

Genetic algorithm driven optimization of a guided mode resonator coupled to an infrared nanocrystal film

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ABSTRACT

Nanocrystal (NC)-based devices suffer from a trade-off between absorption, pushing for thicker films, and charge conduction that is only efficient over a few NC sizes. Therefore, the design of detectors based on NCs strongly benefits from the introduction of optical resonators that engineer the light-matter coupling. Traditionally, the design of such a cavity relies on analytic scaling rules. However, the design becomes far more complex when multiple resonances are targeted, requiring alternative design strategies. In this study, we explore how infrared nanocrystals can be coupled to a guided mode resonator to achieve a broadband enhancement of absorption. A genetic algorithm is used to systematically investigate how geometric factors can be tuned to increase the absorption \times bandwidth product. In comparison to the pristine film, a significant increase in absorption was demonstrated across the targeted spectral range (1–2.5 μm). The detector based on the design metasurface exhibited high detectivity (2×10^{10} Jones @ 200 K) and fast time response (18 ns).

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Nanocrystals (NCs) provide a unique platform for optoelectronics by combining broad spectral tunability with charge conduction, enabling electrical addressing of their optical features. However, their polycrystalline nature leads to a hopping conduction process, which is often viewed as a limitation due to moderate carrier mobility and charge diffusion length.¹ This leads to a trade-off when the film is used as an active layer for light sensing. Charge collection is only efficient over a short distance, typically a few NC, while their absorption depth exceeds 1 μm . This results in an overall poor coupling of the light with the NC film, leading to weak absorption for detectors and limited coupling to the far field for light-emitting devices.

To avoid such bottlenecks, the field has been very active in coupling the NC layer with a photonic structure. This ensures that the light-matter interaction is not solely driven by the material itself. Various photonic designs have been tested,² including plasmonic metallic structures,^{3,4} Fabry-Perot cavities,⁵ either at macroscopic scale or subwavelength scale using metal insulator metal cavities^{6–8} and dielectric counterparts based on Bragg mirrors,^{9–11} nanoantennas,^{12,13}

bound states in the continuum resonance,^{14,15} and more exotic concepts^{16,17} inspired by acoustic or millimeter waves. Thanks to the local Kirchhoff's law,^{18,19} which states that emission is proportional to absorption at the same wavelength, this concept can be used to shape either the absorption or emission signal. Significant performance enhancement has been made by coupling the NC layer to a photonic cavity,⁶ resulting in increased absorption and photoluminescence signals,^{11,20} spectral shaping, directivity of light emission,^{9,11,21} and the design of active devices^{1,4,22} with spectra that can be reversibly reconfigured through the application of a bias.

This approach aims to maximize the modulus of the electric field within the NC layer (excluding the contact that does not generate photocurrent or photoluminescence) by utilizing the cavity design. In practice, and for the case of a single resonance, geometrical factors of the cavity are tuned to match its resonance wavelength to the NC cut-off wavelength.

Further scaling of the cavity factor will then depend on the targeted application. For instance, luminescence, lasing, and gas sensing

spectroscopy benefit from strategies based on a high-quality factor cavity, resulting in spectrally narrow features. Conversely, imaging is a broadband application that requires an increase in the system gain \times bandwidth product. Therefore, the electric field modulus should be enhanced over a broad spectral range, which may require combining several resonances. Several examples have already been reported of combining two types of cavities²³ (Fabry–Perot and Plasmon) or using a multiresonant cavity.^{24,25} However, the idea of combining multiple cavities to achieve several resonances quickly encounters fabrication challenges. In solar cells, Chen *et al.*²⁶ propose aggregating up to nine resonances to cover the entire visible range. Although the device achieves strong broadband absorption, the challenges raised by fabrication strongly balance the benefit of the concept. Conversely, a broadening of the linewidth may also be obtained by stacking diodes with various band edges similar to multi-junction solar cells. This however also leads to a more complex device design and may expose the NCs to damaging fabrication steps (annealing²⁷). Using a polydisperse NC film is also likely to broaden the band edge. However, this approach will come at the cost of increased dark current, as it is driven by the narrowest bandgap material.

Therefore, alternative design strategies are necessary for the development of such a broadband detector. Here, we adopt a resonator design strategy based on a genetic algorithm that does not make assumptions about the nature of the injected mode, other than the fabrication constraints that are initially set. Thanks to this approach, we are able to design a structure that maximizes the product of the absorption magnitude by its bandwidth. The nature of the various resonance is then discussed and we finally quantify the performances of a detector based on such resonator.

