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Spontaneous emission near the band edge of a three-dimensional photonic crystal: a fractional calculus approach

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Abstract

We suggest a better mathematical method, fractional calculus, for studying the behavior of the atom–field interaction in photonic crystals. By studying the spontaneous emission of an atom in a photonic crystal with a one-band isotropic model, we found that the long-time inducing memory of the spontaneous emission is a fractional phenomenon. This behavior could be well described by fractional calculus. The results show no steady photon–atom bound state for the atomic resonant transition frequency lying in the proximity of the allowed band edge which was encountered in a previous study (Woldeyohannes and John 2003 *J. Opt. B: Quantum Semiclass. Opt.* **5** R43). The correctness of this result is validated by the ‘cut-off smoothing’ density of photon states (DOS) with fractional calculus. By obtaining a rigorous solution without the multiple-valued problem for the system, we show that the method of fractional calculus has a logically concise property.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The performance of photonic devices in various fields is greatly limited by the spontaneous emission rate. In light-emitting diodes and lasers, for example, spontaneous emission that is not extracted from the devices will contribute to loss and noise. Inhibiting undesirable spontaneous light emission and redistributing the energy into useful forms becomes important in these fields [1–5]. It has been demonstrated theoretically and experimentally [6, 7] that photonic bandgap (PBG) materials could be effectively used to inhibit spontaneous emission. Near a photonic band edge, the photon density of state (DOS), which determines the rate of spontaneous emission, is significantly different from that of free space. Singularity [8] of the DOS near the PBG leads to the strong atom–field interaction and formation of photon–atom bound states [9, 10], where the spontaneous emission rate is inhibited. The Markov approximation [11] of spontaneous emission in free space is no longer valid in this near-PBG region, where the atomic

decay becomes non-exponential and the emission spectrum becomes non-Lorentzian. By using two-dimensional (2D) photonic crystals, Fujita *et al* [12] successfully inhibited and redistributed the spontaneous light emission by a factor of 5 as a result of the 2D photonic bandgap effect.

The experimental data clearly indicate that the spontaneous emission rate has non-exponential decaying behavior when the emission peak is located near the band edges (the cases of lattice constant $a = 480$ and 390 nm in figure 3 of [13]). This non-Markov behavior of the PBG reservoir had been studied by John *et al* [9, 10, 14] using the Laplace transform method to solve the time evolution integral equation of the excited probability amplitude of an atom in a high- Q microcavity with singular DOS. They showed that the time evolution of the excited-state population exhibits decay and oscillatory behavior before reaching a nonzero steady-state value due to photon localization [9, 10]. This bound dressed state leads to a fractionalized steady-state atomic population in the excited state. This behavior is observed as the prolonged

lifetime effect in [12]. However, John *et al* [14, 15] predicted that the unphysical bound state is present even when the resonant atomic frequency lies outside the band gap. This is inconsistent with the experimental result that the prolonged lifetime effect will disappear when the emission peak lies outside the PBG region. This inconsistency may be caused by the multiple-valued problem encountered in [14] and by the singular DOS which is appropriate for a high- Q microcavity but not suitable for a PBG reservoir with more smooth DOS near the band edge.

The time evolution of the probability amplitude of the excited level of an atom is related to the delay Green function or memory kernel $G(t - t')$ [8, 11], which is a measure of the reservoir memory on the excited atom. The resultant Green function depends very strongly on the photon DOS of the relevant photon reservoir. The DOS near the band edge in the isotropic one-band model has the form of $\rho(\omega) \propto (\omega - \omega_c)^{-1/2}$, where the square-root singularity is a characteristic of a one-dimensional phase space. The corresponding memory kernel [8] has the same form of a square-root singularity: $G(t - t') \propto (t - t')^{-1/2}$. Such a square-root singularity means that the resultant memory kernel possesses the long-time memory effect. That is, there is no timescale to separate the microscopic levels from the macroscopic levels.

Recently, long-time memory phenomena have also attracted a great attention in statistical physics. Such a long-time memory is intrinsic to all timescales (the corresponding memory function has no characteristic scales) of the phase space of a system, provided that the number of divisions generating a fractal set tends to infinity. It has been shown that the relationship between the fractal set and the fractional integral reduces the generalized Langevin, Kramer–Moyal, Fokker–Planck–Kolmogorov equations to their fractional forms [16–18]. The random nature of microscopic dynamics can transmit to the macroscopic level. The correct description of the macroscopic evolution of such systems has to be expressed in terms of fractional calculus [16–18]. In fact, fractional calculus is a macroscopic manifestation of randomness. Fractional calculus is a useful tool for a diffusion process generated by a fluctuation with no timescale at the macroscopic level, and such a diffusion process can be described by a fractional Langevin equation [19, 20].

