement*	*Spillover*	*Source*
< 2003	Optimization for cavity effects	Reduces self-interference of quantum well	No	I, [53]
1993	Chip surface randomization	Total reflection and absorption	No	I, [54][55]
1993	Thin-film chip architecture	Absorption	No	I, [55]
1996	Patterned sapphire substrate	Total reflection and absorption	Yes	I, [56]
Ongoing	Chip design for high LEE	Total reflection and absorption	No	I, [57]
$\sim$2000	Silver p-contacts	Absorption	No	I, [52]

Internal quantum efficiency (IQE) η_{IQ}

Year	*Innovation*	*Area of improvement*	*Spillover*	*Source*
1994	Double heterostructure	Higher Radiative recombination probability	No	[58]
1996	Multiple quantum well	Higher Radiative recombination probability	No	[59]
Ongoing	Active region Doping	Radiative recombination probability	No	I, [60]
Ongoing	Epitaxy Improvements	Radiative recombination probability	No	I
Ongoing	Chip architecture Improvements	Radiative recombination probability	No	I

Light Conversion efficiency (CE) η_C

Year	*Innovation*	*Area of improvement*	*Spillover*	*Source*
Ongoing	Lower current density	Current density	No	I
Ongoing	Epitaxy Improvements	Charge distribution Defect density	No	I, [61]
Ongoing	Chip architecture improvements	Charge distribution Defect density	No	I, [62]
< 2017	Defect getting underlayer	Defect Density	No	I, [63]

Note: *Year* column indicates the first instance of application of corresponding invention in white LEDs. 'Ongoing' indicates improvements that are incremental in nature and have been ongoing since the earliest days of LED manufacturing, with no individual breakthroughs identified. *Spillover* column indicates if the innovation involved technology spillovers. *Source* column indicates the source of information about the innovation or improvement, with 'I' indicating expert interviews as such a source.

5 Phosphor-Related LED Innovations

This section provides additional details for phosphor-related LED innovations identified in this study as affecting both spectral sub-efficiencies and consumer experience metrics and discussed in section 5.2, Table 1 and Figures 5-7 in the main article.

5.1 History of Identified Phosphor-Related LED Innovations

5.1.1 YAG and YGAG Phosphors

Prior to Nakamura's invention of highly efficient blue LEDs in the 1990s, Japan's Nichia Corporation had not sold commercially successful semiconductor products, instead specializing in phosphors for cathode ray tubes (CRT) and fluorescent lamps [64] . Nevertheless, extensive firm expertise in this area helped Nichia's Yoshinori Shimizu formulate the principles of using CRT phosphors to convert blue light from Nakamura's LEDs into white light in 1994 [65][66]. By 1996, Shimizu and his colleagues developed [67][68] the first practical LED application of a well-known Yttrium Aluminium Garnet (YAG) CRT phosphor activated with cerium [69], enabling the first commercial white LED products manufactured and sold by Nichia since late 1996 [70][66].

Importantly, the YAG phosphor does not exhibit the spectral properties desirable in general illumination applications (see Figure 7 in the main article). An early solution to this problem, which was first discovered in the late 1967 [71] and suggested for LEDs by the same team at Nichia in 1996 [70][68], was to use the gadolinium-doped red-shifted YAG phosphor (YGAG). Used in combination with red-emitting sulfide phosphors, by 2002 it helped bring to the market the first generation of warm white light LED products, e.g., those produced by Lumileds [72]. However, sulfide phosphors led to accelerated deterioration of sensitive parts of LED devices and became less efficient as operating temperatures increased. New chemically stable and non-toxic red phosphors were needed.

5.1.2 258, SLA and SALON phosphors

In 1997, Hubert Huppertz and Wolfgang Schnick, working at the University of Bayreuth in Germany, synthesized the first compound in a new class of rare earth nitridosilicate materials [73] later dubbed "258" due to a proportion of elements in its chemical formula. The luminescent properties of these materials were identified by the Schnick's group, by then at Ludwig-Maximilians University of Munich, in 2000 [74] after a suggestion made to Schnick at a conference following earlier reports of good luminosity properties of europium-doped compounds [75]. U.S.-based LED manufacturer Lumileds applied for a patent for the first class of red LED phosphors based on the 258 nitridosilicate chemistry in 2002 [76]. The first use of the 258 phosphor in a commercial "Luxeon" LED package was then reported in a joint publication co-authored by inventors from Lumileds and researchers from the Schnick's group in 2005 [77].

Further efforts in LED phosphor development were directed towards synthesizing a red narrow-band phosphor. Narrow LED emission peak widths yield the highest luminous efficacy of radiation, as in this case less light is emitted in the far-red range of the spectrum in which the human eye is not very sensitive. After synthesizing several narrow-band phosphors emitting in yellow [78] and cyan [79], the Schnick's group identified the local cubic cation coordination structure of the cyan phosphor compound as the reason for its narrow band width [80]. A search for a structurally analogous nitride compound with the narrow red instead of the cyan emission was undertaken. After several unsuccessful attempts, the sought-after cuboidal nitride compound was found in a 2008 publication led by Francis DiSalvo [81]. Based on information provided in this work, Schnick and colleagues synthesized and studied the spectral properties of a new narrow band red SLA phosphor in 2013 [82][83][84]. The material was introduced in commercial LED devices by Lumileds in 2015 [85].

The most recent red narrow-band phosphor innovation included in Table 1 and Figure 7 in the main article, indicated as SALON, has been under development during the late 2010s by a group of Austrian and German researchers that included Huppertz, the discoverer of the "258" material, working in collaboration with Osram, another major LED manufacturer [86][87][88]. The first U.S. patent application for this phosphor was filed in 2017 [86]. The SALON phosphor is a derivate of the SLA phosphor. Therefore, it is the only innovation related to consumer experience metrics identified in our study that seemingly not involved technology spillovers.

5.1.3 PFS Phosphor

Down conversion with ultra-narrow-band phosphor can achieve the highest spectral efficiency. However, few such phosphors have been identified, with even less exhibiting desirable material properties such as thermal stability [89]. The first commercially successful ultra-narrow-band red phosphor was developed by General Electric (GE). It is based on a potassium fluorosilicate (PFS) compound activated with manganese ions. Its luminescence was first recorded by Adrian Paulusz at GE in 1972 [90]. In the early 2000s, while searching for potential new LED phosphor materials for GE's lighting business at GE Lumination, Emil Radkov rediscovered Paulusz's findings in the literature. Following extensive research on PFS chemical synthesis and material properties conducted in collaboration with the University of Sofia in Bulgaria, Radkov's Alma Mater, the PFS phosphor had been under development at GE since 2005 [91][92]. This work, supported by public funding from the U.S. Department of Energy (DOE) Solid-State Lighting program [93], resulted in a series of critical improvements in the PFS phosphor properties [94][95], eventually enabling its commercialization under the "TriGain" brand in 2015 [96][97][98].

5.1.4 Quantum Dots for Light Down-Conversion

Quantum dots (QD) are semiconductor nanocrystals whose quantum size effects make QDs behave as "artificial atoms". Semiconductor quantum dots were first synthesized in the Soviet Union in 1981 [99] and at Bell Labs in the U.S. in 1983 [100]. Luminescent properties of quantum dots were first empirically observed in 1984 [101] and extensively studied in the early 1990s. The key feature of QD luminescence discovered in those studies is that its colour is determined by the QD particle size, making it possible to create pure monochromatic blue, green and red light sources just by tuning the QD size. The first application of QDs in LEDs was reported in 1994 in an electroluminescent hybrid QD-polymer LED. However, this LED type could not be used in general illumination due to its very low luminous efficacy. An alternative application of QDs as a kind of a "phosphor" for light down conversion from an LED light source was proposed in the early 2000s as part of the U.S. Department of Energy (DOE)-funded "A Revolution in Lighting" project at Sandia National Laboratory [102]. This concept was successfully demonstrated by Sandia researchers on a commercial LED in 2003 [103][104] and was swiftly taken up and advanced further by a group in Taiwan [105][106] The first commercial application of QDs in an LED lamp was brought about by a collaboration between an MIT-born startup QD Vision and the U.S.-based luminaire manufacturer Nexxus Lighting in 2009 [107], [108]. However, rapid advances in the spectral and conversion performance of down-conversion phosphors and high manufacturing cost of quantum dots resulted in the discontinuation of this product. After finding market success in display backlighting first demonstrated by Samsung in 2010 [109] and commercialized by QD Vision in Sony television sets in 2013 [108], QDs returned to the general lighting market in products offered by Lumileds [110][111] around 2017 and Osram in 2019 [112] in the form of mid-power LED packages that combined QDs with traditional phosphors for light down conversion.

5.2 Spectral Data for Identified LED Phosphor Innovations

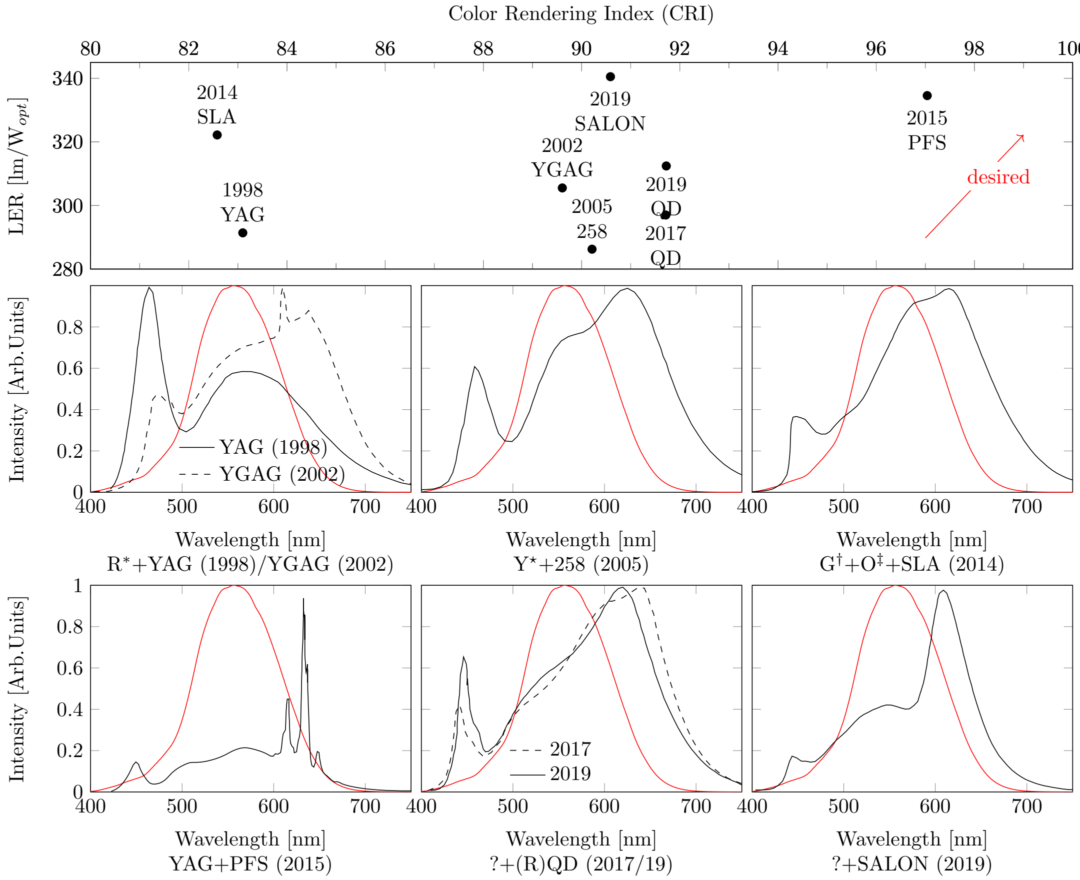

Figure SI11: Spectral data and additional consumer experience metrics for the earliest identified representative white LED products with published spectral data, shown in Figure 7 in the main article, that used phosphor innovations listed in Table 1 in the main article, each indicated by the phosphor label and spectral data publication year. Top panel: Luminous efficacy of radiation (LER) and colour rendering index (CRI) of white LED devices represented in Figure 7 in the main article. The desirable direction of improvements towards higher luminous efficacy at higher CRI is indicated by a red arrow. Metrics were calculated from spectral data shown in the two bottom panels using the `colour-science` package for Python [113]. Bottom two rows of panels: Corresponding spectral data. The luminosity function [114], describing the wavelength-dependent sensitivity of the human eye, is shown for reference in red in each panel. Note that peaks or large tails of the device emission spectrum at the far ends of the luminosity function are not desirable, as the photons of the corresponding energy are lost to the human eye and count towards the spectral loss channel. Plot legends indicate the years of publication of the spectral data and phosphor mixtures used in corresponding LED devices, with the following designations for additional phosphor mix components: ? - other parts of phosphor mixture not disclosed, G* - CaSrS:Eu^{2+}, Y* - β−SiAlON:Eu^{2+}, G† - $Lu_3Al_5O_{12}$:Ce^{3+}, O‡ - $(Ba,Sr)_2Si_5N_8$:Eu^{2+}. Source (top panel): own elaboration based on spectral data. Arb. Units - Arbitrary Units, defined as the ratio of radiation intensity at a wavelength compared to the highest point in the spectrum. Sources: top panel - own elaboration based on spectral data; bottom two panels: adapted from published spectral data for LEDs with the following phosphors: YAG [70], YGAG [72], 258 [77], SLA [83], PFS [96], QD [115][116], SALON [87].

