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Enhanced and inhibited spontaneous emission from a pointlike quantum dot embedded in a finite dielectric sphere are treated classically. Numerical results given for a GaAs quantum dot embedded in an $Al_{0.4}Ga_{0.6}As$ sphere show the spontaneous emission rate is decreased by a factor of 80 when the radius of the sphere is small compared to the wavelength of emitted light. When the radius of the sphere is comparable to the wavelength, the spontaneous emission rate depends strongly on the radius. Sharp resonances, related to Mie resonances, with enhancements up to a factor of 100, are found in some cases.

While the dimensions of quantum-confined semiconductor systems are typically of the order of nanometers, the photons they emit have wavelengths around one micron. Thus, there is a fundamental mismatch between the length scales of the electromagnetic disturbance and that of the system creating it. In quantum well microcavity structures this mismatch can be overcome by embedding the active region in a superstructure, such as a distributed Bragg reflector,[1–3] which enhances the photon density of states for a selected optical transition.[4,5] Spontaneous emission rate enhancements up to a factor of 40 have been reported in quantum well devices,[6] showing the sensitivity of the spontaneous emission rate to the local dielectric environment.[7] In principle, the spontaneous emission rates of 1-D and 0-D confined systems are also influenced by their dielectric environment, though this problem has received, so far, relatively less attention.[2,8] The modification of the spontaneous emission rate in reduced dimensionality systems is of interest from both the practical standpoint of measuring quantum dot properties and, more fundamentally, from the standpoint of investigating quantum electrodynamic effects in finite systems.

In this communication, the modification of the spontaneous emission rate of a quantum dot by a spherical dielectric overlayer is discussed within the framework of classical electrodynamics. Numerical results, pertaining to a GaAs quantum dot embedded in a AlGaAs sphere, show that for spherically symmetric structures, with the dot at the center of the sphere (with dielectric constant ϵ), the spontaneous emission rate of the dot can be enhanced by a factor of $\epsilon^{1/2}\sim 3.5$ over that for the dot embedded in an infinite medium. In the case of nonspherically symmetric structures, with the dot off center of the dielectric sphere, stronger, resonant enhancements of the spontaneous emission rate by two orders of magnitude are found.

A model system of a classical, pointlike dipole oscillating in a uniform dielectric sphere of radius a is considered. Letting the center of the sphere define the origin, the position of the dot is αa. This structure chosen as an idealization of a structure which could result, for instance, from the vapor-phase synthesis of GaAs quantum dots[9] with subsequent vapor-phase growth of an epitaxial passivation layer. The assumption of a pointlike dipole is a reasonable approximation considering the radii of quantum dots, typically 5–10 nm, are well below optical wavelengths. The classical treatment gives the correct modification of the local electric fields and thus the correct behavior of the relative spontaneous emission rate when compared to quantum mechanical treatments. However, it does not treat interactions with the vacuum and thus cannot give an accurate indication of absolute emission rates.

The simplest case is the spherically symmetric one, with the dot at the center of the dielectric sphere (i.e., $\alpha=0$). The fields due to the dipole can be derived from the vector potential[10]

$$\mathbf{A}(\mathbf{r}) = -ik\mathbf{p}e^{ikr}/r,$$

where $\mathbf{p}$ is the dipole moment and $k=2\pi/\lambda$. By matching the fields inside and outside the sphere using the standard boundary conditions and then integrating the power emitted over all angles, the total power, P, radiated by the dipole as a function a can be derived. Normalizing P to the power emitted by the same dipole imbedded in an infinite medium of dielectric constant ϵ, P_0, the relative semiclassical emission rate $\Gamma_0=P/P_0$ is

$$\Gamma_0=\frac{\epsilon^{1/2}}{|\epsilon^{1/2}yj_1(y)[xh_1^{(1)}(x)]'-xh_1^{(1)}(x)[yj_1(y)]'|^2}. \qquad (1)$$

The primes refer to total differentiation with respect to the arguments, $x=ka$ and $y=ka\epsilon^{1/2}$. The functions $j_1(x)$ and $h_1^{(1)}(x)$ are spherical Bessel and Hankel functions, respectively.

The result of a numerical calculation of Γ_0, with $\lambda=700$ nm and $\epsilon=12.25$ is shown in Fig. 1 as a function of the radius, a. The values of λ and ϵ approximately correspond to the exciton energy of a 7 nm GaAs quantum dot passivated by $Al_{0.4}Ga_{0.6}As$. For small a, spontaneous emission is strongly inhibited whereas for larger values the emission rate oscillates with a regular period given by the asymptotic limits of the Bessel functions. In the limit of large a, the maximum of Γ_0 is $\epsilon^{1/2}$ and the minimum is $\epsilon^{-1/2}$.

