Quantum Electrodynamics near a Photonic Band Gap: Photon Bound States and Dressed Atoms

Sajeev John and Jian Wang

Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario, Canada M5S 1A7

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It is shown that in dielectrics exhibiting a complete photonic band gap, quantum electrodynamics predicts the occurrence of bound states of photons to hydrogenic atoms. When the atomic transition frequency lies near a photonic band edge, the excited atomic level experiences an anomalous Lamb shift and splits into a doublet. One member of this doublet exhibits resonance fluorescence whereas the other level is dressed by the emission and reabsorption of near-resonant photons whose amplitude decays exponentially from the vicinity of the atom.

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Electromagnetism is the fundamental force governing much of the low-energy phenomena of atomic, molecular, and condensed-matter systems. Any alteration of this fundamental force will have important observable consequences in molecular quantum electrodynamics. In a periodic array of nondissipative high dielectric constant scatterers of size and spacing comparable to the wavelength of light it is possible to controllably alter the energy-momentum relation of photons by the creation of a gap in the total photon density of states over a narrow frequency range. In this paper, we describe the quantum electrodynamics of a single atom placed within such a dielectric when the first-excited to ground-state transition of the atom lies in or near the photonic band gap. In particular, we demonstrate the existence of a novel state of the excited atom coupled to the electrodynamic vacuum of the dielectric in which the atomic level is dressed by its own radiation field. The resulting eigenstate of the coupled system has an energy within the photonic band gap and consists of radiation from the atom which is exponentially localized in the vicinity of the atom. The radiated photon may tunnel many lattice constants of the dielectric host before being reabsorbed by the atom. This state is stable with respect to single-photon spontaneous emission but is subject to very slow decay by multiphoton spontaneous emission. This photon-atom bound state is the quantum electrodynamic analog of an electron-impurity-level bound state which occurs in the gap of a semiconductor. Near the photonic band edges, the atom is resonantly coupled to photons with vanishing group velocity. This strong coupling leads to a quantum electrodynamic splitting of the atomic level into a doublet exhibiting an anomalous Lamb shift. One level retains the photon-atom bound state and lies within the gap whereas the other level is shifted out of the gap and exhibits resonance fluorescence.

Systematic attempts to modify the quantum electrodynamic vacuum experienced by an atom date back to the early ideas of Kleppner.[1] Here it was suggested, and subsequently observed,[2] that by placing an atom in a small metal cavity the resulting modification of the photon density of states would be directly observable in the corresponding rate of spontaneous emission. For such an effect to be readily observable, the size of the cavity must be comparable to the wavelength of emitted radiation and resistive losses in the walls of the enclosure must be minimized. An alternative possibility was more recently suggested[3] and implemented[4] in the microwave-frequency regime by Yablonovitch using a lossless periodic dielectric microstructure. For a dielectric material with refractive index 3.5 containing an fcc lattice of spherical air cavities a complete photonic band gap has been observed when the volume fraction of the high dielectric material is approximately 0.15. For a frequency range spanning about 6% of the gap center frequency propagating electromagnetic modes are absent in all directions.

This latter result has a number of important consequences. It first provides both a guide and a proof of feasibility of the experimental observation of the strong localization of photons.[5–7] As in the case of electron localization in an amorphous semiconductor, any small deviation of the dielectric lattice from perfect periodicity will induce strongly localized electromagnetic modes in the gap region. In all previous experimental studies of optical transport in strongly scattering dielectrics,[8–10] emphasis has been placed on disorder rather than order. Numerous wave-interference phenomena such as coherent backscattering,[11–14] fluctuation, and memory effects[15–18] have been observed but no true localization. The importance of short-range order in the observation of strong localization of photons has been emphasized elsewhere.[7] Second, the occurrence of a gap or pseudogap in the photon density of states dramatically affects the quantum electrodynamics of radiation-molecule interactions. Important questions concerning the nature of resonant energy transfer between molecules placed in such a dielectric have been raised.[19] In order to clarify the microscopic consequences of strong localization of photons, we consider in detail the properties of a hydrogenic atom within such a photonic material.