6 List of Interviewees

Table SI3: Anonymized list of LED experts interviewed for this study. Abbreviations: sr. - senior

#	*Sector*	*Role*	*Country*	*Expertise*
1	Academia	Sr. researcher	UK	Epitaxy
2	Industry	Consultant, former sr. researcher	USA	Device architecture
3	Industry	Consultant, former head of R&D	Germany	Epitaxy
4	Academia	Professor	Austria	Phosphors
5	Industry	Consultant, former head of R&D	USA	Device architecture
6	consulting	Consultant, former sr. technical advisor	USA	Device architecture
7	Academia	Professor	Germany	Phosphors
8	government	R&D manager	USA	Device architecture
9	consulting	Consultant	USA	Device applications
10	Academia	Professor	France	Device physics
11	Industry	Sr scientist, former head of R&D	USA	Device architecture
12	Industry	Principal scientist	Germany	Phosphors
13	Industry	Former head of R&D	USA	Phosphors

7 Complete List of Sources for Figures

7.1 Figure 1 (Historical Development of Luminous Efficacy)

The list of sources for Figure 1 in the main article is organized by different technologies:

Technology	*References*
LED	own research, compare [117]
CFL (<1984)	[118][119]
CFL (1984-2011)	[120]
CFL (>2011)	[121]
Fire, Incandescent, HID	[122] augmented by own calculations based on [123]
Max. efficacy	[124]

7.2 Figure 2 (Historical Development of Lamp Prices)

The sources for Figure 2 in the main article are grouped by the provider of data:

Provider of References	*References*
Stiftung Warentest (Germany)	[125][126][127][128][129][130][131][132][133][134][135][136][137][138]
Konsument (Austria)	[139]
Which (UK)	[140]
Industry Periodical	[141]
Government Report	[142]

7.3 Figure 3 (Historical Evolution of LED Chip Architectures)

The sources for Figure 3 in the main article are grouped by their type:

Type of Source	*References*
Scientific Publications	[143][144][145][146]
Patents	[147][148][149][149][150][151][152]
Other Publications	[153][154][155]

7.4 Figure 4 (Historical Developments in Device Sub-Efficiencies)

The sources for Figure 4 in the main article are grouped by sub-efficiencies represented on different panels of the figure:

Panel (Sub-Eff.)	*References*
Panel A1 (V_f)	[156][157][158][159][160][161][162][163][164][165][166][167][168] [169][170][171][172][173][174][175][176][177][178]
Panel A2 (Droop)	Data calculated from luminous intensity curves of respective device datasheets: [179][164][180][181][182][163][169][170][175]
Panel B1 (IQE)	own research, compare [117] [183][184][185][186][187][188][189][190][191][192][193]
Panel B2 (LEE)	[194][195][196][197][198][199][200][201][202][203][204][205][206][207] [208][209][210] [194][211][212][213][214][215][216][217][218][219][211] [212][220][221][222][223][224][225][226][227]

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Journal Name

ARTICLE TYPE

Cite this: DOI: 00.0000/xxxxxxxxxx

Rapid technological progress in white light-emitting diodes and its sources in innovation and technology spillovers

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Received Date
Accepted Date

DOI: 00.0000/xxxxxxxxxx

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The first commercial white light-emitting diodes (LEDs) were introduced to the market in 1996. Since then, white LEDs have experienced major improvements in performance and efficiency. Significant decreases in cost have been driven by a rapid expansion of the market share of LED-based solid-state lighting (SSL), which in turn has driven down costs even further. Today, the technology is one of the key solutions in global climate change mitigation efforts. However, despite its significance, the extent and sources of the underlying improvements have not been systematically investigated. Understanding what has driven rapid progress in LEDs can provide lessons for accelerating innovation in a broad range of other demand-side clean energy technologies. With this aim, we gather and analyse systematic evidence on cost and performance improvements in white LEDs from cost and performance modelling, augmented by a literature review and interviews with experts in industry and academia. We find that the overall efficiency of the highest performing warm white LED packages has improved from $\eta_L = 5.8\%$ in 2003 to $\eta_L = 38.8\%$ in 2020. During the same period, we estimate that the cost of manufacturing low-power and mid-power LED packages at a U.S. location using state-of-the-art equipment has dropped from 1.1\$ to 0.05\$ (in 2020 USD) between 2003 and 2020, a 95.5% decrease. We further show that technology spillovers — i.e., knowledge originating in other technologies, sectors, or scientific disciplines — affected all performance dimensions of LEDs, contributing no less than 8.5% to total efficiency improvements and ∼100% to the improvements in consumer experience metrics identified, thereby playing a key role in the widespread adoption of the technology. The increase in wafer size and manufacturing yield improvements were the primary causes of LED manufacturing cost reductions.

1 Introduction

A rapid reduction of global carbon dioxide emissions is urgently required in order to mitigate the effects of climate change[1]. According to the United Nations, by the end of 2021, more than 130 countries have set or are considering setting a net zero emissions target by 2050[2]. The European Union, for example, has set a target of net zero emissions by 2050. It aims to meet this goal with the help of the European Green Deal, alongside other EU and national policies[3]. The United Kingdom similarly has adopted a national strategy to achieve net-zero by 2050[4].

Achieving these ambitious and critically important targets will require both the deployment of new clean energy technologies, and the acceleration of innovation in existing supply-side[5] and demand-side energy technologies[6]. To ensure rapid adoption of these technologies, significant reductions in their costs and improvements in their performance and consumer experience are needed. This requires understanding how these cost reductions and performance improvements can be achieved[7 8].

There are a range of mechanisms that contribute to improvements in a technology over time, including targeted research and development (R&D) efforts, economies of scale, learning by doing[9], and economies of scope[10 11 12]. The role of these factors at different stages of the innovation life cycle[13], from research and technology development to demonstration, market formation and diffusion, is an area of active research[14 15 8]. Innovation is also driven by forces of supply and demand. Various "*technology-push*" drivers reduce the costs of innovating, while "*market-pull*" drivers

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increase the pay-offs from investing in innovation[16].

The stages and drivers of the innovation life-cycle for a particular technology play out within a broader innovation system[13 19]. Among these drivers, the role of external knowledge transfer and technology spillovers in research and development of energy technologies is an understudied area[7]. While the exact definition of spillovers in the literature depends on the context and the research question[20 21], we follow Stephan et al.[7] and consider knowledge to be external - thus a spillover - if it has been developed for application in other technologies, sectors, or scientific disciplines. There is emerging evidence that understanding spillovers and the knowledge network outside a technology may be an important factor in understanding[22] and shaping[23 7 24 25] the future evolution of technologies.

Among demand-side technologies, the provision of lighting is a particularly important area for climate change mitigation efforts, as it currently accounts for 15-19% of global electricity consumption[26 27]. It is also an area of rapid recent technological change: since the introduction of the first commercial white light-emitting diodes (LEDs) in 1996, lighting technology has experienced dramatic efficiency improvements. As shown in Figure 1, thanks to the introduction of LED-based solid-state lighting (SSL), the efficacy of lighting sources has increased by three orders of magnitude in just over 20 years, which is significantly faster than the historical progress observed in previous lighting technologies[28]. For comparison, the highest performing light-emitting devices today reach efficacies of 220 lm/W[29], while an incandescent light bulb can only reach efficacies of up to 18 lm/W. Moreover, this rapid improvement in efficiency has been accompanied by a similarly impressive decrease in LED manufacturing costs and retail prices. Figure 2 shows how LED retail prices have fallen by two orders of magnitude, at an annual price per flux decline during the period of from 2008-2020 of 27.3%, in line with previous estimates[30].

These dramatic improvements in lighting technology, supported by the introduction of lighting efficiency regulations phasing out incandescent lightbulbs and targeted policies stimulating LED adoption in many countries, led to the rapid expansion and diffusion of SSL technologies[33 34 35 36]. As a result, by 2020, highly efficient LED luminaires were saving an estimated 131 TWh/year in the EU[37] and 442 TWh/year for the US[38], which is on par with the amount of energy produced annually by all solar photovoltaic installations in these regions. Notably, market adoption of LED lighting is not limited to developed economies[39]. For example, durability, low up-front cost and high efficiency of LED light sources have led to their widespread adoption in rural West African communities without access to grid electricity[40]. LEDs have also been used in a wide range of applications beyond lighting, such as personal health monitors[41 42], watches and smartphones[43], potable water treatment[44], high-bandwidth wireless data transmission[45], and augmented reality eye wear[46].

Despite this impressive history, the sources of LED innovation have not received as much attention from researchers as innovation in supply-side energy technologies, such as solar photovoltaics[15] or wind energy[47 48], or in lithium-ion batteries for transportation[8 7]. To the best of our knowledge, no study has comprehensively discussed the sources or extent of progress across various metrics of LED cost or performance since the introduction of first commercial white LED products. Understanding the extent to which individual innovations and knowledge spillovers contributed to improvements in LED technology, how this effect compares to other sources of improvements such as economies of scale, and how these innovations occurred (i.e., by what mechanisms and actors) will provide valuable lessons both for other demand-side technologies and overall for accelerating clean energy innovation.

To address these questions, in this paper we identify a set of metrics suitable for tracking the historical progress of LED lighting technology. We then trace device efficiency and cost improvements, as well as changes in relevant consumer experience metrics from the time of introduction of the first commercial warm white LEDs in 2003 to 2020, the year with the most recent data available at the time of writing. Given the proprietary nature of knowledge in the SSL industry, we collect corresponding information using a multi-method approach combining a systematic literature review of scientific literature and industry reports with patent analysis and a series of elite interviews[49] with eminent experts from academia, public research institutions and industry. This approach allows us to identify innovations in white LED technology and examine their origins to discern technology spillovers among them. Data on performance improvements associated with individual innovations identified in patents, scientific publications, industry reports, and interviews is then augmented by our calculations of device efficiency, including contributions of specific spillovers to the overall LED efficiency (see Supplementary Information for more detail). We further calculate LED manufacturing costs using a bottom-up cost model with process-step resolution that we developed. Finally, we discuss what our observations may mean for our understanding of the innovation process in innovation process in LED technology and for broader policy and industry efforts to accelerate clean energy innovation.

2 Previous Literature

The historical development development of light-emitting diodes from a novelty semiconductor experiment into powerful lighting devices has received a considerable amount of attention following the recognition of pioneering work on blue LED by Japanese researchers Shuji Nakamura, Isamu Akasaki and Hiroshi Amano with the 2014 Nobel Prize in Physics[50 51]. The sources and disaggregated contributions of subsequent improvements in LED technology, however, have not been documented in the literature in a systematic fashion, though there were notable publications reporting on the progress in the design of devices[52 53 54 55], or the technological improvements underlying the improvement in overall device efficiency for best performers in 2009[56] and 2016[57].