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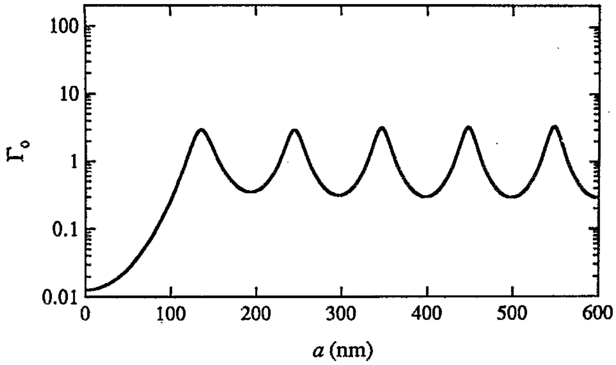

FIG. 1. The normalized spontaneous emission rate Γ_0 of a 7 nm quantum dot (transition wavelength 700 nm) at the center of an $Al_{0.4}Ga_{0.6}As$ dielectric sphere of radius a. For small a emission is inhibited. For larger values of a, emission is alternatively enhanced and inhibited.

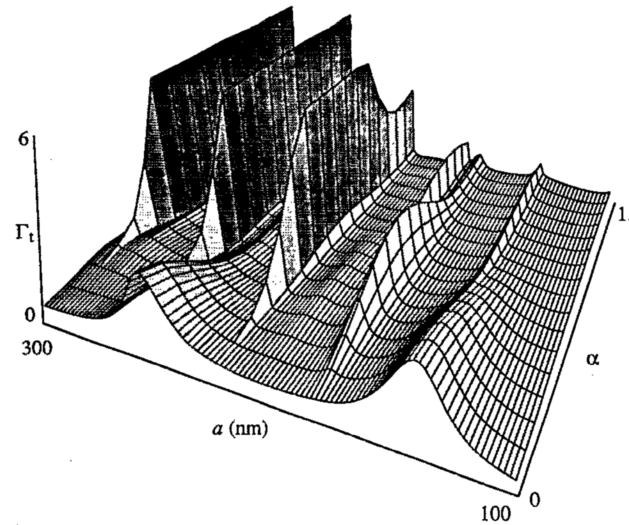

FIG. 2. The relative total spontaneous emission rate, Γ_t, as a function of the radius a and the asymmetry parameter α. Some of the peaks are cut off to emphasize the low lying structure. Along $\alpha=0$, the broad resonances of Fig. 2 are apparent. As α increases, resonances appear which are sharp functions of the radius a, but depend only weakly on α.

In the limit that $a\rightarrow 0$, $\Gamma_0\rightarrow 9/(\epsilon+2)^2\epsilon^{1/2}\cong 0.013$ and, to first order in a, Γ_0 is independent of a. This agrees with the quantum mechanical result.[7] Emission is inhibited since the dielectric lies entirely in the near, or static zone of the radiation field, where retardation effects are not important. The induced polarization of the dielectric screens the dipole and thus destructively interferes with the dipole's radiation. The suppression of Γ_0 may also be interpreted more broadly, within the framework of quantum electrodynamics (QED).[2,4,8] The dielectric support no modes when its diameter is much smaller than a wavelength and consequently suppresses the density of states. From the Golden Rule, the decrease in the spontaneous emission rate follows immediately.

The more general case, $1>\alpha>0$, can also be treated. In this case the solution contains higher order terms in the spherical harmonic expansion. The details of the calculation are given by Chew.[11] The relative emission rates for the dipole perpendicular and parallel to the dielectric surface are

$$\Gamma_{\perp}=\frac{3}{2}\,\epsilon^{1/2}\sum_{n=1}^{\infty}n(n+1)(2n+1)\frac{j_n^2(z)}{z^2}\frac{1}{|d_{e,n}|^2},\tag{2}$$

$$\Gamma_{\parallel}=\frac{3}{4}\,\epsilon^{1/2}\sum_{n=1}^{\infty}(2n+1)\left[\left(\frac{[zj_n(z)]'}{z}\right)^2\frac{1}{|d_{e,n}|^2}+\frac{j_n^2(z)}{|d_{m,n}|^2}\right],\tag{3}$$

where $z=\alpha ka$ is the position of the dot in the sphere and the functions $d_{e,n}$ and $d_{m,n}$ are

$$d_{e,n}=xh_n^{(1)}(x)[yj_n(y)]'-\epsilon^{1/2}yj_n(y)[xh_n^{(1)}(x)]',\tag{4}$$

$$d_{m,n}=\epsilon^{1/2}xh_n^{(1)}(x)[yj_n(y)]'-yj_n(y)[xh_n^{(1)}(x)]'.\tag{5}$$