For the purpose of discussion we introduce a simple model Hamiltonian for electromagnetic waves in a three-dimensional periodic dielectric. The photon dispersion relation $\omega_{\mathbf{k}}$ is chosen to be isotropic and satisfy the transcendental equation

$$4n\cos(kL) = (1+n)^2\cos[(2na+b)\omega_{\mathbf{k}}/c] - (1-n)^2\cos[(2na-b)\omega_{\mathbf{k}}/c]. \quad (1)$$

The photon energy $\hbar\omega_k$ obtained by solving this equation is in fact the exact solution of the scalar wave equation in one dimension,

$$-\nabla^2\phi-(\omega^2/c^2)\epsilon(x)\phi=(\omega^2/c^2)\phi\,, \tag{2}$$

with dielectric constant $\epsilon(x)=\sum_{m=-\infty}^{\infty}u(x-mL)$, where $u(x)=n^2-1$ for $|x|<a$ and zero otherwise. Here n is the refractive index of the scatterer, a is its radius, and $2a+b=L$ is the lattice constant. To complete the definition of the electromagnetic properties of the dielectric, we assume that the normal modes are plane waves of the form $\eta_{\lambda\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}}/\sqrt{\Omega}$, where Ω is the volume of the dielectric sample and the polarization vectors satisfy the transversality condition $\sum_{\lambda=1}^{2}\eta_{\lambda\mathbf{k}}^{i}\eta_{\lambda\mathbf{k}}^{j*}=\delta_{ij}-\hat{k}_i\hat{k}_j$. By symmetrizing $\omega_{\mathbf{k}}$ given in (1) to all directions in $\mathbf{k}$ space, we produce photonic band gaps at the spheres $|\mathbf{k}|=m\pi/L$, $m=1,2,3,\ldots$. This procedure artificially increases the true phase space available for photon propagation near a band edge and the corresponding density of states becomes singular. In a real three-dimensional crystal, the gap is highly anisotropic. We defer a discussion of quantitative corrections to our model near a band edge since it leads to qualitatively correct physics. The dispersion relation (1) exhibits many features of the observed as well as computed band structure in 3D. For example, an approximate value of the optimum high-refractive-index volume fraction for the creation of a large band gap follows from a simple argument. The dielectric "potential" $u(x)$ has the analog of a Mie resonance when precisely a quarter wavelength $(2\pi c/\omega n)/4$ fits into the diameter $2a$. A "Mie" resonance here is defined as a minimum in the transmission coefficient. On the other hand, a Bragg scattering resonance occurs when $\omega/c=\pi/L$. The condition that both the Bragg and Mie resonance occur at the *same* frequency is the condition that the volume filling fraction $f\equiv 2a/L=1/2n$. An optimum of this nature has been observed experimentally[4] and in scalar wave band structures.[20]

For the special case $2na=b$, Eq. (1) can be solved analytically. ω as a function of the complex variable k has branch-cut singularities for $k=m\pi/L$ with the gap vanishing for even values of m.

We consider the quantum electrodynamics of a hydrogenic atom which is minimally coupled to the radiation field defined by Eq. (1). The Hamiltonian for the coupled system may be written as $H=H_{\text{atom}}+H_{\text{rad}}+H_{\text{int}}+H_{\text{ct}}$, where

$$\begin{aligned} &H_{\text{atom}}+H_{\text{rad}}=\mathbf{p}^2/2m+V(\mathbf{r})+\sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}}a_{\mathbf{k}\lambda}^{\dagger}a_{\mathbf{k}\lambda}\,,\\ &H_{\text{int}}=(e/mc)\mathbf{p}\cdot\mathbf{A}(\mathbf{r})+(e^2/2mc^2)\mathbf{A}^2\,, \end{aligned} \tag{3}$$

and $H_{\text{ct}}=(\delta m/m)p^2/2m$ is a mass-renormalization counterterm for an electron of observable mass m. We assume bare atomic eigenstates with energies E_n, $n=0,1,2,\ldots$. In the electric dipole approximation we neglect the electron coordinate $\mathbf{r}$ dependence of the vector potential in the evaluation of any matrix elements.

