Spontaneous emission from excitons in thin dielectric layers

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Spontaneous emission from quantum-well excitons in a thin, high-refractive-index dielectric layer embedded in a low-refractive-index bulk dielectric is studied as a function of the layer thickness and refractive-index ratio. Total spontaneous emission rates for a thin layer decrease linearly with the index ratio for dipoles in the plane of the layer and as the fifth power of the index ratio for dipoles with orientations perpendicular to the layer. These results are of interest in the design of optical microstructures that control the exciton emission dynamics of semiconductor quantum wells.

Modification of the spontaneous emission rates for semiconductor excitons has recently been demonstrated for a number of structures and materials.[1-3] Optical structures have been suggested that have photonic band gaps where no spontaneous emission can occur.[4,5] In addition, the recent success in the fabrication of surface-emitting lasers using molecular-beam epitaxy and etching techniques to form optical microstructures[6,7] has awakened interest in the physics of spontaneous emission in dielectric structures. It has been suggested that by controlling spontaneous emission rates laser thresholds can be reduced to very low levels.[8,9] For example, reduction of laser excitation currents by more than an order of magnitude may be possible.

We analyze spontaneous emission from a thin dielectric layer with refractive index n_L that can be fabricated by recently introduced molecular-beam epitaxy and selective etching technique.[10] This thin layer is surrounded by a bulk dielectric material with refractive index n_l. A $(n_l/n_l)^5$ scaling of spontaneous emission rate for dipoles oriented in a direction perpendicular to the layer (vertically) is found for layer thicknesses smaller than one tenth of a wavelength in the material. For these thin layers the spontaneous emission rate into guided modes propagating in the plane of the layer decreases to a small fraction of the total emission rate, and the surrounding dielectric and layer boundary effects dominate the spontaneous emission rates. The strong reduction for vertical dipoles found in the present analysis can be thought of as a cancellation between the vacuum fields incident from the surrounding bulk medium and the fields that are due to the induced charges on the layer boundaries. The reduced spontaneous emission rates found in the present analysis of thin layers can play a key role in the design of new optical structures that effectively control exciton emission dynamics, e.g., lowering the laser threshold of a quantum-well vertical-cavity laser in which the vertical dipole radiation is a waste of power.

An example of a thin dielectric layer that can be fabricated including a quantum well is shown in Fig. 1. A 5-nm GaAs quantum well is sandwiched between two $Al_xGa_{1-x}As$ well barrier layers with a total thickness L and a refractive index n_L typically equal to 3.4. The quantum well along with the well barrier material is embedded in a surrounding dielectric with thickness $l \gg L$ and refractive index $n_l < n_L$. An example of the surrounding material is an acrylic fabricated as described in Ref. 10 with $n_l = 1.5$. In order for this surrounding dielectric to act as a bulk medium we assume that $l \gg \lambda$.

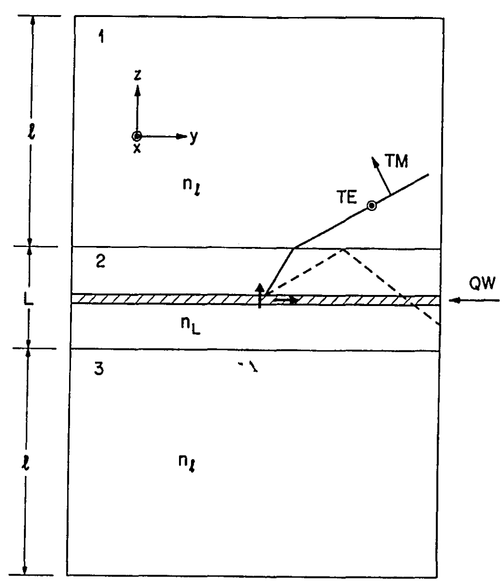

Fig. 1. Schematic of a thin quantum-well structure with index of refraction n_L embedded in a dielectric with index n_l. The quantum-well structure can be a 5-nm GaAs well (QW) sandwiched between two $Al_xGa_{1-x}As$ barriers, resulting in a total thickness L. The solid slanted line shows a free mode propagating from region 2 into region 1. The dashed line shows the guided modes in region 2. Polarization directions are shown for the TE and TM modes.

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We assume that L is large compared with w, the extent of the exciton wave function. If L is of the order of the exciton radius in the well (~10 nm), another interesting regime investigated by Keldysh[11] is obtained.