In this study, our interest had been focused on extended short-wave infrared (SWIR) NCs, with a $2.5\ \mu\text{m}$ cutoff wavelength. Compared to traditional InGaAs alloy active in the SWIR, the NCs lift the constraint of the epitaxial growth, which eases the spectral tunability (i.e., no need to be lattice matched with the substrate) and simplifies the coupling to a readout circuit as in the case of cameras.

This publication focuses on guided mode resonator²⁸ (GMR) geometry, specifically a type of resonator that uses a grating to transform the incident plane wave of light into a propagative mode along the substrate. This approach enables multiple passes of the incident light into the absorbing layer, resulting in increased absorption. The concept has already been applied to nanocrystal-based devices with photoconductive,²⁹ phototransistor,³⁰ and photovoltaic³¹ configurations. This highlights the method's effectiveness in generating devices that absorb most of the incident light.

A way to generate such GMR relies on the structure schematized in Fig. 1(a). A back side mirror is first deposited onto a substrate and coated with a silica spacer. Then a metallic grating is fabricated on top of the silica layer. In its classical form the grating is periodic.^{29–31} It can also serve as interdigitated electrodes, providing a dual role in both enhancing the electromagnetic field and collecting charges. With a periodic structure, the period (p) sets the resonance wavelength (λ_r) based on the equation $p = \lambda_r/n$, where n is the refractive index of the active material. For HgTe NCs with a $2.5\ \mu\text{m}$ cutoff wavelength, the value of n has been determined to be around 2.3 through ellipsometry.³¹ The extinction loss was estimated to be around 0.1 at the exciton, see Fig. S1 and the associated section for the dielectric parameter of each layer.

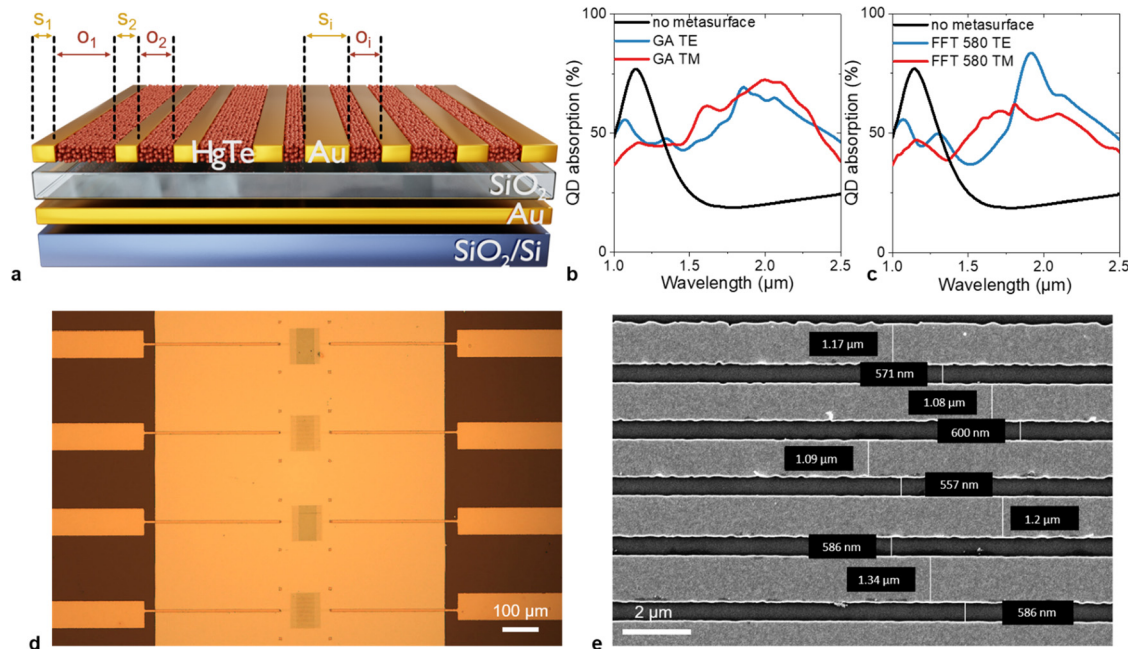


FIG. 1. Guided mode resonator coupled to an infrared NC film. (a) Schematic of the guided mode resonator under optimization through a genetic algorithm. (b) Simulated absorption spectra for the pristine NC film and for the film coupled to the optimized GA structure. (c) Simulated absorption spectra for the pristine NC film and for the film coupled to the optimized GA structure after an FFT filtering and setting all values for $O_1 = 580\ \text{nm}$. (d) Optical microscopy image of the structure (GMR GA FFT580) before NC film deposition. (e) Scanning electron microscopy image of the structure (GMR GA FFT580) before NC film deposition.

