

## Microscopic Theory of Modified Spontaneous Emission in a Dielectric

P. R. Berman

*Michigan Center for Theoretical Physics, FOCUS Center, and Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1120, USA*

P.W. Milonni

*Theoretical Division (T-DOT), Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*  
(Received 3 September 2003; published 6 February 2004)

The modification of the radiative decay rate of a source atom embedded in a uniform, isotropic dielectric is calculated to first order in the density of the dielectric atoms using a microscopic approach. In contrast to the recent results of Crenshaw and Bowden [Phys. Rev. Lett. **85**, 1851 (2000)], the decay rate is found to be consistent with macroscopic theories based on quantization of the field in the dielectric.

DOI: 10.1103/PhysRevLett.92.053601

PACS numbers: 42.50.Ct, 03.65.Ge, 32.80.-t, 42.25.Bs

The theory of radiative processes in dielectric media continues to attract considerable attention, not only for its fundamental importance but also because of its relevance to various applications [1]. Most theories can be characterized as “macroscopic” in that they are based on field quantization in the presence of the dielectric [2]. In a recent development Crenshaw and Bowden [3] have taken a microscopic approach to the modification of the spontaneous decay rate of an atom embedded in a dielectric medium. They obtain the decay rate  $\Gamma = \ell\Gamma_0$ , where  $\Gamma_0$  is the rate in free space,  $\ell = (\frac{n^2+2}{3})$ , and  $n$  is the index of refraction at the frequency of the emitting atom. Their enhancement factor differs markedly from both the “virtual” [ $\ell = n(\frac{n^2+2}{3})^2$ ] and “real” [ $\ell = n(\frac{3n^2}{2n^2+1})^2$ ] cavity results of macroscopic theories [1,2,4]. The quantity  $\ell$  in the macroscopic theories is the product of  $n$  and a local-field correction factor. The familiar Lorentz-Lorenz correction factor  $(\frac{n^2+2}{3})^2$  is obtained using the concept of a virtual cavity surrounding the emitter in an otherwise continuous dielectric, whereas the factor  $(\frac{3n^2}{2n^2+1})^2$  is obtained if it is assumed that the emitter is surrounded by a real spherical cavity. As reviewed by Toptygin [1], most of the experiments reported thus far on spontaneous emission in a dielectric support the real-cavity correction factor. The recent experiments by Kumar *et al.*, Rikken and Kessener, and Schuurmans *et al.* [4] appear to provide particularly compelling evidence for the real-cavity correction factor, although different forms of the correction factor should apply under different circumstances [1]. In light of these results, it is important to reconcile the microscopic and macroscopic theories.

In this Letter we present a different approach to the microscopic theory and calculate corrections to the decay rate that are linear in the dielectric density  $N$ . Our results are perfectly consistent with the macroscopic theories to this order. We identify the point at which our approach differs from Crenshaw and Bowden’s, and we show that our result for the enhancement factor also follows from a

classical microscopic theory of a dipole oscillator in a dielectric medium. Our first-order theory does not distinguish between the virtual-cavity and real-cavity formulas for the local-field correction. However, the calculations can be extended to include corrections of order  $N^2$ , which can be compared with these local-field formulas when they are expanded to second order in  $\delta n$ .

The physical system we choose is a source atom located at  $\mathbf{R} = \mathbf{0}$  in a uniformly distributed bath of atoms constituting the dielectric medium. The source atom is modeled as a two-state atom (upper state 2 and lower state 1) having a transition frequency  $\omega_0$  and a transition dipole moment aligned along the  $z$  axis. The bath atoms have  $J = 0$  ground states separated by frequency  $\omega$  from  $J = 1$  excited states. It is assumed that  $|\omega - \omega_0|/\omega \ll 1$ , allowing us to make the rotating-wave approximation (RWA). At  $t = 0$  the source atom is in its excited state, all the bath atoms are in their ground states, and there are no photons in the field. We calculate the decay rate of the source atom including corrections that are linear in the density of the bath atoms.

