Temperature-dependent radiative decay of localized excitons in a type-II GaAs/AlAs superlattice

J. F. Angell and M. D. Sturge
Department of Physics and Astronomy, Dartmouth College, Hanover, New Hampshire 03755-3528
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The low-temperature luminescence intensity decay following pulse excitation of localized excitons in a type-II GaAs/AlAs superlattice is nonexponential. The decay obeys a previously derived theory in which excitons experience a distribution of decay rates corresponding to a Gaussian distribution of X to Γ scattering matrix elements. At high temperature the decay is exponential with a rate equal to the mean of the distribution. The transition is attributed to excitons motionally averaging the distribution as temperature is increased and thermal delocalization occurs. This paper develops a theory of radiative decay which takes into account motional averaging and is valid at all temperatures. The theory has one fitting parameter γ which is the ratio of exciton lifetime to jump time and determines the extent of averaging performed. Good agreement is found between the theory and experimental data; however, at high excitation intensity and low exciton energy the theory must be modified to take saturation of localized states into account. The temperature dependence of the fitting parameter γ reveals two delocalization processes, the activation energies of which are interpreted as the localization energy and the exciton binding energy.

I. INTRODUCTION

The time dependence of low-temperature luminescence intensity decay of localized excitons in a type-II GaAs/AlAs superlattice is nonexponential.[1,2] The form of the decay is the same as that derived for Brillouin-zone-boundary excitons in an indirect-gap semiconductor alloy such as $Al_xGa_{1-x}As$.[3] In this alloy the potential fluctuations created by the random placement of the Al and Ga atoms mix states at or near the zone boundary X with those at the zone center Γ. The scattering matrix element V is a random variable with a Gaussian distribution for excitons located exactly at the zone boundary and with a Rayleigh distribution for excitons located off the zone boundary. The exciton decay rate is proportional to $|V|^2$ and, therefore, also has a distribution. It has been observed in $Al_xGa_{1-x}As$ that the low-temperature decay is nonexponential and corresponds to that of localized excitons located off the zone boundary ($k \neq k_X$).[3,4] At higher temperatures ($T > 8$ K) the decay is exponential, with a rate approximately equal to the sum of the mean of the distribution and a nonradiative rate determined from the measured quantum efficiency. The transition from nonexponential decay at low temperature to exponential decay at high temperature is attributed to excitons becoming thermally delocalized and motionally averaging the distribution of decay rates.

As stated above, the low-temperature decay of excitons in a type-II superlattice is also nonexponential. The high-temperature ($T \sim 25$ K) decay has been measured[5] and found to be exponential due to the domination of nonradiative processes. It has also been found that the exciton decay can be made exponential by increasing excitation intensity.[2,6] The effect of high intensity is presumably to saturate localized states, causing the observed decay to be from the more numerous delocalized states.

The purpose of this paper is to construct a theory of exciton decay which is valid at all temperatures, and to make measurements on a sample in which nonradiative decay is not important in the relevant temperature range. The theory is developed in Sec. II. It is based on a hopping process in which, as temperature is raised, localized excitons begin to hop to neighboring sites, and in so doing average over the distribution of decay rates. The theory contains one temperature-dependent fitting parameter, the ratio of exciton lifetime to jump time. The temperature dependence of this parameter will tell us about the thermal activation processes involved in the delocalization. We will find that there are two thermally activated processes. The activation energy of the lower-temperature process will be interpreted as the localization energy. This same quantity has been measured in direct-gap GaAs/AlAs multiple quantum wells and we will make a comparison with these results.[7,8] Section III will present experimental data along with a comparison to theory, and Sec. IV will give a discussion of the results and conclusions.

II. THEORY

In 1982 Klein, Sturge, and Cohen derived the low-temperature radiative intensity decay of excitons in an indirect-gap semiconductor alloy, based on a distribution of radiative rates.[3] It is our purpose in this section to develop a model of exciton localization which includes the effects of temperature, and with which we can derive an intensity decay curve which is valid at all temperatures. This radiative decay will be found to contain two parameters. One of these is the mean of the distribution of rates which can be found from the low-temperature result. The other is defined as the ratio of exciton lifetime to jump time and will be the only fitting parameter in the theory. Since the theory developed here is an extension

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of that developed by Klein, Sturge, and Cohen, we will begin with a summary of it.

Klein, Sturge, and Cohen considered the radiative decay of excitons in an indirect-gap semiconductor alloy AB_xC_{1-x}. In such a material the potential fluctuations created by the random placement of B and C atoms mix exciton states at X with those at Γ and cause the no-phonon transition to become allowed in the same manner as in an impurity system such as GaP:N.[9] The scattering matrix element which serves to mix the states is $V(\mathbf{k})$, where $\mathbf{k}$ is a vector in reciprocal-lattice space, and is given by

$$V(\mathbf{k})=\sum_{\mathbf{L}} e^{i\mathbf{k}\cdot\mathbf{L}}V_{\Gamma k}(\mathbf{L})\psi(\mathbf{L}) \ , \tag{1}$$

where

$$V_{\Gamma k}(\mathbf{L})=f_L v_B+(1-f_L)v_C \ , \tag{2}$$

with

$$v_{B(C)}=\int_{\text{cell}} u_C^{\Gamma}(r)V_{B(C)}(r)u_C^{k}(r)d^3r \ . \tag{3}$$