A genetic algorithm (GA) optimization is then initiated for this structure, subject to the following constraints: the thickness of the gold backside mirror is set to 80 nm, a value above the metal skin depth, and the bottom spacer is made of 120 nm of SiO₂ to prevent a short circuit between the bottom gold mirror and the top grating. The grating consists of 5 nm of Ti (later modeled as an additional 5 nm of gold) and 80 nm of gold. These values were directly taken from previous works on GMR coupled to NC films.³² However, the size of the digit (S_i) and the opening size between digit (O_i), see Fig. 1(a), are set as free parameters. We initially set a minimum value for O_i above >200 nm to prevent shorts during the e-beam process. The size of the grating array is set to 100 μm to avoid stitching issues, which can be patterned from a single field of view in most standard scanning electron microscopes. The deposition conditions of the NC layer are optimized, including concentration and deposition speed, to achieve a thickness as close as possible to 300 nm.

Each structure is represented by a 1-by- n vector, where $n \leq 250$, that describes all values for O_i and S_i parameters. The generation size is set at 400, with each generation comprising 200 unique structures. In each subsequent generation, 100 structures are generated through mutation [i.e., one or more O_i/S_i parameters are randomly replaced with new value(s)], while the remaining 100 are produced through breeding (i.e., merging a series of S_i/O_i parameters to form a new structure). Mutation occurs at a rate of 10%, meaning that $0.1 \times n$ elements within a structure are randomly altered. Breeding is performed through two-point scattering breeding, where two elements from two structures are randomly selected and exchanged between the structures.

Then for each structure, we evaluate the absorption spectra relative to the structure using FDTD software (Lumerical). This absorption map is then spatially integrated over the volume of the NC film (V_{NC}) only (i.e., excluding the contact that does not generate photocurrent, but only thermal heat),

$$Abs_{\text{NC}}(\omega) = \frac{P_{\text{NC}}(\omega)}{P_{\text{incidence}}} = \frac{\iiint 0.5\omega |F|^2 \epsilon_0 \text{Im}(\epsilon) dV_{\text{NC}}}{P_{\text{incidence}}}. \quad (1)$$

We then extract from the absorption spectra the parameter S , which is the integral of the absorption from 1 to 2.5 μm . The 1 μm limit is set to focus on the infrared spectral response, while the value of 2.5 μm matches the NC cutoff.

$$S = \int_{1\mu\text{m}}^{2.5\mu\text{m}} Abs_{\text{NC}}(\omega). \quad (2)$$

Finally, the S value is normalized by the value obtained for the same stack without the gold stripes (i.e., the NC layer with the same thickness deposited on the gold backside mirror on top of the silica spacer). Figure 1(b) displays the absorption spectra obtained for the reference (i.e., without nanostructure) and for the GA-optimized structure. Even without patterning, the absorption displays a clear resonance, that we can attribute to a vertical Fabry-Pérot (see Fig. S2). However, the resonance appearing at 1.2 μm does not match the band edge. The linewidth of this resonance is typically around 300 nm.

The performance of a given structure is evaluated using the normalized S parameter as a figure of merit. The optimal structure was identified in the 384th generation, as shown in Fig. S3. The associated absorption spectra can be found in Fig. 1(b). The resonance compared

to the pristine film is shifted toward longer wavelengths (i.e., closer to the band edge) and clearly broadened compared to a periodic structure (Fig. S4). The magnification of the absorption occurs over 1500 nm, stressing the benefit of this approach compared to the single resonance method resulting from Fabry-Pérot, for which enhancement occurs only over 300 nm. Additionally, several sub-features are visible, indicating that multiple resonances are involved, contributing to a significant broadening of the overall absorption enhancement. The absorption maps, at the point of maximum absorption, are displayed in Fig. S5. The modes remain close to the expected mode for the periodic structure, although we observe modulations of the field intensity depending on the exact localization within the structure.

