Calculation of spontaneous emission from a V-type three-level atom in photonic crystals using fractional calculus

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Fractional time derivative, an abstract mathematical operator of fractional calculus, is used to describe the real optical system of a V-type three-level atom embedded in a photonic crystal. A fractional kinetic equation governing the dynamics of the spontaneous emission from this optical system is obtained as a fractional Langevin equation. Solving this fractional kinetic equation by fractional calculus leads to the analytical solutions expressed in terms of fractional exponential functions. The accuracy of the obtained solutions is verified through reducing the system into the special cases whose results are consistent with the experimental observation. With accurate physical results and avoiding the complex integration for solving this optical system, we propose fractional calculus with fractional time derivative as a better mathematical method to study spontaneous emission dynamics from the optical system with non-Markovian dynamics.

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I. INTRODUCTION

Fractional calculus, the differentiation of noninteger order, has attracted increasing attention in physical systems such as physical kinetics [1], quantum theory [2,3], and thermodynamics [4] this decade because this approach is suitable to depict the dynamics with nonlocal quantum phenomena [5,6]. Recently, Iomin interpreted the physical meaning of the fractional time derivative as an operator describing an effective interaction of a quantum system with its environment which leads to the non-Markovian dynamics of the system [7]. The spontaneous emission (SE) of excited atoms embedded in nonlinear dispersive media, such as photonic crystals (PCs), would survive the non-Markovian dynamics in the evolution of the excited-state population, which can be well described by the fractional calculus.

PCs are periodic dielectric structures [8] providing a photonic band gap (PBG) and redistribution of photon density of state (DOS) to control the SE [9–15]. Within the PBG, light is forbidden to propagate in a PC so that the SE is depressed. Near the PBG, however, the anomalous photon DOS makes the Markovian approximation of SE invalid. The non-Markovian photon-atom interaction gives rise to rapid multiatom switch with low quantum noise and laserlike collective atomic emission [16,17]. It also offers the key technology for manipulating light, such as light emitting devices [18] and solar cells [19].

The SE in three-level atom systems, including V-type [20–22], cascade-type [23], and Λ-type arrangements [11,12,24,25], is of particular interest due to the quantum interference between two allowed transitions. The quantum interference between different atomic transitions or atomic coherence in the V-type system can lead to population trapping, phase-sensitive amplification, and lasers without inversion [21,26,27].

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The study of V-type atom systems embedded in PCs was focused mainly on their emission spectra [22,28,29], but rarely on the evolution of the excited-state population due to the requirement for dealing with complex integration in the inverse Laplace transformation [21,30]. In order to simplify the computation, the band-edge frequency of the PC was usually assumed midway between two excited levels of the V-type atom [31] in the isotropic models [30]. However, the band structures of PCs are quite anisotropic in general, and the band-edge frequencies lying exactly between two excited levels are far from a generalized case. In addition, the analytical expression of the excited-state population is hard to obtain through the complex integral due to the calculation of inverse Laplace transformation. The results of the anisotropic model even conflict with the experimental results [32] in the system of a two-level atom [13]. Therefore, a more precise approach with analytical results [33] is needed to deal with the quantum interference between the excited-state populations of a three-level atom embedded in a PC. Fractional calculus not only provides a good approach to solving such a problem but can be also extended to deal with the SE of the atom placed in the microcavity in which the photon DOS exhibits threshold cutoff.

In this paper, we use the fractional calculus with fractional time derivatives to analytically solve the kinetic equations governing the time evolution of the excited states in a V-type three-level atom embedded in a PC. The results reveal that the population of excited states is a linear combination of four dressed states (DSs) in the anisotropic model. By analyzing the properties of four DSs, the dynamical behavior of the excited atom is discussed. In order to verify the accuracy of our derivation, we reduce the system into special cases, whose results can be compared with the experiments. Our analytical results obtained from the fractional calculus agree well with the experimental observation, while a contradiction exists between the results from the complex integral method and the observation.
II. THE FRACTIONAL CALCULUS APPROACH TO DESCRIBE THE DYNAMICS OF QUANTUM INTERFERENCE