Here $V_{B(C)}$ is the potential in a cell containing a $B(C)$ atom. f_L is a random variable due to the random placement of the B and C atoms in the crystal lattice with vector $\mathbf{L}$ and has a mean of x. The u are conduction-band Bloch functions, and ψ is the center-of-mass-dependent portion of the exciton envelope function. When $k=k_X=2\pi/a$, where a is the lattice spacing, the quantity $e^{i\mathbf{k}\cdot\mathbf{L}}=\pm 1$, and since $V_{\Gamma k}(\mathbf{L})$ is a real random variable, $V(\mathbf{k})$ is real with a Gaussian distribution. When $k\neq k_X$ but is close to $2\pi/a$, Eq. (1) contains a sum of terms of random phases with a uniform distribution. The distribution of $|V|$ is then Rayleigh, as in the distribution of amplitudes in laser speckle.[10]

The resulting probability density function of $|V(\mathbf{k})|^2$ is, for $k=k_X$,

$$p(V^2)=(2\pi V^2\langle V^2\rangle)^{-1/2}e^{-V^2/2\langle V^2\rangle} \tag{4}$$

while, for $k\neq k_X$,

$$p(|V|^2)=\langle V^2\rangle^{-1}e^{-|V|^2/\langle V^2\rangle} \ . \tag{5}$$

Here $\langle x\rangle$ is the mean or ensemble average of x. The radiative decay rate w is proportional to $|V|^2$ and has the same probability density. If all the excitons are localized then each one samples only a small environment of the alloy, and a population of excitons exhibits this same distribution of decay rates. For a δ-function excitation, the luminescence decay curve is then found to be[3]

$$I(t)=e^{-w't}(1+2\langle w\rangle t)^{-3/2} \ , \quad k=k_X \ , \tag{6}$$

$$I(t)=e^{-w't}(1+\langle w\rangle t)^{-2} \ , \quad k\neq k_X \ . \tag{7}$$

Here w' takes into account any nonstochastic processes such as phonon-assisted transitions.

We are concerned with type-II GaAs(n)/AlAs(m) superlattices in which the lowest-energy excitons are indirect and also have momentum parallel to the growth direction, z. The X to Γ scattering potential is now the superlattice potential, and the scattering centers are essentially confined to the interface planes. The lattice vector $\mathbf{L}$ in Eq. (1) now has zero or at most a very small z component and, since we are only considering excitons with momentum parallel to z, the phase $\mathbf{k}\cdot\mathbf{L}$ is zero regardless of the value of k_Z. This was first pointed out by Minami *et al.*[5] The matrix element $V_{\Gamma k}(\mathbf{L})$ is still written as in Eq. (2), where f_L now has a mean value of one-half. $V_{\Gamma k}(\mathbf{L})$ is therefore still a random variable with a random sign due to the random placement of the Ga and Al atoms along an interface. $V(k)$ then has a Gaussian distribution leading to Eq. (6) for the intensity decay. From now on we will refer to this case as the $k=k_X$ case, to which it is mathematically equivalent. Note however, that while it is applicable only to zone-boundary excitons in the alloy, it applies to both zone-boundary or off-zone-boundary excitons in the superlattice.

We now consider the effects of temperature by constructing a simple model of exciton mobility. We begin by creating (with a laser pulse) a population of excitons in an indirect-gap semiconductor with some sort of random potential. At low temperature, say <4 K, all the excitons will very quickly localize in the deepest potential wells. When temperature is increased slightly, to about 8 K, the excitons begin to overcome the potential barriers which confine them and they hop to other nearby potential wells. At high enough temperatures, about 30 K, the excitons freely hop from site to site, i.e., they are completely delocalized. Note that here excitons are delocalized in the sense that they are able to hop from one localized state to another. They are not necessarily entering a state with an extended wave function such as a state with energy higher than a mobility edge.

When the excitons are completely localized, the population will exhibit the distribution of radiative rates discussed above and decay nonexpotentially according to either Eq. (6) or (7). If the excitons are sufficiently delocalized, they will motionally average the distribution of decay rates, and a single rate will result, namely $\langle w\rangle$, or something close to it. The decay then becomes exponential. Of course the changeover from nonexponential to exponential decay is not instantaneous, and at intermediate temperatures, where the excitons begin to move around but are not yet mobile enough to fully average over the distribution of decay rates, a different form is expected. We thus seek a formula for the radiative decay which is valid at all temperatures. This will involve a different form of the probability density of the squared scattering matrix element, V.

An effective method of solving this problem is based on the Karhunen-Loeve expansion method.[11] This method has also been used by Jakeman and Pike in their calculation of the intensity fluctuation distribution of Gaussian light,[12] as well as by Slepian in determining fluctuations of random noise power.[13] Here we calculate the integrated square of the scattering matrix element V,

$$A(T)=\int_0^T V^2(t)dt \ , \tag{8}$$

where T is the mean exciton lifetime. It is our purpose to determine the probability density function $p(A)$. We begin by expanding $V(t)$ in an orthonormal series over the

interval 0 to T as follows:

$$V(t)=\sum_n b_n \varphi_n(t) \ , \quad 0<t\le T \ , \tag{9}$$

where

$$b_n=\int_0^T V(t)\varphi_n^*(t)dt \tag{10}$$

and

$$\int_0^T \varphi_n^*(t)\varphi_m(t)dt=\delta_{nm} \ . \tag{11}$$

We choose an expansion such that the coefficients b_n are uncorrelated, that is,

$$\begin{aligned} E[b_n b_m^*] \equiv \langle b_n b_m^* \rangle &= \int_0^T \int_0^T \langle V(t_1)V^*(t_2)\rangle \varphi_n(t_2)\varphi_m^*(t_1)dt_1 dt_2 \\ &= \int_0^T \left[\int_0^T J_V(t_1;t_2)\varphi_n(t_2)dt_2 \right] \varphi_m^*(t_1)dt_1 \\ &= \lambda_n \quad \text{for } m=n \ , \\ &= 0 \quad \text{for } m=n \ , \end{aligned} \tag{12}$$

