Nonradiating and radiating modes excited by quantum emitters in open epsilon-near-zero cavities

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Controlling the emission and interaction properties of quantum emitters (QEs) embedded within an optical cavity is a key technique in engineering light-matter interactions at the nanoscale, as well as in the development of quantum information processing. State-of-the-art optical cavities are based on high quality factor photonic crystals and dielectric resonators. However, wealthier responses might be attainable with cavities carved in more exotic materials. We theoretically investigate the emission and interaction properties of QEs embedded in open epsilon-near-zero (ENZ) cavities. Using analytical methods and numerical simulations, we demonstrate that open ENZ cavities present the unique property of supporting nonradiating modes independently of the geometry of the external boundary of the cavity (shape, size, topology, etc.). Moreover, the possibility of switching between radiating and nonradiating modes enables a dynamic control of the emission by, and the interaction between, QEs. These phenomena provide unprecedented degrees of freedom in controlling and trapping fields within optical cavities, as well as in the design of cavity opto- and acoustomechanical systems.

INTRODUCTION

Cavity quantum electrodynamics (QED) is the field of research that investigates the interaction between quantum emitters (QEs), such as atoms and quantum dots, and a resonant cavity (1). This interaction is fundamentally interesting, and it could be the basis for quantum information processing (2–4). We emphasize that not only coherent but also dissipative interactions can be exploited with this purpose (5–7). QE-cavity interactions are also relevant for single-photon nonlinearities (8), lasing (9–11), quantum many-body systems (12, 13), and cavity optomechanics (14). At infrared and optical frequencies, dielectric microcavities (15), photonic crystals (16–20), and Anderson-localized modes (21) are commonly used owing to their low losses and associated high-quality factors. Despite losses, plasmonic systems (22–25) are also attractive because they provide subwavelength confinements, with cavity sizes well below the diffraction limit.

Aside from this spectrum of conventional cavities, more sophisticated responses in the emission and interaction properties of QEs could be obtained with cavities carved in more exotic materials. For instance, zero-index metamaterials [for example, epsilon-near-zero (ENZ) (26) or epsilon-mu-near-zero media (27–29)] exhibit a decoupling between spatial and temporal field variations (28, 30), which enables numerous wave phenomena, including tunneling (26, 31) and geometry-invariant eigenfrequencies (32). In terms of tailoring the emission properties of QEs, the phase uniformity in zero-index metamaterials has been exploited to enhance the directivity (33, 34) and Purcell factor (35–37) of single emitters, as well as to construct collective interference effects among multiple emitters (29, 38–40). The wealth in wave phenomena related to metamaterials with near-zero parameters, as well as their potentiality in enhancing emission properties, has motivated us to investigate the emission properties of QEs embedded in open ENZ cavities, with a view toward their future application in cavity QED. Note that far from being a theoretical curiosity, there are several experimental demonstrations of zero-index metamaterials based on naturally available materials (41, 42), dispersion engineering in waveguides (31, 43, 44), photonic crystals (45, 46), and artificial electromagnetic materials (47, 48).

RESULTS

Nonradiating modes in open ENZ cavities

We start by considering the emission properties of a QE located at the center of a spherical vacuum bubble of radius $r_0$, which is itself immersed in an unbounded ENZ environment (see Fig. 1A). The insulating bubble is required to avoid the singularity that arises from the direct contact of a source with an absorbing medium (56, 57). The QE is modeled as a point-like two-level system with dipole moment $p = p_z$, intrinsic nonradiative decay rate $\Gamma_{NR}$, and transition frequency $\omega_0$ (58), which is assumed to be centered at the ENZ frequency of the background medium ($\omega_0 \approx \omega_p$). Moreover, we set $\omega_0 = \omega_p = 2\pi \times 29.08 \times 10^{12}$ rad/s (that is, $\lambda_0 \approx \lambda_p \approx 10.32$ μm), which corresponds to the plasma frequency of silicon carbide (SiC) (41), to facilitate future experimental demonstrations. Naturally, either SiC or any other practical implementations of ENZ media must be necessarily lossy and dispersive, and the effects predicted for the ENZ frequency will only be observed for sufficiently small losses and on a bandwidth of finite extent. Therefore, although the theory introduced here is derived in the ENZ limit for the sake of simplicity, numerical simulations, including the effect of losses and frequency dispersion, are included later in the paper.
By solving the pertinent boundary value problem, we have shown (see Supplementary Note 1) that the spatial distribution of the classical fields excited by the aforementioned insulated dipole immersed in an unbounded ENZ medium is equal to those of an electrostatic dipole [although the fields oscillate in time at frequency \( \omega_0 \), \( \exp(-i\omega_0 t) \) time convention], with effective dipole moment \( p_{\text{eff}} \), that is

\[
E(r) = \frac{1}{4\pi\varepsilon_0} \frac{\hat{r} \cdot 2 \cos \theta + \hat{\theta} \cdot \sin \theta}{r^3} p_{\text{eff}}
\]

and

\[
H(r) = 0
\]