The Hamiltonian of the system is  $H = H_0 + V$ , where

$$H_0 = \frac{\hbar\omega_0}{2}\sigma_z + \sum_j \sum_{m=-1}^1 \frac{\hbar\omega}{2}\sigma_z^{(j)}(m) + \hbar\omega_k a_k^\dagger a_k, \quad (1)$$

$$V = \hbar g_k (\sigma_+ a_k - a_k^\dagger \sigma_-) + \hbar [g'_k(m)\sigma_+^{(j)}(m)a_k e^{i\mathbf{k}\cdot\mathbf{R}_j} + g'_k(m)^* a_k^\dagger \sigma_-^{(j)}(m) e^{-i\mathbf{k}\cdot\mathbf{R}_j}], \quad (2)$$

$$g_k = -i \left( \frac{\omega_k}{2\hbar\epsilon_0 \mathcal{V}} \right)^{1/2} \mu(\epsilon_k)_0, \quad (3)$$

$$g'_k(m) = -i \left( \frac{\omega_k}{2\hbar\epsilon_0 \mathcal{V}} \right)^{1/2} \mu'(\epsilon_k)_m^*,$$

$\sigma_\pm$  are raising (+) and lowering (−) operators for the source atom;  $\sigma_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)$ ,  $\mu$  is the dipole moment matrix element of the source atom (assumed to be

real);  $\sigma_{\pm}^{(j)}(m)$  are raising and lowering operators for the  $m$  sublevel of the  $J = 1$  excited level of bath atom  $j$ ;  $\sigma_z^{(j)}(m) = (|m\rangle^{(j)}\langle m| - |g\rangle^{(j)}\langle g|)$  is the population difference operator between the excited state ( $J = 1, m$ ) and the ground state ( $g, J = 0$ ) of bath atom  $j$ ;  $\mu'$  is  $\sqrt{3}$  times the reduced matrix element (assumed to be real) of the dipole operator for the bath atoms;  $a_k$  and  $a_k^\dagger$  are the usual field annihilation and creation operators;  $\mathcal{V}$  is the quantization volume; and  $k$  denotes a plane-wave mode of the field. The polarization index on  $\epsilon_k$  has been suppressed since only the  $\theta_k$  component of the polarization vector enters into the calculation. Explicitly,

$$(\epsilon_k)_0 = -\sin(\theta_k) \quad (\epsilon_k)_{\pm 1} = \mp \frac{\cos(\theta_k)e^{\pm i\phi_k}}{\sqrt{2}}. \quad (4)$$

A summation convention is used in Eqs. (1) and (2) and subsequent equations, such that any repeated subscript on the right-hand side of an equation is summed over unless it also appears on the left-hand side. Equations (1) and (2) describe a system in which the source and bath atoms interact via the *free-space* quantized radiation field. The source atom radiates and this radiation is scattered back to the source atom by the bath atoms, modifying the source atom's decay rate. Interactions involving scattering

at *two or more* bath atoms are neglected, since they contribute terms that are not linear in the bath density. Our goal here is to obtain a result that is “exact” to first order in the bath-atom density.

In an interaction representation and the RWA, the equations of motion for the relevant state amplitudes are

$$i\hbar\dot{b}_{2,0} = V_{2,0;1,k}e^{-i(\omega_k-\omega_0)t}b_{1,k}, \quad (5a)$$

$$i\hbar\dot{b}_{1,k} = V_{1,k;2,0}e^{i(\omega_k-\omega_0)t}b_{2,0} + V_{1,k;m_j,0}e^{i(\omega_k-\omega)t}b_{m_j,0}, \quad (5b)$$

$$i\hbar\dot{b}_{m_j,0} = V_{m_j,0;1,k}e^{-i(\omega_k-\omega)t}b_{1,k}, \quad (5c)$$

subject to the initial condition  $b_{2,0}(0) = 1$ . The amplitude  $b_{2,0}$  is associated with the state in which the source atom is excited, all the bath atoms are in their ground states, and there are no photons in the field. Similarly,  $b_{m_j,0}$  is the amplitude for the state in which bath atom  $j$  is in excited sublevel  $m$ , all the other bath atoms and the source atom are in their ground states, and there are no photons in the field, while amplitude  $b_{1,k}$  refers to the state in which all atoms are in their ground states and there is a photon in mode  $k$ . By formally integrating Eq. (5b) and substituting the result into (5a) and (5c), one obtains the coupled equations