where

$$J_V(t_1;t_2)=\langle V(t_1)V^*(t_2)\rangle$$

and is the autocorrelation function of the scattering matrix element, V. Multiplying each side of Eq. (12) by φ_n^* and using (11), we obtain the following eigenvalue equation:

$$\int_0^T J_V(t_1;t_2)\varphi_n(t_2)dt_2=\lambda_n \varphi_n(t_1) \ , \tag{13}$$

and, from (8) and (9),

$$A(T)=\sum_n |b_n|^2 \ . \tag{14}$$

For the case $k=k_X$, V in Eq. (10) is real, and, since the φ_n are real valued, the b_n are thus real, independent, normally distributed random variables with mean zero and variance λ_n.[14] The probability density of b_n^2 is then similar to that of (4):

$$p(b_n^2)=(2\pi\lambda_n b_n^2)^{-1/2}\exp\left[-\frac{b_n^2}{2\lambda_n}\right] , \tag{15}$$

which has the Fourier transform or characteristic function

$$\begin{aligned} M_{b_n^2}(i\xi)=\langle e^{i\xi b_n^2}\rangle &= \int_0^\infty e^{i\xi b_n^2} p(b_n^2)d(b_n^2) \\ &= (1-2i\xi\lambda_n)^{-1/2} \ . \end{aligned} \tag{16}$$

Because of Eq. (14) and the independence of the b_n^2, we can write the characteristic function of A as the product of the individual characteristic functions of the b_n^2. That is,

$$M_A(i\xi)=\prod_{n=0}^{\infty}(1-2i\xi\lambda_n)^{-1/2} \ . \tag{17}$$

This characteristic function, along with the eigenvalue equation (13), completely describes the problem as long as we specify what $J_V(t_1,t_2)$ is. The probability density of A is the inverse Fourier transform of this characteristic function.

Before continuing, note that if we assume that all the excitons hop at the same rate Γ, then the correlation function takes the form

$$\begin{aligned} &J_V(t_1-t_2)=J_V(t;t) \ , \quad 0<|t_1-t_2|<1/\Gamma \ , \\ &J_V(t_1-t_2)=0 \ , \quad |t_1-t_2|\ge 1/\Gamma \ . \end{aligned}$$

This is equivalent to saying that an exciton visits an average of m sites in its lifetime, where $m=(\Gamma/\langle w\rangle)+1$. From the integral equation (13), we find that all the eigenvalues are equal in this case, and if we cut the number of eigenvalues off such that $\lambda_n=\lambda_0$ for $n\le m$ and $\lambda_n=0$ for $n>m$, we see that the characteristic function becomes

$$M_A(i\xi)=(1-2i\xi\lambda_0)^{-m/2} \ . \tag{18}$$

The probability density of A is the inverse Fourier transform of $M_A(i\xi)$, and is

$$p(A)=\frac{\left[\dfrac{1}{2\lambda_0}\right]^{m/2} A^{(m/2-1)}e^{-A/2\lambda_0}}{\Gamma\left[\dfrac{m}{2}\right]} , \tag{19}$$

where $\Gamma(m/2)$ is the Γ function. From this we can determine the mean of A:

$$\langle A\rangle=\int_{-\infty}^{+\infty} Ap(A)dA=m\lambda_0$$

or

$$\lambda_0=\frac{\langle A\rangle}{m} \ .$$

The decay rate is proportional to A so we replace A by w and $\langle A\rangle$ by $\langle w\rangle$. The intensity decay is then

$$\begin{aligned} I(t) &= \langle w\rangle^{-1}\int_0^\infty we^{-wt}p(w)dw \\ &= \left[1+\frac{2\langle w\rangle t}{m}\right]^{-(m/2+1)} , \end{aligned} \tag{20}$$

which is, again, an approximation because we have assumed that the hopping rate is the same for all excitons.

In actuality we should use a distribution of exciton

jump times. The jump time is the time an exciton occupies a site before jumping to another. We assume that the exciton coherence time is given by the average jump time $1/\Gamma$, where Γ is the average hopping rate. The normalized autocorrelation function is then given by

$$g_1(t_1;t_2)=\frac{J_V(t_1;t_2)}{J_V(t;t)}=\exp(-\Gamma|t_1-t_2|)\ , \tag{21}$$

where

$$J_V(t;t)=\langle V^2\rangle=\frac{\langle A\rangle}{T}\ . \tag{22}$$

This autocorrelation function corresponds to an exponential distribution of hopping times where Γ is the width of the Lorentzian broadened line. We are here assuming that for any particular exciton there is a constant hopping probability Γ per unit time so the probability that the exciton has not made a hop after time t is $\exp(-\Gamma t)$. In fact, different sites will have different Γ's, so this is an approximation, but as long as Γ is not spatially correlated with w it should be a good one. The integral equation (13) now becomes

$$\int_0^T \exp(-\Gamma|t_1-t_2|)\varphi_n(t_2)dt_2=\eta_n\varphi_n(t_1)\ , \tag{23}$$

where

$$\eta_n=\frac{T\lambda_n}{\langle A\rangle}\ .$$

This integral equation was considered by both Slepian[13] and Jakeman and Pike.[12] The eigenvalues λ_k are given by

$$\lambda_k=\frac{2\langle A\rangle\gamma}{\gamma^2+y_k^2} \tag{24}$$

where the y_k are given by the roots of the following:

$$y_k\tan(\tfrac{1}{2}y_k)=\gamma \quad \text{where } 0>y_0>\pi \tag{25a}$$

and

$$y_k\cot(\tfrac{1}{2}y_k)=-\gamma \quad \text{where } \pi<y_1<2\pi\ . \tag{25b}$$

Here $\gamma=\Gamma T$ and is the ratio of exciton lifetime to jump time.