In this paper, we applied fractional calculus to study the dynamics of the spontaneous emission of an atom in a photonic crystal. We derived a fractional Langevin equation for this system and solved it to obtain the excited-state probability density. The solution produced by the fractional (inverse) Laplace transform [21–23] is expressed in terms of the square complex variables. There is no multiple-valued problem like that encountered in the previous studies [14, 15]. This rigorous mathematical method shows that no steady photon–atom bound state exists for the atomic resonant transition frequency lying in the allowed band. We verified the correctness of this by using the ‘cut-off smoothing’ DOS [24] with fractional calculus for the atomic transition frequency lying in the proximity of the allowed band edge. The excited-state probability of this result still shows decaying characteristics. Fractional calculus gives the correct description of the behavior

for the system near the band edge either with or without the ‘cut-off smoothing’ DOS. It not only resolves the multiple-valued problem but also avoids choice of the smoothing parameter. Therefore, we suggest that the behavior of the atom–field interaction in photonic crystals should be expressed in terms of fractional calculus.

2. The dynamics of spontaneous emission

The system we investigate is a two-level atom coupled to the radiation in a photonic crystal with a one-band isotropic model. In the rotating-wave approximation, the total Hamiltonian for the coupled atom–field system can be written as

$$H = \hbar\omega_{21}\sigma_{22} + \sum_{\vec{k}} \hbar\omega_{\vec{k}} a_{\vec{k}}^{\dagger} a_{\vec{k}} + i\hbar \sum_{\vec{k}} g_{\vec{k}} (a_{\vec{k}}^{\dagger} \sigma_{12} - \sigma_{21} a_{\vec{k}}) \quad (1)$$

where $\sigma_{ij} = |i\rangle\langle j|$ ($i, j = 1, 2$) are the atomic operators for a two-level atom with excited state $|2\rangle$, ground state $|1\rangle$, and resonant transition frequency ω_{21} ; $a_{\vec{k}}$ and $a_{\vec{k}}^{\dagger}$ are the annihilation and creation operators of the radiation field. $\omega_{\vec{k}}$ is the radiation frequency of mode \vec{k} in the reservoir, and the atom–field coupling constant $g_{\vec{k}} = \frac{\omega_{21} d_{21}}{\hbar} [\frac{\hbar}{2\epsilon_0 \omega_{\vec{k}} V}]^{1/2} \hat{e}_{\vec{k}} \cdot \hat{u}_d$ is assumed to be independent of atomic position with the fixed atomic dipole moment $\vec{d}_{21} = d_{21} \hat{u}_d$. V is the sample volume, $\hat{e}_{\vec{k}}$ is the polarization unit vector of the reservoir mode \vec{k} , and the Coulomb constant is ϵ_0 .

In the single photon sector, the wavefunction of the system has the form

$$|\psi(t)\rangle = A(t) e^{-i\omega_{21}t} |2, \{0\}\rangle + \sum_{\vec{k}} B_{\vec{k}}(t) e^{-i\omega_{\vec{k}}t} |1, \{1_{\vec{k}}\}\rangle \quad (2)$$

with initial condition $A(0) = 1$ and $B_{\vec{k}}(0) = 0$. Here $A(t)$ labels the probability amplitude for the atom in its excited state $|2\rangle$ with an electromagnetic vacuum state and $B_{\vec{k}}(t)$ for the atom in its ground state $|1\rangle$ with a single photon in mode \vec{k} with frequency $\omega_{\vec{k}}$.

