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Rydberg Physics

Nikola Šibalić and **Charles S Adams** discuss new possibilities for quantum optics, simulation and sensing offered by highly-excited, weakly-bound atomic states.

Rydberg Physics

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Abstract

Rydberg physics is the land of gentle giants—highly-excited electronic states where an electron is, on average, far from the nucleus. They are created with a lot of energy, have unusually large size and live relatively long. They are useful because they are extremely sensitive to their environment; so much so that a single photon of light can change the behaviour of not only one, but dozens or hundreds of them, over distances we can easily see under an optical microscope. Wherever Rydberg states are created—in atoms, molecules or solids—they all share common features that are exemplified by the physical description of highly-excited hydrogen atoms, with energy levels described by the Rydberg formula.

In this ebook, we outline the universal properties of Rydberg systems and then present a selection of ideas that demonstrate their increasing relevance to applications, including sensing, the manipulation of light on the single photon level (quantum optics) and in engineering the interactions between individual quanta (quantum simulation).

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Charles S Adams studied physics at the University of Oxford and received a PhD on laser physics from the University of Strathclyde in Glasgow, UK. He completed postdoctoral work in Germany and the United States before starting a research group at Durham University in 1995. He is Director of JQC Durham-Newcastle and was awarded the 2014 Institute of Physics Thomson medal for his pioneering work in the field of Rydberg quantum optics.

Rydberg Physics

Nikola Šibalić and Charles S Adams

Rydberg Physics

1 Introduction

In Sweden—the land of the northern lights—Anders Jonas Ångström (1814–74) pioneered the field of atomic spectroscopy, identifying the spectral lines of hydrogen in 1862. In 1885, Swiss physicist Johann Jacob Balmer (1825–98) wrote down a formula that described Ångström’s hydrogen spectra. Then a few years later (1890), another Swedish physicist Johannes (Janne) Robert Rydberg (1854–1919) penned a more general formula that described the spectral lines emitted by atoms that have a single outer shell (valence) electron.

According to Rydberg’s formula, the energy of the n th energy level of hydrogen is given by

$$E_n = \frac{-R_H}{n^2},$$

where R_H is known as Rydberg’s constant and n is known as the principal quantum number. Any system with a ‘delocalized’ electron plus a more localized positively charged core looks somewhat hydrogen-like, and a modified form of Rydberg’s formula is applicable where the energy levels are arranged in a specifically ordered series. We illustrate these Rydberg series with two very different examples—a hot gas and a cold semiconductor where excited electrons are bound by the negative hole left behind, forming an exciton—in figure 1. As many systems display these properties, any atom (molecule or semiconductor) with a highly-excited electron in a state with high n is known as a Rydberg atom. We shall use the generic word ‘atom’ from now on.

Ångström’s measurements and Rydberg’s formula became hugely significant because of what followed: first, the birth of quantum physics with Niels Bohr’s model of the atom, and then the birth of scattering theory developed by Enrico Fermi, built on the study of the broadening of Rydberg spectral lines by Edoardo Amaldi (1908–89) and Emilio Gino Segré (1905–89). These initial experimental

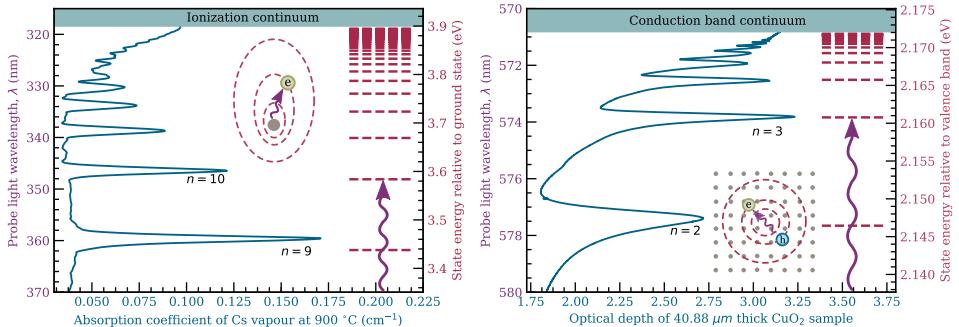


Figure 1. Rydberg states are ubiquitous. Energy levels of highly-excited bound states of many, at first sight very different, systems can be described with Rydberg's formula. This is because the wave function is mostly localized away from the binding potential centre, and hence is insensitive to the short range structure of the potential source. Thus, Rydberg's formula not only describes excitations of hydrogen, but also other atoms and molecules. Even weakly-bound excitations in solid-state systems, like Wannier–Mott excitons in CuO_2 consisting of bound electron–hole pairs (e and h, respectively, in inset) exhibit the same fundamental physics. Data sources: Goran Pichler, Institut za fiziku, Zagreb and Kuwait University (caesium spectra) and Matthew Jones, Durham University (cuprous oxide spectra).

observations of Rydberg series were made in hot gases, excited with broadband lamp light sources. Broadening of the spectral lines due to collisions, and weaker absorption of the lamp light by high-lying states, allowed clear observation only of states with low principal quantum numbers. As technology developed, physicists were able to observe Rydberg atoms with higher and higher principal quantum numbers. In 1965, astronomers at the National Radio Astronomy Observatory in West Virginia, USA, detected the microwave radiation emitted by hydrogen atoms in Orion making a transition beginning in a state with initial principal quantum number $n = 110$; see figure 2. Such highly-excited Rydberg atoms are enormous—of the order of a micron across—larger than a virus, and 10 000 times larger than a ground-state atom. More recently, states with $n = 1000$ have been observed.

The fact that the outer electron is on average so far from the core makes it very weakly bound, and hence very sensitive to external electric fields, including fields induced by nearby Rydberg atoms. This exaggerated sensitivity makes Rydberg atoms ideal for sensing electric fields. The large coupling between Rydberg atoms and microwave fields was exploited by Serge Haroche (1944–present)—winner of the Nobel Prize for Physics in 2011—and colleagues in pioneering light–matter interactions at the level of single quanta.

Current theoretical and experimental efforts are broadly directed towards applications in three areas: sensing, quantum optics and quantum simulation. Below we provide a fundamental description of Rydberg state properties, and then describe their applications with a focus on current research.

2 Background

Rydberg states with high principal quantum number n are observed in high-energy environments such as interstellar media in space, as well as in plasmas on Earth.

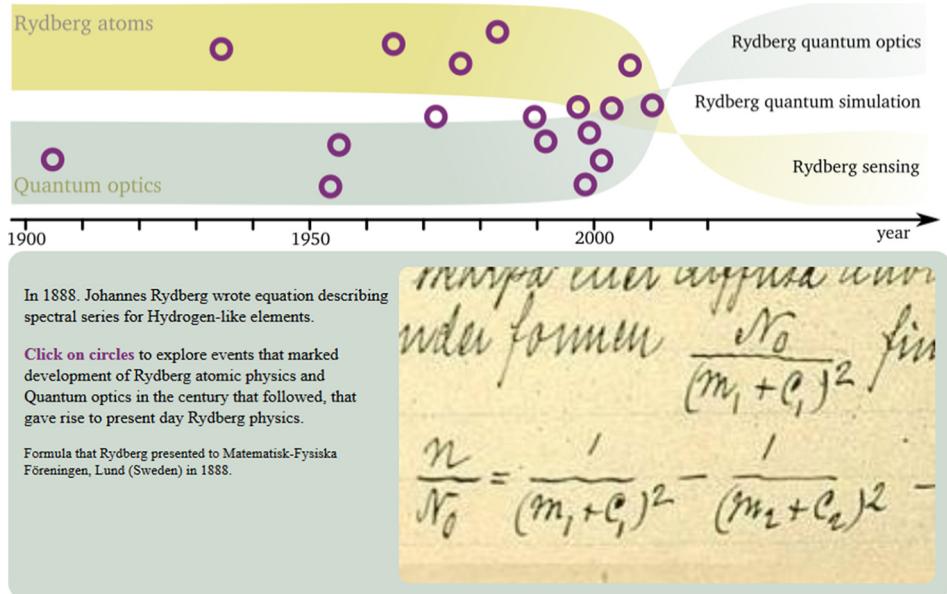


Figure 2. Timeline of some of the early developments. Points mark selected conceptual advances in quantum optics and Rydberg atomic physics. Together with new experimental capabilities provided by reliable coherent driving with wide frequency tuning range, they led to the accumulation of ideas and experimental possibilities for coherent control of atom–light interactions, both in the microwave and visible spectra, paving the way for modern Rydberg physics research. This figure is interactive in the online version, and available to download from <http://iopscience.iop.org/book/978-0-7503-1635-4>.

More controllable excitation of selected Rydberg states has become possible in recent decades with laser technology. A timeline—necessarily incomplete—mapping some of the important discoveries, experimental demonstrations and illuminating concepts that shaped our understanding is shown in figure 2. Below, we shall mainly focus on laser excitation of Rydberg atoms.

Laser excitation

Excitation of Rydberg states from the ground state requires high energy. In alkali metals, for example, the photon energy to excite atoms initially in the ground state is in the ultraviolet region of the electromagnetic spectrum. However, multi-stage, multi-photon excitation schemes involving light in the infrared or visible region are also used. Precision laser excitation first became possible with the invention of tunable dye lasers in the 1970s. More recently, the development of other narrow linewidth lasers and techniques to prepare ultra-cold atoms using laser cooling and trapping—the topic of the 1997 Nobel Prize for Physics awarded to Steven Chu, Claude Cohen-Tannoudji and William D Phillips for the ‘development of methods to cool and trap atoms with laser light’—enabled a new level of precision.

Atom–light interactions: basic concepts

To understand atom–light interactions, we need to know the coupling strength between different atomic states. The coupling originates from interaction of atoms with the electromagnetic field. Often the electromagnetic field comes from a laser, and then we speak of *laser driving*, that forces a *transition of atomic population* between coupled levels, corresponding to cycles of *absorption* and *stimulated emission*. There are many possible spatial directions for laser driving of a single atom. Each of these directions corresponds to a particular spatial mode. Remarkably, even if there is no light present in these modes an excited atom still feels their presence and couples to them: in this case we call them *vacuum modes*. Excited atoms decay via coupling to the vacuum modes, releasing energy in the form of the *spontaneously emitted photons*. The energy of the photons absorbed or emitted into the modes of the electromagnetic field matches the energy difference of the two states involved, and can correspond to different parts of the electromagnetic spectrum: from high-energy ultraviolet, over visible and infrared, to low-energy terahertz and microwave radiation (see figure 3). Transitions in ultraviolet, visible and infrared spectra are often called *optical transitions*. For these we use optical technology: lasers, glass lenses and dielectric mirrors, optical fibres, etc. Transitions in microwave and terahertz regions are driven and detected with technologies developed in high-frequency electronics, like microwave generators, horns, antennas, etc.

