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Exciton-polariton picture of the free-exciton lifetime in GaAs

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The recently determined nanosecond free-exciton lifetime of GaAs is interpreted in terms of radiative decay of exciton-polaritons through coupling to longitudinal acoustic phonons.

Recently, 't Hooft, van der Poel, Molenkamp, and Foxon[1] have made the first determination of the free-exciton lifetime in GaAs using ultrapure molecular-beam-epitaxy (MBE) grown material and saturation of the bound-exciton states. Quite to the surprise of the authors the result turned out to be 3.3 ns, over three orders of magnitude smaller than expected from earlier work of Hwang.[2] From theoretical arguments, Hwang predicted a free-exciton lifetime of 500 μs. His experiments indicated an increasing lifetime with decreasing impurity concentration; the longest lifetime measured was 2.9 ns. Hwang's prediction was based on the expression for the lifetime in terms of an oscillator strength per atom as can be deduced from equations given by Dexter[3] in the context of atomic transitions and an expression for the oscillator strength per atom as derived in the context of the calculation of the absorption strength, cf. Knox.[4]

In this Rapid Communication we show that the ns free-exciton lifetime of GaAs can basically be explained by realizing that the exciting light generates exciton-polaritons in the crystal. As is well known from the work of Hopfield[5] and Fano[6] the coherent nature of the interaction between a free exciton and a photon leads to an intricate coupling, resulting in the formation of exciton-polaritons (in short, polaritons). It is therefore inappropriate to calculate the lifetime, as described above, from an oscillator strength per atom. Time-proportional perturbation theory that takes the coherent nature of the interaction into account yields a lifetime which is inversely proportional to the number of atoms in the crystal and thus ridiculously short, cf. Ref. 4.

We have calculated the free-exciton lifetime of GaAs by studying the phonon-induced conversion of excitonlike polaritons into photonlike polaritons and the radiative "decay" of the latter. The calculations follow the rate equations scheme for the description of the interaction between exciton-polaritons and acoustical phonons as introduced by Sumi[7] and Askary and Yu.[8] It is assumed that the "decay" is dominated by the heavy-hole exciton, since it has a larger density of states than the light-hole exciton. For simplicity we therefore neglect the light-hole exciton, and the transverse polaritons are thus represented by a two-branch model.[9] We do, however, include the longitudinal polariton. The radiative escape is described by the additional boundary condition introduced by Pekar.[10] Calculations have been performed for $T=0$ K and also for temperatures comparable to the transverse-longitudinal exciton splitting.

The dielectric response of GaAs as a function of wave number $\mathbf{k}$ and energy E is described by

$$\epsilon(\mathbf{k},E)=\epsilon_b+\frac{4\pi\beta E_T^2(\mathbf{k}=\mathbf{0})}{E_T^2(\mathbf{k})-E^2}, \tag{1}$$

where ϵ_b is the background dielectric constant which contains contributions from all interactions except the exciton in question, $E_T(\mathbf{k})$ is the dispersion of the exciton $E_T(\mathbf{k})=E_0+\hbar^2\mathbf{k}^2/2m^*$, where m^* is the (isotropic) effective mass of the heavy-hole exciton, and the coupling constant β is the polarizability of the exciton. The values of the parameters are given in Table I. The dispersion of the transverse polaritons is described by $\epsilon(\mathbf{k},E)=(\hbar c\mathbf{k})^2/E^2$ and that of the longitudinal polaritons by $\epsilon(\mathbf{k},E)=0$. Figure 1 shows the dispersion for GaAs.

Upon absorption photons are converted into polaritons which thermalize rapidly, i.e., on a ps time scale the momentum becomes distributed isotropically and both excitonlike and photonlike polaritons are created. The radiative decay time of the free exciton is determined by the conversion of the excitonlike polaritons into photonlike polaritons. For example, the lower-branch polaritons are scattered by phonons into the energy region below or near the exciton energy where radiative decay of polaritons can take place efficiently through their photon components. In the course of this process bottlenecking may occur, as first noticed by Toyozawa.[13]

We will consider only scattering by longitudinal acoustic phonons, and the electron-phonon interaction is described by the deformation potential only. The transition rate for scattering from phonons of a polariton belonging to branch i with momentum $\mathbf{k}_i$, polarization λ, and energy E_i into a polariton belonging to branch j with momentum $\mathbf{k}_j=\mathbf{k}_i-\mathbf{q}$, polarization λ', and energy E_j, is given by

TABLE I. Parameters used in the calculation.