Several descriptive publications provide a very useful overview of selected scientific breakthroughs that have contributed to LED progress[58 59 60 61 62 63]. Additional studies provide more detail regarding the origin and impact of specific advances in LED technology as well as their integration into device manufacturing were published by authors working for key industry actors Lumileds[64 52 65 66] and Osram (now amsOsram)[67 68 54 55]. How-

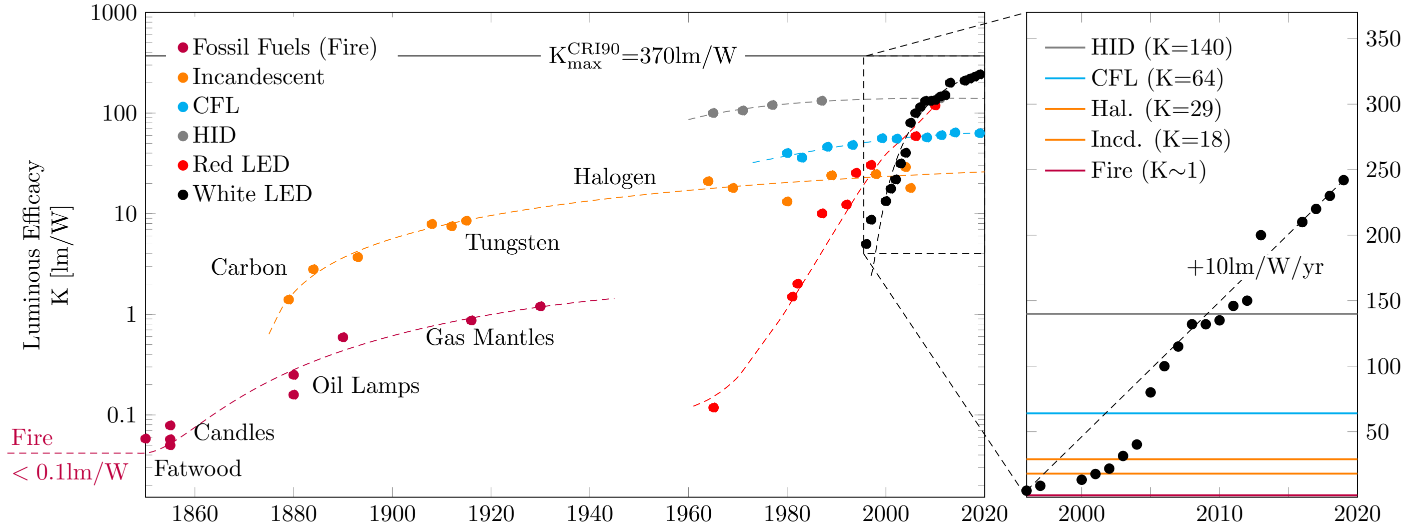

Fig. 1 Historical development of the luminous efficacy (K) of the most widely-used lighting technologies in human history. Data points indicate best performers by year of market introduction. Luminous efficacy is the measure of how efficiently a light source converts electrical energy into visible light that can be perceived by the human eye, taking into account the wavelength sensitivity of the eye. Dashed lines indicate an average improvement for each technology, computed from a 3rd-order polynomial fit to the data. The physical limit for an ideal light source with a colour rendering index of CRI=90, denoted as $K_{m}ax^{CRI90}$, is shown as a black horizontal line, as per calculations by Murphy et al.[17]. The magnified plot shows the progress in cool white LEDs from 1996 to 2020, with the dashed line indicating a linear rate of efficacy improvement of 10lm/W per year. For comparison, efficacies of best performers in legacy lighting technologies for 2020 are shown as coloured horizontal lines. Note the logarithmic scale of the vertical axis on the main plot and the linear scale on the magnified plot. Abbreviations: HID - High-Intensity Discharge; CFL - Compact Fluorescent Lamp; Hal. - Halogen, Incd. - Incandescent. Source: own synthesis of published data based on a visual approach proposed by Azevedo et al.[18]. See Supplementary Information, Section 5 (Complete List of Sources for Figures), for the full list of sources and references.

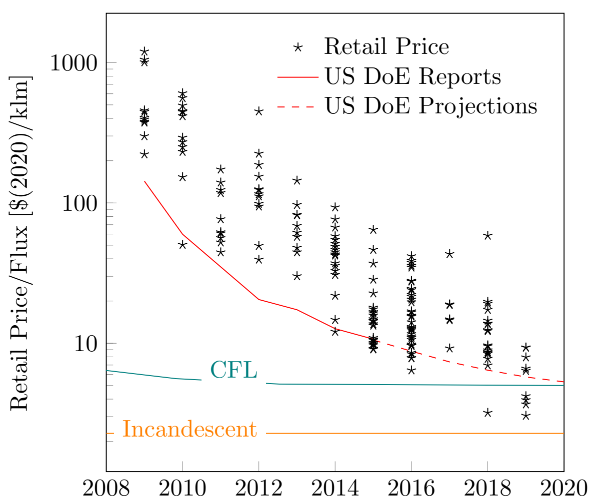

Fig. 2 Historical development of retail sales prices (in 2020 USD) per luminous flux of LED-based luminaires, including light bulbs, spotlights and recessed lights from 2008 to 2020. Red curved and dashed lines represent average retail sales prices and price projections for LED based luminaires published by the U.S. Department of Energy (DOE)[31]. Shown for reference are the average prices for compact fluorescent (CFL) and incandescent light bulbs. Source: own synthesis of data on LED sales prices collected from various consumer watchdog databases and publications. Data on CFL sales prices collected from[32] as well as various consumer watchdog databases and publications. Incandescent light bulb price is assumed constant based on the average in the covered time period. See Supplementary Information, Section 5 (Complete List of Sources for Figures), for the full list of sources and references.

ever, these studies typically include limited information on the macro-level chip design and cover disparate aspects of the technology over different time periods. Another important source of historical information are patents, which cover the entirety of the device architecture or manufacturing process, including those by Lumileds[69], Samsung[70 71] and Soraa[72].

We summarize the information collected, analysed and classified on the progress in LED chip architectures and manufacturing processes from these and other disparate sources in Figure 3, where we show the evolution of LEDs from classical chips with lateral current spreading to chip-scale package flip-chip architectures. Despite the amount of literature published on the topic of LED history, we are not aware of a prior publication that comprehensively and consistently aggregates and analyzes known chip design, manufacturing, and material improvements to show the overall effect of these improvements on device efficiency or manufacturing cost over time. In addition, the effect of individual innovations and technology spillovers, which has been investigated in solar PV[15 73 74], and to some extent in lithium-ion batteries[7], has not yet been studied in the context of lighting. This is consistent with previous observations regarding the marginalization of end-use technologies in the analysis of energy innovation for climate change impact mitigation[75 76].

3 Metrics for technological progress in LEDs

Investigating the sources of rapid progress in LED technology over the past decades, in which white LEDs have come to dominate the lighting market[77], requires selecting appropriate metrics for tracking and quantifying this progress. The choice of metrics af-

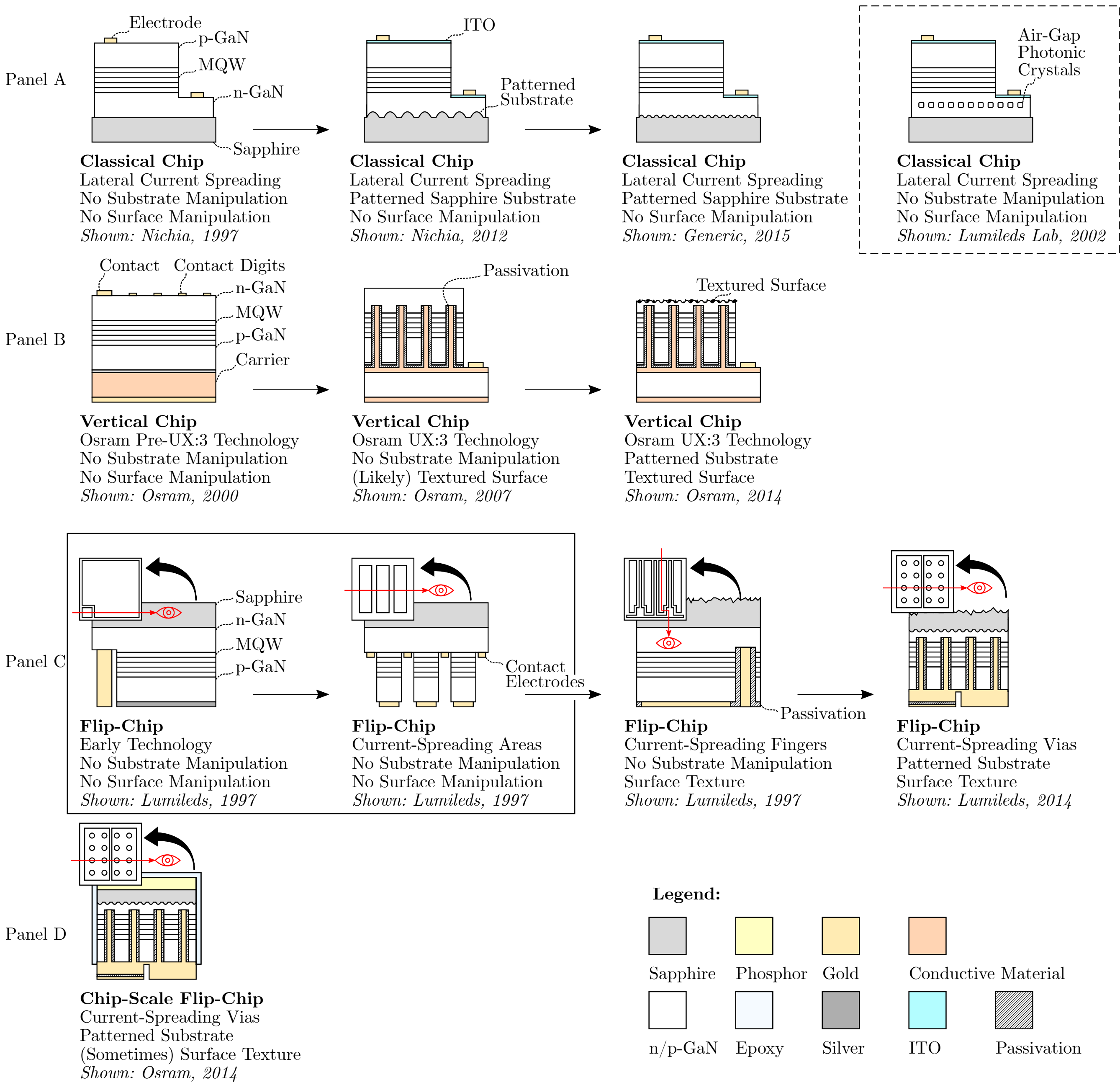

Fig. 3 Historical evolution of light-emitting diode chip architectures. Panel A: classical chip with lateral current spreading; Panel B: Osram's thin GaN flip-chip (vertical) architecture; Panel C: flip-chip architecture; Panel D: chip-scale package flip-chip architecture. Shown are side views of chips without packages, along a cutaway line best suited to the features of each architecture. The cutaway surface is indicated by a red arrow with an eye on the overlaid top view of each chip. Note that the dimensions are not to scale, and smaller features are greatly exaggerated for clarity. Years indicated correspond to the earliest identified patent priority date. Black frames around certain designs indicate chip designs not brought to large-scale production. Chip architecture abbreviations: TF - Thin-Film; FC - Flip-Chip; CSP - Chip-Scale Package; Material abbreviations: GaN - Gallium Nitride, ITO - Indium Tin Oxide, MQW - Multiple Quantum Well. Source: adapted and compiled from multiple patents and industry publications. See Supplementary Information, Section 5 (Complete List of Sources for Figures), for the full list of sources and references.

fects both what data sources can be used in the analysis and which research methodologies can be used to calculate and analyse such metrics. We select the metrics based on the following two general criteria. First, we focus only on metrics widely accepted and reported in industry because metrics proposed in the scientific literature but not reported by device manufacturers cannot be used to compare the performance of commercial LED devices over time. Second, the chosen metrics must be useful for understanding the impact of individual technological improvements on relevant performance and cost characteristics.

Historically, the progress in LED technology has commonly be described by pointing to impressive improvements in LED device performance, more specifically device brightness, electrical efficiency and manufacturing cost reductions[63]. However, metrics related to consumer experience, including the perceived temperature of a white light source and its ability to faithfully render colours, have also played a significant part in the market adoption of new lighting technologies[78 79 80 81] and received substantial attention in LED research and development efforts[18 82]. Therefore, a comprehensive analysis of the evolution in white LED technology must take into consideration advances in: 1) physical device performance; 2) consumer experience; and 3) LED device manufacturing costs. Next, we introduce and discuss the metrics that we use to track progress in each of these three areas.

3.1 Device Performance Metrics

During the years following the market introduction of white LEDs, the primary metric of progress in solid-state lighting was typically luminous flux. This was because the luminous flux (total brightness) of early white LEDs was too small to allow for the economical combination of multiple LEDs into lamps for general illumination purposes. In 2000, the highest performing devices yielded around 10lm, just below the output of a candle as defined in the unit candela (1cd=12.57lm)[83]. Progress in this metric was commonly rendered together with the associated reduction in retail prices as "Haitz's Law"[84 83]. Today, however, the devices with the highest performance yield in excess of 1600lm, the equivalent of a 100W incandescent bulb[85]. LED brightness has thereby become sufficient to enable the construction of lamps from multiple LED devices, with contemporary improvements focusing instead on higher efficiency and quality of light instead of brightness. Even though a large number of scientific publications and industry periodicals continue to focus on brightness as a metric of progress in lighting, we find that, at this point in time, this metric is insufficient to capture the complexity of the multitude of efficiency improvements that have been driving overall LED efficiency[86]. Specifically, the stagnating levels of luminous flux in highest performing devices does not capture a major research focus, which is improving electrical efficiency at constant brightness.

We therefore use a combination of the total device efficiency ("lamp efficiency") and the sub-efficiencies that describe the different physical loss channels within the device to describe progress in light-emitting diode technology.