For most transitions, the strength of the coupling of two states is proportional to the size of the *dipole operator* $e\mathbf{r}$, where e is electron charge and \mathbf{r} is electron position, that forms between these states. For the two coupled states with electron wave functions $|a\rangle = \psi_a(\mathbf{r})$ and $|b\rangle = \psi_b(\mathbf{r})$, respectively, solving the integral over space $\langle a|e\mathbf{r}|b\rangle = \int \psi_a^*(\mathbf{r})e\mathbf{r}\psi_b(\mathbf{r})d\mathbf{r}$ would give the so-called *dipole matrix element* that determines the strength of coupling between these two states and the electromagnetic field. The radiation pattern of such a dipole corresponds to the electromagnetic field distribution of an oscillating classical dipole.

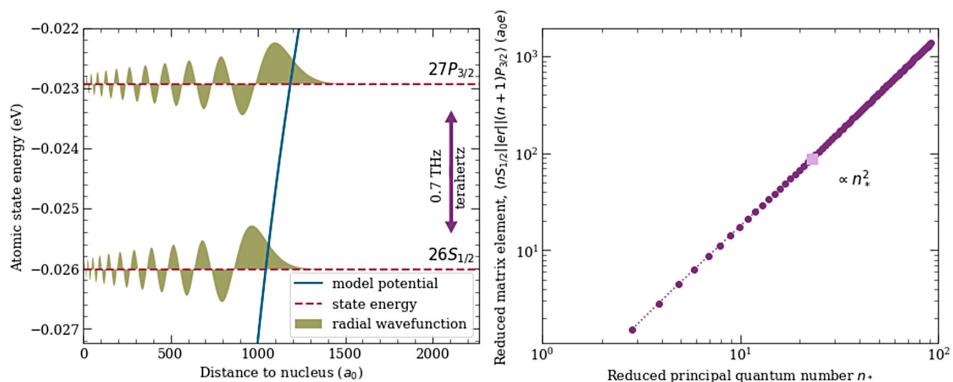


Figure 3. Rydberg state wave functions and scaling of transition strengths between nearby, dipole-coupled states. In alkali atoms, transitions between states close to the ground state (low n) typically correspond to photons in the near-infrared range. However, for intermediate- and high- n states, the transitions are in the terahertz and microwave regions of the spectrum, respectively. The long radiation wavelength for high n means that interactions between nearby Rydberg atoms are typically modelled with equations describing near-field dipole radiation. *Weakly-bound* states also have *large spatial extent*. This leads to the characteristic scaling of the interaction and lifetime properties. Calculation shows data for rubidium atoms. This figure is interactive in the online version, and available to download from <http://iopscience.iop.org/book/978-0-7503-1635-4>.

Rydberg lifetimes

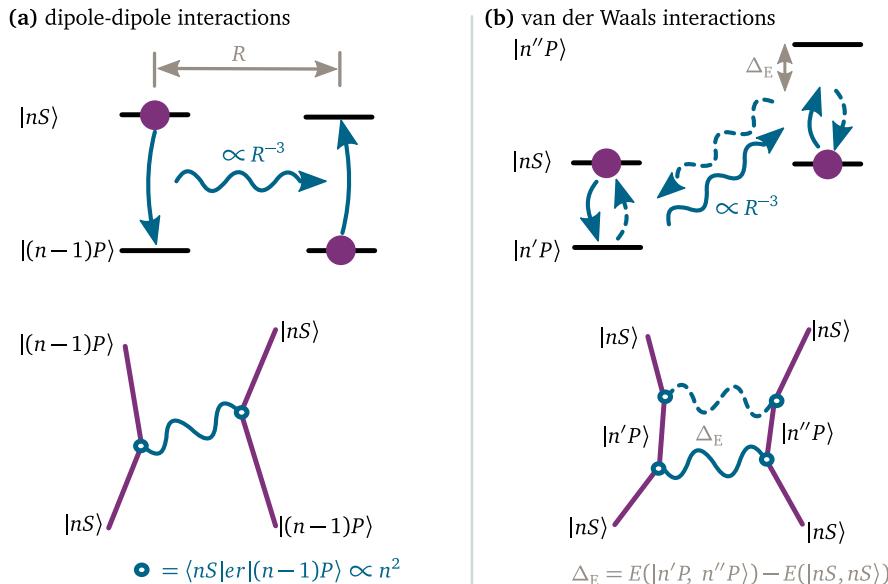
The size of the electron cloud in a Rydberg atom is proportional to the principal quantum number squared ($\propto n^2$; see figure 3). This size scaling property leads to particular n scalings for other properties, such as lifetime, and sensitivity to either electric fields or other Rydberg atoms. The spatial overlap between the small ground-state wave function and a large high- n Rydberg state wave function gives a small value of the dipole operator between these two states (for a review of atom–light interactions and terminology, see box), and hence weak coupling via electromagnetic field. This makes direct laser excitation difficult but also means that Rydberg states are relatively long lived, as decay back to the ground state is slow. One might initially expect that the increased coupling between neighbouring highly-excited states—both with large wave functions $\propto n^2$ (see figure 3)—would cause rapid spontaneous decay to other highly-excited states. After all, the dipole operator for these transitions, which is in the microwave and terahertz regions of the electromagnetic spectrum, scales with the size of the Rydberg atom, i.e. also $\propto n^2$. However, luckily for the stability of Rydberg systems, the number of vacuum modes (see box) available for spontaneous decay in the microwave and terahertz regions goes down even faster. Taking into account all possible decay paths of the initial states, one finds that Rydberg state lifetimes scale as n^3 or even n^5 (for a state with high angular momentum), reaching hundreds of microseconds (μs) for $n \sim 60$. This allows longer interaction times compared to the low-lying excited states (which have lifetimes of only several tens of nanoseconds), making Rydberg states an excellent resource for practical applications.

Detection

The relatively long lifetimes of Rydberg states produce very narrow spectral lines for transitions between them, which were measured in the 1970–1980s using precisely stabilized state-of-the-art microwave sources, like backwards wave oscillators and klystrons. These measurements provided very detailed knowledge of Rydberg energy levels, and that knowledge is still used today in precise experimental modelling. As lifetime increases, and hence the rate of the optical transition back to the ground state decreases, it becomes ever more difficult to detect Rydberg states via fluorescence decay, and the measurement of Rydberg state populations becomes a nontrivial task. In the 1970s, it was shown that applying static electric fields ionizes Rydberg atoms and the charges produced are easily detected. The higher the Rydberg state, the lower the ionization threshold becomes, so this method becomes easier at higher n . Also, by increasing the electric field over time, it is possible to resolve different Rydberg levels in the time domain (see figure 2, 1977). Nowadays, other methods for detecting Rydberg states based on laser spectroscopy, such as the technique of electromagnetically induced transparency (EIT; see figure 2, 1991 and below), and precisely timed, power-stabilized laser pulses have been developed. Laser pulses can be used to transfer Rydberg state populations into lower-lying atomic levels that can subsequently be detected via fluorescence.

Interactions

Strong interactions between two atoms excited into Rydberg states originate from *virtual* photon exchange between them. This redistributes the atomic population between highly-excited states. For example, figure 4(a) illustrates that if one atom is in a $|nS\rangle$ state, where S is the label used for a zero angular momentum state, while the other is in a $|(n-1)P\rangle$ state (with angular momentum, \hbar), the first atom can emit a photon, decaying to state $|(n-1)P\rangle$ in the process, while the second atom absorbs that photon, being excited in the process. Overall, the effect is a transfer of state and energy between the atoms $|nS, (n-1)P\rangle \rightarrow |(n-1)P, nS\rangle$. The rate of the first decay process is proportional to the optical dipole operator that couples these two states via decay to the vacuum mode, i.e., spontaneous emission, $\langle nS | er | (n-1)P \rangle \propto n^2$ (see box). The rate of the inverse process, i.e., absorption of a single photon from a given mode, is determined by the same operator. Hence, this type of interaction scales with principal quantum number as n^4 . The dependence of



Example: Rubidium atoms

$|nS_{1/2}, (n-1)P_{3/2}\rangle$ pair-state

Interactions $V(R) = C_3/R^3 \propto n^4/R^3$

$n = 20$	$C_3/h = 83 \text{ MHz } \mu\text{m}^3$
$n = 80$	$C_3/h = 40 \text{ GHz } \mu\text{m}^3$

Example: Rubidium atoms

$|nS_{1/2}, nS_{1/2}\rangle$ pair-state, all $|n'P, n''P\rangle$

Interactions $V(R) = C_6/R^6 \propto n^{11}/R^6$

$n = 40$	$C_6/h = -1.0 \text{ GHz } \mu\text{m}^6$
$n = 80$	$C_6/h = -4.2 \text{ THz } \mu\text{m}^6$

Figure 4. Interactions between Rydberg states. (a) Resonant dipole–dipole and (b) off-resonant van der Waals interactions between Rydberg states. Top panel shows the relevant energy-level scheme, with the initial population indicated by circles. Middle panels show schematic representations of the process with a Feynman diagram, highlighting the origin of the different scaling of the interaction strengths.

the interaction strength on the interatomic distance, R , can be determined by considering the interaction between two static dipoles. This is because we are typically interested in interactions over distances much smaller than the wavelength, λ , of the photons mediating the interaction, i.e. $R \ll \lambda$. For example, strong interactions between Rydberg atoms in an $n = 60$ state typically occur at distances of up to $R \sim 10 \mu\text{m}$, while the wavelength of radiation mediating this interaction is $\lambda \gtrsim 1 \text{ mm}$. Therefore, retardation of the field can be neglected, and calculations are done in the near field of emitters. As dipole–dipole interactions scale as R^{-3} , the overall strength of these *dipole–dipole interactions* is $\propto n^4/R^3$.