We consider a three-level atom with a ground state $|b\rangle$ and two excited states $|a_1\rangle$ and $|a_2\rangle$ (shown in Fig. 1) that is embedded inside a PC. The allowed transitions from the two excited states $|a_1\rangle$ and $|a_2\rangle$ to the ground state $|b\rangle$ have transition frequencies $\omega_1$ and $\omega_2$, respectively, lying near the PC band-edge frequency $\omega_c$. The total Hamiltonian of the system is $\hat{H} = \hat{H}_0 + \hat{H}_I$, with the noninteraction term

$$\hat{H}_0 = \hbar \omega_1 \sigma_{a_1} + \hbar \omega_2 \sigma_{a_2} + \sum_k \hbar \omega_k \sigma_k^\dagger \sigma_k,$$

and the interaction Hamiltonian

$$\hat{H}_I = i\hbar \left[ \sum_k (g_{1k} \sigma_{a_1}^\dagger \sigma_{b_1} + g_{2k} \sigma_{a_2}^\dagger \sigma_{b_2}) - g_{1k} \sigma_k^\dagger \sigma_{a_1,b} \sigma_{b_1}^\dagger - g_{2k} \sigma_k^\dagger \sigma_{a_2,b} \sigma_{b_2}^\dagger \right].$$

Here, $\sigma_{i,j} = |i\rangle \langle j|$, where $i,j = a_1,a_2$, or $b$ are the atomic operators; $\omega_k, \sigma_k^\dagger$, and $\sigma_k$ are the radiative frequency, creation, and annihilation operators of mode $k$ of the reservoir; and the coupling coefficients between the two transitions and the PC reservoir, $g_{1k}$ and $g_{2k}$, are [34]

$$g_{nk} = \frac{\omega_k d_n}{\hbar} \left( \frac{\hbar}{2 \pi \omega_k V} \right)^{1/2} \mathbf{e}_k \cdot \mathbf{u}_j.$$

We have assumed a fixed atomic dipole moment $d_n = d_\nu u_\nu$ which is independent of the atomic position; the field has a quantization volume $V$; $\mathbf{e}_k$ is the unit vector of the electric field.

The dispersion relation of an anisotropic PC near the band edge $\omega_c$ can be approximately expressed as $\omega_k = \omega_c + D(k - k_c)^2$, where $D = f_k(\omega_c/k_c^2)$ is the curvature near $\omega_c$ with the scaling factor $f_k$ for the different $k$ directions.

We now consider the two dipoles with vectors parallel to each other because the interference between these two dipoles is similar to that of the parallel and antiparallel dipoles, while that of the two orthogonal dipoles does not deliver important results because no interaction exists between the two transitions [21]. Having $\omega_1 - \omega_c = \Delta_1$ and $\omega_2 - \omega_c = \Delta_2$, with $|\Delta_1|,|\Delta_2| \ll \omega_c$, we can further assume $g_{1k} = g_{2k} = g_k$.

The state vector $|\psi(t)\rangle$ at a time instant $t$ thus can be expressed as the superposition of bare states $|a_1,0\rangle$ and $|b,1\rangle$ of the one-photon sector,

$$|\psi(t)\rangle = A_1(t)e^{-i\omega_1 t}|a_1,0\rangle + A_2(t)e^{-i\omega_2 t}|a_2,0\rangle + \sum_k B_k(t)e^{-i\omega_k t}|b,1_k\rangle.$$  

Here $|a_1,0\rangle$ describes the atom in its excited state $|a_1\rangle$ or $|a_2\rangle$ with no photon present and $|b,1_k\rangle$ represents the atom in its ground state $|b\rangle$ with a single photon in mode $k$.

From the time-dependent Schrödinger equation, we obtain

$$A'_1(t) = -\sum_k g_{1k} B_k(t)e^{-i(\omega_k - \omega_1)t},$$

$$B'_k(t) = g_k[A_1(t)e^{-i(\omega_1 - \omega_k)t} + A_2(t)e^{-i(\omega_2 - \omega_k)t}].$$

with $n = 1$ or 2. Integrating Eq. (6), we obtain

$$B_k(t) = g_k \int_0^t [A_1(\tau)e^{-i(\omega_1 - \omega_k)\tau} + A_2(\tau)e^{-i(\omega_2 - \omega_k)\tau}] d\tau.$$  

Substituting Eq. (7) into Eq. (5), we have

$$\ddot{A}(t) = -\int_0^t \ddot{G}(t - \tau) \cdot \dot{A}(\tau) d\tau,$$

where $G(t-\tau)$ is the memory kernel given by

$$G(t) = \sum_k |g_k|^2 \left[ e^{-i(\omega_k - \omega_1)t} + e^{-i(\omega_k - \omega_2)t} - e^{-i(\omega_k - \omega_1)t} - e^{-i(\omega_k - \omega_2)t} \right],$$

and $\dot{A}(\tau) = iA_1(\tau)$.