Figure 1B depicts the electric and magnetic field magnitude distributions obtained with a full-wave numerical solver (see Materials and Methods), for a QE embedded in a spherical vacuum bubble of radius \( r_0 = 0.25 \mu\text{m} \). The figure ratifies that the magnetic field in the ENZ region is zero, and it is trapped within the vacuum bubble. Therefore, we find that insulated QEs embedded in ENZ media can be effectively treated as sources of spatially electrostatic fields with a time-harmonic variation. The strength of the effective electrostatic dipole moment \( p_{\text{eff}} \) is determined by the properties of the vacuum bubble (see Supplementary Note 1), and it is given by

\[
p_{\text{eff}} = \frac{(k_0 r_0)^2}{J_1(k_0 r_0)} p
\]

with \( k_0 = \omega_0/c \) and \( J_1(x) \equiv \sqrt{\pi x/2} J_{3/2}(x) \), where \( J_n(x) \) is the cylindrical Bessel function of the first kind and order \( n \) \([59, 60]\). For example, the effective dipole moment is three times the original dipole moment for deeply subwavelength bubbles, that is, \( k_0 r_0 \ll 1 \rightarrow p_{\text{eff}} \approx 3p \), in consistence with the quasi-static solution to the problem (see Supplementary Note 2). Furthermore, it is apparent from Eq. 3 that the cavity conformed by the vacuum bubble becomes resonant at the zeros of \( J_1(k_0 r_0) \). Moreover, because the spatially electrostatic nature of the fields prevents radiation losses, the magnitude of \( p_{\text{eff}} \) is only limited in practice by dissipation losses. By taking the appropriate limiting procedure (see Supplementary Note 3), we find that at resonance [that is, at \( J_1(k_0 r_0) = 0 \)], \( p_{\text{eff}} \) can be asymptotically written as

\[
p_{\text{eff}} \approx -i \frac{1}{e^{\pi/2}} \frac{(k_0 r_0)}{J_1(k_0 r_0)} p \approx -i \frac{4.6}{e^{\pi/2}} p
\]
The electric field is trapped within the open cavity, whereas the magnetic field is confined to the vacuum bubble. The electric field in the ENZ region can be written as the gradient of a scalar potential \( \nabla \phi \). Therefore, when the background medium is ENZ, \( \epsilon_1 \approx 0 \), all material bodies behave as effectively perfect electric conductors, \( \epsilon_2/\epsilon_1 \rightarrow \infty \), for spatially electrostatic fields. Consequently, the fields excited by the QE will be unable to either escape the cavity or penetrate any dielectric body within this ENZ region. This effect is illustrated in Fig. 2B, which depicts the electric and magnetic field magnitude distributions in the XZ-plane cut. The magnetic field is again trapped within the vacuum bubble containing the QE at its center. By contrast, the electric field penetrates within the ENZ cavity in the form of a time-varying electrostatic field, but it is nonetheless unable to either escape the cavity or enter the other dielectric bodies.

This geometry-invariant confinement can also be understood by noting that the bound charges excited at the interface of ENZ and vacuum have distributions identical to those that would be excited at the interface of vacuum and a perfect electrical conductor (PEC) of analogous geometry. The former are given by \( \sigma_0 = -\nabla \cdot \mathbf{P} = \epsilon_0(\epsilon - 1)\mathbf{E}_{\text{In}} = -\epsilon_0\mathbf{E}_{\text{In}} \), whereas the latter are given by \( \sigma_1 = \nabla \cdot \mathbf{D} = -\epsilon_1\mathbf{E}_{\text{In}} \), where \( \mathbf{E}_{\text{In}} \) is the electric field normal to the interface in the first medium. Because identical charge distributions give rise to the same electric field, it is clear that, in terms of finding the solution to the scattered field, all boundaries in contact to the ENZ medium effectively behave as PEC boundaries.