One may think of the spontaneous emission from excitons in the quantum-well region as being induced by vacuum fluctuations in the optical modes defined by the structure. Each mode of the field has an electric-field amplitude $\xi_m(\mathbf{r})$ determined by the total mode energy condition

$$2\int_V \mathrm{d}\mathbf{r}|n(\mathbf{r})\xi_m(\mathbf{r})|^2 = \hbar\omega\,, \tag{1}$$

where V is the volume used to specify the modes of the system, $\hbar\omega$ is Planck's constant times the mode frequency, $n(\mathbf{r})$ is the refractive index at position $\mathbf{r}$, and $m = 1, 2$ denotes the two polarization modes corresponding to the TE and TM modes for each wave vector $\mathbf{k}$ as shown in Fig. 1. These modes are significantly modified from the modes in a uniform dielectric. For guided modes, radiation propagates in the plane of the quantum-well layer and decays exponentially in the low-index regions. All other modes are free from guiding. These free modes propagate into the low-index region but are modified by the boundary conditions at the interface between the dielectrics. As seen in Fig. 2, these mode modifications result in significant changes in the total spontaneous emission rates for both vertically and horizontally oriented dipoles at small layer thicknesses. Our analysis is particularly interesting for layer thicknesses in the range $w < L < \lambda/4n_L$, which is typically realized in semiconductors for 10 nm < L < 100 nm. This range is now accessible experimentally with the required high index ratios.[10]

We model the spontaneous emission from an exciton in the quantum well as originating from three orthogonal, radiating dipoles: two horizontal dipoles in the x and y directions and one vertical dipole in the z direction, as depicted in Fig. 1. The dipole moment matrix element is $\boldsymbol{\mu} = \langle u|\mathbf{d}|g\rangle$, where $\langle u|$ and $|g\rangle$ are the excited and ground states of a nondegenerate two-level system representing the exciton and $\mathbf{d}$ is the dipole moment operator. These dipoles interact with both the free and the guided modes.

First we calculate the spontaneous decay rate into free modes. This is done by propagating vacuum field modes from the surrounding medium n_l into the layered medium n_L. We use the standard perturbation expression to obtain the incremental contribution to the decay rate that is due to a particular incident mode with wave vector $\mathbf{k}$,

$$\mathrm{d}\gamma_f(\Omega) = \frac{2\pi}{\hbar^2}|\xi_m(0)\cdot\boldsymbol{\mu}|^2\rho_f\sin(\theta_l)\mathrm{d}\theta_l\mathrm{d}\phi_l\,, \tag{2}$$

where θ_l and ϕ_l are the spherical coordinates for the wave vector $\mathbf{k}$ in the surrounding medium n_l, $\mathrm{d}\Omega = \sin(\theta_l)\mathrm{d}\theta_l\mathrm{d}\phi_l$ is the solid angle at $\mathbf{k}$, and $\rho_l = [V_Q/(2\pi)^3]\omega^2(n_l/c)^3$ is the density of states for modes in $n_l \cdot \xi_m(0)$ is the vacuum field evaluated at the dipole. As every free mode ξ_m in n_L actually comes from a quantized mode ξ_m^l in n_l, we can obtain ξ_m from ξ_m^l by the standard Fabry–Perot analysis.[13] ξ_m^l is given by the standard result that a quantized mode in a spatially uniform system having index n_l has the amplitude $|\xi_m^l| = \sqrt{\hbar\omega_A/2\epsilon V_Q}$ ($\epsilon = \epsilon_0 n_l^2$). Finally, we integrate $\mathrm{d}\gamma_f(\Omega)$ over all the incident angles θ_l and ϕ_l in Eq. (2) in order to obtain the free-mode contribution to the spontaneous emission rate γ_f. Note that horizontal dipoles are coupled to both TE and TM modes, while vertical dipoles are coupled only to TM modes. These free-mode rates are shown as dashed curves in Fig. 2. The variations in rate with L are associated with the Fabry–Perot cavity resonances.

Contributions to spontaneous emission from the guided modes are obtained form the equation

$$\mathrm{d}\gamma_s = \frac{2\pi}{\hbar}|\xi_s(0)\cdot\boldsymbol{\mu}|^2\rho_s\mathrm{d}\phi\,, \tag{3}$$

where $\xi_s(0)$ is the vacuum electric field at the radiating dipole layer for a guided mode s, ρ_s is a two-dimensional density of modes per unit energy in the x–y plane, and s denotes the mode index, e.g., the TM1, TM3, ... modes for the vertical dipoles and the TE1, TM2, TE3, TM4, ... modes for the horizontal dipoles. The guided-mode amplitudes and density of states are obtained from a straightforward but lengthy analysis[13] that matches the propagating guided wave in the high-index region with the exponentially decaying fields in the low-index region.