3.1.1 Forward Voltage Efficiency η_{V_f}

Device forward voltage efficiency* (VfE) describes all electrical losses at the interface between the electrodes and the semiconductor and in the bulk. These losses can be due to tunneling and Ohmic resistance at the interface, as well as Ohmic resistance and other electrical losses within the bulk of the semiconductor. It is defined as

$$\eta_{V_f} = \frac{E_{h\nu}}{V_f} \tag{1}$$

where $E_{h\nu}$ denotes the photon energy and V_f is the diode forward voltage[87 56].

3.1.2 Light Extraction Efficiency η_{LE}

Device light-extraction efficiency (LEE) describes the losses due to absorption in the material after electron-hole recombination and the associated emission of a photon. It is defined as

$$\eta_{LE} = \frac{\#\text{ of photons out}}{\#\text{ of photons created}} = \frac{P_{opt}}{P_{int}} \tag{2}$$

where P_{opt} is the optical power of the device and P_{int} is the internal power of the device[87 56].

3.1.3 Internal Quantum Efficiency η_{IQ}

Internal quantum efficiency (IQE) describes non-radiative recombinatory processes in the semiconductor bulk. It is related to the external quantum efficiency through

$$\eta_{EQ} = \eta_{IQ} \times \eta_{LE} \tag{3}$$

and defined as

$$\eta_{IQ} = \frac{\#\text{of photons created}}{\#\text{of electron-hole pairs in}} \tag{4}$$

and depends on the current density of the device[87 56].

3.1.4 Droop η_{droop}

Efficiency droop describes the decrease of device internal quantum efficiency at high current densities, which is caused by a number of different physical effects[88]. This is often treated separately from internal quantum efficiency in the literature due to its importance in high-power devices. It is defined as

$$\eta_{droop} = 1 - \frac{\eta_{IQE}}{\eta_{IQE}(A \to 0)} \tag{5}$$

where $\eta_{IQE}(A \to 0)$ denotes the internal quantum efficiency at low current densities. In practice, droop is often given as the percentage difference between the ideal luminous intensity curve ϕ_{ideal} and the real luminous intensity curve ϕ at a set diode test current A_{test} as

$$D = \frac{\phi_{ideal}(A_{test}) - \phi(A_{test})}{\phi_{ideal}(A_{test})/100} \tag{6}$$

Therefore, according to this definition, a lack of droop corresponds to a droop efficiency of $\eta_{droop} = 100\%$[87 56].

* "Joule Efficiency" ε_J in Tsao et al.[56]

3.1.5 Conversion Efficiency η_C

Light conversion efficiency (LCE) describes losses in the conversion process of blue light that is the basis for all phosphor-converted white LEDs. These losses are the sum of the Stokes loss and well as scattering/absorption losses. It is defined as

$$\eta_C = \frac{E_B}{\sum_{i=R,O,Y,G} E_i} \tag{7}$$

where E_i denotes the total energy of light at the color corresponding to the down-converted photon wavelength[87 56]. Since every individual phosphor component in the device has its own associated conversion losses, the denominator sums over all components, i.e. Red, Orange, Yellow, Green.

3.1.6 Spectral Efficiency η_S

Spectral efficiency (SE) describes losses in the conversion process due to the wavelength-dependent efficiency of the human eyes. Photons converted into the infrared or ultraviolet are lost to illumination purposes. It is defined as

$$\eta_S = \frac{K}{K_{max}(CRI, CCT)} \tag{8}$$

where K is the luminous efficacy of radiation of the light source, which can be computed from the device spectrum and the luminosity function, which describes the sensitivity of the human eye. K_{max} is the maximum luminous efficacy of radiation of a perfect light source with the same color rendering performance and correlated color temperature as the light source in question[87 56].

3.1.7 Overall Efficiency ("Lamp Efficiency") η_L

Finally, the overall efficiency of a light-emitting diode package is the product of all considered sub-efficiencies

$$\eta_L = \prod_{i=(V_f,\dots,S)} \eta_i \tag{9}$$

It thereby describes the electrical and optical losses within the device, as well as the conversion losses in the phosphor layer. It further considers the mismatch between the device spectrum and an optimal spectrum, accounting for the wavelength dependent sensitivity of the human eye.

3.2 Consumer Experience Metrics

The perceived quality of light is entirely determined by the emission spectrum of a light source[89]. Any metric relevant to customer experience can thus be calculated from the spectrum alone. The spectrum of an LED light source is determined by the emission wavelength of the LED itself and the absorption and emission spectra of the down-conversion phosphor used in the device. It is typically included in the product datasheets provided by manufacturers, which enables the calculation of all relevant spectrum-based metrics for these devices. Based on the prevalence in scholarly literature and industry publications, for this study we choose two consumer experience metrics: Colour Rendering Index (CRI) and Colour Temperature. We do not consider flicker, the unintended high-frequency temporal modulation of light, which is another important consumer experience metric for lighting and a subject of recent regulation by the European Union[33]. This effect is caused not by LEDs themselves, but rather by inadequately designed electrical ballasts[90]. As a result, it is beyond the scope of this work.

3.2.1 Color Rendering Index (CRI)

The Colour Rendering Index (CRI) of a light source describes its ability to render the colours of an object faithfully when compared to illumination under a reference light source, such as standard daylight[91]. The way it is calculated is defined by the International Illumination Commission (CIE)[92]. CRI has certain limitations when applied to solid-state light sources[93]. However, despite repeated attempts at constructing more elaborate colour rendering metrics[94], CRI has remained the de facto industry standard for describing colour fidelity of light sources[95]. High colour rendering performance of lighting is a requirement in workplace environments, retail stores, clinical operating environments and art exhibitions[96]. It should be noted that some niche applications prioritize high colour saturation over high CRI, for instance, in food display or fabric retail applications[93]. However, these niche applications remain outside our focus on general illumination. Due to a broad availability and importance of CRI data for consumers of various LED lighting sources, we adopt CRI as the key metric to track progress in consumer experience in LED lighting, despite its limitations.

3.2.2 Colour Temperature

The Colour Temperature of a light source describes the equivalent temperature of an ideal black body which emits light of a colour comparable to that of the light source[97]. Warm white light sources are widely used in general illumination, while cold white light sources are used in workplace illumination and outdoor lighting. Early white LEDs produced only cool white light[98]. The introduction of first commercial warm white LED light bulbs played a significant part in increasing adoption of LED-based lighting among consumers, as their spectrum more closely resembled the warm white colour temperature of incandescent light bulbs[99]. For this reason, we also adopt colour temperature as a metric for tracking progress in consumer experience in LED lighting.

3.3 Manufacturing Cost

In selecting metrics for tracking the progress in manufacturing cost reductions in LED, we must highlight the complexity of this task. Access to manufacturing cost data at the chip level is usually restricted due to its proprietary nature and is available only for selected products. Using sales price information instead of the cost for the same purpose seems a promising alternative, as prices can be easily obtained directly from manufacturers for current products. However, historical data on prices for different chip architectures and different years is similarly difficult to obtain. In addition, sales price includes components not relevant to progress in the technology, such as profit margins and overhead costs, and is affected by policies such as rebates and purchase subsidies. Historical information on these factors and price components, as well as their impact on the manufacturing cost for each LED product under consideration is even harder to obtain, making the use of LED sales prices as a direct metric of technology progress very difficult in practice. Instead, we describe progress in the economics of manufacturing devices by using yielded cost as the primary metric.

The cost model we developed and describe in Section 4.4 relies on a cumulative approach to yielded cost[100]. In this approach, the yielded cost of process step 1 is defined as the ratio between the total cost of step 1 C_1 and the yield of step 1 Y_1:

$$C_{Y_1} = \frac{C_1}{Y_1},\ C_{Y_2} = C_{Y_{2\to 3}} - C_{Y_1} = \frac{C_1(1-Y_2)+C_2}{Y_1 Y_2},\ C_{Y_3} = \dots \quad (10)$$

This cost metric is cumulative by definition, thus

$$\sum_i C_{Y_i} = \frac{\sum_i C_i}{\prod_i Y_i} \quad (11)$$

Yielded cost per step is dependent on the step order and blind to downstream information[100]. Additional information on details of the computation of cost metrics and the different manufacturing steps is presented in Section 2 of the Supplementary Information.

4 Methods and Data Collection

The evolution of LED device architecture and performance as well as the progress in understanding the underlying physical phenomena are well covered in the scholarly literature and patents, as described in Section 2. However, information provided in such sources is insufficient for our goals on at least three accounts: First, existing work focuses only on selected performance parameters or overall device efficiency, rather than on providing a comprehensive coverage of the whole device sub-efficiencies for a particular LED product or design. Scientific publications also do not always disclose the underlying device architecture or the features responsible for the gains in performance. Second, not all relevant innovations are patented[101 102]. In the case of LED patents in particular, our interviews with industry experts suggest that the propensity to patent is the highest for knowledge related to macroscopic device architecture and chemical composition of phosphors, and the lowest for knowledge related to manufacturing process improvements and microscopic chip architecture that is difficult to reconstruct by reverse engineering. This means that relying only on patent literature would bias results by unduly emphasizing some focus areas and de-emphasizing others. Third, scientific publications and patents typically focus on experimental devices, rather than commercial products. While new LED features, designs and manufacturing methods reported in these sources can potentially result in significant performance gains or cost reductions, it is difficult to ascertain if these improvements have since been adopted in industry. Furthermore, information on LED manufacturing cost and the effect of process improvements on the total cost is highly proprietary. Estimates are occasionally reported in the scientific literature and company publications, but these often do not disclose which parts of the manufacturing process are responsible for the largest contribution to the overall cost, or which improvements led to cost reductions.

To overcome the limitations of these different methods for understanding technological progress, in this study we rely on a multi-method approach to data collection and analysis that combines information obtained from a review of the primary scientific literature, device datasheets and relevant patents with our own computations of device sub-efficiencies, information gained from industry publications, semi-structured interviews with experts from academia and industry, and bottom-up manufacturing cost modelling. We provide the details of implementation of each of these methods below.

4.1 Systematic Literature Review

We collected data on LED performance and characteristics in a systematic literature review that included scientific publications, patents, conference proceedings from the largest semiconductor and optoelectronics conferences, industry periodicals and roadmaps, as well as company presentations and reports. This review was structured around the three main goals: 1) tracking the evolution of LED technology over time as indicated by three groups of progress metrics selected above in Section 4.3; 2) identifying individual innovations that contributed to this evolution and whether or not they could be spillovers, and quantifying their impact on device performance and manufacturing cost; and 3) determining whether these innovations had originated within the LED technology domain, or in a field of research or technology outside of solid-state lighting, making them a technology spillovers.

Relevant sources for the review were found in an iterative search process that involved two components. The first was the search in specialized patent and publication databases as well as company websites. The second component was the analysis of backward citations in the identified sources, starting from the reviews mentioned in the Section 2 and then iteratively repeating it for all newly identified sources, until no further relevant and significant new sources were found. We also relied on backward citations for the identification of technology spillovers, considering cited documents as indicators of knowledge origins of an innovation and analyzing whether those documents belonged to the LED technology domain or not.

4.2 Semi-Structured Interviews

To supplement our data collection efforts, verify our findings and identify additional spillovers, we conducted a series of elite semi-structured interviews with eleven eminent experts from academia, industry and the public research sector. Experts were selected based on their engagement in different sub-fields of LED research and manufacturing, as well as the recommendations from other interviewed experts, in essence expanding the list of experts that emerged from the initial literature review. All interviews were conducted between November 2019 and April 2022 by means of video conferencing and lasted for about one hour. A summary of the background of interviewed experts is provided in Table SI 3 in Section 6 of the Supplementary Information.

The primary, structured part of the interviews explored which innovations were deemed most relevant to the evolution of device performance, consumer experience and manufacturing cost of LED packages. Thereafter, interviewees were asked to consider the extent to which these innovations may have originated outside of their respective field of expertise and the LED industry more broadly—i.e., which of the innovations may be considered spillovers. The remainder of the interview was focused on learning about particular aspects of the manufacturing processes relevant to cost and performance modelling, the current state of industry, and the circumstances surrounding the innovations and spillovers identified in the first part of the interview. Specific quantitative data was also provided by experts, helping fine-tune the parameters of the manufacturing cost model (described in Section 4.4) and verify device performance data.

4.3 Performance Metrics Calculations

The contribution of individual technology innovations and spillovers to the progress in overall device efficiency over time is estimated by index decomposition analysis. Mathematically, this involves breaking down a chosen performance indicator into its constituent components, each representing a specific factor that contributes to the change in the indicator [103]. Specifically, we use the additive logarithmic mean Divisia index method I (LMDI-I), also known as the Additive Sato-Vartia indicator [104]. It was developed by Boyd in 1987 [105] on the basis of Divisia Index, a method in statistical economics [106], and subsequently refined.