Another notable case of Rydberg state interactions occurs between two atoms in the *same* Rydberg state. For example, for two atoms in state $|nS\rangle$ (see figure 4(b)), we can proceed as in the previous example, and consider a process where one atom emits a photon transferring it to state $|n'P\rangle$, while the other atom absorbs that photon and goes to the state $|n''P\rangle$. However, it is often the case that no pairs of states $|n'P\rangle$ and $|n''P\rangle$ exist whose total energy E is exactly the same as that of the original pair-state $|nS, nS\rangle$. An *energy defect* $\Delta_E = E(n'P, n'P) - E(nS, nS)$ effectively forbids such a process from happening. However, the process can still occur if the coupling is sufficiently strong, because the energy is imprecisely defined over a short time (on short time scales δt , energies of states cannot be defined better than $\delta E \sim \hbar/\delta t$, as can be seen from the Fourier time–energy uncertainty relation). From the virtual state, $|n'P, n''P\rangle$, the photon can be exchanged again (dashed line in figure 4), returning the atomic population back to the $|nS, nS\rangle$ pair-state. The strength of this second-order process, which involves two pairs of photon emission–absorption events, each with rate $\propto n^2$, can be calculated in the second-order perturbation theory and it scales as $n^8/(\Delta_E R^6)$. Note that now the interatomic distance dependent factor is R^6 , which appears since there are two photon exchanges involved in the process, each contributing one R^{-3} . Through this second-order process called the *van der Waals interaction*, even two atoms in the same Rydberg state can interact. Since for neighbouring states $\Delta_E \propto n^{-3}$, the overall strength of these interactions is $\propto n^{11}/R^6$, scaling extremely quickly with n . The value of the energy defect Δ_E can be changed by using external static fields that shift the energy levels of atoms, providing a means to tune the Rydberg interaction strength in experiments.

In contrast to resonant dipole–dipole interactions, the off-resonant van der Waals interactions do not necessarily cause a state change. Yet still they make the energy of atomic Rydberg states strongly dependant on interatomic distance, R . Such interaction-induced level shifts of Rydberg states can have a dramatic influence on the dynamics of the system. This can be seen by considering how the atom-pair dynamics for laser excitation change dramatically depending on the pair separation, R (see figure 5).

First, consider the case where a laser is tuned to the Rydberg state resonance for a single atom, i.e., laser detuning $\Delta = 0$ (blockade regime in figure 5). Now imagine a pair of atoms: one in the Rydberg state, the other in the ground state. The latter's Rydberg state energy level will be shifted in energy by amount V in the presence of the existing Rydberg atom. This is due to the interaction V that could be of either dipole–dipole ($V \propto n^4/R^3$) or van der Waals ($V \propto n^{11}/R^6$) type, as explained above.

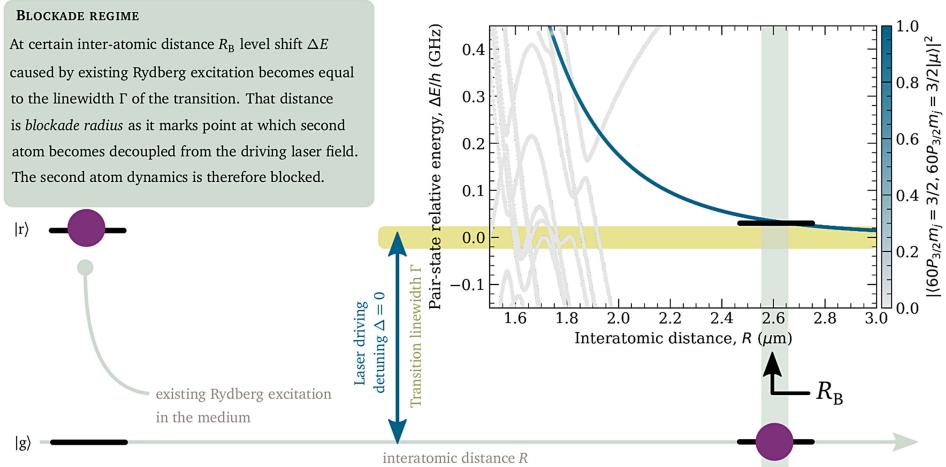


Figure 5. Influence of Rydberg interactions on two-body dynamics under laser driving. Depending on the laser detuning and Rydberg atom locations, Rydberg excitation may be either suppressed or facilitated. Some characteristic distances at which this blockade or facilitation of laser driving to Rydberg states happens are introduced and labelled as *Rydberg blockade radius* and *facilitation radius*, respectively. The interactions between Rydberg states may influence Rydberg excitation even when the laser is far detuned from resonance. In this so-called *Rydberg dressing regime*, interactions will affect the AC Stark shift (the shift in an atom's energy levels in response to the applied electromagnetic field) experienced by the atoms. The pair-state calculations (top right panel) are presented for the $|60P_{3/2}, 60P_{3/2}\rangle, M = 3$ pair-state of rubidium ($M = 3$ means that the angular momenta are parallel), with the highlighting marking the contribution of the original pair-state to the obtained eigenstates. This figure is interactive in the online version, and available to download from <http://iopscience.iop.org/book/978-0-7503-1635-4>.

At large interatomic separations R , the shift in energy is small compared to the transition linewidth, Γ , and the second atom can be excited with the same dynamics as expected for the single-atom case. However, at a certain distance R_B known as the *blockade radius*, the interaction strength, V , is sufficient to shift the energy level of the second atom by more than the transition linewidth, Γ . Below that blockade distance, given by $V(R_B) = \Gamma$, the second atom is completely decoupled from the driving field, and any laser-induced dynamics bringing it to the given Rydberg state is blocked.

Consider now a case where the driving laser is detuned in the direction of the interaction-induced level shift (facilitation regime in figure 5). Here, the second atom is not coupled to the driving laser except at a particular distance, R_F , where the interaction-induced level shift brings it into resonance with driving field $V(R_F) = \Delta$. At this distance R_F , the excitation dynamics are *facilitated*, allowing excitation of pairs of Rydberg atoms.

Finally, consider the case where the driving laser is detuned by a frequency Δ away from the interaction-induced level shift (dressing regime in figure 5). Imagine the situation where *both* atoms are initially in the ground state. Now, the resonant process of exciting the atoms is energetically forbidden, yet the presence of the Rydberg state is still felt through the laser-induced second-order process $\propto \Delta^{-1}$, which

admixes (i.e., adds) a small amount of Rydberg state into the steady state of the system. A consequence of this admixing is a shift in the energy (a so-called *AC Stark shift*). However, in the region of small interatomic separations (below the blockade radius), interactions shift the energy levels of both atoms to such an extent that they prevent the admixing of Rydberg states. As a consequence, at short distances only one of the atoms can experience an AC Stark shift, while at large distances ($\gtrsim R_B$) both atoms can independently experience the effect. This dressing can therefore be summarized as an effective soft-core potential (see U_d in figure 5 for $\Delta < 0$), with a smooth crossover between two regions happening at R_B . In this way, Rydberg–Rydberg interactions are felt among atoms that are almost completely in the ground state, apart from a small Rydberg admixture brought by dressing. This ground-state character of such interacting states is advantageous for realizing quantum simulation protocols (to be discussed in the following section), as ground-state atoms can be trapped and controlled over long time scales. The soft-core shape of the potential is also of fundamental interest, as it creates new phases of matter.

Now that we have reviewed the properties of Rydberg atoms, we shall move on to consider specific applications: first sensing, then nonlinear optics and finally quantum simulators.

3 Current directions

Rydberg sensors

The extreme sensitivity of Rydberg states to electric fields is one reason we called them ‘gentle’ in the introduction. Sensitivity stems from the large spatial extent of the wave function of the weakly-bound valance electron. It was recognized early on that this sensitivity could be used to measure weak electric fields. Static fields, originating for example from patches of charged particles, as well as fields originating from propagating electromagnetic waves in the microwave and terahertz spectra of frequencies, can be measured and imaged. Atoms are particularly advantageous as measurement devices; first because they are the same everywhere, and second because their properties do not evolve over time. This makes them ideal calibrated meters, that are distributed all around the Universe and do not suffer from any drift. If the measurement of one such atom is calibrated once against the standard definition of electric field strength, then we can use identical atoms, wherever they are—forever—for subsequent measurements, and not have to worry about calibration ever again.

To understand how atoms can be used for measuring static electric field, imagine a small static electric field E acting on an atom. We quantitatively describe the action of the electric field by adding a new term into the system Hamiltonian that accounts for the energy shift electrons with charge $-e$ experience in such a field. For an electric field directed along the z -axis, the new term is $-ezE$. If two opposite parity atomic energy eigenstates are labelled $|a\rangle$ and $|b\rangle$, then the effect of the field is to couple them together. The strength of this coupling $eE\langle a|z|b\rangle$, directly reflects the size of the wave functions corresponding to the states $|a\rangle$ and $|b\rangle$. If we pick one atomic state $|a\rangle$ and introduce the electric field, the new eigenstates will have a

contribution from the other state $|b\rangle$ (as illustrated in figure 6(c)). This admixture of other states is responsible for a change in the energy of the initial state $|a\rangle$. For small electric fields, for which we can perform a second-order perturbation calculation, the shift is proportional to $|eE\langle a|z|b\rangle|^2/\Delta_E$, where Δ_E measures the energy difference between the levels $|a\rangle$ and $|b\rangle$.