In the continuum limit, one can replace the discrete summation $\sum_k$ by the continuous one $V \int_0^\infty \rho(\omega) d\omega$ [17] so that the memory kernel for the anisotropic PC is

$$\ddot{G}(t) = \frac{r_d e^{i\pi/4}}{2\sqrt{\pi}(t-\tau)^{3/2}} \begin{bmatrix} e^{i\Delta_1(t-\tau)} & e^{i\Delta_2(t-\tau)} \\ e^{-i\Delta_1(t-\tau)} & e^{-i\Delta_2(t-\tau)} \end{bmatrix},$$

with the coupling constant $r_d = \omega_0^2 d^2/(8\hbar\varepsilon_0 D_0^{3/2})$. Here we have applied the photon DOS of the anisotropic model, $\rho(\omega) = \sqrt{\omega - \omega_c} \delta(\omega - \omega_0)/(4\pi D_0^{3/2})$, with $\delta(u)$ being the Heaviside step function, to the memory kernel.

By substituting Eq. (10) into Eq. (8) and making a transformation of $A_1(t) = e^{i\Delta_1 t}C_1(t)$ and $A_2(t) = e^{i\Delta_2 t}C_2(t)$ to simplify the derivation, we have

$$\frac{d}{dt} C_n(t) + i \Delta_n C_n(t) = -\frac{r_d e^{i\pi/4}}{2\sqrt{\pi}} \int_0^t C_1(\tau) + C_2(\tau) \frac{d}{dt} C_n(t) + i \Delta_n C_n(t)$$

From the definition of the Riemann-Liouville fractional differentiation operator [33]

$$\frac{d^\alpha}{ds^\alpha} u(t) = \frac{1}{\Gamma(-\alpha)} \int_0^t (t-s)^{\alpha-1} u(s) ds,$$

FIG. 1. (Color online) Frequency diagram of a three-level atom placed in a photonic band-gap structure. The two excited states, $|a_1\rangle$ and $|a_2\rangle$, detune from the photonic band edge ($\omega_c$) with $\Delta_1$ and $\Delta_2$.
Eq. (11) can be expressed as a fractional calculus form:
\[
\frac{d}{dt} C_n(t) + i \Delta_n C_n(t) = -r_d e^{i \gamma t/4} \frac{d^{1/2}}{dt^{1/2}} [C_1(t) + C_2(t)],
\]

(13)

where \( \Gamma(x) \) is the gamma function. Here \( \alpha \) can be any real number and \( d^{1/2}/dt^{1/2} \) becomes an integral operator with negative \( \alpha \). We further apply the integral operator \( d^{1/2}/dt^{1/2} \) followed by the differential operator \( d^{1/2}/dt^{1/2} \) to Eq. (13) to get the fractional quantum Langevin equation,
\[
\frac{d^{1/2}}{dt^{1/2}} C_n(t) + i \Delta_n \frac{d^{-1/2}}{dt^{-1/2}} C_n(t) + r_d e^{i \gamma t/4} [C_1(t) + C_2(t)]
= \frac{C_n(0)}{\Gamma(1/2)} t^{-1/2}.
\]

(14)

This fractional Langevin equation with the presence of the fractional time derivative indicates a subordinated stochastic process directing to a stable probability distribution. This equation governs the SE dynamics of the current optical system and appears as a form of the Langevin equation because of the existence of non-Markovian dynamics in the system.