Then, the structure was fabricated following the procedure outlined in Fig. S8. After fabrication, the metasurface was coated with HgTe NCs presenting a band edge at around 2.5 μm and grown using the Keuleyan's procedure.³³ However, a limited number of devices were viable due to the presence of shorts. The difficulty comes from the optimization of the electron dose for the lithography procedure. The random organization of large and small openings resulting from the GA method makes this dose optimization challenging. Thus, we set an additional degree of constraint by setting all the openings equal to $O_i = 580$ nm, which corresponds to the average value over the entire structure obtained after the GA procedure. The choice of this value appears noncritical since a 20 nm change in this value barely impacts the absorption spectra (Fig. S6). We further truncate the FFT distribution to a given threshold ($0.25 \mu\text{m}^{-1}$), which results in the structure becoming periodic with a super-period that includes seven digits. The simulated spectra are shown in Fig. 1(c). The next structure, called GMR GA FFT 580, was imaged using optical and electron microscopy as shown in Figs. 1(d) and 1(e), respectively.

The benefits of the metasurface are evident from the absorption measurements taken over and away from the grating using infrared microscopy, as depicted in Fig. 2(a). The first expected result is a clear

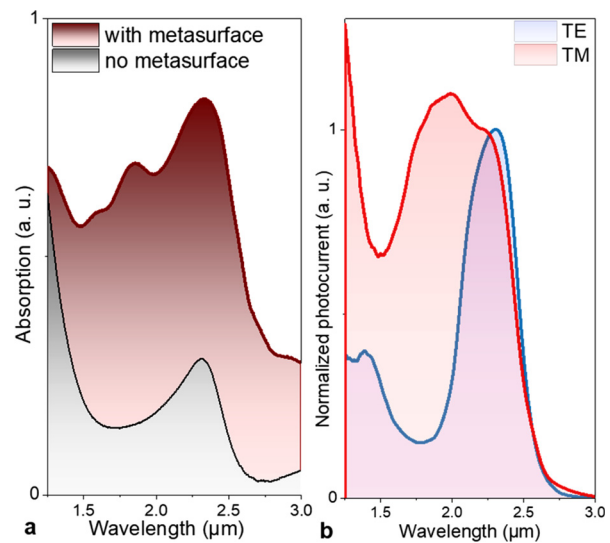


FIG. 2. Spectra from the GMR GA FFT580. (a) Absorption spectrum from the GMR GA FFT580 coupled to HgTe NC film. (b) Photocurrent spectra from the GMR GA FFT580 coupled to HgTe NC film in TM and TE polarization.

TABLE I. Strategies for multiresonant infrared detector based on HgTe NCs.

Number of resonances involved	Nature of the resonances	Maximum absorption	Linewidth of the resonance	References
3	Plasmon+vertical Fabry-Perot+planar Fabry-Perot	60%	1200 cm ⁻¹	6 and 34
3	Plasmon+vertical Fabry-Perot+planar Fabry-Perot	55%	2000 cm ⁻¹	22 and 24
2	Double Fabry-Perot	25% at band edge + 50% on second	≈800 cm ⁻¹	35
2	Plasmon+vertical Fabry-Perot	80%	500 cm ⁻¹ @2250 cm ⁻¹ + second peak @5800 cm ⁻¹	23
4 main	GMR+Fabry-Perot+high order mode	80%	3000 cm ⁻¹	This work

rise of the absorption magnitude, not only at the band edge but also away from it. Additionally, the absorption is enhanced for wavelengths beyond the band edge, which we attribute to the extra metallic losses resulting from the increased amount of metal within the metasurface. In the photocurrent spectra [Fig. 2(b)], we recover the different features seen in absorption and find that the band edge mostly matches with the TE polarization, which dominates in terms of amplitude. Meanwhile, the TM polarization strongly contributes to broaden the overall signal at higher energy. Table I makes a comparison of various strategies of design for multiresonant photonic structure coupled to HgTe NC film and shows that the current strategies are indeed well suited to generate a broad absorption enhancement.

The GA procedure leaves the exact nature of the involved electromagnetic modes unclear, requiring further investigation. Dispersion maps [Figs. 3(a) and 3(b)] provide initial insight in this direction. Around the NC band edge (2.3 μm peak), the dispersion in TE

polarization clearly shows a strongly dispersive mode that splits into two branches under oblique incidence. Its absorption is both located on top of the digit of the grating and in the opening between digits, but very weakly localized in the metal, see Fig. 3(c). This mode matches the expected behavior for a GMR. The absorption in the TM polarization close to the band edge is dominated by a non-dispersive mode, in which absorption is mostly localized on top of the metal digit.