Thus, QEs embedded in open ENZ cavities excite nonradiating modes that are confined within the extent of the cavity independently of its geometry and even when the cavity itself is open to an unbounded vacuum space. We emphasize that although nonradiating modes have already been predicted in open spherical plasmonic cavities (51–53, 55), these modes exist in open ENZ cavities independently of the geometry of its external boundary and that this effect is empowered by the spatially electrostatic nature of the fields, as demonstrated here.

**Switching between nonradiating and radiating modes**

The above analysis is valid as long as the fields excited by the QE-vacuum bubble system are dominated by the electric dipole mode in the bubble. This is exactly the case when the QE is at the center of a spherical bubble, and it is a very accurate estimation for subwavelength bubbles. At the same time, bubbles with larger sizes can efficiently excite different, possibly radiating, modes. Far from being a limitation, this opens up the possibility of switching between nonradiating and radiating modes, and hence activating/deactivating the interaction of QEs with a system external to the cavity. Take for example, as schematically depicted in Fig. 3A (see also fig. S2), an open ENZ cavity with arbitrarily shaped external boundary containing a spherical vacuum bubble, whose radius has been tuned to be resonant with a radiating mode (for example, a magnetic dipolar mode); because of the symmetry of the fields, only the electric dipolar mode is excited when the QE is at the center of the sphere, resulting in a nonradiating cavity mode. However, this symmetry is broken as the QE is shifted from the center of the bubble, and the QE excites the resonant radiating mode. In practice, the position of the QE can be controlled with different mechanisms, for example, sound waves, microelectromechanical systems (MEMS) (62), and optomechanical techniques. One of these options is schematically illustrated in Fig. 3A, in which the QE is assumed to be attached to a membrane in the cavity. Thus, if the cavity were excited by an external optical or acoustic wave, then the membrane would vibrate and the position of the QE and, hence, its emission properties would oscillate in synchrony with the vibrational mode of the cavity.

This effect can be appreciated in Fig. 3B, which depicts the simulated electric field distribution in the XZ-plane cut for the QE emitter at two orientations: one with the QE at the center of the bubble (nonradiating mode) and the other with the QE shifted from the center of the bubble, demonstrating the switching between nonradiating and radiating modes.
different emitter positions (see Materials and Methods). As anticipated, a nonradiating mode is excited for $\Delta x = 0$, and the field is confined within the cavity. By contrast, the field is strongly radiated outside the cavity for $\Delta x = 3.5 \mu m$. In our numerical simulation, the cavity has been considered with an arbitrary (not particularly designed) external boundary, although the radius of the internal spherical vacuum bubble has been numerically optimized to $r_0 = 5.27 \mu m$ to trigger the magnetic dipolar resonance when the QE is displaced away from the bubble’s center (see Supplementary Note 4 and fig. S3).

We make use of the nanoantenna formalism developed by Novotny and van Hulst (63) and Bharadwaj et al. (64) to quantitatively assess the emission properties of a QE embedded in this specific cavity. In particular, we calculate the quantum yield $\eta_{\text{rad}}$ (or radiation efficiency, that is, the ratio of radiative to total decay rates, where the latter includes both the dissipation decay in the cavity and the intrinsic nonradiative decay rate), as well as the normalized excitation rate $\Gamma_{\text{exc}}/\Gamma_{\text{exc,free}}$ (that is, the rate of excitation via spontaneous emission of a receiver located outside the cavity, at the position schematically depicted in Fig. 3A, and normalized to the free-space excitation rate) with $\Gamma_{\text{exc}} = \Gamma_{\text{rad}} D_{\text{rad}}$ where $\Gamma_{\text{rad}} = (\hbar \omega)^{-1} S \cdot \mathbf{n} dS$ and $D_{\text{rad}} = 4\pi r^2 \hat{r} \cdot S = (\hbar \omega) \Gamma_{\text{rad}}$ are the radiative decay rate and radiation directivity, respectively, and $S = 1/2 \text{Re}[\mathbf{E} \times \mathbf{H}^\ast]$ (63, 64). Both quantities are depicted in Fig. 3 (C and D) as a function of the emitter position displacement, $\Delta x$, and for different amounts of loss of the ENZ host medium $\varepsilon''$. For illustrative purposes, we assume that the intrinsic quantum yield is 0.5. It is evident from Fig. 3 (C and D) that both the excitation rate and the radiation efficiency are suppressed when the dipole is at the center of the vacuum bubble, consistent with the excitation of nonradiating modes. Moreover, both figures of merit increase as the QE is shifted from the center, and they reach a maximum at the specific displacement $\Delta x = 3.5 \mu m$. This optimal value corresponds to the maxima of the magnetic dipolar coefficient as computed from the addition theorem (see Supplementary Note 5 and fig. S4) (65). Naturally, the excitation rate and radiation efficiency are limited by the losses of the ENZ medium. However, even with losses $\varepsilon'' = 0.1$, the excitation rate is enhanced by a factor of 5 with respect to that of free space (see Fig. 3C). This result indicates that the effect of switching between radiating and nonradiating modes could potentially be experimentally validated at $\lambda \sim 10 \mu m$ using SiC (41) or at telecommunication wavelengths ($\lambda \sim 1550 \text{ nm}$) using transparent conducting oxides (42, 66). Better performances and even higher frequencies of operation might be obtained with synthetic ENZ media [such as waveguides at cutoff frequencies (31, 43, 44), photonics crystals (45, 46), and/or metamaterials (47, 48)]. Furthermore, we emphasize that a similar effect takes place for near-field interactions because, as shown in Fig. 3B, the near field outside the cavity is also suppressed when a nonradiating mode is excited.