According to this method, for an overall device efficiency function F that is the product of variables a, b that represent sub-efficiencies, the contribution of the change in a single sub-efficiency variable a between times $t = 0$ and $t = T$ can be estimated as [107]

$$\Delta a = \frac{a_{t=T} - a_{t=0}}{\ln(a_{t=T}) - \ln(a_{t=0})} \times \ln\left(\frac{a_{t=T}}{a_{t=0}}\right) \tag{12}$$

$$\overset{a_{t=0} \neq a_{t=T}}{=} L(F_{t=T}, F_{t=0}) \times \ln\left(\frac{a_{t=T}}{a_{t=0}}\right) \tag{13}$$

where $L(F_{t=T}, F_{t=0})$ is the logarithmic mean of F values at times $t = 0$ and $t = T$. These terms contain no residuals, therefore it can be shown that the overall improvement in the device efficiency due to improvements in individual sub-efficiencies is equal to the sum of these improvements in individual sub-efficiencies:

$$\Delta a + \Delta b = \Delta F \tag{14}$$

To document historical improvements in LED device performance accurately, we need data on all sub-efficiencies for the selected device architectures and periods covered. However, the scope of data reporting in scientific literature and industry publications is typically limited to selected metrics of interest, rather than the full ensemble of sub-efficiencies that determine the overall device performance. For this reason, our data collection efforts were supplemented by performance calculations for individual sub-efficiencies where possible and necessary. Of the device sub-efficiencies, those related to the emission spectrum were computed from the spectral data often reported in LED device specifications. In particular, we used the `colour-science` package for Python [108] to calculate the luminous efficacy of radiation, colour rendering performance and luminous efficacy of radiation of phosphor down-conversion of blue light on the basis of available LED emission spectra. This approach allowed us to quantify the improvements related to phosphor development in LEDs.

4.4 Bottom-Up Manufacturing Cost Model

We address the limitations of data availability for the LED manufacturing cost by developing and applying a bottom-up LED manufacturing cost model with process-step resolution. In our case, it is enabled by the general availability of historical data on prices of relevant raw materials, components, and manufacturing equipment, and the relative cost data on manufacturing processes, which is occasionally published as part of industry press releases [109 110].

We identified two existing cost models for LED manufacturing in the prior literature. The first is the LED Cosim model developed by the technology consultancy System Plus Consulting [111 112]. Due to prohibitive licensing costs, this model could not be used in our study. The second model is the publicly available 2012 LEDCOM cost model prepared for the U.S. Department of Energy by Stephen Bland [113]. It was set up to capture the manufacturing process for phosphor-converted LEDs of different power levels. However, given the remarkable changes in the performance, cost and architecture of LEDs since 2012, as shown in Figures 1-3, this model cannot fully capture the recent advances in LED technology, necessitating a significant modification to bring it up to date with the current state-of-the-art in LED manufacturing.

Given this situation, we developed our own bottom-up LED manufacturing cost model. The model structure is generally based on the 2012 LEDCOM cost model, but we expand it significantly both in scope and in its ability to capture historical trends. The model captures three historical time periods corresponding to different "eras" in LED manufacturing: the early period of the first high-power white LEDs around 2003, the period of accelerating consumer adoption of LED lighting around 2012, and the most recent period around 2020, the year of our main data collection efforts. For each of these three years, the most prevalent manufacturing equipment was identified through industry periodicals, archived website data from the *Internet Archive*, and expert interviews. Because the architecture of LED chips has changed significantly since the introduction of the first commercial devices in 1996, three different chip architectures were initially considered in the model: classical chips, flip chips, and chip-scale package flip chips. The details of the manufacturing process for each architecture were collected from the scientific literature, textbooks and relevant patents. In addition, two LED life cycle analyses [114 115] were used to validate the model structure and extract some of the necessary quantitative model inputs. These studies captured a large number of LED manufacturing process steps and included the details on the use of metals, chemicals and electricity for each manufacturing step.

The aggregate result of the cost model is the manufacturing cost per LED package for each of the three years considered, which includes all costs associated with producing the chip, including running costs of the factory. Costs associated with research and development, administrative overhead of the manufacturer or other investment costs are not considered. We also note that the purpose of our cost model is not to give specific estimates of LED manufacturing cost for a factory of any size, specific geographic location or total annual manufacturing volume. It instead assumes an hypothetical factory with an assumed location in the United States and associated overhead costs related to the operation of the factory. It also assumes the use of the most up-to-date equipment for that year. Even with these simplifying assumptions, the model reasonably identifies the impact that changes in single process steps can have on the total LED manufacturing cost. An important limitation of our cost modelling efforts is that, even though the model captures three different chip architectures in its structure, in the present study we were able to collect, estimate and present the full set of quantitative inputs and outputs only for the classical chip architecture of low- to mid-power devices. Populating the model with data for the remaining two architectures requires access to proprietary industry information, which we have not been able to get thus far.

5 Results

5.1 Improvements in Sub-Efficiencies and Overall Efficiency

The historical development of a selection of the sub-efficiencies introduced in Section 3.1 is shown in Figure 4. Using this data, together with data on the remaining sub-efficiencies not shown in this figure, we calculated the overall LED efficiency in for four years: 2003, 2010, 2016 and 2020. Figure 6 shows how improvements in sub-efficiencies led to improvements in overall white LED lamp efficiency from $\eta_L = 5.8\%$ in 2003 to 12.7% in 2010, 32.5% in 2016 and finally to 38.8% in 2020. As is evident from the figure, no single loss channel dominates in terms of its contribution to the overall efficiency, in line with previous observations[56]. We note, however, that the loss channels with a fixed physical limit, e.g., Stokes loss that contributes to the conversion efficiency by phosphors, has become more dominant in 2016 and 2020 compared to 2003 and 2010. This is a direct result of the large efficiency improvements of upstream sub-efficiencies.

Figure 5 shows the overall magnitude of contributions of identified LED innovations and technology spillovers to improvements in LED efficiency over time across different sub-efficiencies. The list of technology spillovers identified in our study is provided in Table 2 and discussed in Section 6.3. The complete list of all LED innovations is provided in Table SI 2 in Section 4 of the Supplementary Information. Through the index decomposition analysis described in Section 4.3, we find that out of the overall LED efficiency increase of 32.9% from 5.8% to 38.8% between 2003 and 2020, at least 2.8% can be attributed specifically to technology spillovers identified in this study, corresponding to 8.5% of the total LED efficiency improvements between 2003 and 2020.

In Figure 5 we also compare, for the first time, efficiency improvements across sub-efficiencies over time, contrasting them with the physical limits of the corresponding loss channels. We find that there has been consistent progress across all device sub-efficiencies in the recorded period. Specifically, between 2003 and 2020, forward voltage efficiency increased from 70% to 99.5%, internal quantum efficiency from 55% to 90%, electrical droop from 65% to 90%, light extraction efficiency from 60% to 90%, spectral efficiency from 74% to 83%, conversion efficiency (red) from 11% to 45%, conversion efficiency (green) from 19% to 61%. Notably, some sub-efficiencies for the most recent devices considered in our study are now within $\sim 10\%$ of their respective physical limits. The exception is spectral efficiency which, at $\sim 17\%$ below the physical limit, shows larger potential for further improvements.

5.2 Improvements in Consumer Experience Metrics

Historical improvements in consumer experience metrics for phosphor-converted warm white LEDs are shown in Figure 7. In general illumination applications, a high colour rendering index (CRI) in combination with a specific, tunable range of possible colour temperatures is desirable. Both metrics are determined by LED device spectra, which, in turn, depend on the properties of the down-conversion materials in the device. We were thus able to establish the links between all major improvements in the two consumer experience metrics identified in this study and individual LED inventions associated either with phosphors or quantum dots. The list of these inventions is provided in Table 2, while details of the device spectra are provided and discussed in Section 5 in the Supplementary Information.

Notably, from detailed descriptions of the history of inventions in this list, which we provide in Section 5 of the Supplementary Information, we find that only a single invention related to LED consumer experience improvements was originally developed specifically for application in solid-state lighting: the 2016 SALON phosphor compound[130 129]. All other inventions in the list were either originally developed for non-LED applications or prominently used knowledge from areas of science and technology beyond LED or SSL. We discuss the details of the corresponding technology spillovers in Section 6.3 below.

5.3 Improvements in Manufacturing Cost

Figure 8 shows key results of our manufacturing cost modelling for low-to-mid-power classic chip GaN phosphor-converted white LED packages. We find that the manufacturing cost of a single such LED decreased from 1.11$ (in 2020 USD) in 2003 to 0.11$ (2020 USD) in 2012 and 0.05$ in 2020, a 95.5% overall decrease.

Among the factors contributing to the LED cost reductions over time, improved manufacturing yields and increases in the wafer size rather than particular LED innovations are found to be responsible for the largest contribution to the overall cost reduction. In the case of manufacturing yield, the higher it is, the less inputs are wasted on the production of a single LED package. With the total manufacturing yield dramatically improving from $\sim 25\%$ in 2003 to $\sim 75\%$ in 2020 (compare Figure 8, Panels A-C), it is not surprising that the total yielded LED manufacturing cost significantly declined over this period. In the case of wafer diameter, the larger the wafer, the more LED chips can be produced from a single wafer. The wafer diameter commonly