We are presently considering the case in which $k=k_X$ and the characteristic function is given by (17). The probability density is then given by

$$p(A)=F^{-1}\{M_A(i\xi)\}=\int_{-\infty}^{\infty}\frac{e^{i\xi A}}{\prod_{n=0}^{\infty}(1-2i\xi\lambda_n)^{1/2}}d\xi\ , \tag{26}$$

for which an analytical solution cannot be found. Slepian, however, has devised a method of simplifying the problem so that a numerical computation can be done rather easily. We will simply describe the results here since any details do not add to the understanding of the problem at hand. The probability density of (26) becomes a sum of an infinite number of line integrals joining the singularities of (17) in the complex plane. The integrand of each of these is similar to that of (26). The denominator in these integrands is then rewritten as a function which does not contain a product of an infinite number of terms. It does, however, have identical properties to the original, including having zeros located at the same points.

The calculation is performed by first deciding how many terms to include in the sum referred to above and calculating the roots of Eq. (25). It was found that the number of terms required depends on the size of γ. For small values ($\sim .01$), including more than ten terms made a negligible difference; however, for larger values (~ 5), up to 50 terms were necessary. For each term of the sum a numerical integration is then performed. This yields the probability density of A as a function of $\langle A\rangle$ and γ. Recall that A is the motionally integrated value of the square of the scattering matrix element, and that the exciton radiative decay rate is proportional to it. As with the approximate derivation described above, we then substitute the decay rate w for A and $\langle w\rangle$ for $\langle A\rangle$. We then integrate over $p(w)$ as in Eq. (20) to arrive at a formula for the radiative decay. It was actually found to be much easier computationally to perform this integration by hand first, followed by the above-mentioned numerical integration, and then the summation.

When $k\neq k_X$, V in Eq. (10) is complex and the b_n are independent, complex random variables with variance $\lambda_n/2$. The determination of the probability density of A can then be performed exactly as in Jakeman and Pike.

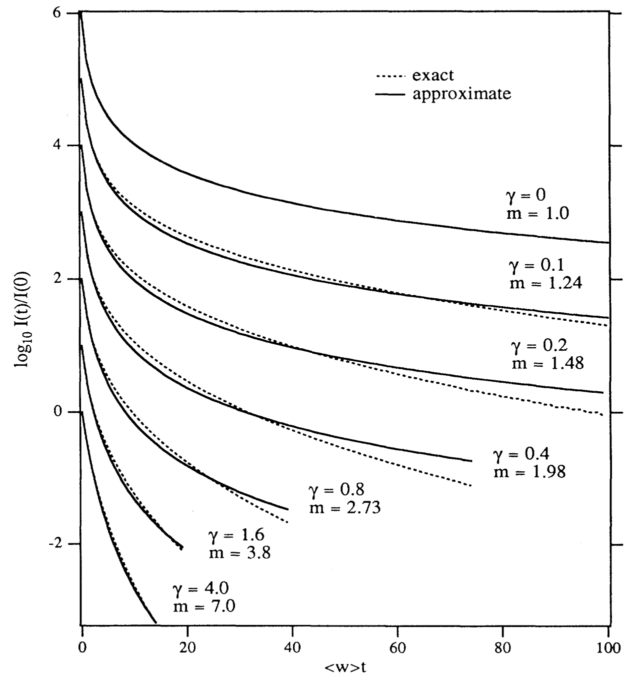

FIG. 1. Comparison of theories of zone-boundary ($k=k_X$) temperature-dependent radiative decay. The dashed lines are from the theory, which includes a distribution of exciton jump times, while the solid lines are fits to these curves using formula (20), which does not include a distribution. The curves are displaced from each other by 1.0 for clarity.

The result for the intensity decay in this case is

$$I(t)=\frac{e^{\gamma}}{\langle w\rangle^2}\sum_{k=0}^{\infty}(-1)^k\frac{2y_k^2}{y_k^2+2\gamma+\gamma^2}\times\left\{t+\left[\frac{\gamma^2+y_k^2}{2\langle w\rangle\gamma}\right]\right\}^{-2}, \tag{27}$$

where the y_k are the roots of Eq. (25). This formula is for the intensity decay of localized off-zone-boundary excitons in an alloy, and does not apply to a superlattice.

Before concluding this section, we note that the formula for intensity decay which does not include a distribution of residence times (20) turns out to be a poor approximation. This is illustrated in Fig. 1, where the dotted lines are the exact $k=k_X$ theory and the solid lines are best fits using the approximate theory. The exact and approximate theories are in agreement only in the limits of complete localization ($m=1,\gamma=0$) and complete delocalization (large m,γ).