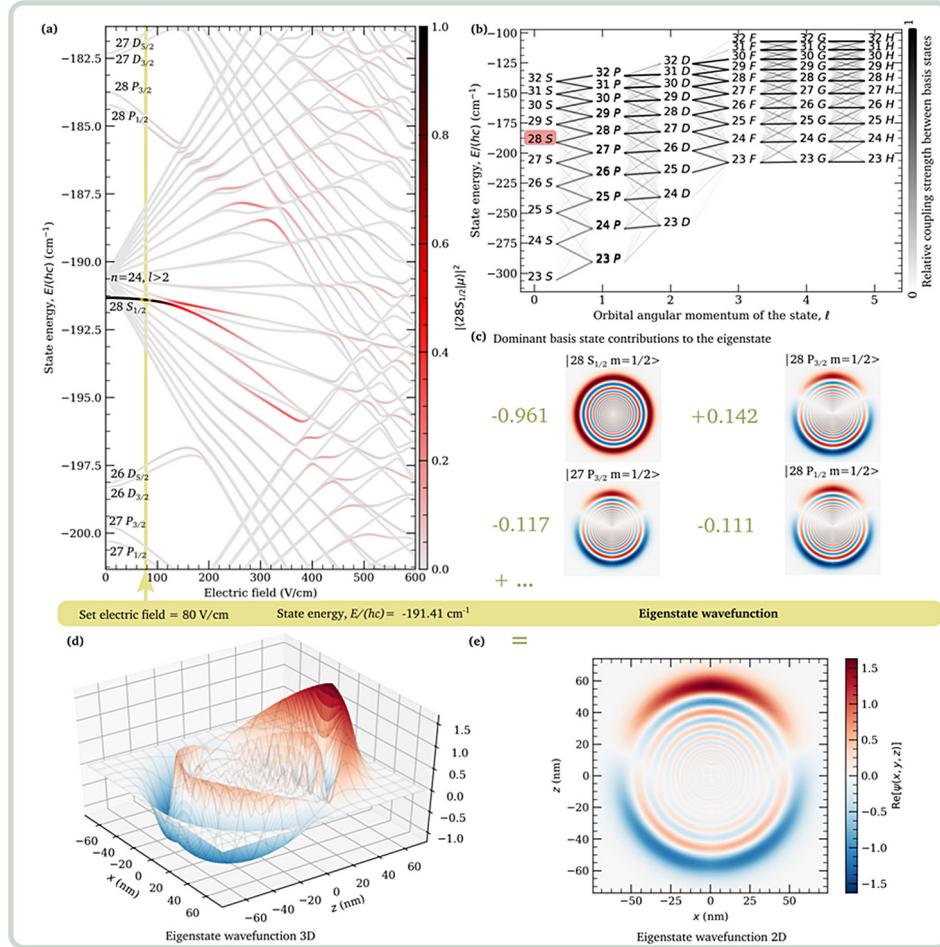


Figure 6. Changes in eigenstate composition and energy for an atom in a static electric field. (a) State energies calculated for caesium atoms in a static electric field. The calculation (in this example, for the caesium $28 S_{1/2}$ state) proceeds as follows. Initially, a basis is formed from the states of the atom in the absence of electric field that lie close in energy to this target state, shown in (b) only up to $\ell=5$. Static electric field Hamiltonian $-eEz$ couples states $|n,\ell,j,m\rangle$ and $|n',\ell',j',m'\rangle$ with $\ell-\ell'= \pm 1$. This is indicated in (b) by lines whose shading maps the relative strength of the coupling between different elements. The Hamiltonian evaluated in this basis produces a sparse matrix that can be diagonalized for every value of the electric field. Resulting eigenstates will be a combination of the basis states, as shown in (c) for the state marked in (a) by a yellow circle. Only four basis states with the largest absolute amplitude contribution to the eigenstate are shown. The resulting electron wave functions (panels (d) and (e)) clearly show how atoms acquire permanent dipole electric moment in a static field. This figure is interactive in the online version, and available to download from <http://iopscience.iop.org/book/978-0-7503-1635-4>.

Now we can see what makes Rydberg states so extremely sensitive to fields compared to the low-lying atomic states. As we have seen in the previous section, the size of the excited electronic wave function and hence the coupler term $eE\langle a|z|b\rangle$ increase as n^2 . At the same time, the energy difference between neighbouring states closes as $1/n^3$, leading to level shifts in the applied electric field, proportional to $|eE\langle a|z|b\rangle|^2/\Delta_E$, that scale as n^7 overall. Take a moment to comprehend this scaling: if one goes from states with principal quantum number of around 15, to states with principal quantum number 80, energy shifts of states in the electric field with the same magnitude will increase by over *six orders of magnitude*!

Sensing electromagnetic fields corresponding to microwave and terahertz frequency radiation is important for a wide array of applications. If we could see them, the room around us would be full of these waves dancing around, allowing communication between many devices through WiFi, Bluetooth and mobile phone networks. They are used for ranging and communication in the aerospace industry and self-driving cars, and for terahertz security scanners in airports. A collective of Rydberg atoms measures microwave and terahertz radiation in real time, and can even visualize the signal, converting this invisible low-energy radiation to the visible range (light).

If the energy (and hence frequency) of such an electromagnetic field matches a transition between the two Rydberg states, the field will be strongly captured by the atoms due to the large dipole matrix elements that scale as n^2 ; see figure 3. The field is absorbed, causing a redistribution of the population of atomic energy levels. If more field arrives, it can cause stimulated emission of the initially captured field photons out of the medium, allowing the medium to capture new photons over the next time step. These cycles of absorption and stimulated emission repeat (oscillate) causing a so-called *Rabi oscillation* with a frequency Ω that is proportional to the product of the amplitude of the applied electromagnetic field and huge dipole matrix element of the Rydberg atoms used for sensing.

Rabi oscillations due to applied field are interesting fundamental phenomena that occur within atoms, but how can one exploit them to build a simple electromagnetic field sensor? The sensing medium—for example, room temperature alkali metal atoms—is held in a small evacuated transparent container called a spectroscopic cell. A tiny chunk of the required alkali metal is placed inside the evacuated cell. The metal evaporates until saturated atomic vapour is formed inside, and it is this vapour that forms our sensing medium. The read-out of the sensor is provided by energy levels, which can be measured precisely by recording the laser absorption that occurs when laser photons are absorbed while exciting some atoms from low- to high-energy states. If we pass only one laser beam through the cell and measure its absorption, the sensor is not very precise because at room temperature the atoms move around at speeds close to the speed of sound, and the Doppler effect produces a distribution of energies in the hot cloud. However, by combining multiple laser beams to shuffle the population between energy levels within the same atom, using techniques known as population shelving and EIT, one can create spectroscopic peaks that are significantly narrower than the Doppler-broadened resonance. Although we are using this sharper sub-Doppler energy scale, we still need a needle

that points to the amplitude of the measured microwave field. This is provided by the Rabi oscillations that move back and forth between two close-by Rydberg levels $|a\rangle$ and $|b\rangle$. If we try to set our laser at the frequency where the atomic level (for example, $|a\rangle$ in figure 7(a)) is in the absence of microwave field, when we turn the microwave field on we will see not only one absorption line, but two of them separated by a small energy distance around the originally expected line. Under constant redistribution of population due to applied microwave field, the two energy eigenstates now consist of a combination of the two Rydberg states $(|a\rangle + |b\rangle)$ and $(|a\rangle - |b\rangle)/\sqrt{2}$, up to normalization factor), each allowing absorption of the probe laser beam due to populating $|a\rangle$ (top panel of figure 7(a)). The energies of these eigenstates are different by Planck's constant times the Rabi frequency of the microwave field-induced population redistribution. As the Rabi frequency Ω is directly proportional to the microwave field amplitude, this splitting in the spectrum (right panel in figure 7(a)) provides a pointer to read out the field strength.

Efforts are ongoing to improve the sensitivity of such microwave and terahertz Rydberg-based sensors. There are also other real-life measurement scenarios that require a somewhat different approach. For example, if the wavelength of radiation is smaller than the detection medium—as is the case, for example, with terahertz radiation whose wavelengths are in the sub-millimetre domain—the field may be inhomogeneous inside the sensing medium, and we may want to spatially image the

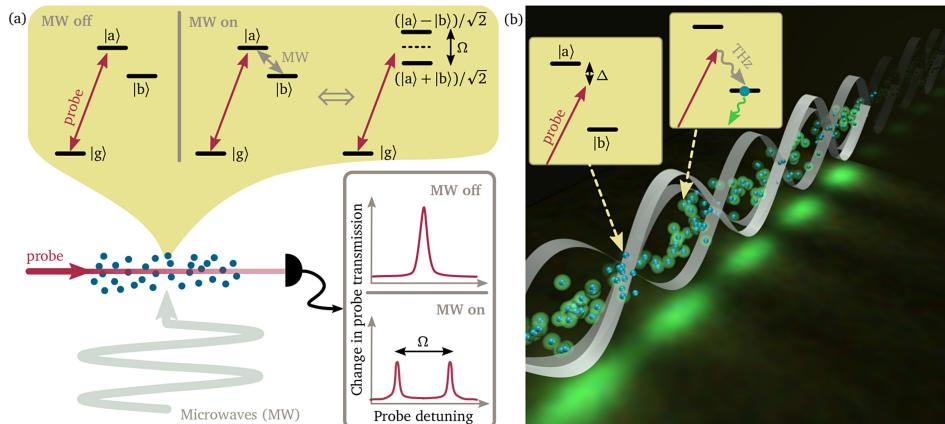


Figure 7. Measuring and imaging microwave and terahertz electromagnetic fields with Rydberg atoms. (a) Detection of microwave field. The probe laser frequency is scanned across the atomic resonance frequency for transition $|g\rangle \leftrightarrow |a\rangle$ in atomic vapour. When the microwave (MW) field is turned on, the transmission peak is split by the amount of the microwave field Rabi frequency Ω that is proportional to the microwave field amplitude. Inset above shows atomic energy levels, where states $|a\rangle$ and $|b\rangle$ are two Rydberg states resonantly coupled by the incoming microwave radiation. (b) Terahertz imaging using Rydberg states. Atoms (blue dots) in the off-resonant probe laser beam can be excited (green spherical clouds) to the Rydberg state only if they are in areas where the field of the terahertz standing wave (grey oscillating strip) is non-zero, as the two-photon resonance for exciting to state $|b\rangle$ is fulfilled (insets). Fluorescence from decay of excited states $|b\rangle$ can be recorded with a camera, uncovering bright and dark spots along the probe laser beam, corresponding to peaks and nodes of the terahertz standing wave. Bottom image is experimentally recorded atomic fluorescence. Data by Christopher Wade (Durham University).

terahertz field using the Rydberg medium. In this case, instead of measuring laser light absorption, we can measure spatially resolved atomic fluorescence in the medium. The low-lying Rydberg states that couple strongly to terahertz radiation decay fast enough to emit visible light from the location where they were first excited. Therefore, fluorescence can be used as a local measure of the population of atoms in the Rydberg state. The only remaining task for spatially resolved imaging of terahertz radiation through fluorescence imaging is to map information about terahertz intensity into the atomic Rydberg state population. Luckily, this can be achieved if terahertz radiation is slightly off-resonant, detuned by energy Δ from resonance for the transition between states $|a\rangle$ and $|b\rangle$. If we also detune the probe laser by the exact same amount Δ , then in regions where no terahertz field is present, atoms cannot be excited to the Rydberg state $|a\rangle$ (node of standing wave in figure 7(b) and corresponding inset). On the other hand, at points of space where there is a terahertz field, since detunings of the terahertz and laser fields match, the resonance condition for a direct transition from ground state $|g\rangle$ to $|b\rangle$ is fulfilled. In this two-photon (Raman) process, one optical and one terahertz photon are simultaneously absorbed, and atoms are directly transferred into state $|b\rangle$, without populating intermediate state $|a\rangle$ in the process. In the subsequent decay of highly-excited state $|b\rangle$, an optical photon is emitted, providing terahertz to optical conversion. Since the rate of this two-photon absorption process is directly proportional to the intensity of the terahertz field and large dipole matrix element between the two Rydberg states (see figure 3), one achieves a linear mapping between the emitted fluorescence and terahertz intensity. Emitted optical photons can be directly imaged with a camera (figure 7(b)), visualizing terahertz intensity in space. Just as in the early 20th Century fluorescent screens were used for detecting electrons, x-rays and gamma radiation, now we can use atoms laser excited into Rydberg states to detect lower energy quanta corresponding to microwave and terahertz radiation.

