Spin flip of excitons in GaAs quantum wells

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We report measurements of the rates of conversion from $J=2$ excitons to $J=1$ excitons, as well as conversion from right-handed $J=1$ excitons to left-handed $J=1$ excitons, and measurement of the intrinsic radiative lifetime of $J=1$ excitons, in very-high-quality GaAs quantum wells. Since the experiments are performed at very low temperature and with resonant excitation, the effects of energy relaxation are absent. [S0163-1829(97)03120-2]

I. INTRODUCTION

How excitons can lose or gain angular momentum is a fundamental question of carrier dynamics. Free atoms or electrons can only lose angular momentum by coupling to photons in the electromagnetic field; carriers in solids couple not only to radiation, however, but also to the phonon field of the entire lattice. This leads to much faster spin relaxation, but the relative contributions of different spin-lattice relaxation processes are still not fully understood.

The theory of carrier spin relaxation has concentrated on three mechanisms: the Elliot-Yafet (EY) mechanism,[1] the Dyakonov-Perel (DP) mechanism,[2] and the Bir-Aroniv-Pikus (BAP) mechanism[3] (for a review see Ref. 4). The EY mechanism notes that coupling between conduction-band states with opposite spin is allowed because of the mixing of conduction-band states at nonzero k with the valence band, in $\mathbf{k}\cdot\mathbf{p}$ theory. Any process that leads to momentum relaxation will therefore give rise to spin relaxation; consequently, the spin relaxation rate for this mechanism is predicted to be *proportional* to the carrier momentum relaxation rate. This process is generally considered to be weak in bulk semiconductors compared to the DP mechanism,[3,4] because $\mathbf{k}\cdot\mathbf{p}$ theory gives a denominator proportional to the band gap, but this process may be stronger in quantum wells.[5,6]

The DP mechanism takes into account the fact that in the zinc-blende symmetry a k^3 term can exist in the carrier Hamiltonian that gives an effective magnetic field, which leads to a spin splitting of the electronic states. As in an externally applied magnetic field, the electron spin will precess between spin states and dephasing processes will lead to an equilibration. The spin relaxation rate for this mechanism is predicted to be *inversely proportional* to the carrier momentum relaxation rate due to the peculiarities of ''motional narrowing.''[7] Since the coupling arises from the k^3 terms in the Hamiltonian, this mechanism is expected to predominate at high temperature in bulk semiconductors.

The BAP mechanism invokes exchange between a free electron and hole to yield a one-step spin flip of both. Since this mechanism is not strongly k dependent, it is expected to predominate at low temperature and high impurity concentration in bulk semiconductors.[8] Maialle, de Andrada, and Sham[5,6] have recently elaborated a similar theory for excitons, taking into account the exciton wave-function overlap. In this case, the spin relaxation is similar to the DP mechanism since the electron-hole exchange interaction can be viewed as an effective magnetic field. The spin relaxation rate for this mechanism is predicted to be *inversely proportional* to the carrier momentum relaxation rate, just as for the DP mechanism.

An important fact to realize for all of these mechanisms is that they do not conserve the total angular momentum of the carriers. This is because the carriers do not exist in a vacuum, but in a crystal medium, and only the momentum of the whole crystal must be conserved. *Any* dephasing process, even elastic scattering processes that conserve total carrier momentum, will couple the carriers to the crystal field, therefore allowing transfer of angular momentum to the crystal as a whole.

The rate of angular momentum relaxation of the free electron-hole gas in bulk GaAs has been measured in several experiments.[8–10] The spin relaxation of excitons in bulk GaAs is much harder to measure since the free exciton appears only weakly in the luminescence spectrum. In a quantum well, the opposite is true: it is much easier to look at the excitonic states. Each of the above mechanisms can occur for excitons in quantum wells, but the rates are affected by the altered wave-function overlap. Recent work[11,12] has shown that the angular momentum relaxation rate in quantum wells is substantially increased relative to bulk GaAs; Munoz *et al.*[13] have argued that substantial deviations in the absolute rates measured in different experiments can be attributed to

localization effects that arise since the quantum wells have rough surfaces; Frommer *et al.*[14] have attempted to distinguish between the localized and delocalized contributions of excitons in quantum wells.