$$\dot{b}_{2,0} = -\frac{1}{\hbar^2} \int_0^t V_{2,0;1,k}V_{1,k;2,0}e^{-i(\omega_k-\omega_0)(t-t')}b_{2,0}(t')dt' - \frac{e^{i\Delta t}}{\hbar^2} \int_0^t V_{2,0;1,k}V_{1,k;m_j,0}e^{-i(\omega_k-\omega)(t-t')}b_{m_j,0}(t')dt', \quad (6a)$$

$$\dot{b}_{m_j,0} = -\frac{1}{\hbar^2} \int_0^t V_{m_j,0;1,k}V_{1,k;m_j,0}e^{-i(\omega_k-\omega)(t-t')}b_{m_j,0}(t')dt' - \frac{e^{-i\Delta t}}{\hbar^2} \int_0^t V_{m_j,0;1,k}V_{1,k;2,0}e^{-i(\omega_k-\omega_0)(t-t')}b_{2,0}(t')dt', \quad (6b)$$

where  $\Delta = \omega_0 - \omega$ . We have dropped terms involving the coupling of the bath atoms to one another, as discussed above. In the Weisskopf-Wigner approximation the first term on the right-hand side of Eq. (6a) becomes  $-\gamma b_{2,0}(t)$  and that in Eq. (6b) becomes  $-\gamma' b_{m_j,0}(t)$ , where  $\gamma = [(\frac{1}{4\pi\epsilon_0})2\mu^2\omega_0^3/3\hbar c^3]$  is half the decay rate of state 2 and  $\gamma' = [(\frac{1}{4\pi\epsilon_0})2\mu'^2\omega^3/3\hbar c^3]$  is half the decay rate of level  $m$  (and is independent of  $m$ ). The summations over  $\mathbf{k}$  in the remaining terms can be carried out in a straightforward manner by transforming them to integrals using the prescription  $\sum_{\mathbf{k}} \rightarrow [\mathcal{V}/(2\pi)^3] \int d\mathbf{k}$ . For example, using Eqs. (2)–(4), one finds

$$\frac{1}{\hbar^2} \sum_{\mathbf{k}} V_{2,0;1,k}V_{1,k;m_j,0}e^{-i(\omega_k-\omega)(t-t')} = \frac{\gamma}{\omega_0^3} (\mu'/\mu)\pi^{-1} \int_0^\infty d\omega_k \omega_k^3 e^{-i(\omega_k-\omega)(t-t')} f_m(k, \mathbf{R}_j), \quad (7)$$

where

$$f_0(k, \mathbf{R}) = \frac{3}{8\pi} \int d\Theta_k \sin^2(\theta_k) e^{-i\mathbf{k}\cdot\mathbf{R}}, \quad (8)$$

$$f_{\pm 1}(k, \mathbf{R}) = \pm \frac{3}{8\pi} \frac{1}{\sqrt{2}} \int d\Theta_k \sin(\theta_k) \cos(\theta_k) e^{\pm i\phi_k} e^{-i\mathbf{k}\cdot\mathbf{R}},$$

and  $k = \omega_k/c$ . The integral over the solid angle is most conveniently carried out using

$$e^{i\mathbf{k}\cdot\mathbf{R}} = 4\pi \sum_{m=-\ell}^{\ell} i^\ell Y_{\ell m}(\theta_k, \phi_k) Y_{\ell m}^*(\theta, \phi) j_\ell(kR), \quad (9)$$

where  $(\theta, \phi)$  are the polar angles associated with  $\mathbf{R}$  and  $j_\ell$  is a spherical Bessel function. Thus one can replace Eqs. (6) by

$$\dot{b}_{2,0} = -\gamma b_{2,0} - \gamma(\mu'/\mu)e^{i\Delta t} \times \int_0^t G_m(\mathbf{R}_j, \tau, \omega) b_{m_j,0}(t-\tau) d\tau, \quad (10a)$$

$$\dot{b}_{m_j,0} = -\gamma' b_{m_j,0} - \gamma(\mu'/\mu)e^{-i\Delta t} \times \int_0^t H_m(\mathbf{R}_j, \tau, \omega_0) b_{2,0}(t-\tau) d\tau, \quad (10b)$$