III. EXPERIMENT

The data reported on in this section were taken on a type-II GaAs/AlAs superlattice consisting of 60 periods of 23-Å-thick GaAs layers and 19-Å thick AlAs layers. These thicknesses correspond approximately to eight and seven monolayers, respectively. The sample was grown by molecular-beam epitaxy (MBE) at Philips Research Laboratories, where the layer thicknesses were measured through x-ray diffraction.[15]

The sample was mounted in a variable-temperature liquid-helium cryostat. The temperature was stabilized with feedback to within ±0.2 K, and the temperature indicator was calibrated to ±0.25 K. The excitation source was an argon-ion laser which was pulsed with an acousto-optic modulator. The laser pulses were about 0.2 μs long, and the repetition rate was 25 kHz. The luminescence from the sample, after passing through a spectrometer, was detected by a cooled photomultiplier tube. Intensity decay was measured with a photon-counting apparatus.

Figure 2 displays the photoluminescence spectrum at various temperatures in the same temperature range that decay measurements were made. The vertical lines indicate energies, relative to the peak, at which decay was measured. Energies closer to the peak were not studied because many higher-energy excitons migrate to lower-lying states during the initial portion of the decay.[6] This process of spectral diffusion spoils the agreement with theory at low temperature [Eq. (6)], which is necessary before one can expect agreement at higher temperatures. The spectrum is noticeably asymmetric at lower temperatures, with more intensity being located on the low-energy side. This characteristic of the spectrum has been observed before in other indirect-gap disordered semiconductors such as $GaAs_{1-x}P_x$.[16] As temperature is raised, the line broadens somewhat and becomes more symmetric. At the same time the peak shifts slightly to lower energy.

In Fig. 3, exciton luminescence intensity decay is plotted logarithmically for five temperatures between 6 and 18 K. These measurements were made at an exciton energy of 1.851 eV, which is about 3.5 meV below the peak in the low-temperature luminescence spectrum, and at an excitation intensity of 0.6 J/cm^2 which corresponds to 3×10^{11} photons/cm^2 per pulse. The data clearly reveal a transition to exponential decay as temperature is raised, and by 18 K the decay is almost entirely exponential. At this temperature the change in slope with time indicates that the decay rate decreases by only about 30% over four decades of decay. The solid lines were calculated using the theory developed in the previous section. $\langle w\rangle$ is held constant at 7.0×10^6 s^{-1} for each temperature. This rate was found by fitting the low-temperature decay formula (6) to the 6-K data. At this low temperature there is a small exponential component to the decay with rate 1×10^4 s^{-1} which is due to phonon-assisted processes. This small contribution is not detectable at higher temperatures.

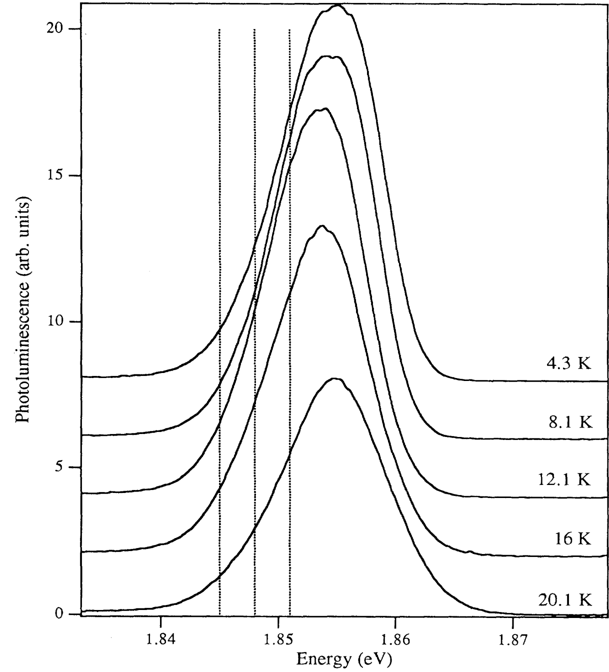

FIG. 2. Photoluminescence spectra measured at five temperatures showing the no-phonon line of the X-Γ transition. The dotted lines indicate the exciton energies at which the luminescence decay measurements were made.

The values of γ indicated in Fig. 3 are results of best fits to the theory. Recall that γ is the ratio of exciton lifetime to jump time. The lower curve in Fig. 4 displays $\ln(\gamma)$ vs $1/T$ for the decay data of Fig. 3. The solid line is a fit of the following:

$$\gamma=A\exp(-E_a/kT)+B\exp(-E_b/kT), \tag{28}$$

with $A=250\pm100$ and $E_a=4.8\pm0.5$ meV. The errors involved in B and E_b are so large (nearly as large as the parameter value itself) that a quantitative discussion is almost meaningless. The γ error bars at the lowest (8 K)

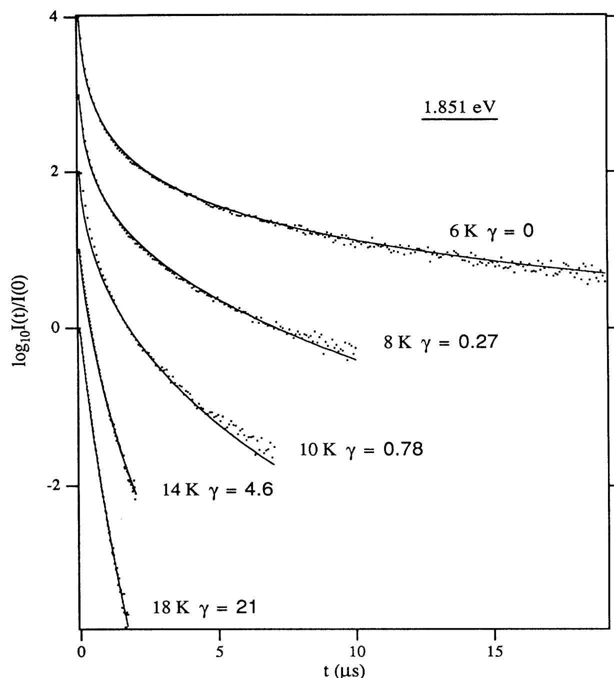

FIG. 3. The temperature-dependent luminescence decay plotted on a $\log_{10}$ scale. The excitation intensity is 0.6 J/cm^2 and the exciton energy is 1.851 eV. The solid lines were calculated using the theory developed in Sec. II. The values of γ used are indicated on the graph, and $\langle w \rangle = 7.0 \times 10\ s^{-1}$ at all temperatures. The curves are displaced from each other by 1.0 for clarity.