Most of the experiments on excitons in quantum wells have concentrated on the conversion of excitons in the $J=1$ state that couples to the photon field, from a ''right-handed'' ($m_J=1$) circular polarization state to a ''left-handed'' ($m_J=-1$) state. Less work has addressed the issue of coupling to ''dark'' exciton states that exist in GaAs quantum wells, which do not couple directly to the dipole electric field. These states, which are known to exist from group-theoretical considerations and have been seen, for example, in two-photon absorption measurements,[15] have been accounted for in various ways in rate equations as channels for the decay of the $J=1$ excitons.

It is instructive to review the symmetries of the states.[16] The conduction-band s states and the valence-band p states are represented in the cubic zinc-blende T_d group as $^1\Gamma_1$ and $^3\Gamma_5$, respectively. In the double group representation, which takes into account electron spin, the $^1\Gamma_1$ conduction band becomes $^2\Gamma_6$ and the $^3\Gamma_5$ transforms to $^2\Gamma_7 \oplus {}^4\Gamma_8$. Spin-orbit coupling splits off the $^2\Gamma_7$ band, leaving the $^4\Gamma_8$ as the ground state of the holes. In a quantum well, which lowers the T_d symmetry to D_{2d}, this band is split into $^2\Gamma_6$ (light hole) and $^2\Gamma_7$ (heavy hole) bands. Since the lower mass of the light hole gives it greater zero-point energy, the heavy-hole state is the ground valence state in the quantum well. Excitons formed from the $^2\Gamma_6$ conduction-band electron and $^2\Gamma_7$ valence-band hole have symmetry $^2\Gamma_6 \otimes {}^2\Gamma_7 = {}^2\Gamma_5 \oplus {}^1\Gamma_3 \oplus {}^1\Gamma_4$, i.e., a doubly degenerate $^2\Gamma_5$ exciton with (e_x,e_y) symmetry and two nondegenerate dark exciton states. Optical experiments with varying magnetic field[17] have given an estimation of the splitting between these states. The $^2\Gamma_5$ exciton states couple directly to light propagating perpendicular to the well, with an in-plane electric field along either e_x or e_y, acting as a $J=1$ state. Of the remaining two states, the $^1\Gamma_3$ exciton has e_x^2-e_y^2 symmetry, while the $^1\Gamma_3$ exciton has e_xe_y symmetry.

Each of the dark states couples only to the two-photon operator, which has $^2\Gamma_5 \otimes {}^2\Gamma_5$ symmetry in D_{2d}. Therefore, two circularly polarized photons of the same handedness, or two linearly polarized photons, will excite a linear combination of these two exciton states. As seen in the above group-theoretical argument, neither of these states is a pure $m_J=2$ ($|2\rangle$) or $m_J=-2$ ($|-2\rangle$) state: the splitting occurs between eigenstates that correspond to ($|+2\rangle+i|-2\rangle$) and ($|+2\rangle-i|-2\rangle$). This means that two photons of the same circular polarization will excite each of these states, no matter what the handedness of the circular polarization. This fact is borne out in the experiments, as discussed below.

In this paper, we present experimental results for the rate of conversion from the dark exciton states to either of the $J=1$ exciton states. In order to understand these results, we must first review the results of our measurement of the rate of ''spin flip'' between $m_J=1$ and $m_J=-1$ states of the $^2\Gamma_5$ $J=1$ exciton, as well as the radiative lifetime of the $J=1$ excitons in quantum wells.

II. $J=1$ SPIN FLIP AND RADIATIVE LIFETIME

Several previous works have looked at the coupling of $J=1$ spin states of excitons in quantum wells under various conditions. In several papers (e.g., Refs. 11 and 12), spin polarizaton of excitons was observed in the direct recombination luminescence of excitons following *nonresonant* (above-gap) excitation. Vinattieri *et al.*[18] used resonant excitation of the heavy-hole excitons, but since their sample had a temperature of $\sim$10 K in helium vapor, they had to account for coupling of the excitons out of the resonantly excited states into higher-energy states. Frommer *et al.*[14] used a sample held at 1.6 K in an immersion cryostat and excited the excitons resonantly, but observed the exciton luminescence at slightly lower energies, i.e., from localized states. Munoz *et al.*[13] also studied spin flip following resonant excitation at low temperature, using a sample with a Stokes shift of 2.5 meV attached to a cyrostat cold finger at 4 K, which could have allowed the sample to rise to higher temperatures.