All the sensing examples we have mentioned so far have been for fields that contain a large number of photons. The last remaining questions regarding sensing applications are, what is the ultimate sensitivity of Rydberg sensors? And can they be used for sensing fields at the single photon level?

The answer is yes. They have been used to detect the presence of single photons, amazingly *without absorbing them in the process*—this is called a quantum non-demolition measurement. For this, we need to introduce a new experimental setup. First of all, we need another container to hold the photon. The photon container is based on superconducting cavities where single microwave photons can bounce back and forth more than a billion times before being lost. The second experimental ingredient is a beam of Rydberg atoms that pass in sequence through the sides of the cavity. The speed of the atoms is selected by various techniques such that they have a small spread in velocities before they enter the cavity. This allows a well-defined interaction time between each of the atoms and the photon field inside the cavity.

So how can these atomic sensors measure the number of photons in the cavity *without* photon absorption? Usually, measurements are done via resonant processes: the photon has an energy that is resonant with a transition between two atomic states. In this case, the presence of the field is recorded by counting the number of

excited atoms. If atoms are prepared in the Rydberg state $|a\rangle$, but are later detected in state $|b\rangle$, we know we had a photon in the cavity, but we also know that we destroyed (demolished) that photon by absorbing it in a process $|a\rangle \rightarrow |b\rangle$. Off-resonant measurements, however, offer the opportunity to conduct non-destructive measurements. The idea is that the process of absorption of an off-resonant photon is energetically forbidden. However, although this first-order process does not happen, the second-order process, where a photon is absorbed only for a short time—shorter than the Heisenberg time–energy uncertainty relation—and then emitted again, can still happen. The consequence of this process is that atomic state $|a\rangle$, due to this off-resonant coupling to state $|b\rangle$, will have slightly different energy δE . That means that if the photon is in the cavity, and the atom passes in state $|a\rangle$ through the cavity during time τ , the state will acquire an additional phase of $\delta E \cdot \tau$ relative to the case when there is no photon in the cavity. To measure this phase, an interferometer is constructed from internal atomic states. Atoms are initially prepared into a superposition of $|a\rangle$ and $|b\rangle$ via a precisely timed microwave pulse that couples them. Then they pass through the cavity where the two states acquire different phase shifts depending on whether the photon is in the cavity. Finally, the two states are again recombined, and the population of one state is measured to determine the accumulated phase shift. Continuous measurement of this phase shift can track photon number in the microwave cavity, without destruction of the measured photons.

There are several crucial features of Rydberg states that allow experiments like this. First the preparation of a superposition state relies crucially on the fact that transitions between Rydberg states are in the microwave domain, where precisely timed microwave sources—coherent sources of electromagnetic radiation—can be used for manipulation of the atomic population. Microwave sources are similar to lasers in the optical domain, but their stability—both in frequency and power—is usually better. To achieve significant phase shift during the time the atom flies through the cavity, large dipole matrix elements of the Rydberg states are crucial. Second, to allow enough time for preparation, sensing and detection of coherent recombinations of the atomic Rydberg states, long Rydberg lifetimes are also essential.

The above described approach combines perfect control between individual quanta of light and quanta of matter to non-destructively measure the smallest units of electromagnetic radiation (photons), without destroying them. For this and related work with Rydberg atoms, Serge Haroche and David J Wineland were jointly awarded the 2012 Nobel Prize in Physics ‘for ground-breaking experimental methods that enable measuring and manipulation of individual quantum systems’.

Yet, the ultimate sensitivity does not stop there. Thanks to quantum mechanics, there is one additional trick that researchers can use. To understand this, let us first consider the sensitivity of the above-described interferometer, through which individual probe (sensing) atoms travel; see figure 8(a). Atoms are prepared in such a way that if they are in state $|a\rangle$ they will feel some perturbing field that induces a phase shift ϕ relative to the atoms in state $|b\rangle$ that do not experience that perturbing field. At the exit of the interferometer, two states are recombined and we

make a projective measurement between the two possible states $|a\rangle$ and $|b\rangle$. The binary outcome means that we need to perform many measurements in order to determine the phase shift ϕ . The number of counts at one output of the interferometer is proportional to $q = \cos^2(\phi/2)$, and therefore the sensitivity to small shifts in phase $\delta\phi$ is $\delta q = |\partial q/\partial\phi|\delta\phi = |\cos(\phi/2)\sin(\phi/2)|\delta\phi \simeq \delta\phi/2$, where initial ϕ is selected to maximize sensitivity. Upon repeating the measurement N times, by passing single atoms through such an interferometer each time, the error of our phase shift estimate scales as $1/\sqrt{N}$.

Now let us consider the improvement in sensitivity that can be obtained by using entangled quantum states for probing, i.e., performing *quantum metrology*. Now we pass all N spins at once, but crucially instead of the first beam splitter we have a special engineering device through which atoms are prepared in a highly-correlated state, such that either all N of them are in the upper arm of the interferometer, or all of them are in the lower arm; that is, the probing state $|a\dots a\rangle + |b\dots b\rangle$; see figure 8(b). In the interferometer, each atom in state $|a\rangle$ acquires phase shift ϕ , achieving a total phase shift for the $|a\dots a\rangle$ state relative to $|b\dots b\rangle$ of $N \times \phi$. The probability of detecting something at the output of the interferometer is now $q = \cos^2(N \times \phi/2)$, and we see that the sensitivity to phase shifts $\delta\phi$ scales as $\delta q = |\partial q/\partial\phi|\delta\phi = N|\cos(N\phi/2)\sin(N\phi/2)|\delta\phi \simeq N \times \delta\phi/2$. This overall scaling of $1/N$ with the number of entangled measurements enhances sensitivity of quantum measurements by a factor of \sqrt{N} relative to classical (unentangled) measurements.

The entangled states described above that are so useful for quantum metrology are called *Schrödinger cat states*. Analogous states have been achieved within a single atom, again thanks to the exquisite coherent microwave control of the atomic populations possible among the highly-excited Rydberg states. These Schrödinger cat states have been used to improve ultimate sensitivity for weak static electric field measurements (see Additional resources for Rydberg sensors at the end of this book).

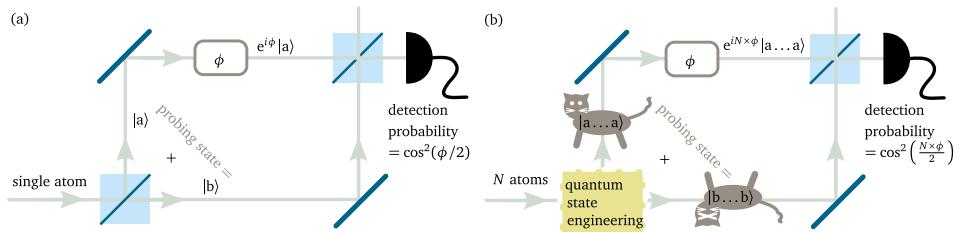


Figure 8. Detecting phase with single atoms and entangled states for improved sensitivity. (a) Ramsey interferometer setup used to measure the relative phase that atoms in state $|a\rangle$ acquire, for example, due to an off-resonant coupling to the electromagnetic field (AC Stark shift). One can use a microwave pulse to create an initial superposition between states $|a\rangle$ and $|b\rangle$ at the interferometer input, and a second pulse to recombine the states at the interferometer exit. As in a light interferometer, the output state is sensitive to the relative phase accumulated by each part of the superposition. Note that in practice, if only atomic state $|a\rangle$ is sensitive to the field inducing phase shift ϕ , there is no need to spatially separate atoms in state $|a\rangle$ and state $|b\rangle$. (b) With N atoms, it is possible to engineer an entangled state, called a Schrödinger cat state, that achieves \sqrt{N} enhancement in sensitivity compared to using the measurements with setup (a) repeated N times.

Rydberg state-based sensors are continuing to be improved and developed, combining different technological approaches with fundamentally new ideas from quantum metrology.

Rydberg quantum optics

In general, optical media like glass have a linear response to light, meaning that optical properties do not depend on whether we send through one, two or even dozens of photons at the same time. This is because a single optical photon somewhere in the medium does not induce a significant change in the energy levels of nearby atoms, leaving other atoms free to respond as if there were no other photons present. Without interactions between atoms to transfer information, they simply do not have a clue that there is another photon inside the medium. This linear response of most materials to light is something that is exploited, for example, in long distance optical communications.

However, for some applications we would like to directly control one photon with another photon. For this, we need a medium that is nonlinear at the level of two photons. For example, we might imagine an all-optical transistor, where the presence of a single photon blocks the transmission of other incoming photons. Additionally, one can encode some information into the states of the photons; for example, by using polarization states of the photon or their relative phase. Photons can be in superposition states too, realizing in this way quantum carriers of bits of information, or qubits. If one manages to realize transmission switching of the second photon depending on the presence or absence of the initial photon, *without leaving information about the photon state anywhere other than maybe the two photons themselves*, then photon states will not be projected (measured) in the process. Photons can then maintain their quantum superposition of states, allowing such a transistor to be used in quantum logic gates. However, such a transistor operation, dependent on photon number, is strongly nonlinear and requires manipulation on the level of single quanta of light.

One way a nonlinear response of the medium can arise is when two photons try to interact with the same atom. But as the scattering cross section of one atom is smaller than a tightly focused laser beam (limited by diffraction), the probability that two photons will interact with the same atom is relatively small. Practically, this problem has been addressed using mirrors in the form of an optical cavity to keep reflecting the same photon back onto the same atom. This field is known as cavity quantum electrodynamics (cQED). However, the small cavity volume required to increase the probability of interaction brings new technical challenges.