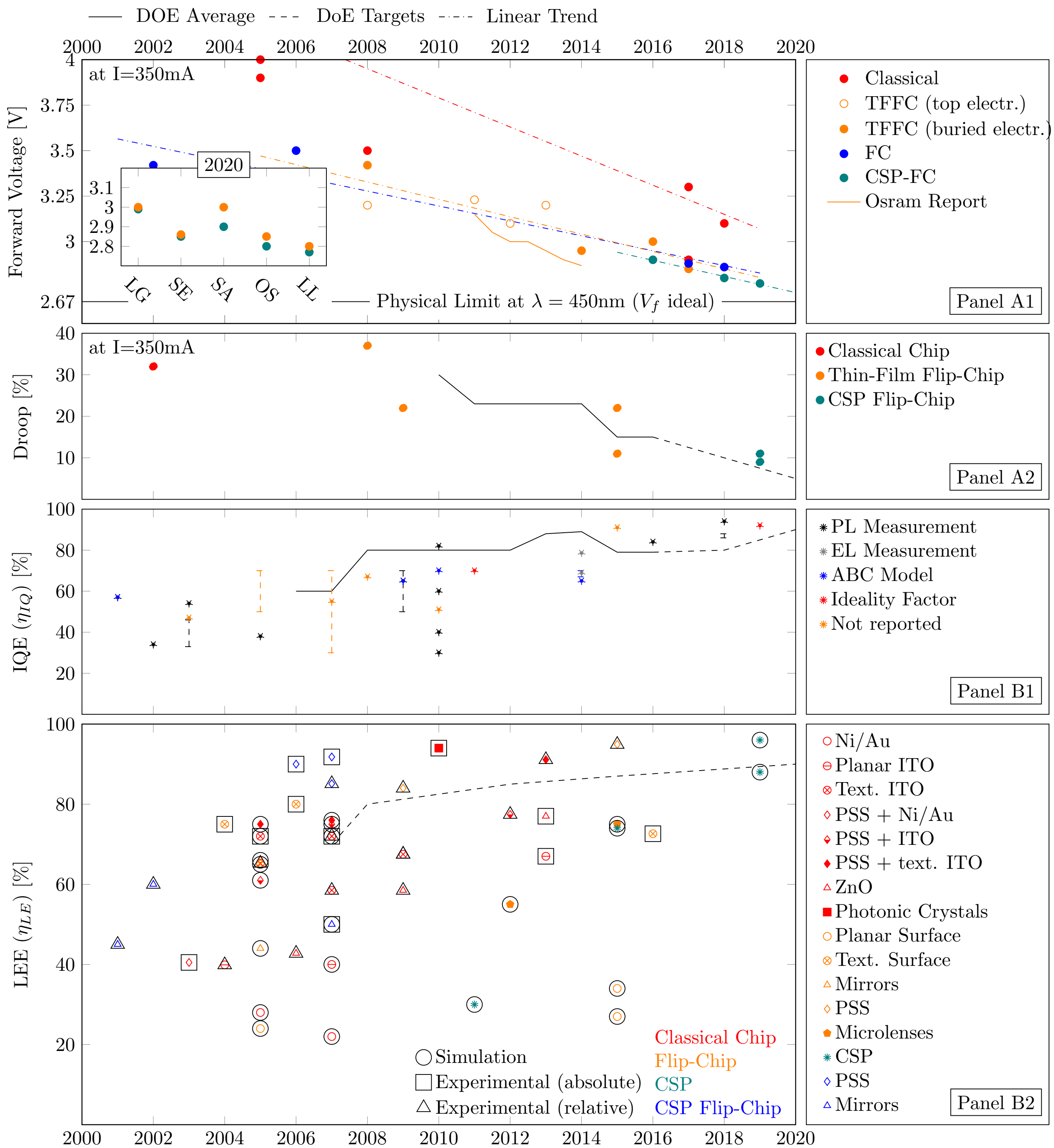

Fig. 4 Historical improvements in light-emitting diode technology. Panels A1-A2 show they key metrics used to compute forward voltage efficiency η_{Vf} and droop efficiency η_{droop}. Panels B1-B2 show the sub-efficiencies internal quantum efficiency η_{IQ} and light extraction efficiency η_{LE}.
Panel A1: Device forward voltage at a test current of I=350mA. The physical limit for a blue light wavelength of 450nm without electric pumping is shown for reference. Data points for devices released in 2020 by various manufacturers are shown in an inset plot. Panel A2: Efficiency droop at the test current of I=350mA. Panel B1: Internal quantum efficiency, for different chip architectures, by type of measurement used. Note that the artifact in the DOE Average around 2013 is due to a change in definition for internal quantum efficiency laid out in [116]. Measurement methods: PL - Photoluminescence [117], EL - Electroluminescence [118]; ABC Model [119], Ideality Factor [120]. "Not reported" denotes data points where the IQE measurement technique was not reported. Whiskers indicate reported ranges, where applicable. Panel B2: Light extraction efficiency for different chip architectures, by type of specific technology used to improve light-extraction efficiency. The source of data points is shown for reference: simulation - ray-tracing computer simulation; experimental (absolute) - light-extraction efficiency given directly; experimenal (relative) - relative improvement over baseline chip architecture given. Abbreviations: TFFC - Thin-Film Flip-Chip; FC - Flip-Chip; CSP - Chip-Scale Package; ITO - Indium Tin Oxide; ZnO - Zinc Oxide; PSS - Patterned Sapphire Substrate; electr. - electrode; text. - textured. Source: own synthesis of data from the full list of sources provided in the Supplementary Information, Section 5. Data on average device performance adapted from U.S. Department of Energy (DOE) Reports [121 122 116 123 124] and an Osram company report [125].

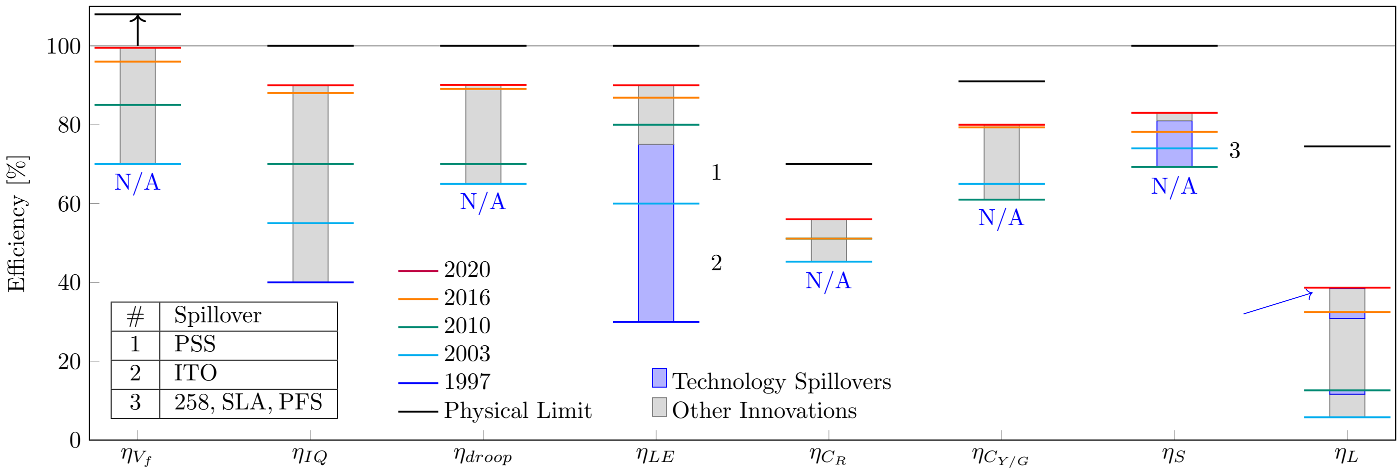

Fig. 5 Contribution of innovations and technology spillovers to the historical progress in sub-efficiencies of phosphor-converted warm white LEDs with test currents of at least 350mA. Vertical bars represent LED technology innovations, with purple bars indicating innovations driven by technology spillovers (annotated and listed in an inset table) and grey bars indicating all other improvements identified in this study. Horizontal coloured lines indicate state-of-the-art sub-efficiency levels for the four years used in Figure 6: 2003, 2010, 2016, and 2020. To provide additional historical context, data for 1997 is included for selected sub-efficiencies. "N/A" denotes sub-efficiencies where 1997 data could not be calculated to different reasons: η_{Vf}, η_{droop} depend on the device current, which was below 350mA in 1997, making comparison with contemporary devices difficult. $\eta_{C(R)}$,$\eta_{C(Y/G)}$ and η_S are relevant only to warm white spectrum LEDs, which were not available in 1997. Note that indium tin oxide (ITO) affects different sub-efficiencies depending on the chip architecture in question. For instance, in modern flip-chip architectures, light-extraction efficiency no longer depends on ITO, as can be seen from Figure 3. Physical limits on sub-efficiencies are indicated by black horizontal lines. The overall LED lamp efficiency, η_L, is displayed in the rightmost column. η_{V_f} - forward voltage efficiency; $\eta_I Q$ - internal quantum efficiency; η_{Droop} - efficiency droop; η_{LE} - light extraction efficiency; $\eta_{C(R)}$ - conversion efficiency for red phosphors; $\eta_{C(Y/G)}$ - conversion efficiency for yellow/green phosphors; η_S - spectral efficiency; PSS - patterned sapphire substrate, ITO - indium tin oxide current spreading layer; 258, SLA, PFS - different phosphors used in white LEDs, see Table 1. Note that physical limit on η_{V_f} is above 100% due to quantum effects which depends on electrical device parameters[126]. Source: own elaboration based on data in Figure 6 and Figure 4, as detailed in Section 4.

Year	Desig.	Chemical formula	Description	Significance	SI Sec.
1996	YAG	$Y_3Al_5O_{12}:Ce$	Yttrium aluminium garnet (YAG) phosphor activated with cerium	First LED phosphor, enabled white LEDs	SI 5.1.1
1996	YGAG	$(Y_{1-x}Gd_x)_3Al_5O_{12}:Ce$	Gadolinium-doped YAG phosphor	First red-shifted phosphor, enabled warm white LEDs	SI 5.1.1
2002	258	$(Ba,Sr)_2Si_5N_8:Eu^{2+}$	Europium-doped nitridosilicate phosphor	First red LED phosphor	SI 5.1.2
2003	QD	N/A	Quantum dot-based phosphor	First use of QD for LED light down conversion	SI 5.1.4
2005	PFS	$K_2SiF_6:Mn^{4+}$	Manganese-activated potassium fluorosilicate (PFS) phosphor	First ultra-narrow-band red LED phosphor	SI 5.1.3
2013	SLA	$Sr[LiAl]_3N_4]:Eu^{2+}$	Europium-doped cuboidal nitridolithoaluminate phosphor	Improved narrow-band red phosphor	SI 5.1.2
2016	SALON	$Sr[Li_2Al_2O_2N_2]:Eu^{2+}$	Europium-doped oxonitride phosphor	High-performance ultra-narrow-band red phosphor	SI 5.1.2

Table 1 LED down-conversion materials (phosphors and quantum dots) related to improvements in consumer experience metrics identified in this study. The *year* column represents the earliest reported application of invention in white LEDs. These differ from the years used in Figure 7 which correspond to the earliest publication of spectral data for representative LED products that relied on those materials. The *SI Sec.* column refers to the corresponding section in the Supplementary Information that contains references and a detailed description of the history of corresponding invention. Abbreviations: Desig. - Designation, Sec. - Section.

used in LED manufacturing has been steadily increasing 2003 from 51mm (2 inch) in 2003 to 200mm (~8 inches, referred to as "8 inch") in 2020. We capture this in the model by assuming the following wafer diameters and calculating the associated number of die per wafer (DPW)[136]: $d(2003) \sim 50mm \rightarrow 851DPW$, $d(2020) \sim 200mm \rightarrow 26,838DPW$. With more than a thirty-fold increase in the number of die per wafer between 2003 and 2020, the contribution of the whole-of-wafer processing steps to the total cost of manufacturing an individual LED chip and package has dramatically declined over time. As the number of die per wafer increases, the packaging steps, which in the classic chip architecture must be performed separately for each individual LED chip, carry a significantly larger share of the total cost in 2020 than in 2003 (compare Panels A-C in Figure 8). However, as our interviewees noted, while growing LEDs on larger wafers is economically desirable, it is associated with engineering and epitaxy challenges as well as high up-front cost of new equipment.

Our findings are further supported by a preliminary sensitivity analysis, presented in Section 2 of the Supplementary Information section, where we find that the sensitivity of the cost model

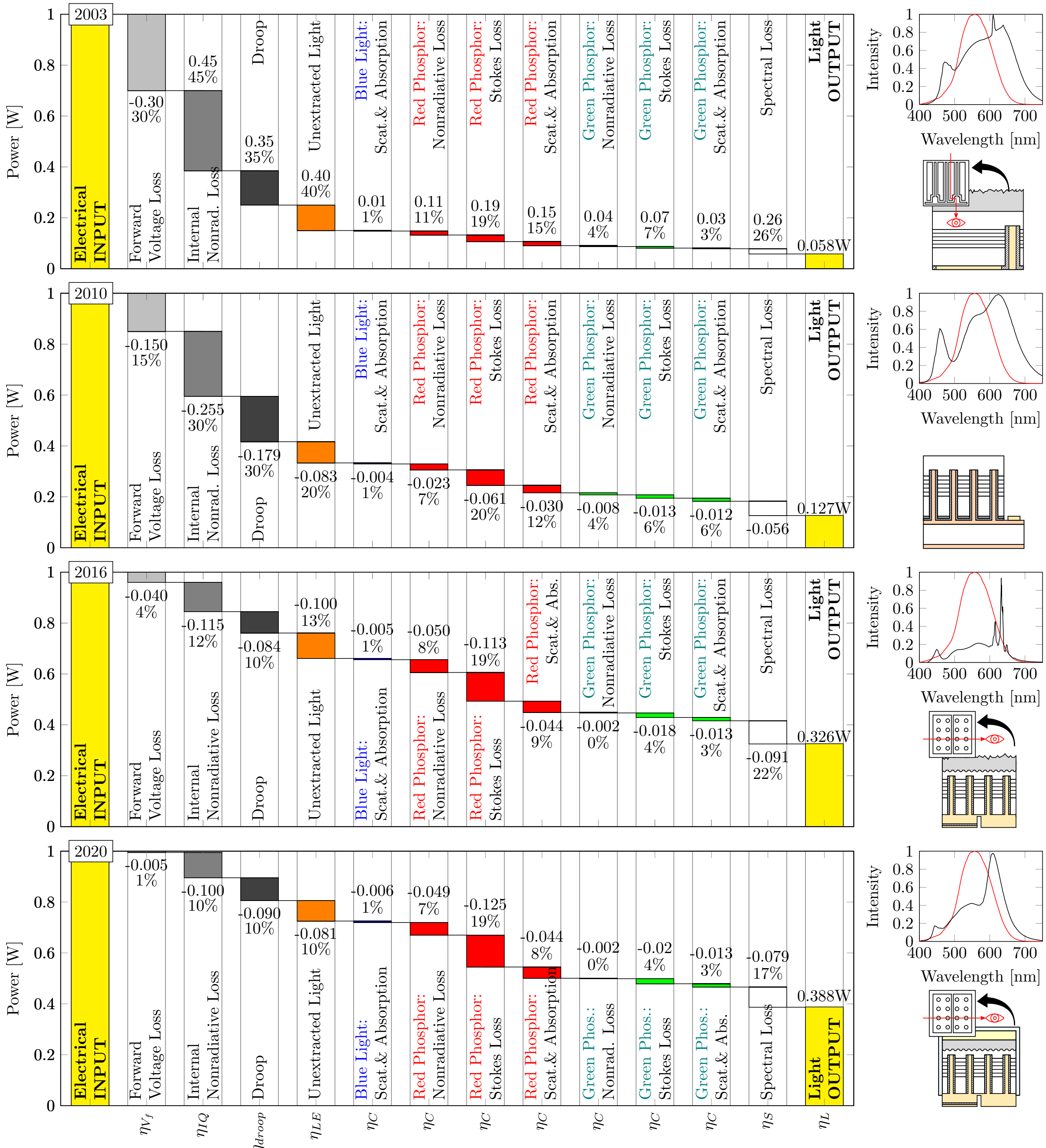

Fig. 6 Waterfall diagrams of the loss channels in a generic mid/high-power LED package for 2003, 2010, 2016 and 2020 (top to bottom panels), normalized to 1 Watt of electric power input (yellow bar on the left). Sub-efficiencies corresponding to each loss channel are listed below each column and described in Section 3.1. Numbers for each loss channel indicate energy losses both in relative terms of input power (in percent) at the point of the channel and absolute values (in Watts). Percentages for red, green and blue loss channels indicate losses of remaining red/green/blue light energy. The corresponding LED architectures and associated down-conversion phosphors are shown for reference: 2003 - flip-chip with YGAG phosphor; 2010 - flip-chip with 258 phosphor; 2016 - flip-chip with PFS phosphor; 2020 - flip-chip with SALON phosphor. A more complete overview of LED chip architectures is provided in Figure 3. Details on phosphor chemical composition are provided in Table 1 with additional data on the spectral performance and development history collected in Section 4 in the Supplementary Information. Overall LED package efficiency is $\eta_L = 5.8\%$ in 2003, $\eta_L = 12.7\%$ in 2010, $\eta_L = 32.6\%$ in 2016 and $\eta_L = 38.8\%$ in 2010. Abbreviations: Scat. = Scattering. Source (Efficiency): own elaboration based on data in Figure 4, as detailed in Section 4. Source (spectral data): Adapted from publications on the respective phosphors: YGAG (2003)[127], 258 (2010)[64], PFS (2016)[128], SALON (2019)[129].