We have performed a similar experiment, but with a very-high-quality sample with no detectable Stokes shift between the photoluminescence excitation and photoluminescence spectra,[19] immersed in liquid helium at 1.6 K, in which we observe luminescence at the same energy as the resonant exciting photon energy. Since at this temperature k_BT is comparable to the linewidth, we detect the luminescence from the majority of the population of $J=1$ excitons at all times. This greatly simplifies the analysis of the data. Circularly polarized light from a picosecond Ti:sapphire laser was tuned to the heavy-hole resonance in the quantum well and luminescence was detected with a Hamamatsu streak camera. The specular reflection of the laser was eliminated with a beam stop so that overflow of the streak camera was not a problem.

Using the standard[12] method of fitting a straight line on a semilogarithmic plot of the ratio $(I_+-I_-)/(I_++I_-)$ as a function of time, where I_+ and I_- are the intensities of left- and right-circularly polarized luminescence, respectively, following excitation with a left-circularly polarized laser, we find a coupling time of 215 $\pm$ 10 ps at an estimated density of 10^9 cm^{-2} and 38 $\pm$ 4 ps at an estimated density of 10^{10} cm^{-2}. These results are consistent with earlier measurements of the spin-flip time in quantum wells by Munoz *et al.*,[13] which found a spin-flip rate proportional to density in the range 10^9–10^{10} cm^{-2}; a spin-flip rate increasing with increasing density was also reported in Ref. 20. In contrast, Ref. 12 reported a decreasing spin relaxation rate with increasing density, but that experiment used nonresonant excitation at higher lattice temperature; as seen in Sec. III, scattering into higher, nonradiative states substantially affects the measured decay rates.

No temperature dependence of the spin-flip time was observed in the range 1.6–9 K, which is also consistent with the experiments of Munoz *et al.*[13] At the estimated density of 10^{10} cm^{-2}, the momentum relaxation time due to exciton-exciton scattering in a quantum well is expected to be around 0.2 ps, by extrapolation from Ref. 21. By comparison, at a temperature of 9 K, the momentum relaxation time of excitons due to absorption of phonons is 20 ps, as seen in Fig. 2 below. Therefore, exciton-exciton scattering will dominate the momentum relaxation at these temperatures.

A density dependence of the spin-flip rate may at first seem surprising since in a vacuum, elastic collisions between particles will not change the total angular momentum of the

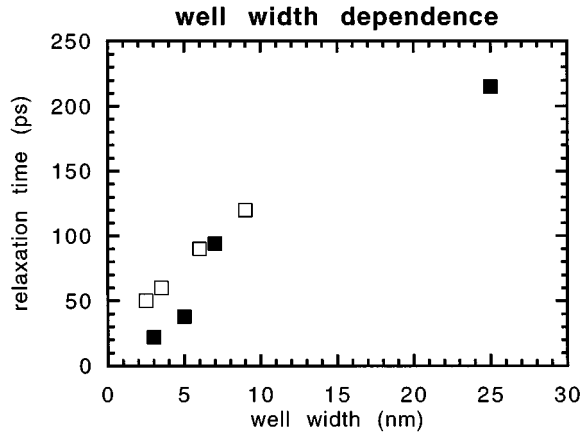

FIG. 1. Solid squares, a summary of the spin-flip time constants found in this study; open squares, spin-flip time constants from Ref. 11.

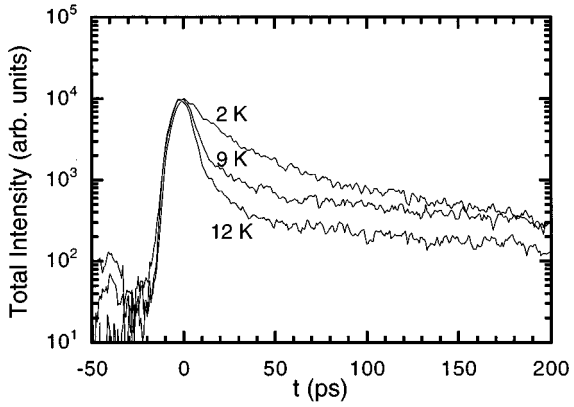

FIG. 2. Total (left- plus right-circularly polarized) recombination luminescence from the $J=1$ spin state of $1s$ heavy-hole excitons from a 25-nm GaAs quantum well, at three bath temperatures.

gas. Elastic collisions do contribute to momentum relaxation, however, and therefore to quantum state dephasing, as seen, e.g., in Ref. 22. As discussed in the Introduction, the EY mechanism allows any dephasing process to couple the carriers to the crystal momentum field. A spin relaxation rate proportional to the momentum relaxation rate is inconsistent with the DP and Maialle–de Andrada–Sham (MAS) mechanisms, however, which predict spin relaxation rate *inversely* proportional to momentum relaxation rate.