where

$$G_m(\mathbf{R}, \tau, \omega_0) = \pi^{-1} \omega_0^{-3} \int_{-\infty}^{\infty} d\omega_k \omega_k^3 e^{-i(\omega_k-\omega_0)\tau} \times f_m(k, \mathbf{R}); \quad (11a)$$

$$H_m(\mathbf{R}, \tau, \omega_0) = \pi^{-1} \omega_0^{-3} \int_{-\infty}^{\infty} d\omega_k \omega_k^3 e^{-i(\omega_k-\omega_0)\tau} \times f_m^*(k, \mathbf{R}); \quad (11b)$$

$$f_0(k, \mathbf{R}) = \sqrt{4\pi} \left[ j_0(kR)Y_{00}(\theta, \phi) + \frac{1}{\sqrt{5}}j_2(kR)Y_{20}(\theta, \phi) \right]; \quad (12a)$$

$$f_{\pm 1}(k, \mathbf{R}) = -\sqrt{4\pi}\sqrt{3/20}j_2(kR)Y_{2\pm 1}(\theta, \phi), \quad (12b)$$

and the integrals over  $\omega_k$  in Eqs. (11) have been extended to  $-\infty$  [5]. In what follows we drop the distinction between  $\omega_0$  and  $\omega$ , which is consistent with the RWA. Equations (10) describe the coupling of the excited-state amplitudes of source and bath atoms.

The integral over  $\omega_k$  can be carried out by replacing  $\omega_k$  by  $\omega_0$  and  $k$  by  $k_0 = \omega_0/c$  in the integrand *except* where these quantities appear as arguments of exponentials or trigonometric functions. Thus

$$G_m(\mathbf{R}, \tau, \omega_0) = [e^{ik_0R}\delta(\tau - R/c)M_m(k_0, \mathbf{R}) + e^{-ik_0R}\delta(\tau + R/c)M_m^*(k_0, \mathbf{R})]; \quad (13a)$$

$$H_m(\mathbf{R}, \tau, \omega_0) = [e^{ik_0R}\delta(\tau - R/c)P_m(k_0, \mathbf{R}) + e^{-ik_0R}\delta(\tau + R/c)P_m^*(k_0, \mathbf{R})], \quad (13b)$$

where

$$M_0(k_0, \mathbf{R}) = P_0(k_0, \mathbf{R}) = -\sqrt{4\pi} \left[ \frac{i}{\rho}Y_{00}(\theta, \phi) + \frac{1}{\sqrt{5}} \left\{ i \left( \frac{3}{\rho^3} - \frac{1}{\rho} \right) + \frac{3}{\rho^2} \right\} Y_{20}(\theta, \phi) \right]; \quad (14a)$$

$$M_{\pm 1}(k_0, \mathbf{R}) = \sqrt{4\pi}\sqrt{3/20} \left[ i \left( \frac{3}{\rho^3} - \frac{1}{\rho} \right) + \frac{3}{\rho^2} \right] Y_{2\pm 1}(\theta, \phi); \quad (14b)$$

$$P_{\pm 1}(k_0, \mathbf{R}) = \sqrt{4\pi}\sqrt{3/20} \left[ i \left( \frac{3}{\rho^3} - \frac{1}{\rho} \right) + \frac{3}{\rho^2} \right] Y_{2\pm 1}^*(\theta, \phi); \quad (14c)$$

and  $\rho = k_0R$ . Equation (10b) can be integrated formally, assuming that  $b_{2,0}$  varies slowly on a time scale of order  $1/\Delta$ . Substituting the resulting equation into Eq. (10a), one finds

$$\begin{aligned} \dot{b}_{2,0} = & -\gamma b_{2,0}(t') + \frac{\gamma\gamma'}{\gamma' - i\Delta} \int_0^t G_m(\mathbf{R}_j, \tau, \omega_0) d\tau \\ & \times \int_0^{t-\tau} H_m(\mathbf{R}_j, \tau', \omega_0) b_{2,0}(t - \tau - \tau') d\tau', \quad (15) \end{aligned}$$