Figure S7 shows a change in the magnitude of the absorption spectrum with thickness but no shift, while the resonance condition for a vertical Fabry-Perot cavity is that the thickness is a multiple of λ/n with λ being the resonance wavelength and n the refractive index. Thus, we should look for other possible resonances and, in particular, for the formation of a horizontal Fabry-Pérot cavity.²⁴ The latter results from the discontinuity of the effective index for the NC film on top of the dielectric and on top of the metal digit. Although this combination of GMR and cavity mode are similar to the mode that can be generated

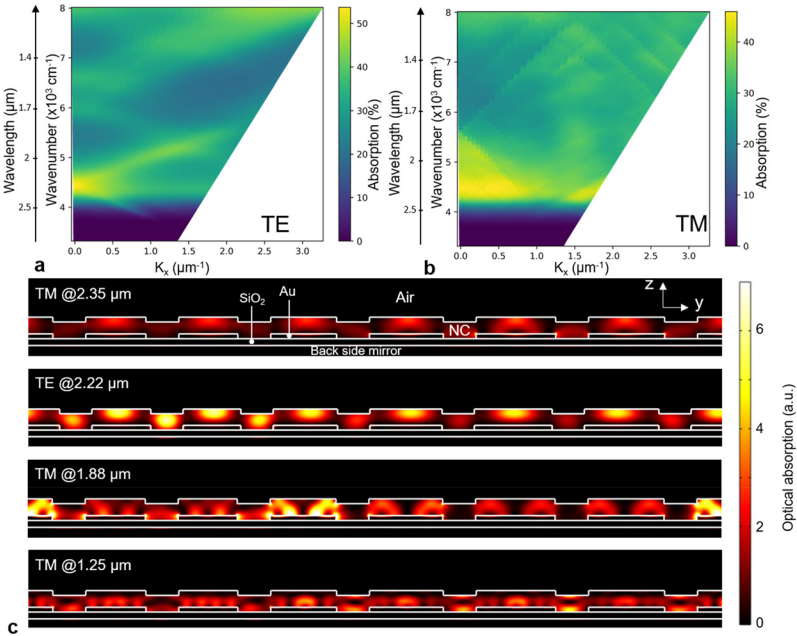


FIG. 3. Optical mode determination from the GMR GA FFT580 coupled to HgTe NC film. (a) [resp (b).] Dispersion map in TE (resp TM) polarization for the GMR GA FFT580 structure. (c) Simulated absorption map at various wavelengths and polarizations corresponding to resonance in the absorption spectra [Fig. 2(a)].

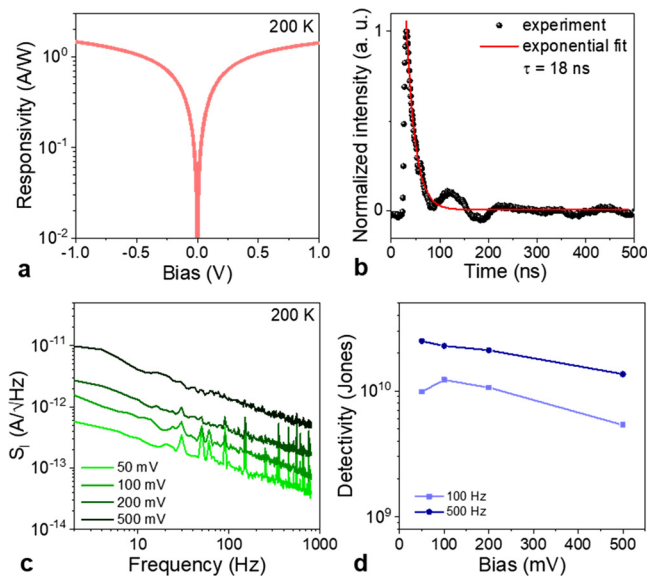


FIG. 4. Detection properties from the GMR GA FFT580. (a) Responsivity as a function of the applied bias, measured at 200 K. (b) Time response after a 1 ns pulse at $1.57 \mu\text{m}$ (bias = 0.2 V). (c) Current spectral density as a function of the signal frequency while various biases are applied over the device. (d) Specific detectivity as a function of the applied bias for two signal frequencies at 200 K.