We obtained the equations of motion for the amplitudes by projecting the time-dependent Schrödinger equation on the one-photon sector of the Hilbert space as

$$\frac{d}{dt} A(t) = - \sum_{\vec{k}} g_{\vec{k}} B_{\vec{k}}(t) e^{-i\Omega_{\vec{k}}t} \quad (3)$$

$$\frac{d}{dt} B_{\vec{k}}(t) = g_{\vec{k}} A(t) e^{i\Omega_{\vec{k}}t} \quad (4)$$

with detuning frequency $\Omega_{\vec{k}} = \omega_{\vec{k}} - \omega_{21}$. By substituting the time integration of equation (4) into equation (3), we have the time evolving equation of the excited-state probability amplitude

$$\frac{d}{dt} A(t) = - \int_0^t G(t - \tau) A(\tau) d\tau \quad (5)$$

with the memory kernel $G(t - \tau) = \sum_{\vec{k}} g_{\vec{k}}^2 e^{-i\Omega_{\vec{k}}(t-\tau)} = \beta^{\frac{3}{2}} \int \rho(\omega) e^{-i(\omega - \omega_c)(t-\tau)} d\omega$. We could observe from this equation that the memory kernel is a measure of the reservoir’s memory in its previous state. The system evolves according to this equation.

The object of our study, the dynamics of the spontaneous emission, could be obtained by solving the time evolving equation (5). Here we applied fractional calculus and fractional Laplace transform to solve this equation, which was shown to have the form of a fractional Langevin equation. We adopted the isotropic one-band model to determine the memory kernel in equation (5), which is given as $G(t - \tau) = \frac{\beta_1^{3/2}}{(t-\tau)^{1/2}} e^{-i[\pi/4 - \Delta_c(t-\tau)]}$ with $t > \tau$ in the long-time limit [8]. Here $\Delta_c = \omega_{21} - \omega_c$ is the detuning frequency of the atomic resonance frequency ω_{21} from the band edge ω_c and $\beta_1^{3/2} = (\omega_{21}^{7/2} d_{21}^2) / (12\pi^{3/2} \hbar \epsilon_0 c^3)$ is the coupling constant. By substituting this memory kernel to equation (5) and making a transformation $A(t) = e^{i\Delta_c t} C(t)$, we obtained

$$\frac{d}{dt} C(t) + i\Delta_c C(t) = -\beta_1^{3/2} e^{-i\pi/4} \int_0^t \frac{C(\tau)}{(t-\tau)^{1/2}} d\tau. \quad (6)$$

Comparing this equation with the Riemann–Liouville fractional differentiation operator [21–23]

$$\frac{d^\alpha}{dt^\alpha} u(t) = \frac{1}{\Gamma(-\alpha)} \int_0^t (t-\tau)^{-\alpha-1} u(\tau) d\tau, \quad (7)$$

we observed that the term on the right-hand side could be expressed as a fractional differentiation operator with order $\alpha = -1/2$. That is, the time evolution equation can be written as

$$\frac{d}{dt} C(t) + i\Delta_c C(t) = -\beta_1^{3/2} e^{-i\pi/4} \Gamma(1/2) \frac{d^{-1/2}}{dt^{-1/2}} C(t) \quad (8)$$

with gamma function $\Gamma(x)$. This fractional differential equation could be solved by manipulating the fractional operator. Here we used the mathematical steps of applying the integral operator (d^{-1}/dt^{-1}) and the fractional differentiation operator $d^{3/2}/dt^{3/2}$ to obey the law of exponents for fractional integrals (from Dirichlet’s formula) and the definition of the fractional derivative [23]. The former step gave

$$C(t) - C(0) + i\Delta_c \frac{d^{-1}}{dt^{-1}} C(t) + \beta_1^{3/2} e^{-i\pi/4} \sqrt{\pi} \frac{d^{-3/2}}{dt^{-3/2}} C(t) = 0. \quad (9)$$

The latter step yielded the fractional form of the time evolving equation

$$\begin{aligned} \frac{d^{3/2}}{dt^{3/2}} C(t) + i\Delta_c \frac{d^{1/2}}{dt^{1/2}} C(t) + \sqrt{\pi} \beta_1^{3/2} e^{-i\pi/4} C(t) \\ = -\frac{1}{2\sqrt{\pi}} t^{-3/2}, \end{aligned} \quad (10)$$

which is defined to be a *fractional Langevin equation* of this atom–field interaction system. This equation describes the evolution of an excited atom in a PBG material whose time degree of freedom becomes stochastic [20] because the occurrence of the temporal fractional operator in the kinetic equation indicates a subordinated stochastic process. The directional process is related to a stochastic process with a stable probability distribution [25]. The parameter characterizing the stable distribution coincides with the index of the temporal fractional operator in the corresponding kinetic equation.

