Inhibited and Enhanced Spontaneous Emission from Optically Thin AlGaAs/GaAs Double Heterostructures

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Inhibited spontaneous emission in atomic physics has been intensively investigated recently. In solid-state physics these effects are no less important. We have studied the spontaneous emission of light from electron-hole recombination in optically thin GaAs double heterostructures. The electron-hole radiative recombination rate coefficient B is not purely a property of the GaAs itself, but depends strongly on the optical-mode density and refractive index of the medium in which it is immersed. The spontaneous-emission rate can be markedly increased or decreased depending on whether the surrounding refractive index is higher or lower than that of GaAs.

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Inhibited spontaneous emission in atomic physics has been intensively investigated recently.[1-3] In solid-state physics these effects are no less important. The concept of a "photonic band structure" has been proposed[4,5] in which a three-dimensional periodic face-centered-cubic dielectric structure has a forbidden gap for photon modes. The influence of one-dimensional periodic layering[6] has also been investigated. In this paper, we will show that simply changing the index of refraction of the surrounding medium can already have a pronounced effect on spontaneous-emission rates.

Double heterostructures of the III-V semiconductors are among the handful of condensed-phase systems[7,8] capable of radiative quantum efficiency approaching unity. In addition, spontaneous emission plays an important role in many types of electronic devices, such as semiconductor lasers. Such solid-state systems can be readily immersed in optical media of widely varying index of refraction. The spontaneous-emission rate will depend on the refractive index $n_{\rm ext}$ of the host medium, which may be greater or less than the $n_{\rm int}$ of the radiating substance itself.

The rate of spontaneous emission is usually given by Fermi's "golden rule":

$$\frac{1}{\tau_R} = \frac{2\pi}{\hbar} |\langle | x E^{\dagger}_{\rm int} | \rangle |^2 \rho(\omega) , \qquad (1)$$

where τ_R is the radiative lifetime which we seek to influence, x is the material dipole operator, $E^{\dagger}_{\rm int}$ is the electric field creation operator inside the luminescent material, and $\rho(\omega)$ is the total density of optical modes per unit energy at the angular frequency ω. Attention is generally directed toward the density of states, which in an external optical medium is $n^3_{\rm ext}$ times greater than in vacuum:

$$\rho(\omega) = \frac{n^3_{\rm ext}\omega^2}{\pi^2 \hbar c^3} V , \qquad (2)$$

where V is the normalization volume of the optical modes. Most of the interesting physical questions will be connected with the electric field creation operator $E^{\dagger}_{\rm int}$ inside the excited luminescent material. This operator creates a single photon; thus the electric field magnitude associated with this operator can be regarded as the "field of a single photon" which is in turn related to the "zero-point electric field." Therefore an investigation of spontaneous-emission properties in condensed matter structures is also an investigation of the local distribution of zero-point electric field fluctuations. (In empty space these would have been called vacuum fluctuations.)

We will show that a determination of the correct index of refraction dependence of the operator $E^{\dagger}_{\rm int}$ in a given dielectric geometry can be nontrivial. The electric field creation operator in the surrounding host dielectric medium will play an important role in our considerations:

$$E^{\dagger}_{\rm ext} = \frac{1}{n_{\rm ext}} \left[\frac{2\pi\hbar\omega}{V} \right]^{1/2} e^{i\mathbf{k}\cdot\mathbf{r}} a^{\dagger} , \qquad (3)$$

where $a^{\dagger}$ is the photon creation operator, $\mathbf{k}$ is the wave vector, and $\mathbf{r}$ is the position vector. In general, $E^{\dagger}_{\rm int}$ will be linked to $E^{\dagger}_{\rm ext}$ by Maxwell's equations and by boundary conditions.

At the outset, let us consider a spherical fluorescent medium of optical dielectric constant $n^2_{\rm int}$, much less than a wavelength in diameter. Such an object has too small a lateral extent to support any purely internal optical modes. The electric field operator internal to such a sphere is determined by the extended normal modes which fill all of space. To calculate the actual modes in this geometry would be rather involved. Instead, let us simply propagate the plane-wave field operators [Eq. (3)] inward from infinity. Because of the famous spherical local field factor the internal field is reduced relative to the external field by

$$E^{\dagger}_{\rm int} = \frac{3}{(n^2_{\rm int}/n^2_{\rm ext}) + 2} E^{\dagger}_{\rm ext} . \qquad (4)$$