from a periodic grating design²⁴ (Fig. S9) close to the band edge, a key difference with the single period design is the presence of extra resonances at high energy. For the sake of illustration, absorption maps at 1.25 and $1.88 \mu\text{m}$ are also shown in Fig. 3(c). These modes appear more complex (i.e., non-repetitive over the whole pattern) over to analyze since they appear more localized (TM at $1.88 \mu\text{m}$) or resulting from a high-order mode as the absorption map depicts several nodes (TM at $1.25 \mu\text{m}$). Those extra modes are responsible for the broad absorption enhancement generated by the GA method.

The device's responsivity exceeds 1 A W^{-1} when operated with a bias of 1 V and at a temperature of 200 K [Fig. 4(a)]. This corresponds to an external quantum efficiency of $\approx 50\%$, since the absorption around the band edge averages at 70%, the internal quantum efficiency is also around 70%. Noise in the system has been measured to be $1/f$ limited [Fig. 4(c)], which is the usual behavior for a photoconductive device based on narrow bandgap NCs. The resulting specific detectivity (in Jones) of the sample is determined using the formula: $D^* = \frac{R\sqrt{A}}{S_i}$, where R (in A W^{-1}) is the responsivity, S_i is the noise ($\text{A}/\sqrt{\text{Hz}}$), and A is the area of the device taken in square centimeters (in this case $100 \times 50 \mu\text{m}^2$). Values above 2×10^{10} Jones, under 0.1 V, have been achieved, see Fig. 4(d). This value is one order of magnitude lower than the values currently achieved for this material when integrated into a vertical geometry photodiode.^{36–39} Since the responsivity values are similar between the diode and our film coupled to the GA-optimized resonator, the main difference arises from the noise magnitude, resulting from the higher bias operation of our photoconductor. Certainly, future efforts should focus on transferring this concept of GA-optimized resonator into a diode device. However, the reduced signal-to-noise ratio is balanced by a higher temporal bandwidth. The time response of the sensor appears very fast with an exponential decay

fit determined to be 18 ns [Fig. 4(b)]. This value is 10 times faster than that reported for a vertical geometry device operating in the short-wave infrared and based on the same materials.^{36–38}

To summarize, we have investigated how the combination of multiple resonances shapes the spectral response of a photoconductive device in a way that increases the gain \times bandwidth product. However, this combination becomes complex when analytic scaling rules are lost or due to increasing fabrication complexity. Therefore, a complementary method based on a genetic algorithm optimization is pursued. The absorption resulting from the metasurface-coupled film is increased in both magnitude and linewidth compared to the uncoupled pristine film. Our findings indicate that the mode near the semiconductor band edge remains similar to the one obtained for a periodic structure, but additional modes at high energy are present, being either localized or resulting from high-order modes. We demonstrate that such an advanced design for the resonator enables a clear enhancement of the absorption \times bandwidth product.

See the [supplementary material](#) for details on (i) nanocrystal growth, (ii) input for electromagnetic simulation, (iii) structure optimized through genetic algorithm, (iv) device fabrication and characterization, and (v) simulation for periodic structure.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Claire Abadie: Conceptualization (equal); Investigation (equal); Methodology (equal); Writing – original draft (equal). **Nicolas Ledos:** Formal analysis (equal); Investigation (equal); Writing – original draft (equal). **Ye Mou:** Conceptualization (equal); Investigation (equal). **Mariarosa Cavallo:** Writing – review & editing (equal). **Erwan Bossavit:** Writing – review & editing (equal). **Huichen Zhang:** Writing – review & editing (equal). **Albin Colle:** Writing – review & editing (equal). **Tommaso Gemo:** Writing – review & editing (equal). **Adrien Khalili:** Writing – review & editing (equal). **Yoann Prado:** Data curation (equal); Resources (equal). **Bruno Gallas:** Investigation (equal); Resources (equal); Supervision (equal); Writing – review & editing (equal). **Grégory Vincent:** Supervision (equal); Writing – review & editing (equal). **Mathieu Mivelle:** Conceptualization (equal); Formal analysis (equal); Resources (equal); Writing – review & editing (equal). **Emmanuel Lhuillier:** Conceptualization (equal); Project administration (equal); Resources (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its [supplementary material](#).

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