Transverse exciton energy[a] $E_T(k=0)$	1.5151 eV
Longitudinal-transverse splitting[b] ΔE_{LT}	0.08 meV
Polarizability[b] β	1.06×10^{-4}
Background dielectric constant[b] ϵ_b	12.56
Exciton effective mass[b] m^*	$0.6m_e$
Sound velocity[b] u	4.8×10^5 ms^{-1}
Density[c] ρ	5.307 g cm^{-3}
Deformation potential strength[c] D	7.8 eV

[a]Reference 1.
[b]Reference 11.
[c]Reference 12.

$$W(\mathbf{k}_i,\mathbf{k}_j)=\frac{2\pi}{\hbar}|V_{ac}(\mathbf{q})\Phi(E_i,E_j)\cos\theta(\lambda,\lambda')|^2\{[n(\hbar\omega_{\mathbf{q}})+1]\delta(E_i-E_j-\hbar\omega_{\mathbf{q}})+n(\hbar\omega_{-\mathbf{q}})\delta(E_i-E_j+\hbar\omega_{-\mathbf{q}})\}, \quad (2)$$

where $\theta(\lambda,\lambda')$ is the angle between the polarization directions, and $V_{ac}(\mathbf{q})$ is the matrix element for scattering by the acoustic phonon with momentum $+\mathbf{q}$ or $-\mathbf{q}$ and energy $\hbar\omega_{\pm\mathbf{q}}$:

$$|V_{ac}(\mathbf{q})|^2=\frac{\hbar|\mathbf{q}|D^2}{2V_c\rho u}. \quad (3)$$

Here V_c is the crystal volume, ρ is the density of the crystal, u is the LA sound velocity, and D the deformation potential strength (values are given in Table I). The function $\Phi(E_i,E_j)$ measures the joint exciton content of the polariton branches i and j. For both i and j transverse, Φ can be expressed in terms of the coefficients introduced by Hopfield,[5] appropriately generalized for the spatially dispersive case. For both i and j longitudinal, $\Phi=1$, while for i longitudinal and j transverse or vice versa, Φ is equal to the appropriate Hopfield coefficient measuring the exciton content of the transverse polariton. In Eq. (3) we have ignored the internal structure of the exciton.[13]

The description of the escape of light from the crystal deserves special care. Outside the crystal we have a transmitted electromagnetic wave. Inside the crystal we have an incident transverse or longitudinal wave, one reflected wave, and up to two converted waves, all of them involving both electromagnetic and material character. Apart from the boundary conditions imposed by Maxwell's equations we use the additional boundary condition introduced by Pekar:[10]

$$\sum_i \mathbf{P}_i|_{\text{surface}}=\mathbf{0}, \quad (4)$$

which requires that the sum of the polarizations of the exciton components of the polariton waves at the surface of the crystal vanishes. This condition at least implies that no energy is dissipated or generated at the interface, cf. Selkin.[14] Using the complete set of boundary conditions we can determine the reflection, transmission, and conversion coefficients $X_m^{m'}$ for an incident wave m in the crystal and outgoing waves m' (in the crystal or vacuum). We define

$$X_m^{m'}=\left|\frac{\hat{\mathbf{z}}\cdot\mathbf{S}_{m'}}{\hat{\mathbf{z}}\cdot\mathbf{S}_m}\right|, \quad (5)$$

where $\hat{\mathbf{z}}$ is the unit vector perpendicular to the surface and $\mathbf{S}_{m'}$ and $\mathbf{S}_m$ are generalized Poynting vectors as introduced by Selkin.

We can now determine the reflection, transmission, and conversion rates by multiplying the fraction of polaritons of type m that actually arrives at the crystal surface by the probability for such a polariton to undergo a certain transfer ($m\rightarrow m'$) process as described by the coefficients $X_m^{m'}$, i.e.,

$$P_m^{m'}(E_m)=\frac{v(E_m)}{L}\int_0^{\pi/2}\sin\theta\cos\theta X_m^{m'}(E_m,\theta)d\theta. \quad (6)$$

Here L measures the crystal thickness and $v(E_m)$ is the polariton group velocity of branch m at energy E_m. Start-

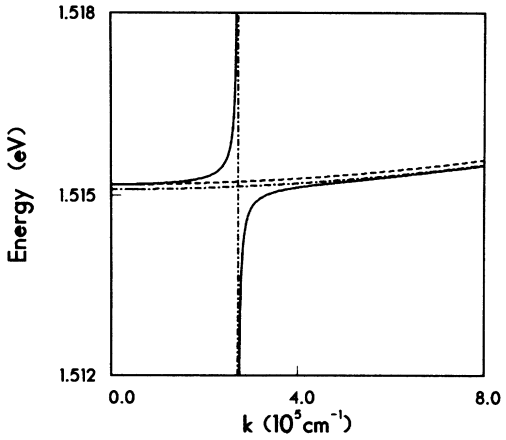

FIG. 1. Dispersion curve of the exciton polariton in GaAs; the solid lines represent the transverse polaritons, the dashed curve is the longitudinal exciton, and the dash-dotted curves represent the unperturbed exciton and photon branches.