We proceeded to solve the probability amplitude $C(t)$ by performing Laplace transform along with the formulae of the fractional Laplace transform and inverse Laplace transform on this fractional Langevin equation. First, the Laplace transform of $C(t)$ was thus given by

$$\tilde{C}(s) = \frac{\sqrt{s}}{s^{3/2} + i\Delta_c s^{1/2} - (i\beta)^{3/2}} \quad (11)$$

with $\beta^{3/2} = \beta_1^{3/2} \sqrt{\pi}$. Then we converted the variable $s^{1/2}$ as X and rewrote this equation as a sum of partial fractions

$$\tilde{C}(X) = \frac{a_1}{(X - X_1)} + \frac{a_2}{(X - X_2)} + \frac{a_3}{(X - X_3)}. \quad (12)$$

Here X_n are the roots of $X^3 + i\Delta_c X - (i\beta)^{3/2} = 0$ and a_n are X_n -related coefficients. X_n and a_n are expressed as

$$X_1 = \beta^{1/2} (\eta_+ + \eta_-) e^{i\pi/4}, \quad (13)$$

$$X_2 = \beta^{1/2} (\eta_+ e^{-i\pi/6} - \eta_- e^{i\pi/6}) e^{-i\pi/4}, \quad (14)$$

$$X_3 = \beta^{1/2} (\eta_+ e^{i\pi/6} - \eta_- e^{-i\pi/6}) e^{i3\pi/4}, \quad (15)$$

with

$$\eta_{\pm} = \left[\frac{1}{2} \pm \frac{1}{2} \sqrt{\left(1 + \frac{4\Delta_c^3}{27\beta^3}\right)} \right]^{1/3} \quad (16)$$

and

$$a_n = \frac{X_n}{(X_n - X_j)(X_n - X_m)} \quad (n \neq j \neq m; n, j, m = 1, 2, 3). \quad (17)$$

We can easily apply the inverse Laplace transform to this form of partial fraction by using the formula for the inverse fractional Laplace transform

$$L^{-1} \left\{ \frac{1}{s^{1/2} - a} \right\} = E_t \left(-\frac{1}{2}, a^2 \right) + a e^{a^2 t}. \quad (18)$$

This procedure gave the probability amplitude

$$C(t) = \sum_{n=1}^3 a_n \left[E_t \left(-\frac{1}{2}, X_n^2 \right) + X_n e^{X_n^2 t} \right], \quad (19)$$

where $E_t(\alpha, a) = t^\alpha \sum_{n=0}^{\infty} \frac{(at)^n}{\Gamma(\alpha+n+1)}$ is the fractional exponential function of variable t , order α , and constant a . We have the dynamics of spontaneous emission for the system by plotting the excited-state probability density $P(t) = |A(t)|^2 = |C(t)|^2$, which has no multiple-valued problem. This probability amplitude could be further expressed by the error function as

$$A(t) = e^{i\Delta_c t} \sum_{n=1}^3 a_n \left\{ X_n + Y_n \left[\text{Erf} \left(\sqrt{X_n^2 t} \right) \right] \right\} e^{X_n^2 t} \quad (20)$$

with $Y_n = \sqrt{X_n^2}$ ($n = 1, 2, 3$), which is the result of previous studies [14, 15]. However, this expression is a multiple-valued problem because the square roots of complex numbers X_n^2 will introduce multiple-valued complex numbers to Y_n . When the numerical results are shown, every complex sheet of X_n^2 has

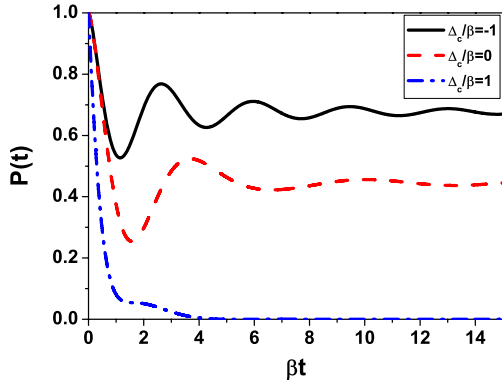


Figure 1. Excited-state probability density, $P(t) = |A(t)|^2$, as a function of βt for various values of the atomic detuning frequency ($\Delta_c = \omega_{21} - \omega_c$), $\Delta_c/\beta = -1$ (inside the band gap), $\Delta_c/\beta = 0$ (at the band edge) and $\Delta_c/\beta = +1$ (within the allowed band).