These constraints are drastically changed if one considers atoms promoted to Rydberg states. As we have seen in the Background section, a single photon that excites an atom into a Rydberg state will interact and significantly shift the Rydberg energy levels of all nearby atoms. The range of this interaction spreads over a volume with size given by the blockade radius and can be up to $(10 \mu\text{m})^3$. This means that to achieve strong nonlinearities on a single photon level, one only need to ensure that the photon interacts with any atom within the blockade radius. The more atoms

contained in this blockade radius, the stronger the nonlinearity. This *nonlocal* impact of Rydberg level excitations is the main reason why there have been many proposals and demonstrations of Rydberg-based quantum optics.

For Rydberg nonlinear optics to work, we would like to couple optical fields to Rydberg states, where we can exploit long-range Rydberg interactions, mediated by microwave and terahertz photons, to achieve effective interactions between optical photons. However, Rydberg states have high energies, and to excite them directly from the ground state often requires ultraviolet frequencies, as discussed above. If we want to manipulate visible and near-infrared frequencies, we need to use a multi-step excitation scheme, such as the one shown in figure 9. Here, a coherent optical field (laser) couples ground state $|g\rangle$ to some intermediate short-lived state $|m\rangle$ with coupling Rabi frequency Ω_p . The intermediate state is in turn coupled to Rydberg high-lying energy level $|r\rangle$ with Rabi coupling frequency Ω_c . In other words, the Hamiltonian \mathcal{H} describing the dynamics of such a three-level system and resonant driving fields is

$$\mathcal{H} = \Omega_p|g\rangle\langle m| + \Omega_c|m\rangle\langle r| + \text{H. c.},$$

where H.c. marks Hermitian conjugate. One might expect that in such a resonantly driven system something will always end up in this intermediate, short-lived state $|m\rangle$. Interestingly, this is not always the case, and the reason lies in an interesting case

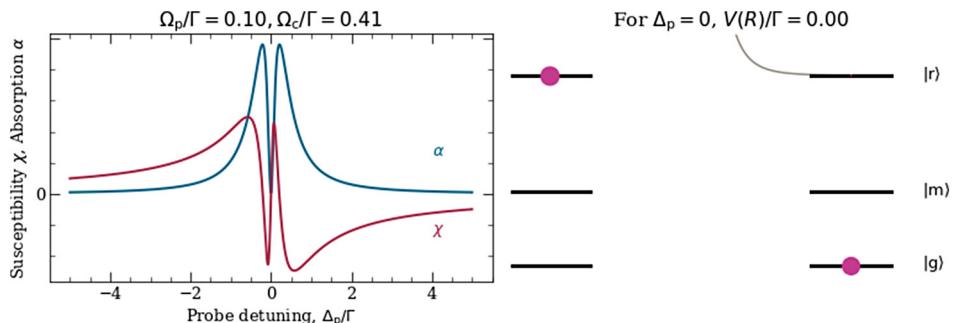


Figure 9. EIT in the presence of Rydberg excitation. When an atom is far away from an existing Rydberg excitation, there is a clear transparency widow for probe light opened by the control laser beam. Relative populations of different atomic states are represented by the size of the circles for each of the states. The population of ground and Rydberg states in this regime can be changed by changing the ratio of control beam Rabi frequency Ω_c to that of the probe coupling frequency Ω_p . Reducing the driving control beam makes the EIT window narrower. At the same time dispersion, as seen from the fast change in (the real part of) the susceptibility χ , becomes stronger, leading to group velocity slowing down for light pulses propagating through the medium. The atomic medium is now excited, with the atom population predominantly residing in the Rydberg state—but, importantly, without any contribution from the short-lived intermediate state. If an atom is now brought closer to the existing Rydberg atom in the medium, an interaction-induced level shift between Rydberg states effectively detunes the coupling laser away from the transition. The position of the transparency window and its dispersion (refractive index) are thus modified. If atoms are very close to each other, the transition to the Rydberg state is effectively decoupled from the control laser. The remaining two-level system absorbs probe light photons, and transfers the photons into a short-lived intermediate state. This state decays on short time scales, causing dissipative loss. This figure is interactive in the online version, and available to download from <http://iopscience.iop.org/book/978-0-7503-1635-4>.

of destructive interference. To see this, consider an atom in a state which up to normalization factor is given by $|\psi\rangle \propto \Omega_c|g\rangle - \Omega_p|r\rangle$. If we next look at what happens in time $d|\psi\rangle/dt = \mathcal{H}|\psi\rangle$, we find that the ground- and excited-state contributions to the middle state probability amplitude vary as $\Omega_p\Omega_c$ and $-\Omega_c\Omega_p$, respectively. They have exactly equal magnitudes but opposite phase, resulting in complete destructive interference ($\mathcal{H}|\psi\rangle = 0$) for excitation of the middle state $|m\rangle$. This is completely analogous to the formation of dark fringes in the double slit interference experiment, where light coming from two slits contributes two fields of equal electric field amplitude but opposite phase. This destructive interference is seen in figure 9 for probe and control fields on resonance ($\Delta_p = 0$, $\Delta_c = 0$) as a dip in probe absorption, which is known as *electromagnetically induced transparency* (EIT).

The transparency created in this medium, however, does not convert the medium into just a clear, transparent window. To understand why, let us imagine a pulse of probe light, consisting of a single photon travelling through this medium, while the continuous laser beam retains the wide transparency window, maintaining Rabi driving frequency Ω_c . A single photon has low intensity ($\Omega_p/\Omega_c \rightarrow 0$) and propagates through the transparency window whose width is given by Ω_c (remember, the spectral line splitting from the Rydberg sensors section). In this scenario, our state $|\psi\rangle$ consists of only ground-state atoms $|g\rangle$. Now let us see what happens if we reduce the intensity of the second laser beam. From the susceptibility plot in figure 9, we see that in the vicinity of the transparency peak, the phase velocity changes rapidly. As we reduce Ω_c , making the transparency window narrower, we increase the slope of this refractive index change with detuning. A light pulse is a superposition of many closely spaced frequencies (given by the Fourier transform), and a large slope in the frequency dependence of the refractive index means the pulse propagates slowly, i.e. has a small group velocity—a phenomenon known as slow light. But what about the atomic state $|\psi\rangle$ during pulse propagation? As Ω_c is reduced, the atomic wave function $|\psi\rangle$ becomes less like the state $|g\rangle$, and acquires a significant component of the Rydberg state $|r\rangle$. The energy to excite $|r\rangle$ is provided by a combination of the probe photon and another photon from the laser beam-driving second transition with Rabi frequency Ω_c . But wait, you might say, does that mean that our probe photon is absorbed after all? The answer is *both* yes and no. The probe photon is coherently coupled to the medium but still continues to propagate. A consequence of this coupling is that there is a probability amplitude that some atom is excited into $|r\rangle$, but there remains an electromagnetic component. The photon becomes a superposition of electromagnetic field and atomic excitation.

Whereas the energy of a photon in free space is contained only in the electromagnetic field, within a medium the energy is shared between the electromagnetic field and an atomic excitation wave that overlaps with the photonic component. This second wave contains the probability that a single atom is excited to the Rydberg state $|r\rangle$. Excitation energy is transferred as a coherent superposition of these two waves, a so-called *polariton*. The weaker the second laser driving field (smaller Ω_c), the narrower the transparency window becomes, and the slower the polariton propagates (it propagates with group velocity $\propto \partial n/\partial \omega$). At the same time, the probability amplitude for the excitation to be in the atomic medium becomes larger.

Indeed, if we gradually turn off the control field ($\Omega_c \rightarrow 0$), the polariton will become just a stationary excitation of a single Rydberg atom delocalized within the medium, with no field in the optical channel. If we then slowly turn the control field back on, the process is reversed—the polariton continuously evolves from a purely atomic excitation field to a purely electromagnetic field, increasing speed in the process. In the end, the photon (since in this extreme limit case where $\Omega_c \gg \Omega_p$, the polariton consists of the light field only) will shoot out of the medium at the full speed of light.

This mechanism provides controllable coherent coupling of the single quanta of the light field (a photon) into Rydberg excitations and back. What about two photons? Here, Rydberg state properties come into play, creating a strong non-linearity at the single photon level. If we try to pass a second photon inside a medium where the first photon has created a Rydberg excitation, then we can get absorption (or scattering). This is because if the photon is interacting with atoms close to the Rydberg excitation—closer than the blockade radius—the Rydberg energy level will be shifted completely out of resonance with the control field that couples the unperturbed $|m\rangle \longleftrightarrow |r\rangle$ transition. That means that photons will interact with a simple two-level system, being absorbed in the process $|g\rangle \rightarrow |m\rangle$, and scattered into the environment once the short-lived intermediate $|m\rangle$ state decays; see figure 9 for small interatomic separations. This *dissipative* mechanism can be exploited to realize a single photon transistor.

If the medium is larger than the single blockade sphere, such that we can have two optical excitations propagating through it, there is another interesting effect. Due to the Rydberg character of these excitations in the medium, if two polaritons are close to each other, but further apart than the usual blockade sphere, they can experience small energy level shifts of the Rydberg state. At these intermediate distances, the level shift is not big enough to completely decouple the control field Ω_c from resonance, but the slight energy change shifts the transparency window and changes the refractive index; see figure 9. This distance-dependent change of refractive index acts as an effective potential between the two photons, causing an attractive interaction (see Additional resources).

New schemes for controlling light at the single photon level, and maintaining its coherent nature, continue to be developed. The challenge is to further increase the probability of photon interactions, allowing for deterministic control, whilst maintaining coherent superpositions of photons or atoms in the medium. This will allow the creation of new states of light fields, previously unobtainable, and promises all-optical computation methods.

Although the problem of weak absorption of a single photon by single atoms was solved using the Rydberg blockade, the fidelity of the process depends on the number of atoms in the blockade volume, or equivalently on optical density within the blockade radius of the medium. However, one cannot simply put as many atoms in this volume as one may wish. The limit is not experimental but fundamental. Namely, there is an interaction between the large electronic wave function and ground-state atoms that sit inside the highly-delocalized Rydberg electron. It is even possible to observe bound states of Rydberg and ground-state atoms due to this

interaction, forming new types of molecules. For quantum optics, these molecular resonances cause broadening and disable excitation of purely atomic resonances, leading to additional dephasing effects that wash away phase information important for quantum superposition states. Possible resolutions can come from protocols that carefully arrange the atoms in space or from a combination of optical cavities and Rydberg atom ensembles, which would effectively increase the photon interaction time with atoms, allowing increased absorption probability with more dilute atomic clouds. An alternative direction is to search for new interaction protocols to realize strong photon–photon interactions that do not rely on initial photon absorption occurring necessarily within one blockade volume.