A numerical calculation of the total emission rate, $\Gamma_t=(2\Gamma_{\parallel}+\Gamma_{\perp})/3$, for the range of values $100<a<300$ nm and $0<\alpha<1$ is shown in Fig. 2. Along the axis $\alpha=0$ broad resonances are evident (cf. Fig. 1). For $\alpha>0$, additional, resonances which are strongly sensitive to the dielectric radius, a, but relatively insensitive to α, are observed. To better emphasize these sharp features, cuts through the surfaces $\Gamma_{\parallel}$ and $\Gamma_{\perp}$ at $\alpha=0.6$ are shown in Figs. 3(a) and 3(b). The resonances in $\Gamma_{\parallel}$ are doubled due to coupling to both the electric and magnetic modes of the sphere. For certain values of a, enhancements of the spontaneous emission rate by a factor of 100 are found.

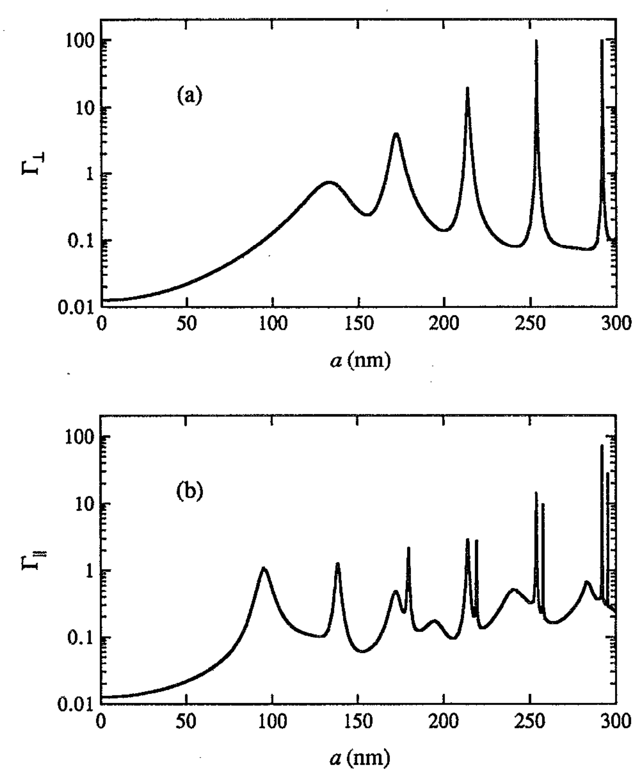

FIG. 3. (a) $\Gamma_{\perp}$ for $\alpha=0.6$. Several sharp resonances, due to a coincidence between the dipole and an internal standing wave in the particle, are clearly visible. (b) $\Gamma_{\parallel}$ for $\alpha=0.6$. The resonant peaks are doubled because the dipole couples to electric and magnetic modes of the sphere.

It should be recognized for a range of photon energies over which the dielectric constant ϵ does not vary too much, Figs. 1–3 may be equally interpreted as the photon energy dependence of the spontaneous emission rate for a fixed cluster size a. This arises from the functional dependence of Eqs. (1)–(5) on ka. Some of the predicted resonances, with Q's up to order 10^3, are sufficiently sharp to resolve individual optical phonons in GaAs.

The denominators, Eqs. (4) and (5), which are responsible for resonances in the Γ's, are the same as those in Mie-scattering coefficients,[12] and appear generally in the theory of waves interacting with a spherical geometry. While there is no direct experimental evidence for the specific effects predicted here, related resonances have been observed in many instances, including the enhanced Raman scattering of molecules on semiconductor clusters[13] and the QED enhancement of stimulated emission from microdroplets.[14] By suitable control of growth conditions, the possibility that the spontaneous emission rate from quantum dots may be precisely controlled appears strong. For many minority carrier devices, the spontaneous emission lifetime of the excited state plays a vital role in determining device performance. The use of resonance-enhanced dot's in high-speed applications or dots with inhibited spontaneous emission where extended carrier lifetimes are important may lead to improvements in the performance of quantum-dot based devices beyond those already anticipated due to quantum size effects.

In summary, the resonance enhancement of the spontaneous emission rate for a quantum dot embedded dielectric sphere has been studied. Numerical calculations for a GaAs dot embedded in a $Al_{0.4}Ga_{0.6}As$ dielectric reveal a complex resonance structure for the spontaneous emission rate as a function of the dielectric radius, a. For small a the spontaneous emission rate is inhibited by a factor of 80. For larger a, the spontaneous emission rate can be enhanced or inhibited, depending on a, and the location of the dipole in the sphere.

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