where the relationship  $\gamma' \approx \gamma(\mu'/\mu)^2$  has been used. The sum over the bath atoms  $j$  is now converted to an integral over all space,  $\sum_j \rightarrow N \int d\mathbf{R}$ , where  $N$  is the bath density. The imaginary part of the integral over  $R$  is perfectly well behaved near  $R = 0$ ; as such, the terms proportional to  $\delta(\tau + R/c)$  can be excluded since they can contribute only at  $R = 0$  and this is a set of measure zero. Retaining only the  $\delta(\tau - R/c)$  terms, neglecting retardation effects [i.e., taking  $b_{2,0}(t - 2R/c) \approx b_{2,0}(t)$  for the slowly varying amplitude  $b_{2,0}(t)$ ], and using the orthogonality of the spherical harmonics, one obtains [6]

$$\dot{b}_{2,0} = -\gamma b_{2,0} + \frac{2\pi N \gamma \gamma'}{\Delta k_0^3} (A_0 + A_1 + A_{-1}) b_{2,0}, \quad (16)$$

where we have used the assumption that  $|\Delta| \gg \gamma'$ . The radial integrals  $A_m$  are given by [7]

$$A_0 = 2\text{Im} \int_0^\infty d\rho \left\{ \frac{-1}{\rho^2} + \frac{1}{5} \left[ i \left( \frac{3}{\rho^3} - \frac{1}{\rho} \right) + \frac{3}{\rho^2} \right]^2 \right\} \rho^2 e^{2i\rho};$$

$$A_1 = A_{-1} = 2\text{Im} \int_0^\infty d\rho \left\{ \frac{3}{20} \left[ i \left( \frac{3}{\rho^3} - \frac{1}{\rho} \right) + \frac{3}{\rho^2} \right]^2 \right\} \rho^2 e^{2i\rho}.$$

A convergence factor must be added to ensure that the integrals are well behaved at infinity [6]. When a convergence factor of the form  $e^{-g\rho}$  is included, the integrals can be evaluated analytically. Setting  $g = 0$  in the final result, one finds  $A_0 = -2$  and  $A_1 = -3/4$ . It then follows that

$$\dot{b}_{2,0} = -\gamma\ell b_{2,0},$$

where

$$\ell = 1 - \frac{1}{4\pi\epsilon_0} \left( \frac{4\pi N \mu'^2}{3\hbar\Delta} \right) \left( 2 + \frac{3}{2} \right). \quad (17)$$

The index of refraction for the dilute dielectric medium considered here is  $n \approx 1 - \frac{1}{4\pi\epsilon_0} \left( \frac{2\pi N \mu'^2}{\hbar\Delta} \right) = 1 + \delta n$ , implying from Eq. (17) that  $\ell = 1 + 7\delta n/3$ . This is the same result that would be obtained if either of the enhancement factors  $\ell = n(\frac{n^2+2}{3})^2$  or  $\ell = n(\frac{3n^2}{2n^2+1})^2$  assumed in macroscopic theories is expanded to first order in  $\delta n = n - 1$ . In other words, our result for the spontaneous emission rate is perfectly consistent with that given by the macroscopic theories involving constitutive relations and field quantization in the dielectric. In our microscopic treatment the modification of the spontaneous emission rate is a consequence of dipole-dipole interactions between the source atom and the bath atoms, and we do not have the same clear separation of local-field effects and ‘‘index’’ effects as in the macroscopic theories. In contrast to macroscopic theories, it is not necessary to exclude a small volume around the source atom to calculate the local-field effects in our approach. The modification of the decay rate resulting from the bath is calculated exactly as an integral over all space.

We can now comment on why our result differs from Crenshaw and Bowden's. The sum over bath atoms of the product of the  $G$  and  $H$  functions in our Eq. (15) is evaluated *directly* by replacing the sum by an integral. At a comparable stage of their calculation, however, Crenshaw and Bowden make an approximation which, in our notation, amounts to integrating the  $G$  and  $H$  factors in Eq. (15) *separately* over the bath atoms, using a spatial averaging and their near dipole-dipole interaction [8]. For this reason, even if we had assumed that the

dielectric consists of two-state atoms or oscillators with  $J = 1, m = 0$  excited states, our result,  $\ell = 1 + 4\delta n/3$  [obtained by setting the  $A_{\pm 1}$  terms in Eq. (16) equal to zero], still differs from the Crenshaw and Bowden result,  $\ell = 1 + 2\delta n/3$ , expanded to first order in  $\delta n$ . The additional contributions from the  $A_{\pm 1}$  terms in Eq. (16) *must* be included, however, if one is to correctly model an isotropic dielectric. A two-state model necessarily results in a dielectric that has different refractive indices in different directions. One must include ground and excited states with all appropriate magnetic sublevels to model an isotropic dielectric. The simplest case is the  $J = 0$  to  $J = 1$  transition considered in this work.