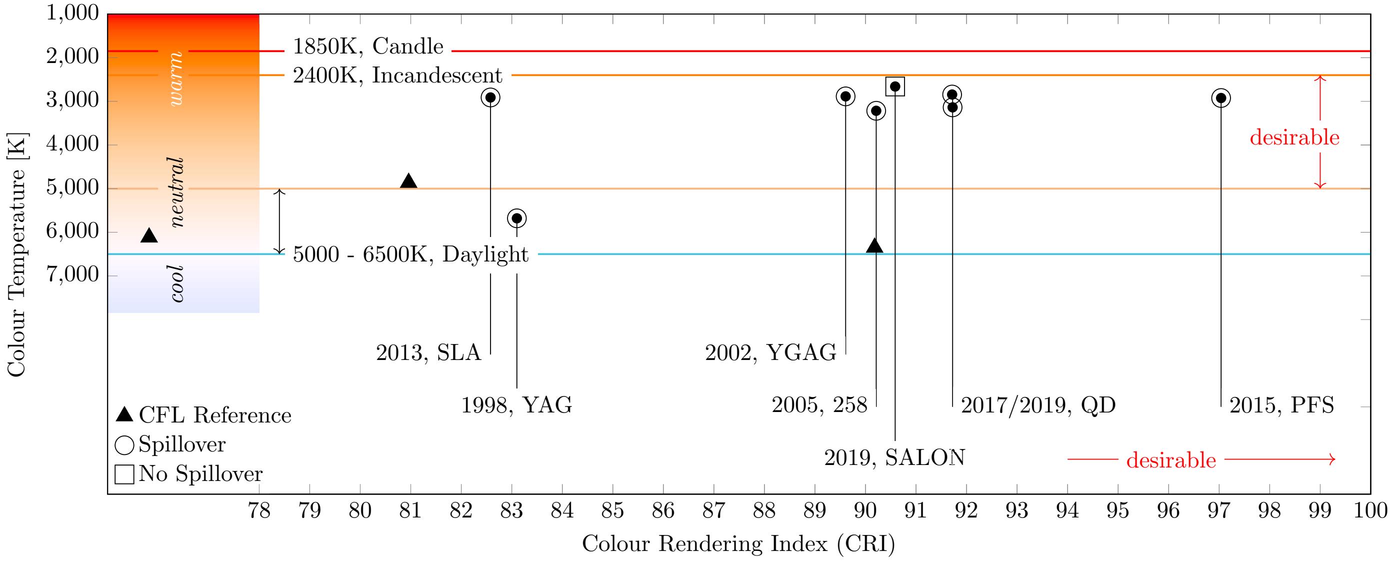

Fig. 7 Historical improvements in consumer experience metrics of phosphor-converted white LEDs. Data points show the color temperature and colour rendering index performance of the earliest identified representative white LED products with published spectral data that used phosphors listed in Table 1, each indicated by the phosphor label and publication year. Corresponding spectral data used to calculate colour temperature values is provided in Figure SI 11 in section 5 of the Supplementary Information. Three data points representing compact fluorescent lamps (CFLs)[131], shown as black triangles, are provided for comparison. Horizontal lines represent typical colour temperatures of "traditional" light sources, shown for reference. The desirable range of colour temperatures for home illumination, indicated by a vertical red arrow, lies between two horizontal orange lines representing typical incandescent light and warm daylight colour temperatures. Horizontal red arrow indicates desirable higher values of colour rendering index (CRI). LED products based on the following phosphor innovations from the following LED manufacturers are represented : YAG - Nichia, 1998[132]; YGAG - Lumileds, 2002[127]; 258 - Lumileds, 2005[64]; SLA - Lumileds, 2014[133]; PFS - GE, 2015[128]; QD - Lumileds, 2017[134], Osram, 2019[135]; SALON - Osram, 2019[129].

to variation in its main parameters decreases over time with the increase of the number of DPW. We also provide a comparison of our model with past cost calculations and projections published by the US DOE on the basis of the LEDCOM cost model[113] and industry data reported to the DOE as part of SSL round tables[137 138 139 140 141 142 143] in Section 2 of the Supplementary Information.

6 Discussion

6.1 Performance Improvements

Our findings show that the overall white LED efficiency has increased from $\eta_L = 5.8\%$ in 2003 to $\eta_L = 38.8\%$ in 2020. This efficiency increase was predominantly the result of a series of innovations in the LED architecture and materials as well as changes in the manufacturing process. Our comparison of improvements in device sub-efficiencies between 2003 and 2020 shows that the overall efficiency improvement was not dominated by any single sub-efficiency channel. Instead, there has been consistent progress across all device sub-efficiencies during the time period under consideration. Some sub-efficiencies, such as forward voltage efficiency, internal quantum efficiency and light extraction efficiency, are now within $\sim 10\%$ of their respective physical limits. A marked trend is the increasing relative contribution of losses in channels with physical limits on efficiency due to the underlying physics, such as light conversion efficiency of phosphors determined by the Stokes shift. Spectral efficiency also remains at $\sim 17\%$ from its physical limit, showing potential for further improvements. Research on improving LED performance across efficiency loss channels continues[82 166].

We further find that there has been significant progress in performance across the set of consumer experience metrics, which was driven by innovations related to the conversion of blue light generated by conventional GaN LEDs into the white light required for general illumination. The first commercial white LEDs produced by Nichia in 1996 used a YAG phosphor that could generate only cool white light[132]. After a series of breakthrough innovations in phosphors shown in Figure 7 and described in Section 4 of the Supplementary Information, LEDs today can be tuned for high colour rendering performance, high colour saturation and a range of desirable colour temperatures.

Overall, we find that LED technology innovations made crucial contributions to the progress in both LED performance and consumer experience metrics across the entire period covered by our study. Our interviews have also revealed an important role of incremental process improvements and learning-by-doing[167 168] in the progress in LED efficiency. However, further research is needed to quantify the contribution of learning by doing to this progress.

6.2 Cost Reductions

Our LED manufacturing cost model shows that the cost of manufacturing low-to-mid power GaN-based white LED packages with classical chip architecture at a U.S. location using state-of-the-art equipment has decreased by 95.5% from 1.11$(2020) in 2003 to 0.05$(2020) in 2020. In contrast with LED performance im-

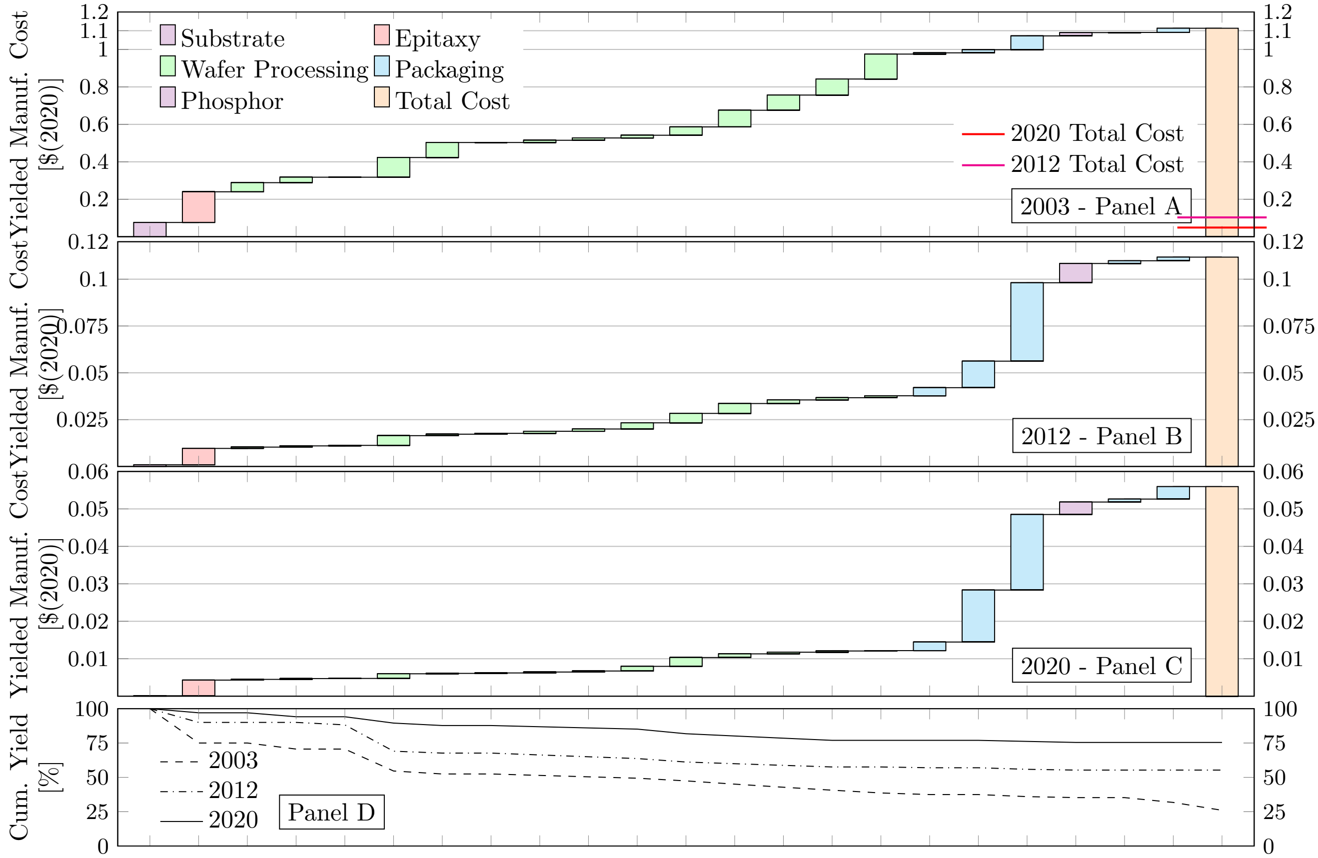

Fig. 8 Manufacturing cost structure modelled for a single low-to-mid power GaN, classic chip, phosphor-converted white LED package, assuming an ideal factory with state-of-the-art equipment at a U.S. location. Consecutive steps shown on the x-axis. Panels A-C: Waterfall diagrams of LED manufacturing cost split by manufacturing process steps for years 2003, 2012 and 2020. Process steps on the horizontal axis are sequenced from left to right in the same order as in the modelled LED manufacturing process. Panel F: Cumulative manufacturing yield after each process step for years 2003, 2012 and 2020. For a visual representation of the overview of the manufacturing process, compare the diagrams in Section 2 of the Supplementary Information. Abbreviations: Litho – Lithographic Process, Insp. – Inspection, Depn. – Deposition, CMP - Chemical-Mechanical Planarization

provements, where progress was driven mostly by LED technology innovations, the dramatic decline in LED manufacturing cost resulted from an increase in the wafer size used in manufacturing and higher yields across manufacturing steps. This can be seen in the reduction of the contribution of wafer processing steps to totaal cost in Figure 8. Both were enabled by advances in manufacturing equipment performance and incremental process improvements from learning-by-doing.

Notably, our bottom-up cost model is constructed to provide process-step resolution across three different key chip architectures: classical chips, flip chips, and chip-scale package flip chips. However, in this study we were able to collect data and compare the outcomes only for the classical chip architecture. Collecting the full set of data needed to populate the model for the remaining two architectures would require access to proprietary information from industry. With this limitation, tracking manufacturing cost declines across three key LED chip architectures remains a topic for future work.