Figure 1 gives a summary of the measured spin-flip times as a function of well width. In addition, earlier data[11] are presented. Both data sets reveal the same trend toward faster spin relaxation with narrower well width, although the relaxation times observed here are consistently shorter than those reported earlier. This dependence on well width is consistent with the MAS theory,[5] although, as discussed above, the dependence on the momentum relaxation rate is not.

Considerable debate continues about the value of the intrinsic radiative recombination time in quantum wells. First-principles theory[23] suggests that the intrinsic lifetime of excitons should be of the order of 20 ps; recent theory[24] has indicated that localization of excitons due to surface irregularities can give a longer lifetime of up to 100 ps. Several recent experiments[18,25,26] have observed a fast initial decay of the exciton luminescence, in some cases followed by a longer late-time decay.

Several effects can give a fast initial decay of the exciton luminescence that have nothing to do with the intrinsic radiative lifetime. Energy and momentum conservation imply that only excitons with low momentum participate in the direct recombination process. For a direct luminesence linewidth of ~ 0.1 meV, this means that at exciton temperatures of greater than about 1 K, a substantial fraction of the excitons cannot decay through radiative recombination.

In the resonant-excitation experiments discussed above, we can determine the total population lifetime τ_0 by adding the intensities of the two polarizations of the luminescence. Since the radiative recombination luminescence only samples excitons in low-energy states, the measured τ_0 is really a sum of all rates out of the low-energy states, including inelastic scattering to high-energy states via phonons, and coupling to dark $J=2$ states. The densities in these experiments are well below the level required for significant Auger recombination.[27]

Figure 2 shows the total luminescence intensity, equal to the sum of both circular polarizations, from the high-quality 25-nm GaAs quantum well, following circularly polarized, resonant excitation of the excitonic ground state at very low density (approximately 10^9 cm^{-2}). As seen in this figure, at higher temperatures an initial fast decay occurs due to scattering of excitons into higher-energy nonradiative states; at lower temperatures this intitial fast decay disappears. The lifetime at the lowest temperature is measured in this sample as 96 ± 4 ps.

As in the study of Deveaud *et al.*[25] under very similar conditions, we find that the lifetime increases sublinearly with increasing density: at a density of approximately 3×10^{10} cm^{-2}, the lifetime increases to 140 ps, i.e., about 40% longer than the above value at 10^9 cm^{-2}. This dependence can be attributed to density-dependent scattering into higher-energy nonradiative states. We have not oberved a luminescence decay time of less than about 100 ps under these conditions, however, in contrast to Ref. 25, which reported decay times in the range 30–40 ps for a high-quality quantum well with width of 4.5 nm, at low density ($2\cdot10^9$ cm^{-2}), and lattice temperature of 1.7 K, and extrapolated these measurements to a value of 10 ps at extremely low density. This extrapolation was based on the assumption that scattering into nonradiative states is strictly proportional to the dephasing rate and that the dephasing at the lowest density was given by the Lorentzian linewidth of 0.34 meV extracted from the Gaussian inhomogenous broadening of 1.7 meV in their sample at the lowest density. If one assumes that the contribution of density-dependent scattering to nonradiative states is negligible at these densities, however, then most of the difference between our measured lifetime of 96 ps and the measured lifetimes in Ref. 25 at comparable densities can be attributed to the difference in excitonic binding energy in the two structures. The well width dependence of the excitonic binding energy implies a value of approximately 6.5 meV in our 25-nm well[28,29] and approximately 12.5 meV in a 4.5-nm well[30,31] with similar $Al_{0.3}Ga_{0.7}As$ barriers. Besides the well width difference, the sample of Ref. 25 had pure AlAs barriers, which gives at least 20%

greater excitonic binding energy[31] than estimated above; Gurioli *et al.*[32] give a value of 16.5 meV for a well with 5 nm width and pure AlAs barriers and extrapolate theoretically to a value of 17 meV for a well width of 4.5 nm. Since the excitonic recombination rate is proportional to the $r=0$ wave-function overlap of the exciton, which is inversely proportional to the square of the excitonic Bohr radius, this implies that the lifetime is inversely proportional to the excitonic binding energy. Within the experimental uncertainties, therefore, the ratio of excitonic binding energies imples a factor of 2.6 difference in the intrinsic lifetimes of our sample and that of Ref. 25, which is approximately the observed ratio of recorded lifetimes at comparable densities. Other factors that may contribute to the discrepancy may be that the sample used by Deavead *et al.* had a fast nonradiative decay due to tunneling into other parts of the structure and that their sample had slightly larger inhomogenous broadening, 1.7 meV compared to 0.3 meV in our sample.