Taking the Laplace transform of these two fractional Langevin equations for \( n = 1 \) and 2,
\[
L \left\{ \frac{d^{1/2}}{dt^{1/2}} A(t) \right\} = \int_0^\infty e^{-s \alpha} \frac{d^{1/2}}{dt^{1/2}} A(t) dt
= s^{-1/2} L \{ A(t) \} - \sum_{k=0}^{m-1} s^{m-k-1} \left[ \frac{d^{k-m}}{dt^{k-m}} A(t) \right]_{t=0},
\]

(15)

with \( m - 1 < \alpha \leq m \) for \( m = 1, 2, 3, \ldots \), we have
\[
A_n(s + i \Delta_n) = \frac{A_n(0) (s + i \Delta_{3-n}) + (-1)^n r_d \sqrt{s} e^{i \gamma t/4} [A_1(0) - A_2(0)]}{s^2 + 2r_d e^{i \gamma t/4} \sqrt{s} + i s (\Delta_1 + \Delta_2) + i r_d e^{i \gamma t/4} \sqrt{s} (\Delta_1 + \Delta_2) - \Delta_1 \Delta_2}.
\]

(16)

With \( X = s^{1/2} \), we can then rewrite Eq. (16) as a sum of partial fractions,
\[
A_n(s + i \Delta_n) = \sum_{m=1}^{4} \frac{n \alpha_m}{(X - X_m)}.
\]

(17)

Note that the parameters \( X_m \) \((m = 1, 2, 3, 4)\) of Eq. (17) are the roots of
\[
X^4 + 2r_d e^{i \gamma t/4} X^3 + i (\Delta_1 + \Delta_2) X^2
+ r_d e^{i \gamma t/4} (\Delta_1 + \Delta_2) X - \Delta_1 \Delta_2 = 0,
\]

(18)

and the coefficients \( n \alpha_m \) are given by
\[
n \alpha_m = \frac{A_n(0) [X_m^2 + i \Delta_{3-n}] - r_d e^{i \gamma t/4} X_m [A_1(0) - A_2(0)] (-1)^n}{4 \prod_{j=1 \left( \text{mod} \ 4 \right)} \left( X_m - X_j \right)}.
\]

(19)

From the formula of the inverse fractional Laplace transformation [33]
\[
L^{-1} \left\{ \frac{1}{s^{1/2} - X} \right\} = E_t \left( -\frac{1}{2}, X^2 \right) + X e^{X^2 t},
\]

(20)

we can obtain the probability amplitudes of two excited states
\[
A_1(t) = e^{i \Delta_1} \sum_{m=1}^{4} \alpha_m \left[ E_t \left( -\frac{1}{2}, X_m^2 \right) + X_m e^{X_m^2 t} \right]
\]

\[
= e^{i \Delta_1} \sum_{m=1}^{4} \alpha_m \left[ X_m^2 E_t \left( \frac{1}{2}, X_m^2 \right) + X_m e^{X_m^2 t} \right]
\]

(21)

and
\[
A_2(t) = e^{i \Delta_2} \sum_{m=1}^{4} \alpha_m \left[ E_t \left( -\frac{1}{2}, X_m^2 \right) + X_m e^{X_m^2 t} \right],
\]

(22)

where \( E_t(\gamma, a) \) is the fractional exponential function of order \( \gamma \) and is defined as
\[
E_t(\gamma, a) = \frac{d^{-\gamma}}{dt^{-\gamma}} e^{at} = t^{-\gamma} \sum_{n=0}^{\infty} \frac{(at)^n}{\Gamma(\gamma + n + 1)}.
\]

(23)

The fractional calculus provides an analytic solution in the inverse fractional Laplace transformation, while the complex integration method only gives an integral formula and has to solve it by the numerical method. The fractional calculus can deal with a realistic or generalized case to give a more complete conclusion as a result of its analytic form.

III. THE PROPERTIES OF DRESSED STATES AND THE EVOLUTION OF SPONTANEOUS EMISSION

In the previous section, the population amplitudes of the two excited states in an anisotropic PC contributed from four DSs have been expressed as analytical forms, such as Eqs. (21)
and (22), by the fractional calculus. These two equations can be further written as [33]

\[
A_1(t) = e^{i\Delta t} \sum_{m=1}^{4} \alpha_m [Y_m \text{Erf}(Y_m \sqrt{t}) + X_m] e^{X_m t},
\]

\[
A_2(t) = e^{i\Delta t} \sum_{m=1}^{4} \alpha_m [Y_m \text{Erf}(Y_m \sqrt{t}) + X_m] e^{X_m t},
\]