6.3 Contribution of Technology Spillovers

We find that nine technology spillovers identified in our study, listed in Table 2, affected all dimensions of white LED performance. Three spillovers associated with the use of YAG/YGAG phosphors in LEDs played the key role in the first commercial white LED lighting products, essentially enabling the solid-state lighting market and industry of today. Spillovers also had a significant effect on physical device performance, cumulatively contributing to 8.5% of the total LED lamp efficiency improvements between 2003 and 2020. Technology spillovers were particularly important for consumer experience performance of white LED lighting sources, with corresponding spillover-driven innovations responsible for ∼100% of the improvements in consumer experience metrics.

Following the framework for the analysis of technology spillovers proposed by Stephan and colleagues[7], we gathered information from the interviews, historical records, literature reviews and industry publications to analyze the sources, mechanisms, and enablers of the identified spillovers into the white LED technology listed in Table 4. Among the spillover sources, we find that all nine spillovers had origins in basic science disciplines such as various branches of chemistry, materials science, optics and photonics, and solid-state physics. Five spillovers also utilized technical knowledge and expertise in cathode ray tubes, fluorescent lighting, optoelectronic devices, nanotechnology, and nature-inspired material design.

Among the spillover mechanisms, six spillovers (involved in all phosphors except PFS and SLA, plus ITO) were a result of application of external scientific and technical knowledge already available to researchers and inventors. Three remaining spillovers (involved in PFS and SLA phosphors, plus PSS) occurred as an outcome of targeted search for relevant external knowledge outside the LED domain. In addition, at least two spillovers (involved in 258 and PFS phosphors) occurred through direct R&D collaboration.

Among important enabling factors for the identified spillovers, we highlight public mission-driven R&D funding; industry-academia partnerships; firm experience in multiple industries; conferences that brought together researchers from academia and industry; cultural and language proximity; freedom of search in academia; and university alumni networks.

We find that, on average, it took 26 years from the initial scientific discovery or invention to the moment of its spillover into the LED domain, with this time varying from 5 to almost 70 years. In contrast, it took much less time – just 6 years on average, varying from just a few months to 19 years – to develop a commercial application for the spillover knowledge in the LED market.

7 Conclusions

Our study has analysed the sources of the dramatic cost reductions and performance improvements in white light-emitting diodes since their introduction to the market in 1996 in a systematic and granular way. We find that the total LED device efficiency increase from 5.8% to 38.8% between 2003 and 2020, as well as improvements in consumer experience metrics, have been predominantly driven by LED technology innovations that affected all physical energy loss channels and corresponding device sub-efficiencies. We also find that among those innovations, at least nine were driven by knowledge spillovers originating in areas of science and technology beyond LEDs or solid-state lighting. These spillover-driven innovations were responsible for 8.5% of the total efficiency improvements and nearly 100% of the improvements in consumer experience metrics.

Our manufacturing cost model shows a 95.5% decrease in the cost of producing white classic-chip LEDs (from 1.11$(2020) to 0.05$(2020)) between 2003 and 2020, driven mostly by increases in the wafer size and yields across different manufacturing steps over time. In contrast with efficiency improvements, these learning by doing, economies of scale and improvements were facilitated by both improvements in the performance as well as policies creating and growing market demand, such as incandescent light bans and subsidies for LEDs.

Our analysis of the sources, mechanisms and enablers of the identified technology spillovers which were significant drivers of improvements in LED efficiency and consumer experience metrics, highlights the critical role played by a deep understanding of the physical, chemical and optical phenomena underlying the operation of LEDs, as well as materials science and technology and nanotechnology involved in the production of LEDs, for past and future advances in LED and solid-state lighting technology. Specifically, deep physical understanding of LED device efficiency loss channels enabled important innovations in LEDs that increased several sub-efficiencies in LEDs and will continue, as expected by eminent experts in the field[166]. This suggests that additional research in these areas and a more deliberate search for relevant external knowledge may accelerate expected future advances in LED technology. These knowledge spillovers can be enabled or even accelerated by knowledge exchange events and long-term partnerships between academia and industry, dedicated mission-driven public R&D funding, and a freedom of search in academia. This further reinforces arguments made against the dichotomy of basic research versus applied research[169 170] and the calls for open, inclusive and flexible research cultures[7].

There are various important avenues of future research that are opened up by our analysis. First, future work could expand the cost model by collecting and including data for a broader set of chip architectures and analyzing the impact of individual innovations and spillovers on the costs as opposed to performance. Second, a deeper dive on the role of learning-by-doing is needed both in the cost and performance analysis. Third, building on the work on LED sub-efficiencies and physical limits, future efforts could focus on identifying priority areas for further efficiency improvements in LEDs and SSL in general. Finally, by comparing the drivers of innovation, technology spillovers, cost reductions and performance improvements at a granular level across different clean energy technologies, we can identify patterns or differences that would help us formulate recommendations for industry and policymakers aimed at accelerating further clean energy innovation for climate change mitigation.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This research was supported by the grant from the Alfred P. Sloan Foundation titled *"What factors drive innovation in energy technologies? The role of technology spillovers and government investment"*. Michael Weinold further gratefully acknowledges support from the Swiss Study Foundation. We would like to thank Venkatesh Narayanamurti, Gabriel Chan, Anna Goldstein, Didier Sornette, and participants of the SPIE West 2021 Conference and C-EENRG Seminar Series of the University of Cambridge for many helpful discussions and feedback. We also express our deepest gratitude to all interviewees for their willingness to participate in this study and share their invaluable insights with us.

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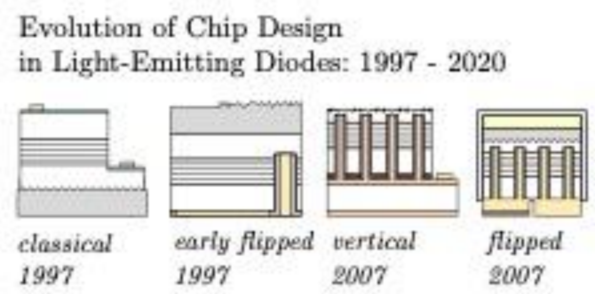
Disc.	S/O	Comm.	LED Innovation	Spillover	Origin	Ref.	Area of Improvement
1926	1994	1996	LED phosphors	Use of phosphors for light down conversion in LEDs	Materials science (S), Cathode ray tubes (T)	[144 145 82]	Enabled light down conversion in LEDs
1967	1996	1996	YAG phosphor	Use of YAG phosphor in a first white LED product	Chemistry (S), Materials science (S), Fluorescent lighting (T), Cathode ray tubes (T)	[146 147 132 148 82]	Enabled white LED products, η_S, η_C
1967	1996	<2002	YGAG phosphor	Use of YGAG phosphor in first warm white LEDs	Chemistry (S), Materials science (S)	[149 132 148 127]	Enabled warm white LEDs, η_S, η_C
1982	1996	<2010	Patterned sapphire substrate (PSS)	Use of anti-reflective properties of substrate patterns in LEDs	Optics and photonics (S), Materials science and technology (S,T), Nature-inspired material design (T)	[150 151 62 152]	η_{LE}, η_{IQ} (depending on the chip architecture, compare Figure **??**)
1971	1999	<2005	Indium tin oxide (ITO) current spreading layer	Use of ITO current spreading layer in white LEDs	Optics and photonics (S), Materials science and technology (S,T), Optoelectronic devices (T)	[153 154 155]	η_{Vf}, η_{LE} (depending on the chip architecture, compare Figure **??**)
1997	2002	2005	258 phosphor	Use of luminescent '258' nitridosilicate compound as LED phosphor	Chemistry (S), Materials science (S)	[156 157 64]	η_S, η_C
1984	2003	2009	Quantum dot-based phosphor	Use of quantum dots for light down conversion in LEDs	Solid-state physics (S), Photochemistry (S), Nanotechnology (T)	[158 159 160 161]	η_S, η_C
1972	2005	2015	PFS phosphor	Use of knowledge in luminescent materials and skills in "wet" chemical synthesis to synthesize PFS compound and optimize it as LED phosphor	Chemistry (S), Materials science (S)	[162 163 128]	η_S, η_C
2008	2013	2015	SLA phosphor	Use of knowledge about existing cuboidal nitride compounds to identify and synthesize structurally similar SLA phosphor	Structural chemistry (S), Materials science (S), Solid-state physics (S)	[164 165 133]	η_S, η_C

Table 2 Technology spillovers involved in white LED technology innovations identified in this study. Note: LED innovations are ordered by the year in which a technology spillover into LED occurred, provided in the Spillover to LED column. Initial discovery is the year of the earliest identified discovery of the original idea or invention outside the LED domain. Commercial application is the year of the first recorded application of that idea or invention in a commercial LED product. Origin column represents knowledge domains in which spillovers initially emerged, where (S) denotes a scientific discipline and (T) is an area of technology. References column lists literature sources for the represented innovations and spillovers. Area of improvement represents the impact of spillovers on different aspects of white LED technology, e.g., improvements in particular sub-efficiencies. Abbreviations: Disc. - Year of Discovery, S/O - Year of Spillover to LED, Comm. - Year of Commercial Application., Ref. - References.



Quantifying the underlying historical drivers of LED efficiency and cost reductions can inform policy to accelerate broader clean energy innovation.

In 1997, a series of material science breakthroughs enabled the manufacturing of bright blue light-emitting diodes, which can be used together with a coating phosphor to generate white light. Since then, the technology has experienced rapid improvements in performance and manufacturing cost, which has resulted in widespread global adoption. This has led to a significant reduction in global electricity consumption from lighting, which accounts for almost 20% of global electricity consumption and 6% of CO2 emissions. However, the underlying drivers of the large improvements in efficiency and decreases in manufacturing cost are still poorly understood. In particular, it remains unclear if any technological breakthroughs have originated in another industry sector or scientific discipline. These “technology spillovers” are crucial for clean energy innovation as they enable the transfer of knowledge, skills, and technology developed in other sectors. To facilitate spillovers and thereby accelerate innovation in other clean energy technologies, we can learn from the technological success story of light-emitting diodes. This analysis provides the first disaggregation of the underlying drivers of efficiency improvements and identifies technology spillovers and their impact on LED development. With climate change mitigation an ever more pressing issue, this analysis fills an important gap in understanding the role of research management, research funding structure, and policy on technological progress in clean energy technologies.

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Neil Scriven
Executive Editor
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Dear Mr. Scriven,

We would like to submit an original analysis article entitled "Rapid progress in light-emitting diodes and its sources in innovation and knowledge spillovers" for consideration by *Energy & Environmental Science*. We confirm that this manuscript has not been published elsewhere and is not under consideration by another journal. All authors have approved the manuscript for submission to E&ES.

The turn of the year has marked the end of the 25th anniversary of the breakthroughs that enabled the manufacturing of the first bright white light-emitting diodes (LEDs) in 1997. Since then, this technology has experienced rapid efficiency improvements, cost reductions and widespread global adoption. The global annual energy savings from the adoption of highly efficient LED light sources are larger than the total annual electricity production by all photovoltaic installations globally. The long lifetimes and low cost of LEDs have contributed to the adoption of LEDs beyond usual early adopter markets: particularly remarkable is the early adoption of LED lighting in rural communities around the globe, presenting the first off-grid lighting alternative to kerosene lamps. There is also evidence to suggest that the importance of LEDs is extending well beyond general illumination, since niche applications have emerged on either side of the electromagnetic spectrum: infrared LEDs are now used in personal health devices for vital sign monitoring and ultraviolet LEDs are used to enable access to drinking water in difficult environments.

The relatively short time period during which the technology has come to establish itself in different markets makes LEDs a useful technology to study to gather insights to help accelerate innovation in other clean energy technologies. With climate change mitigation an ever more pressing issue, our research thus fills an important gap in understanding the role of research management, research funding structure, and policy on technological progress in energy technology.

Efforts to quantify the contribution to overall efficiency of the underlying technological advances in light-emitting diode technology have been previously limited the difficulty in navigating a field of active research in which most relevant breakthroughs are closely guarded trade secrets and all manufacturing data is proprietary and that most efforts have considered shorter time periods and not the full technology.

Our research has, for the first time, been able to disaggregate the underlying drivers of efficiency improvements in light-emitting diodes between 2000-2020. In particular, we show that technology spillovers – knowledge originating in other sectors or scientific disciplines – have played a major part these improvements and what factors enabled them. We show in particular the impact that fundamental research in inorganic chemistry (phosphor materials) has had on first enabling and thereafter continuously improving the most relevant metrics in consumer experience, a vital element in driving rapid adoption of the technology. We also report, for the first time, the breakdown of cost reductions across three time periods showing how advances in metal-organic chemical vapour deposition (MOCVD) have enabled the scale-up of manufacturing volume required to make the technology affordable to general illumination purposes.

We thank you for considering out manuscript!

Kind regards,

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