An approximate eigenstate of the Hamiltonian (3) in the one-photon sector of the entire Hilbert space may be determined by introducing a trial wave function

$$|\psi\rangle=\sum_{n=1}^{\infty}\phi_n|n\rangle+\sum_{n=0}^{\infty}\sum_{\mathbf{k}\lambda}\psi_{\mathbf{k}\lambda}^{(n)}|\mathbf{k}\lambda;n\rangle\,, \tag{4}$$

where $|\mathbf{k}\lambda;n\rangle$ denotes a state in which there is a single photon in mode $\mathbf{k}\lambda$ and the atom is in its nth-excited state. In the state $|n\rangle$ the atom has been excited and the photon absorbed. For a single photon of energy $\hbar\omega_k\simeq E_1-E_0$, the dominant coefficients in the wave-function expansion are ϕ_1 and $\psi_{\mathbf{k}\lambda}^{(0)}$. All other coefficients are higher order in an expansion in the fine-structure constant $\alpha\equiv e^2/4\pi\epsilon_0\hbar c$. If we consider electron-mass renormalization to order α, the approximate Schrödinger equation, $H|\psi\rangle=E|\psi\rangle$, can be expressed as a set of coupled equations for the expansion coefficients:

$$(E-E_n)\phi_n=\frac{e}{m}\sum_{\mathbf{k}\lambda l}\gamma_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}}\eta_{\lambda\mathbf{k}}\cdot\mathbf{p}_{nl}\psi_{\mathbf{k}\lambda}^{(l)}+\frac{\delta m}{m}K_{n1}\phi_1\,, \tag{5a}$$

and

$$(E-E_n-\hbar\omega_{\mathbf{k}})\psi_{\mathbf{k}\lambda}^{(n)}=\frac{e}{m}\sum_{l=0}^{\infty}\gamma_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}}\eta_{\lambda\mathbf{k}}^{*}\cdot\mathbf{p}_{nl}\phi_l\,. \tag{5b}$$

Here $K_{n1}\equiv\langle n|p^2/2m|1\rangle$, $\mathbf{p}_{nl}\equiv\langle n|\mathbf{p}|l\rangle$, and $\gamma_{\mathbf{k}}\equiv(\hbar/2\epsilon_0\omega_{\mathbf{k}}\Omega)^{1/2}$. Also we have chosen units in which the vacuum speed of light $c=1$.

The amplitude $\psi_{\mathbf{k}\lambda}^{(n)}$ may be eliminated by substituting (5b) into (5a). If we then neglect the amplitudes ϕ_n except the dominant one ϕ_1, we obtain an approximate eigenvalue equation for the energy E of the coupled atom-radiation field:

$$E-E_1\simeq\frac{e^2}{6\pi^2\epsilon_0\hbar m^2}\sum_{n=0}^{\infty}|\mathbf{p}_{1n}|^2(E-E_n)g(E-E_n)\,, \tag{6a}$$

where

$$g(E)\equiv\int_0^{\Lambda}dk\,k^2\omega_k^{-2}(E/\hbar-\omega_k)^{-1}\,,\quad \Lambda\equiv mc/\hbar\,. \tag{6b}$$