with \( Y_m \) being the square root of \( X_m^2 \) in the first or second quadrant and \( E_i(1/2, X) = \exp(X t) \text{Erf}((X^2 t)/\sqrt{X}) \). Here \( \text{Erf}(t) \) is the error function. Substituting Eqs. (24) and (25) into Eq. (4), it is obvious that the wave function possesses four DSs with their frequencies equal to \( \omega_c - \text{Im}(X_m^2) \), where \( \text{Im} \) represents the imaginary part. When \( X_m^2 \) is a complex number, the population of the excited state contributed from the DS characterized by \( X_m \) will behave as decaying. On the other hand, when \( X_m^2 \) is a pure positive imaginary number, the population contributed from the DS of \( X_m \) may oscillate initially due to the error function term and then the oscillation decreases as time passes due to \( \text{Erf}(Y_m \sqrt{t}) = 1 \) as \( t \to \infty \). Under this circumstance, the amplitude of the atomic excited state contributed from the \( m \)th dressed state of \( X_m \) is equal to \( 2\alpha_m X_m \exp(X_m^2 t) \) at \( t \to \infty \) as \( X_m \) locates in the first quadrant, but is equal to 0 as \( X_m \) locates in the third quadrant. Therefore, only the DS with \( X_m \) having amplitude \( P_m \exp(i\pi/4) \) \( (P_m > 0) \) contributes a bound state to the \( |a_1, 0\rangle \) or \( |a_2, 0\rangle \) state, otherwise, the DSs are the decaying states. The magnitude of the amplitude of the excited state \( |a_1, 0\rangle \) or \( |a_2, 0\rangle \) contributed from the \( m \)th bound DS is \( 2\alpha_m X_m \).

Therefore, the time evolution of the excited state of a three-level atom in an anisotropic PC would behave differently as a result of different numbers of contributed bound DSs and can be categorized into three regimes, as shown in Fig. 2.

A. Non-Markovian decay regime with no bound DS

When the excited states of the atom are both in the photonic allowed band, the DSs are all unbound. It behaves as spontaneous non-Markovian decay with some oscillation initially contributed from either the exponential terms or the interference of four unbound DSs. The atom transfers all its stored energy to the SE propagating field in the PC; therefore, it shows decaying excited-state populations for \( \Delta_1/r_d = 0.5 \) and \( \Delta_2/r_d = 0.25 \); (b) \( \Delta_1/r_d = 0.5 \) and \( \Delta_2/r_d = -0.5 \); and (c) \( \Delta_1/r_d = -0.5 \) and \( \Delta_2/r_d = -0.25 \). \( P_f \) is equal to \( P_1 + P_2 \).

FIG. 2. (Color online) Regions of SE dynamics in the anisotropic model with the detuning frequencies of atom 1 and 2 being \( \Delta_1 \) and \( \Delta_2 \) with respect to the photonic band edge in the unit of coupling coefficient \( r_d \).

FIG. 3. (Color online) Dynamics of SE in anisotropic PC systems with initial conditions \( A_1(0) = 1 \) and \( A_2(0) = 0 \). \( P_1 = |A_1|^2 \) and \( P_2 = |A_2|^2 \) are the populations of excited states 1 and 2 with (a) \( \Delta_1/r_d = 0.5 \) and \( \Delta_2/r_d = 0.25 \); (b) \( \Delta_1/r_d = 0.5 \) and \( \Delta_2/r_d = -0.5 \); and (c) \( \Delta_1/r_d = -0.5 \) and \( \Delta_2/r_d = -0.25 \). \( P_f \) is equal to \( P_1 + P_2 \).
B. Damped quantum interference regime with one bound DS

When one excited state is above the band edge and the other is below the band edge in the gap, one bound DS exists. The excited-state populations will oscillate initially because of the strong interaction between the photon and the PC reservoir. This oscillation will diminish and finally reach a steady (bound) state, as shown in Fig. 3(b), with \( \Delta_1/r_d = 0.5 \) and \( \Delta_2/r_d = -0.5 \). Although the state \( |\alpha_1\rangle \) is in the allowed band, its population does not completely decay to zero due to the existence of one bound DS and it is much smaller than that of the state \( |\alpha_2\rangle \) lying within the gap. The initial fast damped quantum interference occurring in the time interval having \( r_d t = 0 - 25 \) is caused by energy transfer between the atom and the PC reservoir through interference of two excited states before relaxing the decaying DS population to a photon.