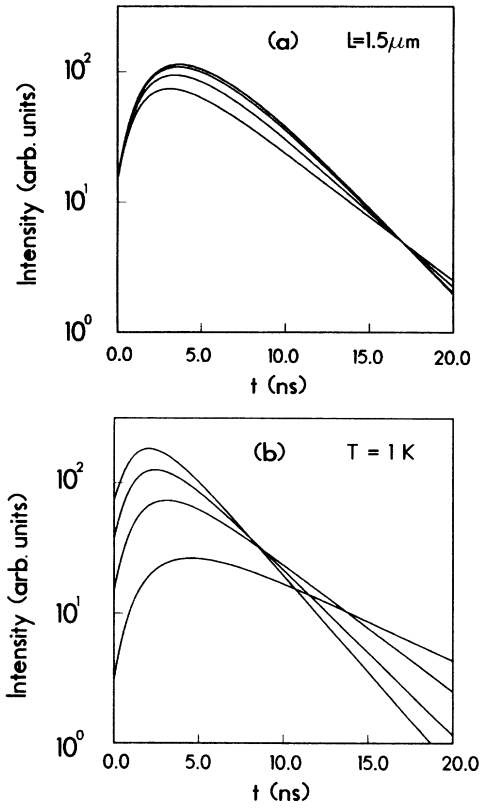

FIG. 2. (a) The rate of photon emission for crystal thickness $L=1.5$ μm and $T=0$, 0.5, 0.75, and 1 K (increasing temperature corresponds with decreasing slope). (b) The rate of photon emission for $T=1$ K and crystal thickness $L=0.75$, 1, 1.5, and 3 μm (increasing crystal thickness corresponds with decreasing slope).

TABLE II. The free exciton-lifetime for several temperatures and a crystal thickness $L = 1.5$ μm.

Temperature T (K)	Lifetime τ (ns)
0	7.5
0.5	7.7
0.75	8.7
1	10.0

TABLE III. The free-exciton lifetime at $T = 1$ K for several values of the crystal thickness.

Crystal thickness L (μm)	Lifetime τ (ns)
0.75	6.6
1	7.9
1.5	10.0
3	16.0

ing from assumed, in $\mathbf{k}$ space isotropic, distributions $\rho_m(E_m(\mathbf{k}_m),t=0)$ for branches m, we can determine the distributions $\rho_m = \rho_m(E_m(\mathbf{k}_m),t)$ using rate equations. We take

$$\frac{d\rho_m}{dt} = \left(\frac{d\rho_m}{dt}\right)_{\text{in}} - \left(\frac{d\rho_m}{dt}\right)_{\text{out}}, \tag{7}$$

where the rates of generation and loss of polaritons are given by

$$\left(\frac{d\rho_m(E_m,t)}{dt}\right)_{\text{in}} = \sum_j \left(\frac{V_c}{(2\pi)^3}\int d\mathbf{k}_j \rho_j(E_j,t) W(\mathbf{k}_j,\mathbf{k}_m)\right) + \sum_{j(\neq m)} [\rho_j(E_m,t) P_j^m(E_m)] \tag{8}$$

and

$$\left(\frac{d\rho_m(E_m,t)}{dt}\right)_{\text{out}} = \sum_j \left(\frac{V_c}{(2\pi)^3}\int d\mathbf{k}_j W(\mathbf{k}_m,\mathbf{k}_j) \rho_m(E_m,t)\right) + \sum_{l(\neq m)} [P_m^l(E_m)\rho_m(E_m,t)], \tag{9}$$

where the sum over j involves the three branches in the crystal and the sum over l involves the three polariton branches in the crystal and the photon wave in the vacuum. Note that in contrast to the work of Askary and Yu[8] we do not disregard the longitudinal polariton.

Starting from a Gaussian distribution with energies centered at the photon energy, 1.5151 eV, and with a width of 0.5 meV, we have numerically calculated the rate of photon emission as a function of temperature and crystal thickness. Results are given in Figs. 2(a) and 2(b) and Tables II and III. The calculated lifetime is somewhat longer than experimentally observed by 't Hooft *et al.*[1] This may be due to our neglect of the piezoelectric interaction and of the light-hole exciton polariton.[9] In addition there is some uncertainty about the values of the parameters used in the calculation. For example, if one takes for the strength of the deformation potential D the more recent estimate[15] of 12 eV, one expects[13] the calculated lifetime to decrease by a factor of roughly $(12/7.8)^{-5/4} \approx 0.6$. The thickness dependence is of course due to the finite transfer time of polaritons through the crystal; the free-exciton lifetime is to some extent a confinement time. Of course the experimental verification of this result will be limited by the finite absorption length of light in the crystal. The lifetime shows a slow increase with temperature. Unfortunately we have not been able to go above 1 K; scattering rates became too large to allow numerical treatment. However, note that for all temperatures investigated the scattering rates near the crossing point of the polariton branches remain much smaller than $\Delta E_{\text{LT}}/\hbar$ corresponding to the longitudinal-transverse splitting, implying that our treatment of the scattering from phonons by perturbation theory is valid.

In conclusion, we feel that radiative decay of a free exciton in GaAs can be treated in the framework of exciton-polaritons, their coupling to phonons, and their escape from the crystal as photons. Our results show that such a description is pertinent even when $k_B T$ is comparable with or exceeds the longitudinal-transverse splitting.

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