Rydberg quantum simulators

Many exotic yet technologically relevant materials such as superconductors have properties that depend crucially on quantum mechanics. These materials pose very difficult questions—questions about their ground states, phase diagrams and dynamics. For example, how do they reach thermal equilibrium and what is their behaviour far from equilibrium? How are energy and information transported from one place to another? Answering these questions is likely to expand the range and scope of these materials’ applications, and allow us to engineer completely new quantum states of matter, whose uses we can only begin to guess. The reason why answers to these questions are lurking in darkness is the very high wall of complexity associated with quantum systems. We have been unable to find a simplified description of these quantum many-body systems, and consequently are unable to simulate them starting from only basic knowledge of their interactions and states. The problem is that for any quantum mechanical system, the number of states scales ridiculously fast with the number of constituents. For a system containing N spin 1/2 particles, there is 2^N possible quantum superpositions, and for each one we have to save a complex number in order to track the general state of the system. Even for small system sizes ($N \gtrsim 50$), memory requirements would quickly grow beyond what one can hope to achieve in any classical computer. Memory requirements are therefore one factor contributing to the height of the computational complexity wall. The other factor is complexity of the actual calculations, i.e., algorithmic complexity, which can be extreme even for solving completely classical problems. Despite these challenges, researchers persist—classical problems solved with quantum hardware offer faster algorithms than those executable on classical machines, as discussed in *How to Build a Quantum Computer* by Barry C Sanders (Physics World Discovery series).

One avenue researchers are exploring looks to nature for answers. The dynamics of natural systems containing more than 50 spins is evaluated all the time without too much effort, simply by following the quantum laws. In one view, any system in nature simulates *itself* following physical (quantum) laws. However, this is not a helpful observation when confronted with a system that puzzles us, because typically we cannot readily observe and record the system dynamics, nor control underlying parameters. However, precise control and observational capabilities are needed in

order to try to exploit a simplified effective simulator to model the dynamics of real-world systems. The hope is that one can find a simulator that mathematically has all the same interactions and elementary states as the real-world system we want to explore, except—and this is the main trick—that the simulator can be scaled to much larger length scales. Thanks to that scaling, instead of having to think about electrons in solids, which we cannot directly measure, we can have atoms on optically resolvable distances, for example, emulating electron states and their quantum dynamics. These are then quantum analogue simulators, whose individual constituents—given, for example, as qubits encoded in atomic states held at $\sim 1 \mu\text{m}$ or larger distances—can be directly measured and individually adjusted through strongly focused optical fields, for example (for more details about different possible routes in, see *Quantum Simulations* by Chad Orzel (Physics World Discovery Series)). Importantly, such quantum simulators can provide valuable answers about system behaviour even before quantum manipulation with very low errors, required for full general quantum computers, is technologically realized.

Using ground-state atoms as basic building blocks for quantum simulation is possible by exploiting direct density-density, i.e., contact, interactions. However, Rydberg states of atoms bring new opportunities. The interaction is both switchable and tunable. Depending on the exact Rydberg state selected, and external fields, it is possible to achieve strong interactions even up to distance scales of order $\sim 10 \mu\text{m}$. Also the distance R dependence of the interaction strength V is tunable between off-resonant $V \propto R^{-6}$ van der Waals interactions and resonant dipole-dipole interactions $V \propto R^{-3}$. The latter are especially important as they are long-range interactions in three dimensions, which qualitatively change the phase diagrams of such systems. Depending on laser detuning with respective Rydberg levels, the dynamics can exhibit either Rydberg excitation blockade or facilitation, or manifest itself as an effective soft-core potential between the *ground-state* atoms, as discussed earlier (see figure 5).

Typically, for a simulation of material properties one would like to start with large arrays of individually addressable and measurable atoms. Imaging of individual atoms kept at distances $\gtrsim 1 \mu\text{m}$, and their addressing with optical beams, is possible with state-of-the-art optics. However, making arrays of sites that will trap only single atoms, and making sure they are all filled has been a significant challenge. Recently, two alternative paths have emerged for making such unity filled arrays: one exploiting a quantum phase transition and the other using real-time adaptive control of optical trapping potentials. Both techniques start with *laser cooling*.

Atoms originating from a heated atomic source, from which they fly away at speeds comparable to the speed of sound, are first slowed down using lasers. By adjusting their internal energy level shift with external fields, and using the fact that moving atoms see the Doppler shift of the driving laser frequency, faster atoms are forced to absorb and scatter more photons. Scattering through atomic decay happens in random directions, while absorbed photons always propagate in the opposite direction of the atomic velocity. In this way, each photon absorption event kicks the atom a bit, slowing it down and allowing it to be trapped. This method earned Steven Chu, Claude Cohen-Tannoudji and William D Phillips the 1997

Nobel Prize for Physics for the ‘development of methods to cool and trap atoms with laser light’.

Next, the two techniques for making unity filled arrays diverge. In the first method, atoms are cooled down further by letting the fastest atoms leave the trap, while the rest thermalize to lower temperature, just as happens with molecules of water in a mug of coffee as water evaporates. As temperature T goes down with reduced average atomic momentum p , the atomic wave function size λ describing atom positions in space becomes very delocalized (‘fuzzy’), $\lambda = \hbar c/p$, where the momentum $p \propto \sqrt{T}$, comparable with the atomic cloud size. If the cooled atoms have integer spin, i.e., they are bosons, at some point the available states for the low-temperature system become so densely populated that the fundamental bosonic indistinguishability of individual atoms will start playing an important role, forcing all the atoms to cram together into a single energy level, lowest in energy. This is how a *Bose–Einstein condensate* of atoms is created—for which Eric A Cornell, Wolfgang Ketterle and Carl E Wieman were awarded the 2001 Nobel Prize for Physics for ‘the achievement of Bose–Einstein condensation in dilute gases of alkali atoms, and for early fundamental studies of the properties of the condensates’—where all the atoms share the same Schrödinger wave function, delocalized over the whole atomic sample. In the final step, the power of standing wave laser beams is ramped up. The frequency of the optical field is kept too low (known as red detuning) to cause atom excitation. Yet, while this far detuned off-resonant driving is unlikely to be absorbed, it still reduces the ground-state atoms’ energy levels via the second-order AC Stark shift. This attracts the atoms into the brightest spots in the red detuned laser beam, condensing the wave function into the lattice. Initially, all the atoms still share the same wave function, being delocalized over all the lattice sites. However, as the laser beam power is ramped up, tunnelling between sites becomes increasingly weaker and at a certain point a phase transition happens: atoms become localized in individual lattice sites. As this new state—called the *Mott insulator*—is created from the lowest-energy state of the Bose–Einstein condensate, the lowest-energy Mott insulator state is created. For an equal number of lattice sites and atoms, it consists of exactly one atom per lattice site.

A unity filled atom lattice prepared in this way has a typical interatomic separation of ~ 0.5 μm , given by the period of the optical standing wave that produces lattice potential. Remarkably, all the atoms are in the absolute ground states of their respective lattice sites, having their location described by the Schrödinger wave function for the ground state. However, this method has somewhat limited lattice geometry tunability, and is also limited to relatively small sample sizes. It also requires a quite complex preparation procedure that is both technologically challenging and time-consuming for every prepared lattice. Given that one often wants to accumulate many realizations to calculate statistical properties of the samples, the long experimental acquisition times are a significant disadvantage. More recently, an alternative approach has emerged.

In the second approach, atoms are also initially laser cooled, but then they are trapped inside individual strongly focused laser beams. Far off-resonant (relative to

the ground state–excited state transition) red detuned optical fields again make an attractive potential for atoms. These strongly focused beams are called *optical tweezers*. Arrays of tweezers, with programmable geometry and interatomic distances ranging from $\sim 3 \mu\text{m}$ onward, can be formed from a single focused laser beam, by imprinting a special mask to the beam that changes the relative phase of different points along the laser beam profile generating a hologram. This is done with programmable mirrors, consisting of tens of thousands of pixels, where light reflecting from different pixels can have controllable relative phase. Focusing such a programmable hologram creates an array of strongly focused beams in the focal plane, each one realizing a single-atom optical tweezer.

These tweezer traps are loaded with atoms from the laser-cooled background atomic gas. Importantly, for small trapping volumes only single atoms in traps can stay for long periods of time, as atom pairs are quickly lost. This is due to so-called *light-assisted collisions*. Here, one of the atoms is excited by the absorbing trapping light, and then speeds up toward the other atom, as they both feel the same strong attractive interatomic potential. This potential is similar to the pair-state potentials calculated for Rydberg states (see figure 5, for example), but occurs at much shorter distances of $\lesssim 1 \mu\text{m}$ for the low excited–ground state atomic pair-states. Under the action of this potential atoms accelerate, gaining energy, until spontaneous emission brings both atoms back to the ground state. If the atoms gain more energy than the depth of the trapping optical potential of the tweezers, set by the intensity of the optical trapping field, then these atom pairs will be quickly lost. In this way, all atom pairs are removed, leaving only the singly occupied tweezers. The net result is that there is a probability of 0.5 that any site is filled with a single atom.

In this way, lattices with arbitrary trapping site geometries are created. However, they are filled stochastically, leaving typically half of them in an unfilled state. For experiments running a pre-programmed sequence of actions, repeating the sequence many times in order to accumulate statistics, this represents a problem since the researchers will have to average over initial atom distributions, preventing simulation of situations where unity filled spin systems are required. This problem can be resolved by inserting an additional preparation step that will depend on the *current stochastic realization* of half-filling of each given lattice. Namely, if the half-filled lattice is imaged, it is possible to analyse this image, find filled sites and plan a sequence of moves by an additional fast optical tweezer beam that will redistribute the atoms one by one, creating a region with a unity filled lattice. This process is demonstrated in figure 10 (video). It ensures that each experimental realization has the same completely filled initial lattice.