As discussed in the following section, the lifetime measured in this experiment corresponds to the total radiative lifetime of the four coupled $J=2$ and $J=1$ states. In principle, if $J=1$ excitons converted quickly to $J=2$ excitons, which then did not convert quickly back to $J=1$ excitons (as, for instance, in the case that the $J=1$ states had substantially higher energy than the $J=2$ states), then the measured lifetime would really give the time of conversion from $J=2$ excitons to $J=1$ excitons, instead of the intrinsic radiative lifetime. As discussed in the next section, however, when the two-photon excitation data are analyzed taking into account the different possible couplings between $J=2$ and $J=1$ excitons, the value of the radiative lifetime is not substantially affected.

III. $J=2$ TO $J=1$ CONVERSION

A different experiment[33,34] allows direct measurement of the conversion rate of $J=2$ to $J=1$ excitons. The experiment is basically as follows. First, $J=2$ excitons are created via two-photon (infrared) excitation. Following the generation of the excitons, the single-photon recombination luminescence (visible or near infrared) from the $J=1$ excitons is detected with a streak camera with an S20 photocathode, which is completely insensitive in the infrared. Since the streak camera does not respond to the exciting laser light, the $J=2$ excitons can be created by resonant excitation and observed immediately thereafter, without unwanted background from the laser light.

This experiment relies on the fact that just as single-photon emission from $J=2$ states is forbidden, two-photon absorption by $J=1$ excitons is forbidden and two-photon absorption by $J=2$ excitons is allowed. Previous studies[15] have shown that two-photon absorption into the $1s$ heavy-hole exciton state in GaAs quantum wells is comparable to two-photon absorption into the $2p$ state. Therefore, in these experiments excitons can be created directly in the $J=2$ ground state of the quantum well.

An optical parametric oscillator (OPO) sychronously pumped by a Ti:sapphire laser was used to generate 100-fs infrared pulses, which could be tuned in the wavelength range 1400–1550 nm to create excitons in a GaAs quantum-well sample via two-photon absorption. RG1000 filters in the exciting laser beam path ensured that no light from the pump laser reached the sample. Single-photon-recombination luminescence from the sample was detected in the wavelength range 700–800 nm by a Hamamatsu streak camera with time resolution of about 10 ps. Several samples with $Al_{0.3}Ga_{0.7}As$ barriers and varying GaAs well width were examined. For a 3-nm well width, heavy-hole excitons in the $1s$ state are generated by laser light at approximately 1460 nm.

Three pitfalls must be avoided in these experiments. First, since the 100-fs exciting laser has a full width at half maximum of 20 meV, when the laser is tuned to the $1s$ resonance, it is also possible to generate excitons in the $2p$ states, which can then drop down into $1s$ exciton states and give a rise time of $J=1$ luminescence unrelated to the $J=2/J=1$ conversion time. To avoid this problem, the exciting laser was tuned to 10 meV *below* the $1s$ resonance. Since the number of $2p$ excitons depends on the *square* of the laser intensity resonant with the $2p$ state, the contribution due to down-conversion from $2p$ excitons can be made negligible.

Second, at high powers it is also possible to generate highly excited excitons via *three-photon* excitation from the substrate or from wider quantum wells in a multiple-quantum-well structure (two photons create an exciton, which is then excited over a barrier into a higher-lying quantum-well states). This was checked in these experiments via observation of the $1s$ luminescence when the exciting laser photon energy was *well below* the ground state, i.e., when no excitons could be created directly by two-photon excitation at all. In this case, weak but measureable luminescence from the $1s$ excitons still occurred, with total intensity proportional to the laser power to the third power. This could only come about due to relaxation of excitons excited over the barrier layers by absorption of a photon. Using low laser power substantially reduces this effect, but, in general, some contribution from this effect always occurs. Since this small contribution does not depend strongly on laser wavelength, the three-photon signal, taken at laser photon energies well below resonance, can simply be subtracted from the signal when the laser is near resonance.