Here we have used the mass-renormalization counterterm to cancel the linearly divergent part of the sum over $\mathbf{k}$. Following Bethe[21] we introduce a cutoff Λ in the photon wave vector since photons of energy higher than the electron rest mass mc^2 probe the relativistic structure of the electron wave packet.[22] The standard nonrelativistic perturbation-theory result[21] for the Lamb shift of the level E_1 follows by setting $E=E_1$ in the right-hand side of (6a) and solving for E. If the atomic level E_1 lies in the allowed spectrum $\hbar\omega_{\mathbf{k}}$, the integral over k is interpreted as a principal value. More generally, we may consider exact solutions of Eqs. (6) for the complex variable E. The function $g(E)$ has branch-cut singularities at energies in the allowed electromagnetic spectrum and care must be taken in defining the multivalued function. In particular, the phenomenon resonance fluorescence appears as a solution to Eqs. (6) in the lower half of the complex E plane on the second Riemann sheet. Analytic continuation to this second sheet is obtained by evaluating $\lim_{\epsilon\to 0}g(E+i\epsilon)$ for real E and letting E be a general complex number in the resulting expression. For

real E in the vicinity of E_1, this means

$$E-E_1=(e^2/6\pi^2\epsilon_0\hbar m^2)\left\{\sum_{n=0}^{\infty}|\mathbf{p}_{1n}|^2(E-E_n)P(g(E-E_n))-i\pi|\mathbf{p}_{10}|^2E\int_0^{\Lambda}dk\,k^2\omega_k^{-2}\delta(E/\hbar-\omega_k)\right\}. \quad (7)$$

Here we have chosen $E_0=0$ and P denotes principle value.

This eigenvalue equation always has a solution for some real value of E which lies in the photonic band gap as depicted graphically in Fig. 1 provided E_1 is in or near the gap. Here we have chosen the refractive index $n=1.082$ yielding a gap center frequency $\omega_0=\pi(1+n)\times c/2nL$ and relative gap width $\Delta\omega/\omega_0=0.05$. The numerical solution of Eq. (7) was facilitated by use of the known dipole matrix elements $\mathbf{p}_{1n}$ for discrete and continuum states of hydrogen.[23] This solution corresponds to a bound state of the photon to the atom and occurs despite the weakness of the minimal-coupling interaction (3) because of the resonant interaction between the radiation field and the atom. In Fig. 2 we plot the Lamb shift, and the tunneling distance (localization length) of the bound photon as E_1 varied from being just below the gap to just above the gap. This is done for E_1 corresponding to the $2p_{1/2}$ state as well as the $2S_{1/2}$ state of hydrogen.

Near the upper and lower band edges an additional strong-coupling effect arises from the resonant interaction of the atom with photons whose group velocity $d\omega_k/dk$ vanishes. It follows that when E_1 is close to a band edge, and provided $|\mathbf{p}_{10}|^2$ is nonzero, the quantum electrodynamic interaction is sufficiently strong to split the atomic level into a doublet. This is apparent from the graph of Fig. 1 since the function $E-E_1$ intersects the real part of the photon propagator at two energies.

It is straightforward to verify from Eq. (7) that for the vacuum case $\omega_k=ck$ the single complex solution on the second Riemann sheet (negative imaginary part for E) describes the correct single-photon spontaneous emission rate $1/\tau_1$ from the Lamb-shifted level E. In particular, $1/\tau_1=2|\mathrm{Im}E|$. Near a band edge of the dielectric material it is instructive to analytically estimate the corresponding solutions. We consider an "effective-mass" approximation to the dispersion relation (1) for $k\simeq k_0\equiv\pi/L$. Near the upper band edge ω_c, $\omega_k\simeq\omega_c+A(k-k_0)^2$. For special case $b=2na$,

$$\omega_c=\frac{c}{4na}\left[2\pi-\cos^{-1}\left(\frac{1+n^2-6n}{1+n^2+2n}\right)\right]$$

and

$$A=-\frac{cL^2}{2a(1+n)^2\sin(4na\omega_c/c)}.$$

The imaginary part of the left-hand side of (7) is singular for $E\to\hbar\omega_c$. Using perturbation theory to estimate the nonsingular parts we obtain