Atom-based quantum simulators using Rydberg states have already been used in a number of pioneering experiments. Slow change of experimental parameters was used to adiabatically and deterministically prepare many-body states and correlations in the system. Energy transport dynamics has been observed between Rydberg states, and even the interactions between atoms that are only off-resonantly coupled to Rydberg states (Rydberg dressing) have been measured. Ongoing efforts are expanding the detectability of various states, and improving the fidelity of atom state control, allowing more complex simulations on larger lattices. On the theoretical

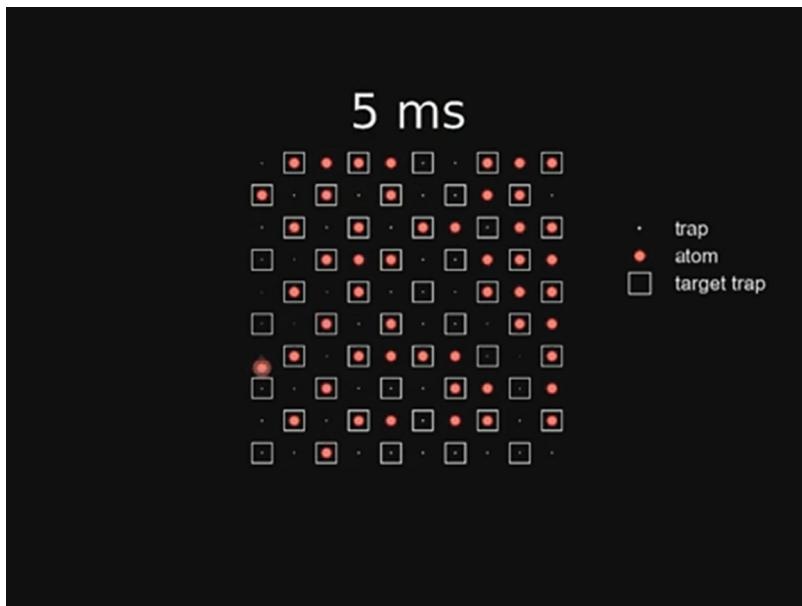


Figure 10. A fast optical tweezers sorts atoms from an initially randomly filled array of single-atom trapping sites into a uniformly filled array, initializing a Rydberg atomic system for quantum simulation. Optical imaging is used to detect which lattice sites are initially filled, and this information is then used in real time to move the atom using a strongly focused laser beam before the atom is lost. A uniformly filled array with a controlled geometry is a good starting point for quantum simulation. Reproduced with permission from Barredo D *et al.* 2016 *Science* **354** 1021. Copyright (2016) AAAS. This figure is interactive in the online version, and available to download from <http://iopscience.iop.org/book/978-0-7503-1635-4>.

side, new schemes for controlling and creating novel many-body states are being developed.

However, there remain factors limiting quantum simulation with Rydberg atoms. For example, blackbody radiation-induced transitions, originating from the finite temperature of the background environment can limit Rydberg-based quantum simulation to short evolution times. Blackbody-induced decays can cause a redistribution of the atomic population among the states in the Rydberg manifold, and cause unwanted optical–microwave two-photon transitions even under off-resonant driving. Each newly populated state combination then has a new interaction potential that can produce additional resonances and processes not accounted for in simplified models. Careful selection of states, smaller atom numbers used for lattices and working with stronger, faster dynamics that allows shorter simulation times, can all mitigate problems of blackbody-induced decays. An alternative experimental approach that is also sometimes used is to shield the atomic sample from the radiation using a cryostat.

In addition to blackbody radiation, another initially unwanted coupling mechanism that has a significant effect on atom dynamics has been coupling of atomic transitions to excitations of various surfaces. Advances are now being made in understanding atom–surface interactions. Understanding interactions of Rydberg

states close to surfaces offers potential for combining Rydberg atoms with other quantum dynamics platforms—from spins in solid-state devices to superconducting circuits. In particular, coupling of Rydberg atoms to microwave superconducting circuits can provide an important interface for information, as Rydberg atoms can capture optical information and feed it through microwave transitions between Rydberg levels to superconducting circuits. The latter is already being successfully used for elementary quantum computing logic gates. All of this requires electromagnetic fields that are confined to planar cavities and waveguides. Such engineering of electromagnetic field modes mediating interactions can also be used in advanced atom quantum simulators of the future by tuning inter-spin interactions.

4 Outlook

The field of Rydberg physics continues to advance rapidly, via experimental progress and new theoretical ideas feeding off one another. Although some limiting factors are often encountered such as blackbody radiation, new technological solutions are always lurking on the horizon, typically using new state control protocols and careful state selection.

In our timeline (figure 2), we have seen how new technologies have been crucial to advancing Rydberg physics. Without microwave technology, dye lasers and later semiconductor lasers, many achievements would not have been possible, or would have been significantly more complicated. These technological advances are continuing apace, allowing the three-dimensional positioning of individually addressed Rydberg atoms, for example. On the hardware side, a new ‘technology’ that is on the horizon is Rydberg excitons—bound electron–hole pairs—in semiconductors. While sharing the amazing optical properties of Rydberg states of atoms and molecules, as illustrated in figure 1, their solid-state nature would naturally lend them to much easier and faster technological exploitation. Currently, clearer absorption and transmission switching and narrower linewidths are still required for applications, but there is an ongoing research effort in improving samples and designing new optical interaction schemes for these materials.

But we don’t want to get lost in promising applications, of which there are many, that could potentially bring to the world many new ‘quantum gadgets’. Instead, let’s go back to why Rydberg physics is so interesting for physicists today. The fact is that we can now combine the control of both single atoms and single photons—this is amazing! Take a moment to contemplate and enjoy it. These feats would have been thought impossible several decades ago. For the founders of quantum mechanics at the beginning of the 20th century they were merely thought experiments, yet the reason they chose these elementary building blocks, atoms and photons, in their thought experiments, brings to light more food for thought: their simplicity beautifully expresses the fundamental nature of quantum laws. It is no accident that efforts to understand simple atomic spectra, pioneered by Ångström, Balmer and Rydberg, first uncovered the quantum nature of the world. And no accident that through further contemplation of these laws, experimental physicists had new dreams for quantum state control, and devised ingenious experimental approaches

to make those dreams come true. It is this possibility to *both* think about *and* control the elementary ingredients of our world where one finds the most scientific joy, with the greatest potential to see the world with new eyes and learn about phenomena in many other fields of physics. Indeed, for the curious physicist, Rydberg physics continues to offer the most exquisite playground to explore the quantum world.

Additional resources

Rydberg sensors

- Sedlacek J A *et al* 2012 *Nature Phys.* **8** 819 explains the measurement of microwave electric fields using Autler–Townes (AC Stark) splitting of Rydberg lines observed in laser transmission.
- Wade C G *et al* 2017 *Nature Photon.* **11** 40 shows how atomic fluorescence from Rydberg states can be used for imaging the intensity profile of terahertz radiation.
- Nogues G *et al* 1999 *Nature* **400** 239 and Gleyzes S *et al* 2007 *Nature* **446** 297 describe quantum non-demolition measurements of microwave photons in a cavity using Rydberg atoms.
- Facon A *et al* 2016 *Nature* **535** 262 describes how a Rydberg atom in a Schrödinger-cat state can be used for precision measurement of static electric fields. This is an example of quantum enhanced metrology, where entanglement of the probing state allows an improvement in sensitivity inaccessible to classical probes.

Rydberg quantum optics

- Pritchard D *et al* 2010 *Phys. Rev. Lett.* **105** 193603 describes the first experimental demonstration of giant Rydberg interaction-induced optical nonlinearity via electromagnetically induced transparency.
- Firstenberg O *et al* 2013 *Nature* **502** 71 shows how dispersive nonlinearity originating from shifts in the electromagnetically induced transparency can induce an effective attractive interaction between photons.
- Tiarks D *et al* 2016 *Sci. Adv.* **2** e160003 shows how dispersive interactions between the single stored Rydberg excitation and propagating slow light pulse can imprint a π phase shift on the propagating light pulse. This is also an important step towards all-optical deterministic gates for optical quantum computing.
- Gorniaczyk H *et al* 2014 *Phys. Rev. Lett.* **113** 53601 and Tiarks D *et al* 2014 *Phys. Rev. Lett.* **113** 53602 show how a single stored photon can induce dissipative interactions in the propagating slow light, realizing a classical optical transistor switched by a single photon.
- Thompson J D *et al* 2017 *Nature* **542** 206 shows an example of realized strong photon–photon interactions that go beyond the idea of the Rydberg blockade, exploiting strong resonant dipole–dipole interactions between a stored Rydberg state excitation and slowly propagating Rydberg polariton.

Rydberg quantum simulators

- Schauß P *et al* 2015 *Science* **347** 1455 shows an example of many-body state preparation through a slow (adiabatic) change of driving parameters. As a consequence of Rydberg interactions, crystalline ground states consisting of a different number of Rydberg excitations can be prepared. An initial unity filled lattice for quantum simulation is prepared through the Bose–Einstein condensate-to-Mott insulator transition.
- Zeiher J *et al* 2016 *Nature Phys.* **12** 1095 shows a demonstration of Rydberg dressing-induced interactions between the ground-state atoms. An initial unity filled lattice for quantum simulation is prepared through the Bose–Einstein condensate-to-Mott insulator transition.
- Labuhn H *et al* 2016 *Nature* **534** 667 shows a realization of two-dimensional Ising models with Rydberg atoms. Filling fraction was not controlled, but was measured, and the experimental sequence would be triggered once the requested filling was achieved stochastically. The problem of stochastic filling of larger arrays was resolved by the same authors with fast real-time reassembly of atoms in an array (see next reference).
- Endres M *et al* 2016 *Science* **354** 1024, Barredo D *et al* 2016 *Science* **354** 1021 and Barredo D *et al* 2018 *Nature* **561** 79 show methods for real-time experimental control that allow assembly of arbitrary one-, two- and three-dimensional unity filled arrays (respectively) from initially stochastically filled single-atom traps.
- Bernien H *et al* 2017 *Nature* **551** 579 shows an example of a quantum Ising model realized in a one-dimensional atomic chain with Rydberg atoms. An initial unity filled lattice for quantum simulation is prepared with atom sorting.

Open source libraries and programs for calculating Rydberg state properties

- Šibalić N *et al*. 2017 *Comput. Phys. Commun.* **220** 319—ARC (Alkali Rydberg Calculator) is a package of routines written in Python, using object-oriented programming to make a modular, reusable and extendable collection of routines and data for performing useful calculations of single- and two-atom properties, like level diagrams, interactions and transition strengths for alkali metal atoms. A preview of basic functionality is available online at <https://atomcalc.jqc.org.uk>
- Weber S *et al* 2017 *J. Phys. B: At. Mol. Opt. Phys.* **50** 133001—Pair Interaction software calculates pair potentials for Rydberg atoms in arbitrary external fields. For more details see <https://pairinteraction.github.io/>