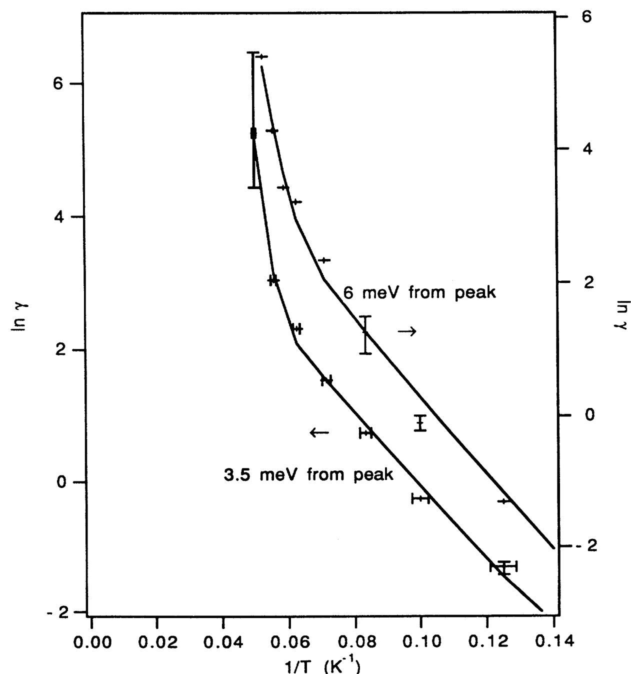

FIG. 4. $\ln\gamma$ vs $1/T$ for two sets of decay data measured 3.5 and 6.0 meV from the peak in the photoluminescence spectrum. The solid lines are fits of Eq. (28). The right and left axes are separated by 1.0 for clarity.

and highest temperatures (20 K) are the result of the standard deviation in the theoretical fit plus an assumption of a 5% inaccuracy in $\langle w \rangle$. The temperature error bars assume a ± 0.25-K error in the calibration of the temperature indicator.

The upper curve in Fig. 4 is from a set of temperature-dependent decay data measured at a lower exciton energy, 6 meV below the luminescence peak, and a smaller excitation intensity of about 0.12 J/cm^2. At this exciton energy we obtain similar results to those above, but there are some significant differences. It was found to be necessary to use the smaller excitation intensity in order to avoid saturation of localized states and achieve good agreement between theory and experiment. This point will be discussed further below. It was also found that the fitted $\langle w \rangle$ varies with temperature. It has a value of $6.6 \times 10^6\ s^{-1}$ at the lowest (4–6-K) and highest (17–20-K) temperatures, but is smaller at intermediate temperatures, being as low as $4.6 \times 10^6\ s^{-1}$ at 12 K. At this low excitation intensity, the same effect was observed at a higher exciton energy about 3 meV below the peak. Therefore it seems to be an intensity effect rather than an energy effect. We have, however, no explanation for it. The solid line is, again, a fit to Eq. (28) with $A = 450 \pm 100$ and $E_a = 5.0 \pm 0.5$ meV. For this set of data there are a few more high-temperature points available and $B \sim 10^9$ and $E_b \sim 30$ meV. The γ error bars at the 10-K and 12-K points represent an estimate of the error due to the lack of knowledge of $\langle w \rangle$ at these temperatures. The magnitude of the error is equal to the difference between the value of γ obtained from the best theoretical fit with $\langle w \rangle$ held at $6.6 \times 10^6\ s^{-1}$ and the value of γ obtained when $\langle w \rangle$ takes on the value to give the very best theoretical fit.

IV. EFFECT OF INTENSITY

As mentioned above, the excitation intensity which gave good agreement between theory and experiment at an exciton energy 3.5 meV below the photoluminescence (PL) peak (Fig. 3) gave poor agreement at the lower energy 6 meV below the PL peak. Figure 5 shows three low-temperature decay curves measured at an exciton energy about 6 meV below the PL peak and at three excitation intensities differing by factors of 10. The bottom curve is the highest intensity and the top curve is the lowest intensity. The solid lines are best fits of Eq. (6). The theory fits the data well at low intensity but becomes increasingly poor as intensity is increased. This is presumably due to a saturation of localized states at high excitation intensity and a subsequent filling of delocalized states.