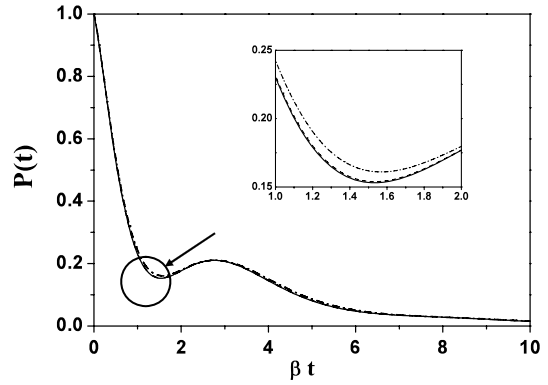


Figure 2. Excited-state probability density, $P^s(t) = |A^s(t)|^2$, for the atomic detuning frequency $\Delta_c/\beta = 0.3$ with three values of smoothing parameter $\epsilon = 0$ (solid line), $\epsilon = 10^{-5}$ (dashed line), $\epsilon = 10^{-3}$ (dot dashed line). The difference of these lines marked by a circle is enlarged and shown in the inset.

to be checked one by one to avoid errors. Obviously, our results with fractional calculus are mathematically rigorous and concise. Besides, our result in figure 1 shows that there is no steady photon–atom bound state for the atomic transition frequency lying in the proximity of allowed band edge, which is very different from the results of previous studies [14, 15].

In order to fortify the accuracy of the result of no unphysical photon–atom bound state in the allowed band, we proceeded to apply the ‘cut-off smoothing’ density of state (DOS) to investigation of the behavior of the system near the allowed band edge [24]. As mentioned before, the DOS in the isotropic single band model $\rho(\omega) \propto (\omega - \omega_c)^{-1/2}\theta(\omega - \omega_c)$ with Heaviside step function θ has a weak singularity for $\omega \rightarrow \omega_c$ (near band edge). This singular behavior is treated by the ‘cut-off smoothing DOS’ $\rho^s(\omega) \propto \lim_{\epsilon \rightarrow 0} \frac{(\omega - \omega_c)^{1/2}}{(\omega - \omega_c + \epsilon)}\theta(\omega - \omega_c)$ in realistic photonic crystals [24]. Here ϵ is the smoothing parameter and the superscript s denotes the ‘cut-off smoothing DOS’ case. The excited-state probability amplitude $A^s(t) = e^{i\Delta_c t} C^s(t)$ could be exactly solved by performing a Laplace transform on the memory kernel and time evolving equation (5). These procedures gave the Laplace transform of probability amplitude $C^s(t)$ as

$$\tilde{C}^s(s) = \frac{1}{s + i\Delta_c + \tilde{G}(s)} \quad (21)$$

with $\tilde{G}(s) = \frac{\beta^{3/2} e^{-i\pi/4}}{\sqrt{s + \sqrt{i\epsilon}}}$. This expression could be further rewritten in terms of the roots, X_n^s ($n = 1, 2, 3$), of $z^3 + \sqrt{i\epsilon}z^2 + i\Delta_c z + (i\sqrt{i\epsilon}\Delta_c - \beta^{3/2}i^{3/4}) = 0$ as

$$\tilde{C}^s(s) = \frac{\sqrt{s + \sqrt{i\epsilon}}}{\prod_{n=1}^3 (\sqrt{s} - X_n^s)} = \sum_{n=1}^3 \frac{a_n^s}{\sqrt{s} - X_n^s}, \quad (22)$$

where

$$X_1^s = \beta^{1/2}(\eta_+^s + \eta_-^s) e^{i\pi/4} - \frac{\sqrt{i\epsilon}}{3} \quad (23)$$

$$X_2^s = \beta^{1/2}(\eta_+^s e^{-i\pi/6} - \eta_-^s e^{i\pi/6}) e^{-i\pi/4} - \frac{\sqrt{i\epsilon}}{3} \quad (24)$$

$$X_3^s = \beta^{1/2}(\eta_+^s e^{i\pi/6} - \eta_-^s e^{-i\pi/6}) e^{i3\pi/4} - \frac{\sqrt{i\epsilon}}{3} \quad (25)$$

with

$$\eta_{\pm}^s = \left(\frac{\chi}{2} \pm \frac{\sqrt{\xi}}{2} \right)^{1/3}, \quad \chi = 1 - \frac{2}{3} \frac{\Delta_c \sqrt{\epsilon}}{\beta^{3/2}} - \frac{2}{27} \frac{\epsilon^{3/2}}{\beta^{3/2}},$$

$$\xi = \chi^2 + \frac{4}{27} \left(\frac{\Delta_c - \epsilon/3}{\beta} \right)^3 \quad (26)$$

and

$$a_n^s = \frac{X_n^s + \sqrt{i\epsilon}}{(X_n^s - X_j^s)(X_n^s - X_m^s)} \quad (n \neq j \neq m; n, j, m = 1, 2, 3). \quad (27)$$