**Coupling/decoupling of QEs**

This effect also empowers a dynamic control of the dipole-dipole interactions between different emitters and their associated collective effects (67, 68). As depicted in Fig. 4A, one could embed two emitters, for example, $\mathbf{p}_1$ and $\mathbf{p}_2$, in different spherical bubbles placed within an open ENZ cavity. When both emitters are at the center of their bubbles, they excite nonradiating modes and are effectively decoupled. However, they become resonantly coupled as their position separates from such a symmetric position. In this case, we can assume that the position of the QEs may be controlled by means of, for example, MEMSs placed outside the cavity (62). To this end, the cavity may again be assumed to be formed by an ENZ host with a noncanonical geometry (see Fig. 4A and fig. S5), whereas spherical bubbles, separated by a distance $d = 6 \mu m$,
can be assumed to be made of silicon (Si), characterized by relative permittivity $\varepsilon_{\text{Si}} = 11.7$, and their radius $r_0 = 1.505 \text{ m}$, whose position may be externally controlled by MEMSs. The cavity is pierced by a Si rod of cross-section $0.25 \text{ mm} \times 0.25 \text{ mm}$, which position may be externally controlled by MEMSs (see Supplementary Note 4 and fig. S4). The cavity is pierced by a Si rod of cross-section $0.25 \text{ mm} \times 0.25 \text{ mm}$ to illustrate that the proposed cavities are robust against the geometrical modifications that could be required to implement position control of mechanisms (for example, MEMSs).

The coupling/decoupling mechanism mediated by the nonradiating and radiating modes is illustrated in Fig. 4B, which represents the simulated electric field magnitude distributions excited by the QE in the left bubble for displacements $\Delta x = 0$ (decoupled) and $\Delta x = 1 \text{ mm}$ (optimal coupling case according to the addition theorem; see Supplementary Note 5). Note that in our simulation, the presence of the Si rod has no appreciable impact on the radiating and nonradiating nature of the fields. A quantitative estimation of the coupling/decoupling performance is gathered in Fig. 4C. To this end, we compute the individual decay rate, $\Gamma_{11}$, decay rate related to coupling, $\Gamma_{21}$, and cooperative Lamb shift, $\Delta \omega_{21}$, normalized to the free-space individual decay rate, $\Gamma_0$, as a function of frequency in the coupled ($\Delta x = 1 \text{ mm}$, first row) and decoupled ($\Delta x = 0$, second row) states. The ENZ host has been modeled with a dispersive Drude model $\varepsilon(\omega) = 1 - \omega_p^2/\omega(\omega + i\omega_c)$, where $\omega_p = 2\pi \times 29.08 \times 10^{12} \text{ rad/s}$, and for three different amounts of loss, characterized by $\omega_c = 0.001\omega_p$ (first column), $\omega_c = 0.01\omega_p$ (second column), and $\omega_c = 0.1\omega_p$ (third column).