This saturation can occur in the following manner. Since excitons are created with photons of energy well above the band gap (the excitation source is the 2.4-eV argon laser), they are initially truly delocalized in the sense that they occupy states with extended wave functions. Lower-energy localized states then serve as exciton traps. If, somehow, localized and delocalized states exist at the same energy, then, if all the localized states become filled, any remaining excitons will occupy delocalized states. Therefore, if a particular excitation intensity creates just enough excitons to saturate localized states,

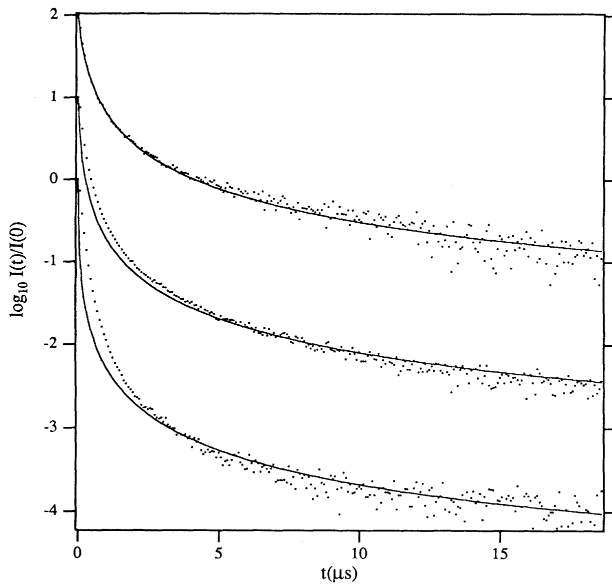

FIG. 5. The luminescence intensity decay measured at exciton energy 1.848 eV and three excitation intensities differing by factors of 10. The excitation intensity is about 0.05, 0.5, and 5 J/cm^2 for the top curve, middle curve, and bottom curve, respectively. The solid lines are best fits to formula (6). The curves are displaced from each other by 1.0 for clarity.

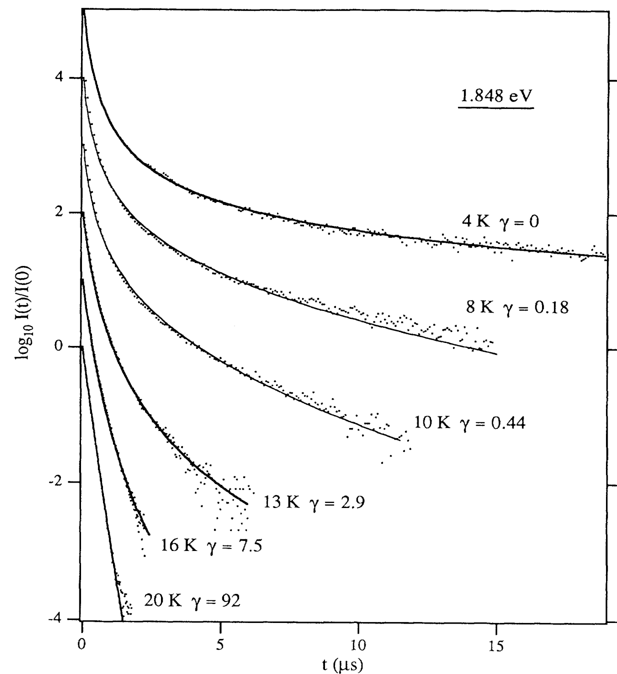

FIG. 6. The temperature-dependent luminescence decay plotted on a $\log_{10}$ scale. The excitation intensity is 0.6 J/cm^2 and the exciton energy is 1.848 eV. The solid lines were calculated using the model which includes both localized and partially delocalized excitons at low temperature. The values of γ for the localized excitons are indicated on the graph, and $\langle w \rangle = 7.0\times 10\ s^{-1}$ at all temperatures. The curves are displaced from each other by 1.0 for clarity.

at higher intensities the delocalized states will begin to fill. Localized and delocalized excitons can exist at the same energy if there are regions of the crystal which are not in communication with each other because they are separated by large energy barriers. An exciton in one region may be delocalized in the sense that its wave function extends throughout that region, but yet there are large energy barriers which prevent it from entering a different region in which, at the same energy, it may be completely delocalized. Note also that the saturation effect is only present at low energy where the density of states is small, the density of localized states being proportional to the excitation intensity which just saturates them.

This saturation effect can be modeled by assuming two populations of excitons to exist at low temperature, one which is localized and another which is partially delocalized. If we use the value of $\langle w \rangle$ obtained from high-temperature data where the decay is exponential, then there are two fitting parameters at low temperature. These are the ratio of the number of delocalized excitons to localized excitons and the value of γ for the delocalized excitons. Recall that γ for low-temperature localized excitons is zero. At higher temperatures (8–12 K), these two parameters are not allowed to vary, but are fixed at their low-temperature values. The only fitting parameter now is γ of the localized excitons. At the highest temperatures (14–20 K), the saturation effect is no longer noticeable since excitons are thermally delocalized anyhow. Therefore, it was only necessary to include the two populations of excitons up to about 12 K. Using a model such as this, we can fit the theory to the data measured at the two lowest exciton energies indicated in Fig. 2 and at the higher excitation intensity. This is displayed for the 6-meV data in Fig. 6. Here the ratio of the number of localized to delocalized excitons obtained from the low-temperature data is 1.35 and the value of γ for the low-temperature delocalized excitons is 2.7. We obtained fitted values of γ for the excitons localized at 4 K with a temperature dependence similar to that shown in Fig. 4, and Eq. (29) yields similar parameter values to those discussed above.

In summary, we have discussed four sets of decay data. Three of these were measured at the exciton energies relative to the peak of the PL indicated in Fig. 2 and at an intensity approximately equal to 0.6 J/cm^2. Of these we found good agreement with theory only at the highest energy. If we assume that the disagreement at the lower energies is due to a saturation of localized states, we can successfully model this saturation by adding a population of excitons which is partially delocalized even at low temperature. The fourth set of data was measured at the middle exciton energy of Fig. 2 and at a lower intensity of about 0.12 J/cm^2. Here we achieved good agreement with theory without invoking saturation.