Here again we used fractional calculus (fractional inverse Laplace transform) to obtain the excited-state probability amplitude

$$A^s(t) = e^{i\Delta_c t} \sum_{n=1}^3 a_n^s \left[E_t \left(-\frac{1}{2}, (X_n^s)^2 \right) + X_n^s e^{(X_n^s)^2 t} \right]. \quad (28)$$

As we study how the system behaves near the allowed band edge, we choose the detuning frequency $\Delta_c = 0.3\beta$ with smoothing parameters $\epsilon = 10^{-3}, 10^{-5}, 0$, respectively, in figure 2. It could be observed that the excited-state probability density $P^s(t) = |A^s(t)|^2$ has a small oscillatory behavior in the short time regime but approaches zero in the long-time limit. This means that there is really no steady photon–atom bound state for the atomic transition frequency lying in the proximity of the allowed band edge. Actually, we have plotted all the behavior of the system inside the allowed band in figure 3. For the atomic resonant transition frequency located deep inside the allowed band ($\Delta_c/\beta_1 = 10$) or very close to the band edge ($\Delta_c/\beta_1 = 0.01$), we found that these probabilities all show decaying characteristics in the long-time limit. That is, the photon located within the allowed band will not strongly interact with atom so a photon–atom bound state will not be formed. This result could be verified analytically from the mathematical expression of the excited-state probability amplitude $A^s(t) = e^{i\Delta_c t} \sum_{n=1}^3 a_n^s [E_t(-\frac{1}{2}, (X_n^s)^2) + X_n^s e^{(X_n^s)^2 t}]$.

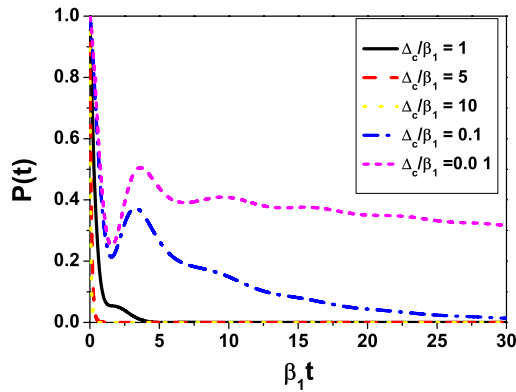


Figure 3. Excited-state probability density, $P(t) = |A(t)|^2$, for various values of atomic detuning frequency inside the allowed band ($\Delta_c/\beta > 0$).

For the positive detuning (inside the allowed band $\Delta_c > 0$), both terms in the square bracket will asymptotically cancel each other out as time approaches infinity ($t \rightarrow \infty$). We get the correct depiction of the dynamics of spontaneous emission in a PBG reservoir through fractional calculus, which is proposed as a better mathematical method for studying the behavior of the atom–field interaction in photonic crystals.

3. Conclusion

The dynamics of the spontaneous emission of an atom in a photonic crystal with one-band isotropic band structure can be treated by fractional calculus using either a singular or ‘cut-off smoothing’ photon DOS. For the first time to our knowledge we show that it is a fractal phenomenon that induces the long-time memory of spontaneous emission in the photonic crystal. Solving the time evolving equation of the probability amplitude for the system governed by the fractional memory kernel described by the singular density of states, we obtained rigorous solutions without encountering a multiple-valued problem. Besides, we found that there is no unphysical state of a fractionalized atomic population in the excited state when the resonant atomic frequency lies in the allowed band, even extremely close to the band edge. This result was validated by the ‘cut-off smoothing DOS’ with fractional calculus. We suggest that the correct description of the dynamics of spontaneous emission in a photonic crystal should be expressed in terms of fractional calculus. This mathematical method, used here with the isotropic model, can be easily extended to the anisotropic case and to study interesting effects

such as the enhancement of the index of refraction with greatly reduced absorption, electromagnetically induced transparency, and optical amplification without population inversion.

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