$$E-E_1'\simeq-\frac{i\pi e^2}{6\pi^2\epsilon_0\hbar m^2}|\mathbf{p}_{10}|^2E_1'\left(\frac{k_0}{\omega_c}\right)^2\left\{\frac{dk}{d\omega_k}\right\}_{\hbar\omega_k=E}, \quad (8)$$

where

$$E_1'\equiv(e^2/6\pi^2\epsilon_0\hbar m^2)\sum_{n=0}^{\infty}|\mathbf{p}_{1n}|^2(E-E_n)P(g(E_1-E_n))$$

is the standard Lamb shift. To study band-edge behavior, we choose $E_1'=\hbar\omega_c$ and substitute the effective-

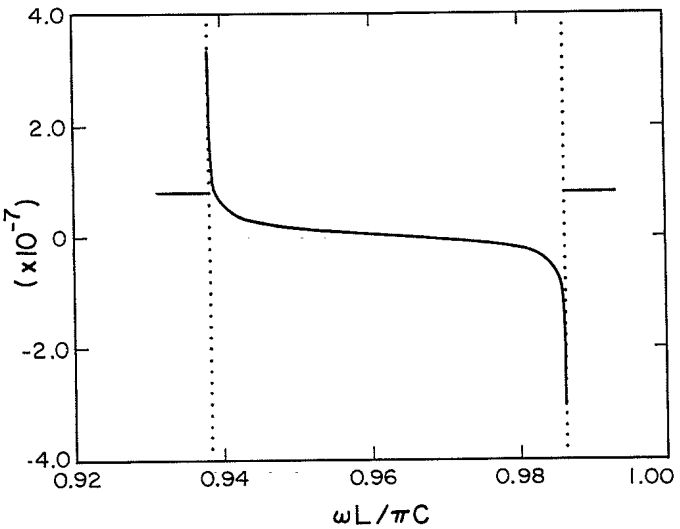

FIG. 1. The right-hand side of Eq. (7) measured in units of $\hbar\omega_0$ as a function of $\omega L/\pi c$, for $\omega\equiv E/\hbar$ near or within the band gap. The dotted lines mark the lower and upper band edges.

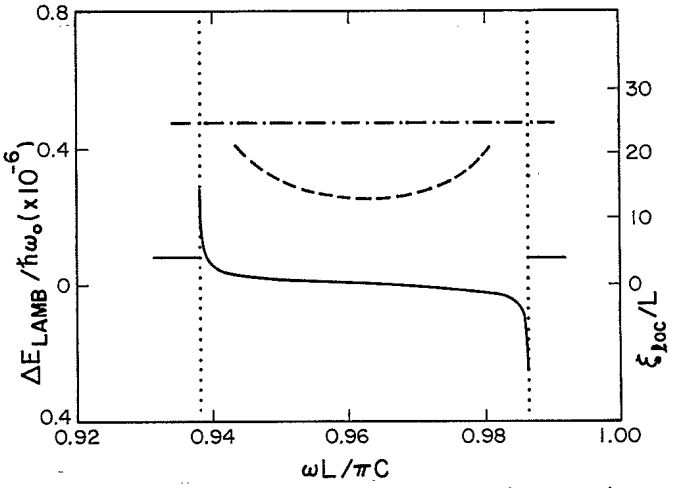

FIG. 2. The Lamb shift for the $2p_{1/2}$ state (solid line) and the $2S_{1/2}$ state (dash-dotted line) measured in units of ω_0 within the band gap. Here $\omega\equiv E_1/\hbar$. At the band edge the Lamb shift for the $2p_{1/2}$ state reaches a finite value. The localization length ξ/L within the band gap (dashed line) is also shown.

mass expression for ω_k in (8). This gives

$$z^{3/2} \simeq (-i\alpha/3)\,|\mathbf{p}_{10}/m|^2 (k_0/\omega_c)^2 (A\omega_c)^{-1/2}$$

for the complex variable $z \equiv (E-\hbar\omega_c)/\hbar\omega_c$ which measures the anomalous part of the Lamb shift and the rate of spontaneous emission. Using the definition $z^{3/2} = |z| \times e^{3i\Theta/2}$ for the complex variable $z = |z|\,e^{i\Theta}$, $0 < \Theta < 2\pi$, we find two solutions of Eq. (9). The solution at $\Theta = \pi$ corresponds to the photon-atom bound state within the photonic band gap whereas the solution for $\Theta = -\pi/3$ (on the second Riemann sheet) lies within the allowed electromagnetic spectrum.