Third, since only excitons with energy less than the homogeneous linewidth can recombine, at high lattice temperature excitons will be excited into higher-energy nonradiating states, substantially complicating the analysis of the conversion times, as seen in Fig. 2. To prevent this, the sample was held at 1.6 K via immersion in a liquid-helium bath.

Since almost no excitons are created in higher states in this experiment, we can write simple rate equations for the ground states of the $J=1$ and $J=2$ states since these should be the only relevant populations. Only $J=1$ excitons are assumed to decay to recombination with lifetime τ_0 since the $J=2$ excitons do not couple to the photon field. Therefore, for the two populations n_1 and n_2 representing the coupled $J=1$ and $J=2$ states, with only the n_1 population undergoing decay, we write the rate equations

$$\frac{\partial n_1}{\partial t}=-\frac{n_1}{\tau_{12}}+\frac{n_2}{\tau_{21}}-\frac{n_1}{\tau_0}, \tag{1}$$

$$\frac{\partial n_2}{\partial t}=\frac{n_1}{\tau_{12}}-\frac{n_2}{\tau_{21}}.$$

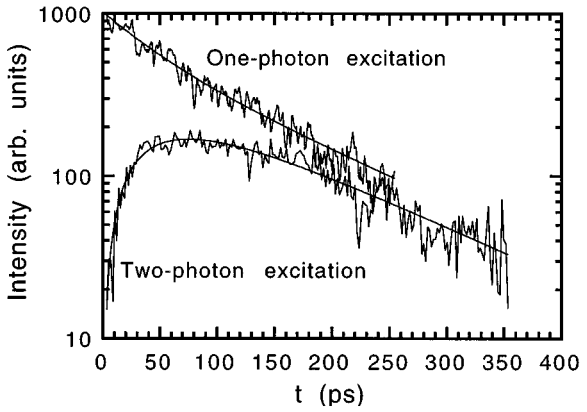

FIG. 3. Lower curve, recombination luminescence from the $J=1$ spin state of $1s$ heavy-hole excitons in a 3-nm GaAs quantum well, following generation in the $J=2$ spin state by a short (100 fs), circularly polarized laser pulse at 1471 nm, slightly below the resonance for two-photon generation; solid line, a fit of the rate equations discussed in the text; upper curve, total (left- plus right-circularly polarized) recombination luminescence from the $J=1$ spin state of $1s$ heavy-hole excitons in the same quantum well, following generation by a short (<5 ps) laser pulse at the resonance for single-photon generation; solid line, a fit of the rate equations discussed in the text. The relative scales are arbitrary since the excitation densities cannot be directly compared.

Since there is an energy splitting between the $J=1$ and $J=2$ states, in principle, there can be two rates: $1/\tau_{12}$ for conversion from $J=1$ to $J=2$ states and $1/\tau_{21}$ for conversion from $J=2$ to $J=1$ states.

For initial conditions $n_1=0$, $n_2=1$, the solution of the above equations gives

$$n_1(t)\propto e^{-t(\Gamma_{\rm tot}+\sqrt{\Gamma_{\rm tot}^2-\Gamma_{2*}^2})}-e^{-t(\Gamma_{\rm tot}-\sqrt{\Gamma_{\rm tot}^2-\Gamma_{2*}^2})}, \qquad (2)$$

where $\Gamma_{\rm tot}=\frac{1}{2}(1/\tau_{12}+1/\tau_{21}+1/\tau_0)$ and $\Gamma_{2*}=\sqrt{1/\tau_{21}\tau_0}$.

The lower curve of Fig. 3 shows as a function of time the luminescence at 730 nm, from a 3-nm quantum well at 2 K, excited by circularly polarized OPO light at 1471 nm (i.e., two-photon excitation about 10 meV below the $1s$ heavy-hole resonance.) The data are reasonably well fit by the two-parameter solution (2), shown as the solid curve.