This reduction of the local field operator strength and the density of states [Eq. (2)], must be substituted into the spontaneous-emission rate [Eq. (1)]:

$$\frac{1}{\tau_R} \propto \left[\frac{3}{(n_{\rm int}^2/n_{\rm ext}^2)+2}\right]^2 \frac{n_{\rm ext}}{n_{\rm int}}. \tag{5}$$

This equation gives some idea of the enormous influence of the surrounding environment on the spontaneous radiative lifetime of the molecules in a dielectric sphere. By immersing that sphere in lower-index surroundings τ_R can be drastically increased. Many common semiconductors have a refractive index ~3.5 and can be immersed in air of index 1. The increase in the spontaneous radiative lifetime is roughly proportional to the fifth power of the index ratio. By use of Eq. (5), the spontaneous lifetime of electrons and holes in a small semiconductor particle, immersed in air rather than an index matching medium, becomes longer by a factor ~80.

The so-called natural spontaneous lifetime can therefore be artificially controlled by changing the density of optical modes in the surroundings and by influencing the coupling of external and internal electric field creation operators. One of the points to be noticed in the procedure outlined above is that no real attempt was made to solve for the actual electromagnetic modes appropriate to the geometry. Instead it was assumed that the plane wave $E_{\rm ext}^\dagger$ at infinity could be propagated in toward the object by Maxwell's equations. In effect the zero-point electric field is regarded as a boundary condition at infinity. This procedure fails when there are purely localized internal optical modes confined to the vicinity of the dielectric object. Then the localized contribution to both $E_{\rm int}^\dagger$ and mode density would have been overlooked. Despite this, there would be no error in the net decay of excited states, since localized modes are reabsorbed and cannot carry any energy away.

The experiments to be described here were done on thin film semiconductors rather than small semiconductor particles. The spontaneous-emission rate for electron-hole recombination is usually written as BNP, where N and P are the electron and hole concentrations and B is called the radiative recombination coefficient. Numerical B values are tabulated[9] in books. Measurements of B on $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ double heterostructure thin films have been performed many[10] times in the past, with a considerable scatter in the results. Invariably those measurements were done with the following dielectric configuration: The thin film would have the original GaAs growth substrate below it and air or vacuum above it. In view of our introductory discussion, the value of B is not purely a material constant of the GaAs but must depend on the surrounding refractive index geometry.

In our laboratory we have developed techniques[11] to float epitaxial films off their growth substrates and then transfer them to any arbitrary substrate. We find that van der Waals forces are sufficient to bond the film and substrate. Since the new substrate can have any arbitrary refractive index, B can be strongly influenced. The experimental geometry is shown in Fig. 1, with an $\mathrm{SiO_2}$ substrate shown as an example. The undoped GaAs film is typically 500 nm thick with undoped 100-nm $\mathrm{Al_{0.3}Ga_{0.7}As}$ carrier-confinement layers on either side. Carriers are injected into the film by means of a doubled Q-switched Nd-doped yttrium aluminum garnet laser. The calibrated carrier conductivity is monitored by contactless inductive coupling using the rf coil at 500 MHz. The transient conductivity decay is then converted to a density decay with known values for the plasma mobility. A typical set of density decays for three different sub-

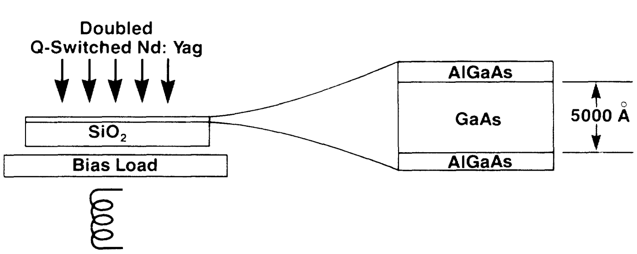

FIG. 1. The carrier density injected into an $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ double heterostructure by a doubled Q-switched Nd-doped yttrium aluminum garnet laser is monitored by 500-MHz inductive coupling. The bias load helps to ensure a linear response of the rf bridge. The epitaxial semiconductor film is shown on an $\mathrm{SiO_2}$ substrate as an example.