Our result for  $\ell$  can be obtained using a *classical* approach in which it is assumed that a dipole oscillator having frequency  $\omega_0$  is embedded in a dielectric consisting of dipoles having frequency  $\omega$ . The equations of motion for the coupled linear dipole oscillators are

$$\ddot{x} + 2\gamma\dot{x} + \omega_0^2 x = \frac{e}{m} \hat{x} \cdot \mathbf{E}(t),$$

$$\ddot{x}_j + 2\gamma'\dot{x}_j + \omega^2 x_j = \frac{e}{m} \hat{x}_j \cdot \mathbf{E}_j(t).$$

Here  $\gamma = \frac{1}{4\pi\epsilon_0} \frac{e^2 \omega_0^2}{3mc^3} \cong \gamma'$ ,  $x$  is the displacement of the charge  $e$  for the source dipole,  $\hat{x}$  gives the dipole direction, and  $\mathbf{E}(t)$ , equal to the sum of the fields produced by dipoles of the dielectric medium, is the field acting on the source dipole. Similarly  $x_j$  and  $\hat{x}_j$  are the corresponding displacement and direction of the dielectric dipole  $j$ , and  $\mathbf{E}_j(t)$  is the field on dipole  $j$  due to all the other dipoles. As in our quantum model we approximate the field on dipole  $j$  by the field due to the source dipole alone to calculate terms that are first order in the dielectric density.

We write  $x(t) = S(t)e^{-i\omega_0 t}$ ,  $x_j(t) = S_j(t)e^{-i\omega t}$  and assume that  $S(t)$  and  $S_j(t)$  are slowly varying compared with  $e^{-i\omega_0 t}$ . Then the equations of motion for our classical model become

$$\dot{S} = -\gamma S + \sum_j F_j S_j, \quad (18)$$

$$\dot{S}_j = -i(\Delta + i\gamma')S_j + F_j S, \quad (19)$$

where  $\Delta = \omega_0 - \omega$ ,

$$F_j = \frac{1}{4\pi\epsilon_0} \frac{ie^2}{2m\omega_0} \left[ [\hat{\mu} \cdot \hat{\mu}_j - (\hat{\mu} \cdot \hat{R}_j)(\hat{\mu}_j \cdot \hat{R}_j)] \frac{k_0^2}{R_j} + [3(\hat{\mu} \cdot \hat{R}_j)(\hat{\mu}_j \cdot \hat{R}_j) - \hat{\mu} \cdot \hat{\mu}_j] \times \left( \frac{1}{R_j^3} - \frac{ik_0}{R_j^2} \right) \right] e^{ik_0 R_j}, \quad (20)$$

$\hat{\mu} = \hat{x}$ ,  $\hat{\mu}_j = \hat{x}_j$ , and  $\hat{R}_j$  is the unit vector pointing from the source dipole to the dipole  $j$  a distance  $R_j$  away. We take  $\omega \cong \omega_0$  except, of course, in the detuning  $\Delta$ . Proceeding now in much the same way as in the quantum calculation (including an average over the relative orien-

tation of the source and dielectric dipoles), and noting that  $\delta n = -\frac{1}{4\pi\epsilon_0} \frac{N\pi e^2}{m\omega_0 \Delta}$  for  $|\Delta| \gg \gamma$  in this classical model, we find the enhancement factor for the decay rate is  $\ell = 1 + \frac{7}{3}\delta n$ , exactly as in our quantum calculation.

In summary, we have shown that a microscopic treatment of the modification of the decay rate of a source atom embedded in a dielectric, calculated to first order in the dielectric density, is consistent with macroscopic treatments based on quantization of the radiation field in the presence of a dielectric.

The research of P.R.B. is supported by the National Science Foundation under Grant No. PHY-0244841 and the FOCUS Center Grant and by the U.S. Army Research Office under Grant No. DAAD19-00-1-0412. We thank R.W. Boyd for informative discussions on the subject of local-field corrections.

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