In order to extract information about the individual rates, the two-photon excitation data must be compared to the single-photon excitation data for the same quantum well. The upper curve of Fig. 3 shows the total (right-circularly polarized plus left-circularly polarized) luminescence following single-photon excitation with circularly polarized light at the heavy-hole exciton resonance, similar to the data of Fig. 2 at low temperature. For the initial conditions $n_1=1$, $n_2=0$, the solution of the rate equations (1) is

$$n_1(t)\propto(\Gamma_{2*}-\Gamma_{\rm tot}+\Gamma_{1*})e^{-t(\Gamma_{\rm tot}+\sqrt{\Gamma_{\rm tot}^2-\Gamma_{2*}^2})} +(\Gamma_{2*}+\Gamma_{\rm tot}-\Gamma_{1*})e^{-t(\Gamma_{\rm tot}-\sqrt{\Gamma_{\rm tot}^2-\Gamma_{2*}^2})}, \qquad (3)$$

where $\Gamma_{1*}=(1/\tau_{12}+1/\tau_0)$. Since the two time constants are already known from the fit to the two-photon excitation data, this solution leads to a one-parameter fit of the single-photon excitation data. As seen in Fig. 3, a good fit is obtained. Using the fit values of the three time constants, we obtain $\tau_{12}=256$ ps, $\tau_{21}=62$ ps, and $\tau_0=92$ ps.

The difference in τ_{12} and τ_{21} is consistent with an energy splitting of 0.2 meV, assuming that $1/\tau_{12}=e^{-\Delta/k_BT}(1/\tau_{21})$, for the bath temperature of 1.6 K in these experiments. This energy splitting is consistent with the absolute value of the number found in magnetic resonance experiments;[17] however, in that work the authors argued that the $J=1$ state should lie *above* the $J=2$ states. The present experiments indicate the opposite conclusion, however. Suppose that the $J=1$ states lay above the $J=2$ states. Then, if the coupling were fast, there would be an initial fast decay in the luminescence following single-photon excitation during the time that $J=1$ excitons convert down to $J=2$ excitons. If the coupling were slow, then the coupling of $J=2$ excitons up to $J=1$ excitons would be even slower, in which case the rise time of the luminescence following two-photon excitation would be much slower than observed here. Without further study of the temperature dependence of the two-photon excitation data, however, we cannot be sure that the asymmetry of the coupling rates τ_{12} and τ_{21} arises due to the Boltzmannian occupation factor $e^{-\Delta/k_BT}$ and therefore the estimation of the energy splitting here is tentative.

Maille *et al.*[6] wrote two matrix elements for the coupling from $J=2$ excitons to the $m_J=+1$ and $m_J=-1$ states of the $J=1$ excitons. In these two-photon excitation experiments, however, no appreciable difference in the data is seen when the visible $J=1$ luminescence is analyzed for linear polarization or for circular polarization of either handedness, although both of the incoming photons in the two-photon excitation are circularly polarized with the same handedness. This is consistent with the group-theoretical considerations discussed in the Introduction, which imply that *both* dark states couple in the same way to two-photon excitation of either handedness.

IV. CONCLUSION

We have arrived at several surprising conclusions. First, the spin flip from the $m_J=+1$ and $m_J=-1$ states of the $J=1$ exciton depends on the carrier density and quantum-well width, but not on temperature, in these experiments. Our measured rates are consistent with earlier measurements[11,13] but not consistent with the proposed DP and MAS mechanisms of spin relaxation; our results do have the same dependence on momentum relaxation as the EY mechanism, although early arguments[3] indicated that the EY mechanism should be weak.

The intrinsic radiative lifetime of the $J=1$ states, which we measure in our experiment in which the reflected laser beam is outside the detection solid angle, is approximately 100 ps in our 25 nm quantum-well samples. Initially fast decay of the luminescence at higher temperatures is found to arise because of scattering of the excitons into higher, nonradiative states.

This experiment provides a direct measurement of the rate of conversion of $J=2$ excitons to $J=1$ excitons. The time scale for the spin-flip process is of the order of 60 ps. The conversion rate seems to be the same for transitions from the $J=2$ states to either the $m_J=+1$ or the $m_J=-1$ spin states,

even when the $J=2$ excitons are created by two circularly polarized photons of the same handedness. This is consistent with group-theoretical arguments that neither of the two $J=2$ states is a pure spin state.

A comparison of the luminescence lifetimes, assuming a Boltzmannian occupation of the spin states, indicates that the $J=2$ states lie *above* the $J=1$ states, in contrast to earlier deductions based on magnetic resonance experiments that placed the $J=2$ states below the $J=1$ states. The amount of splitting estimated here, 0.2 meV, is consistent with those measurements, however.

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