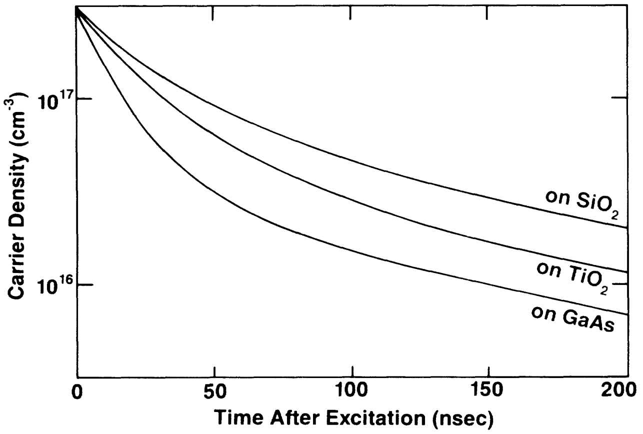

FIG. 2. The transient-carrier density decay from GaAs double heterostructures mounted on substrates of three different indices of refraction. The initial nonexponential decay is due to "bimolecular" radiative spontaneous BNP recombination of electrons and holes in accordance with Eq. (6). The radiative B coefficient is a function of the surrounding refractive index geometry. The exponential decay at lower densities is nonradiative.

strate materials is shown in Fig. 2. We have previously[12] employed this apparatus for a wide variety of minority-carrier lifetime measurements, including particularly nonradiative recombination in bulk and at surfaces. The carrier-density decay rate is given by

$$-\frac{dN}{dt}=\frac{N}{\tau_{\mathrm{nr}}}+BN^2+CN^3\,, \tag{6}$$

where $N \approx P$ since there is no background doping, τ_{nr} is the nonradiative decay lifetime, and C is the Auger recombination coefficient. In this experiment we are concentrating on the intermediate range of carrier densities where spontaneous emission B is a major contributor. For Fig. 2, double heterostructures from the same parent GaAs wafer were mounted on SiO_2 (index = 1.5), TiO_2 (index = 2.6), and left on the original GaAs growth substrate (index = 3.6). At the lower range of densities in Fig. 2, the nonradiative τ_{nr} exponential decay term dominates, and is the same for the different substrates. At the higher densities the BNP (so-called bimolecular recombination) term is important and responsible for the nonexponential character of the decay. Auger recombination remained negligible at the densities in Fig. 2. By fitting Eq. (6) to the nonexponential decay, the values of B and τ_{nr} could be extracted. For reasons to be mentioned later, the B coefficient is plotted against n_{int}^2+1 in Fig. 3, showing a linear dependence.

The thin-film geometry of this experiment can be analyzed in a manner similar to the small spherical particle case. The main unknown is $E_{\mathrm{int}}^{\dagger}$. Once again we will propagate the plane waves $E_{\mathrm{ext}}^{\dagger}$ in from infinity. In principle, it is well known how to solve the interaction of plane waves incident on a thin film. At normal incidence the problem is simple enough, but in general we should include the following effects: incidence angle averaging, polarization averaging, averaging over the volume or depth of the luminescent material, averaging over the band of luminescent wavelengths, and averaging over thickness fluctuations or surface roughness of the luminescent material. To avoid getting bogged down in a specific geometry we will make approximations in two important limits; the film is (a) thinner or (b) thicker than a half wavelength.

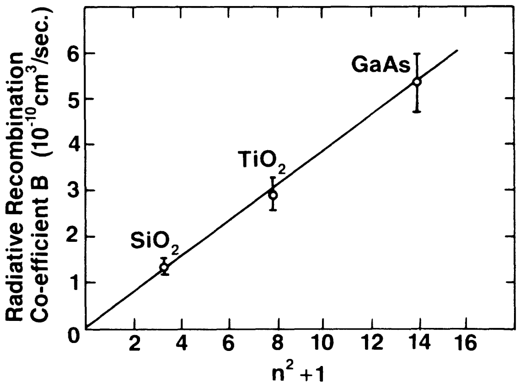

FIG. 3. The dependence of the radiative B coefficient on the refractive index n of the substrate material. In accordance with Eq. (9), B is proportional to the mean square refractive index of the substrate and the air, n^2+1.

Case (a).—At normal incidence by continuity of the tangential component of electric field, $E_{\mathrm{int}}^{\dagger}=E_{\mathrm{ext}}^{\dagger}$. Let us take this as an approximation, ignoring the required averaging over angles and polarizations. Then by use of Eq. (3), $|E_{\mathrm{int}}^{\dagger}|^2 \sim 1/n_{\mathrm{ext}}^2$. The spontaneous-emission rate will also pick up a factor n_{ext}^3 from the density of final states. Compared to being immersed in a medium sharing its own index of refraction, the net rate of spontaneous emission will then be reduced by a factor $n_{\mathrm{ext}}/n_{\mathrm{int}}$. This ratio, though significant, is much milder than Eq. (5) where the local field factors played the major role. In the limit of films much thinner than a half wavelength, localized waveguide modes can be neglected as before.