![Fig. 4. Coupling/decoupling QEs in open ENZ cavities.](image-url)
understood by using a simple circuit model [see Supplementary Note 6, fig. S6, and Liberal et al. (71) for the circuit model analysis of the light scattering of two coupled nanoparticles]. These simulations also serve to illustrate that, although an ENZ response must be necessarily dispersive, the bandwidth provided by one particular implementation can be sufficient for certain applications. Specifically, it is clear from Fig. 4C that the coupling between both emitters in the $\Delta x = 0 \mu m$ (no displacement) configuration is suppressed on a reasonable bandwidth. In addition, if the linewidth of the resonators is smaller than this bandwidth, then resonant coupling ($\Delta x = 1 \mu m$) can still be observed without a detrimental effect because of the ENZ dispersion. Again, we emphasize that the validity of our analysis and the proposed configuration will be limited to those scenarios in which the interaction within the QE and the environment is dominated by this frequency band, in which the environment exhibits a near-zero permittivity response. This will be the case, for example, for spontaneous emission in the weak coupling regime.

DISCUSSION

Our results demonstrate that bubble-insulated QEs embedded in open ENZ cavities present the unique signature of exciting nonradiating modes independently of the geometry of the external boundary of the cavity. We believe that this effect provides unprecedented degrees of freedom in controlling and trapping electromagnetic fields within an open optical cavity. From a practical point of view, the possibility of having an arbitrarily shaped external boundary offers several opportunities in engineering this class of open cavities, also suitable for foldable, moldable, and flexible photonic platforms.

In addition, our study reveals that it is possible to switch between nonradiating and radiating modes. This could provide new venues in controlling the emission properties of QEs, such as enhancing/suppressing the spontaneous emission exiting the cavity, as well as dynamically activating/deactivating the coupling between them. The fact that these effects can take place in cavities with arbitrarily shaped boundaries could be exploited to resonantly couple with other physical processes, such as sound waves, enabling the coupling with specifically designed cavity-induced vibrational modes.

The geometry of the external boundary could also be tailored to boost the emission of radiating modes and/or to facilitate the excitation and manipulation of QEs with electromagnetic waves operating at frequencies where the cavity could be transparent or resonant. We emphasize that the geometries in the examples studied in Figs. 2 to 4 were selected to illustrate the possibility of using an arbitrarily shaped boundary. Thus, these configurations represent specific examples that are by no means optimized. For example, better performances in terms of stronger coupling parameters are obtained with a simple cubic cavity (see fig. S7). The design and optimization efforts required to meet the specifications of one or another application are out of the scope of the present work. Our work has also been restricted to spherical bubbles for the sake of simplicity and to establish a clear comparison with the theory. However, a variety of enhanced dissipative and coherent coupling phenomena could be achieved (and dynamically activated/deactivated) by using (not necessarily identical) multiphase or multimode resonant bubbles.

MATERIALS AND METHODS

The commercially available full-wave electromagnetic simulator software COMSOL Multiphysics version 5.2 (72) was used to compute the dyadic Green’s functions and the field distributions displayed in all figures of the main text and the Supplementary Materials. Specifically, we carried out analysis in the frequency domain solver using a tetrahedral mesh. The QE was modeled as a point dipole source with dipole moment $p$. The Green’s functions and field distributions provided by the numerical solver were used to compute the following related quantities: effective electrostatic dipole moment, excitation rate, quantum efficiency, decay rate associated to coupling, and photonic Lamb shift, which were evaluated according to their definition introduced in the main text.

The effective dipole moment $p_{\text{eff}}$ depicted in Fig. 1C was numerically computed by evaluating the field at the position $(0, 0, 1.05r_0)$, where $r_0$ is the radius of the vacuum bubble containing the QE, and normalizing it relative to that of an electrostatic dipole with the same dipole moment.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/2/10/e1600987/DC1

Supplementary Note 1. Derivation of Eqs. 1 to 3.

Supplementary Note 2. Quasi-static solution to the problem.

Supplementary Note 3. Derivation of Eq. 4.

Supplementary Note 4. Magnetic dipole resonances.

Supplementary Note 5. QEs shifted from the origin of the coordinates.

Supplementary Note 6. Equivalent circuit model.

fig. S1. Sketch and dimensions of the system studied in Fig. 2.

fig. S2. Sketch and dimensions of the system studied in Fig. 3.

fig. S3. Optimal bubble radius for resonant magnetic dipole excitation.

fig. S4. Electric and magnetic dipole excitations as a function of emitter displacement.

fig. S5. Sketch and dimensions of the system studied in Fig. 4.

fig. S6. Equivalent circuit model.

fig. S7. Coupling parameters in a cubic cavity.

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