Since the anomalous Lamb shift described by Eq. (9) for the case of hydrogen affects the odd-parity $2p_{1/2}$ state and not the even-parity $2S_{1/2}$ state, relative shifts of this nature may be detectable using microwaves.[24] Restoring correct dimensions to Eq. (9) we see that the right-hand side is simply $\alpha\,|\mathbf{p}_{10}|^2/3m^2c^2 \simeq 10^{-8}$ multiplied by numerical factors of order unity which may depend on the refractive index n. For the isotropic model described by Eq. (1), the splitting of the $2p_{1/2}$ level is larger than the ordinary Lamb shift of the $2S_{1/2}$ level because the exponent of z is $\frac{3}{2}$. This exponent, however, depends sensitively on the dimension d_b of the phase space occupied by band-edge photons of vanishing group velocity. We have overestimated this phase space by assuming that $d\omega_k/dk$ vanishes over the entire sphere $|\mathbf{k}| = \pi/L$. For a real dielectric crystal in three dimensions with an allowed point-group symmetry the band edge occurs at certain points along the Bragg planes of the lattice. For scalar waves such as electrons the locus of points for which $|d\omega_k/d\mathbf{k}| = 0$ has dimension $d_B = 0$. This would be true, for instance, in a many-valley semiconductor crystal for which the bottom of the conduction band occurs at the center of a Bragg plane. The vector nature of the electromagnetic wave modifies this picture and the phase space near a band edge is enlarged. It was argued previously[7] using the nearly-free-photon approximation for Bragg scattering that the photonic band edge actually lies on a circle of finite radius on the Bragg plane. This suggests that $d_B = 1$ in a real photonic crystal as opposed to $d_B = 0$ for scalar waves or $d_B = 2$ for the isotropic model of Eq. (1). In three dimensions, the photonic density of states $\rho(\omega)$ at a band edge ω_c behaves as $(\omega-\omega_c)^{(1-d_B)/2}$ for $\omega \gtrsim \omega_c$. This argument suggests that for a general photonic band structure with band-edge dimension d_B, the exponent of z in Eq. (9) becomes $(1+d_B)/2$. For the case $d_B = 1$, the left-hand side of Eq. (9) is of the form $z/\ln z$ and it follows that the anomalous splitting of the $2p_{1/2}$ level given by $z \equiv (E-\hbar\omega_c)/\hbar\omega_c$ is of order 10^{-7}. This is comparable to the ordinary Lamb shift of the $2S_{1/2}$ level.

In summary, we have shown using a simple model of photonic band structure that atomic and molecular spectroscopy in a dielectric with a photonic band gap is qualitatively modified from its nature in vacuum. For an isolated atom in the band gap of the dielectric, single-photon spontaneous emission is inhibited and a quantum electrodynamic bound state of the photon to the atom is formed. Near a band edge, the dressing of the atom by photons with a finite effective mass becomes strong enough to split the atomic level by an observable amount. This splitting occurs in the absence of any external radiation field. For a collection of atoms in the dielectric we anticipate that transfer of excitation energy between atoms at energies within the gap will occur by tunneling rather than propagating photons and that the atoms will form a narrow photonic impurity band within the dielectric. The presence of such a narrow impurity band formed by gas of impurity atoms in such a dielectric may be relevant to cooperative phenomena such as laser action within this new class of photonic materials.

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