C. Quantum interference regime with two bound DSs

The populations will always oscillate due to the interference of two bound DSs as the energies of excited states are both within the band gap. The energy would transfer between these two bound DSs, i.e., \( \langle \text{Im}(X^2 - X'^2) \rangle \).

IV. SPECIAL CASES: ASYMPTOTIC TWO-LEVEL ATOM

When one of the unperturbed atom states, e.g., the state \( |\alpha_2\rangle \), locates far above the band edge of the PC, it should hardly interact with the other state and act as in the free space. Therefore, let \( \Delta_2 \) be equal to infinite in Eq. (18); we can rewrite this equation as

\[
X^2 + r_d e^{i\pi/4} X + i \Delta_1 = 0. \tag{26}
\]

The dynamical solution of this anisotropic PC system is

\[
A_1(t) = e^{i(\Delta_1 - i\pi/4)/\sqrt{r_d^2 - 4\Delta_1}} \left[ X_1^2 E_i(1/2, X_1^2) - X_2^2 E_i(1/2, X_2^2) + X_1 e^{X_1^2} - X_2 e^{X_2^2} \right] \tag{27}
\]

for \( r_d^2 \neq 4\Delta_1 \), and

\[
A_1(t) = e^{-i\Delta_1 t} \left[ -r_d^3/4e^{i\pi/4} E_i(1/2, i r_d^2/4) - \frac{r_d^2/2e^{i\pi/4} E_i(1/2, i r_d^2/4) + (1 + i r_d^2/2)e^{i\pi/4}}{\sqrt{r_d^2 - 4\Delta_1}} \right] \tag{28}
\]

for \( r_d^2 = 4\Delta_1 \), with \( X_n \) being the roots of the characteristic equation \( (26) \) [13]. This equation possesses one bound DS when \( \Delta_1 < 0 \) and no bound DS when \( \Delta_1 > 0 \). That is, as the atomic transition frequency lies within the allowed band (\( \Delta_1 = \omega_1 - \omega_c > 0 \)), the anisotropic system shows no photon-atom bound state. The absence of the bound state leads to the appearance of a SE image which is consistent with experimental observations by Barth et al. They found that the anisotropic band structure of the artificial colloidal opals (PCs) brought an extra angular anisotropy of fluorescence (SE) image as the emission frequency \( \omega_1 \) of the embedded quantum dots (two-level atoms) lay near the band edge \( \omega_c \). The consistency between our analytical result and that of the experimental observation illustrates the accuracy of our calculation results.

This fractional calculus approach is also suitable to describe the atoms or other active materials embedded in the microcavity [37,38] with the width of one (or two) dimension comparable to the wavelength of the emitting photon, and the others’ widths are much larger than the wavelength in which the dispersion relation of the cavity can be expressed as \( \omega_k = \omega_c + Dk^2 \). In such a case, the memory kernel is not exactly equal to Eq. (9) because the summation of the \( k \) points is no longer satisfying the continuum limit in three dimensions, which makes the photon DOS in microcavity different from that of the PC case. However, when the DOS can be treated accurately, the spontaneous emission of atoms in the microcavity can also be well described by the fractional calculus approach via similar processes.

V. CONCLUSION

The fractional calculus is used to study the spontaneous emission dynamics of a V-type atom embedded in a PC in which the evolution functions are expressed by a linear combination of four DSs. With different detuning of excited states with respect to the photonic band edge, the dynamic behavior can be classified into three regions: non-Markovian decay, damped quantum interference, and quantum interference. When two excited states are above the band edge, the decay rates of the excited-state populations appear as non-Markovian with fast decay at the beginning, but become slow after evolving for a certain time, which is observed in the relaxation of excited-state electrons in the semiconductor. As only one excited state is within the band gap, the population reveals fast damped quantum interference initially, caused by the interference of decaying states, and finally reaches a stationary bound state in both excited states. In the quantum interference regime, with both of the excited states within the band gap, energy will be exchanged between two excited states due to the interference of two bound dressed states.

By reducing our system into a two-level atom system to compare it with the experimental results and the theoretical results obtained by the complex integral approach, we found that the photon-atom bound state existed only when the excited-state energy was located within the photonic band gap, which corresponds to the experimental results in which the quantum dots are embedded in the photonic crystal.

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