V. DISCUSSION

Plots of the natural log of the values of γ resulting from fits of the theory of Sec. II vs inverse temperature indicate that γ increases with temperature according to

the double exponential of Eq. (28). The low-temperature region ($T < 15$ K) is clearly Arrhenius, and there is evidence of some sort of higher-temperature process contributing to the increased hopping rate or delocalization of excitons with increasing temperature.

The smaller activation energy can be identified with the energy of localization or perhaps the average well depth that localized excitons are situated in. The fits to γ of Eq. (28) showed this to be about 5 meV. This is comparable with the previous results on direct-gap excitons in quantum wells of Hegarty and Sturge,[8] who measured homogeneous linewidth and found E_a to be equal to the difference between the energy of the peak in absorption and the exciton energy. Our peak in the photoluminescence is at 1.855 eV (see Fig. 2) and is about 4 meV lower in energy than the peak in the absorption,[15] so these two results are in agreement within experimental error.

We do not, however, observe a mobility edge. If there were an effective mobility edge as in Ref. 8, one would expect to see a slope of -1 in a plot of activation energy vs exciton energy. We do not, however, see any increase in activation energy with decreasing exciton energy. This is possibly due to the uncertainty involved in our determination of E_a, since we were unable to make measurements close to the luminescence peak because of the spectral diffusion process. It may also be that a true mobility edge does not exist in this system, and that different exciton energies correspond to different regions in the sample between which there is no communication. Our result can be compared with that of the $Ga_{1-x}Al_xAs$ alloy.[4] Here an activation energy for nonradiative decay was measured. The dependence of this activation energy on emission energy also did not indicate a mobility edge. It was further found that at high intensity, nonradiative centers became saturated and decay was from the more numerous delocalized states. If there were a true mobility edge then there should be, in this case, a shift of the spectrum to higher energy which was not observed. The differences (this work versus that of Ref. 8) and similarities (this work versus that of Ref. 4) in results described above are probably not dependent on dimension, but rather on the time scale of the measurement.

The preexponential factor A is the $T=\infty$ ratio of exciton lifetime to jump time. At $T=\infty$ the lifetime is $1/\langle w \rangle$, which is approximately 1.4×10^{-7} s, and $A \sim 250$, so the jump time is on the order of 0.5 ns. From this we can determine the localization time; i.e., the time it takes for an exciton which is initially delocalized to find a localized site. To do this we need the density of localized sites. This can be estimated from the intensity at which localized sites become saturated, and is about 5×10^{8} cm^{-2}. The density of sites is the inverse of the exciton area which, for an exciton Bohr radius of 50 Å, is $\sim10^{12}$ cm^{-2}. The exciton therefore visits $\sim$2000 sites before it localizes and the localization time is ~ 1 μs.

The higher activation energy is larger than the exciton binding energy. If we picture this second process as involving the breakup of the exciton, followed by each particle hopping to neighboring wells and then recombining, this energy would be the exciton binding energy plus the localization energy. The exciton binding energy for a superlattice of comparable layer thicknesses to those of the sample used for this study has been calculated by Salamassi and Bauer[17] and measured by Hodge *et al.*[18] to be about 17 meV. We might therefore expect this activation energy to be about 20–25 meV. The value of 30 meV reported above is slightly larger than one would expect from this oversimplified picture, but not outside experimental error.

When excitons begin dissociating, γ increases dramatically because the individual electrons and holes have many more states available to them, thus increasing the effective exciton lifetime. The prefactor B is determined by mass action and should be the ratio of the product of electron and hole effective densities of states N_c and N_v to the density of excitons.[19] The density of electron (hole) states is given by $2(2\pi m_{c(v)} kT/h^2)^{3/2}$ in three dimensions, where $m_{c(v)}$ is the conduction-(valence-) band effective mass. The density of electron states per superlattice layer at 10 K is approximately 3×10^{9} cm^{-2}, and the density of hole states is approximately 7×10^{8} cm^{-2}. At the excitation intensity of 0.6 J/cm^{2}s the exciton density is $\sim10^{9}$ cm^{-2}, so we would expect B to be $\sim10^{9}$, as observed.

In conclusion, we have shown that the luminescence intensity decay of localized excitons in an indirect gap GaAs/AlAs superlattice is nonexponential at low temperature, and the measured decay curve is well fit by a model in which the excitons experience a distribution of decay rates corresponding to a Gaussian distribution of scattering matrix elements.[3] We have further shown that, as temperature is raised, the exciton decay gradually becomes exponential and is almost purely exponential by 20 K. At an excitation intensity at which localized states are not saturated, the temperature dependence of the exciton decay agrees well with a theory based on a model of excitons thermally hopping between localized sites and motionally averaging the distribution of decay rates. The extent of averaging is determined by a fitting parameter γ which is the ratio of exciton lifetime to jump time.

The temperature dependence of γ indicates that its increase with increasing temperature is due to two thermally activated processes. The first of these has a small activation energy $\sim$5 meV which is interpreted as the localization energy, and the second has a larger activation energy $\sim$30 meV which is of the order of the exciton binding energy. The activation energies and preexponential factors of the thermally activated processes were shown to be in reasonable agreement with other measurements or previously published results. No evidence was found for a mobility edge.

At high excitation intensity there is possibly a saturation of localized states and a subsequent filling of delocalized states. This effect can be modeled by considering two populations of excitons, one which is at least partially delocalized even at low temperature and another which is localized at low temperature and then becomes thermally delocalized.

We conclude that the temperature dependence of the decay of indirect excitons is well described by the model of Klein, Sturge, and Cohen, and that it can give some information on exciton dynamics.

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