Case (b).—The film is thicker than a half wavelength, though still optically thin with respect to absorption. Our GaAs films were roughly 3 wavelengths thick. Rather than solving a coherent multiple reflection problem, we will once again take note of averaging over angles, polarizations, position, wavelengths, and thickness fluctuations. Consistent with such a statistical approach, we will use Kirkhoff's law or equivalently detailed balancing to say that the incoming energy flux into the film is equal to that outgoing from the film:

$$\frac{c}{n_{\mathrm{int}}}n_{\mathrm{int}}^2|E_{\mathrm{int}}^{\dagger}|^2=\frac{c}{n_{\mathrm{ext}}}n_{\mathrm{ext}}^2|E_{\mathrm{ext}}^{\dagger}|^2. \tag{7}$$

This now takes the place of Eq. (4) which leads to $E_{\mathrm{int}}^{\dagger}=(n_{\mathrm{ext}}/n_{\mathrm{int}})E_{\mathrm{ext}}^{\dagger}$. The combination of this with Eqs. (2) and (3) and substitution of them into Eq. (1) leads to the following expression for spontaneous-emission lifetime:

$$\frac{1}{\tau_R}\propto\frac{n_{\mathrm{ext}}^2}{n_{\mathrm{int}}^2}. \tag{8}$$

We see that the spontaneous lifetime can be increased by about a factor of 12 or 13 for GaAs films surrounded by air. In the experiment we have described, the GaAs film had air on one side and the substrate of index n_{ext} on the other side. Repetition of the same reasoning applied on our experimental geometry gives

$$\frac{1}{\tau_R}\propto\frac{n_{\mathrm{ext}}^2+1}{2n_{\mathrm{int}}^2}, \tag{9}$$

which is the functional dependence tested in Fig. 3.

In case (b), our analysis ignores any localized modes which do not couple to the plane waves [Eq. (3)] at infinity. Since our films are ~ 3 wavelengths thick, spontaneous emission into such modes will indeed occur. Nevertheless this would not lead to any error in the net decay rate of excited states, since localized modes cannot carry any energy away and they are eventually reabsorbed. The films in case (b), though optically thin, are totally absorbing for localized modes which cannot escape to infinity. (If $n_{\text{ext}} > n_{\text{int}}$ such modes would not exist, and the spontaneous emission would be enhanced rather than inhibited.) Qualitatively, case (b) represents a different type of spontaneous-emission inhibition from case (a) or the small spherical particle case, and may have more in common with optically thick luminescent slabs. The effect on the population of excited states is the same however.

The excellent agreement between Fig. 3 and Eq. (9) shows that it is the availability of optical modes on either side of the luminescent film which controls the spontaneous-emission rate. The absolute value of B cannot be directly compared with previous measurements[10] which were unclear in their treatment of the local refractive index geometry. It can, however, be checked against the Shockley-van Roosbroeck relation[9] which is derived by detailed balancing:

$$BN_i^2 = \frac{8\pi n_{\text{int}}^2 \nu^2}{c^2} \int_0^\infty e^{-h\nu/kT} \alpha(\nu)\, d\nu , \tag{10}$$

where N_i is the intrinsic carrier density determined from an accurate knowledge of the thermodynamic[13] band gap, $\alpha(\nu)$ is the direct absorption spectrum, kT is the thermal energy, and c is the speed of light. It is implicitly assumed in Eq. (10) that the GaAs is immersed in a matching refractive index n_{int}. Using Eq. (10), we calculate $B = 13 \times 10^{-10}\ \text{cm}^{-3}\ \text{sec}^{-1}$. This must then be multiplied by $(n_{\text{int}}^2 + 1)/2n_{\text{int}}^2$ in accordance with Eq. (9) to be compared with the GaAs data point in Fig. 3. The predicted value is $7 \times 10^{-10}\ \text{cm}^{-3}\ \text{sec}^{-1}$ in comparison with the measured value $\sim 5.5 \times 10^{-10}\ \text{cm}^{-3}\ \text{sec}^{-1}$. In view of the many uncertainties in any absolute measurement the agreement is reasonable, but both are much higher than previously accepted[10] values for B.

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