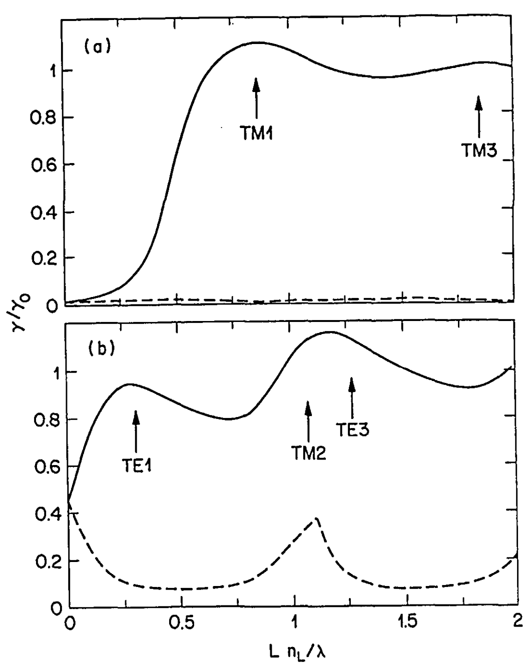

Fig. 2. Spontaneous emission rates γ/γ_0 calculated for the structure in Fig. 1 shown as a function of the normalized thickness Ln_L/γ of the quantum-well structure. These rates are normalized with respect to the rates in a uniform dielectric with index n_L. (a) Rates for vertically oriented dipoles in the quantum-well region, where the solid curve is the total rate and the dashed curve is the contribution for the free modes. (b) Rates for horizontally oriented dipoles, where the solid and dashed curves again correspond to total and free-mode contributions, respectively. The arrows indicate where the contributions from specific modes are maximized.

Equation (3) is integrated over all modes s and over all ϕ to produce the guided-mode contribution to the total spontaneous emission rate γ_g.

Shown in Fig. 2 is the total spontaneous emission rate $\gamma = \gamma_f + \gamma_g$ normalized to γ_0, the rate in a uniform index material with refractive index n_L, where $\gamma_0 = (4\omega_0^3/3\hbar c^3)|\mu|^2 n_L$, with ω_0 being the frequency of the exciton transition. At thicknesses greater than $\lambda/4n_L$ the guided modes dominate the emission rate. We are interested in the thin regime, $Ln_L/\lambda < 0.25$ for a vertical dipole and $Ln_L/\lambda < 0.1$ for horizontal dipoles, where guided modes begin to play a minor role in the spontaneous emission rate. In this thin regime the remaining guided modes—TM1 for the vertical dipoles and TE1 for the horizontal dipoles—extend further into the low-index regions as the thickness L decreases because of the decreasing guiding influence of the thin layer. This increase in the spatial extent of the guided modes results in a decrease in the vacuum field amplitude $\xi_s(0)$ in Eq. (3) [as determined by Eq. (1)] and a corresponding reduction—nearly quadratic with L—in the spontaneous emission rate due to the guided modes.

In the thin regime the free modes dominate the spontaneous emission rate. Especially striking is the small free-mode contribution to the vertical dipole emission rate. Vertical components of the vacuum electric field propagating through the high-index layer are discontinuous at the boundary. When both interfaces of the thin layer are taken into account the vertical components of the vacuum electric field at the quantum well are reduced by a factor of $(n_l/n_L)^2$. Free vacuum field amplitudes in the low-dielectric regions scale with n_l [see the discussion after Eq. (2)], so that the net reduction of the γ/γ_0 ratio is $(n_l/n_L)^5$. This strong dependence of the vertical rate on the index ratio explains the small free-mode contribution for the example shown in Fig. 2, where $(n_l/n_L)^5 \simeq 0.017$. However, the free-mode contribution for the horizontal dipole rates remains at a substantial level. This is because the horizontal component of the vacuum electric field is continuous across both boundaries of the thin layer and a reduction factor of only (n_l/n_L) is obtained for γ/γ_0 in order to account for the vacuum field normalization in the low-index region.

Experiments to confirm these large spontaneous emission rate reductions for thin layers are being designed for the GaAs system. For a GaAs quantum well the vertical and horizontal dipole transitions share a common excited state at the exciton feature approximately 50 meV below the conduction band. At 77 K the spontaneous emission from the 1e–1lh (light-hole ground state) transition and the 1e–1hh (heavy-hole ground state) transition can be spectrally resolved. The heavy-hole transition corresponds to a horizontal dipole, and the light-hole transition corresponds to a combination of three-quarters vertical dipole and one-quarter horizontal dipole.[14] The large suppression of the vertical dipole spontaneous emission rate for the thin regime should be easily demonstrated by measuring the power radiated in these two spectral features as a function of layer thickness L. It is important in the analysis of experiments in this layered regime to take into account the absorption of radiation in the guided modes. This absorption and reemission along the layer can easily lengthen the measured spontaneous emission rate. In an experiment with $L = 0.1\lambda/n_L$ with $n_L = 3.4$ and a 5-nm GaAs quantum well we estimate the absorption length in the $x-y$ plane to be 16 μm at 77 K. Measurement of emission from a region with a diameter of 10 μm or less should ensure that only the single emission events will contribute in the experiment.

We have shown that the total spontaneous emission rates for dipoles associated with exciton in high-index quantum wells can be dramatically reduced by embedding the quantum well structure in a low-index medium. These reductions occur in the range of thicknesses between 100 and 10 nm for GaAs quantum wells, a region that can be explored experimentally at present. Spontaneous emission in semiconductor structures with dimensions smaller than a wavelength in the material is a topic of fundamental interest and will be important for the design of the coming generation of optical microstructures based on new etching and growth techniques.

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