Spectrally resolved Overhauser shifts in single GaAs/Al_xGa_{1-x}As quantum dots

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In this work, the direct observation of spectrally resolved, polarization-dependent shifts in excitonic Zeeman splittings arising from dynamic polarization of lattice nuclei (the Overhauser effect) in single quantum dots is reported. Overhauser shifts corresponding to effective nuclear fields greater than 1.3 T were observed from quantum dots in a 4.2-nm-wide quantum well. These electron-nuclear interactions are an important aspect of the optical properties of quantum dots and may significantly affect recombination dynamics in quantum dots even in the absence of an external magnetic field.
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Early luminescence studies of quantum dots measured ensembles of dots, which resulted in large inhomogeneously broadened spectral features. Recently, however, several groups have reported high-resolution spectroscopic studies of excitonic luminescence from single quantum dots.[1–7] Magnetic field studies of single quantum dots, including an analysis of the Zeeman splitting as a function of applied magnetic field, have been previously discussed.[2,4] However, effects of nuclear spin polarization on the observed excitonic Zeeman splittings were not considered.

Optical pumping of the nuclear spin system is a two-step process involving the transfer of angular momentum from photons to the nuclear spin system.[8,9] The first step, the polarization of the electron spins by absorption of photons, is accomplished by exciting the system with circularly polarized light in a longitudinal external magnetic field. In the second step, the electron spin polarization is transferred to the nuclear system through the hyperfine interaction, preferentially orienting the nuclear magnetic moments. A static effective magnetic field proportional to the degree of nuclear orientation then acts back on the electron system, shifting the electronic energy levels (Overhauser shift). Overhauser shifts in electron spin resonance have been observed from a variety of semiconductor materials,[10–19] with shifts corresponding to effective nuclear fields up to 1.0 T being observed in measurements of a two-dimensional electron gas confined at a GaAs/Al_xGa_{1-x} interface.[18]

In this work, we consider effects of nuclear spin polarization on excitonic recombination from single quantum dots in an external magnetic field. In particular, we report the direct observation of spectrally resolved, polarization-dependent Overhauser shifts of excitonic Zeeman splittings corresponding to effective nuclear hyperfine fields as large as 1.3 T. These shifts demonstrate one important consequence of the hyperfine interaction on the magnetospectroscopy of single quantum dots. Even in the absence of an external magnetic field, the hyperfine interaction may strongly influence spin relaxation and recombination dynamics in these confined systems.[20,21] In addition, the observation of effects of nuclear orientation on excitonic luminescence from a single quantum dot implies that optical nuclear magnetic resonance of a single quantum dot may be possible.[15,22,23]

In this work, luminescence was excited and collected through a 1.5-μm aperture in an aluminum mask deposited on the sample, which consisted of a series of five single GaAs/$Al_{0.3}G_{0.7}$As quantum wells with varying width. Here we will primarily consider luminescence from a 4.2 nm wide quantum well. Details of the sample preparation and experimental setup have been previously reported.[6,7] Luminescence from the sample, shown in Fig. 1, consisted of a number of exceptionally narrow features arising from recombination of excitons localized by potential fluctuations in individual quantum dots within the quantum well.[6,7] From the energies of the excited exciton states, as measured by excitation spectroscopy, the lateral dimensions of the quantum dots are estimated to be roughly 100 nm, with values as low as 40 nm.[7]

The evolution of the luminescence in an external magnetic field when exciting the quantum dots with circularly polarized light is shown in Fig. 2 for applied magnetic fields varying from 0.5–3.0 T. Consider in detail the magnetic field dependence of the exciton with a zero-field energy of ~1.623 eV. For excitation with $\sigma+$ polarized light, the energy levels are resolvably split at 0.5 T into two levels [Fig. 2(a)]. As the magnetic field increases, the splitting of the two

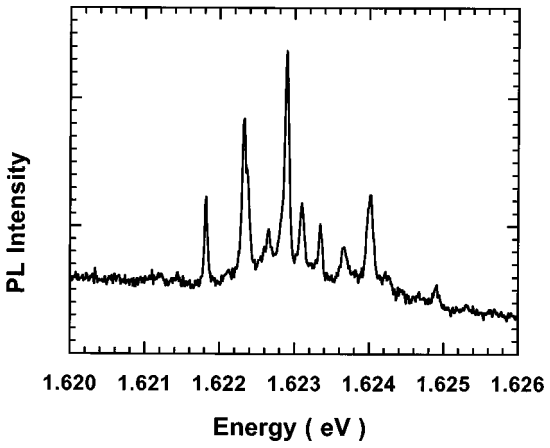

FIG. 1. Luminescence spectrum from a 4.2-nm-wide quantum well. The sample was optically excited through a 1.5-μm aperture with 10 mW of power at 1.637 eV; the laser spot size was ~100 μm. The sample temperature was 5 K.

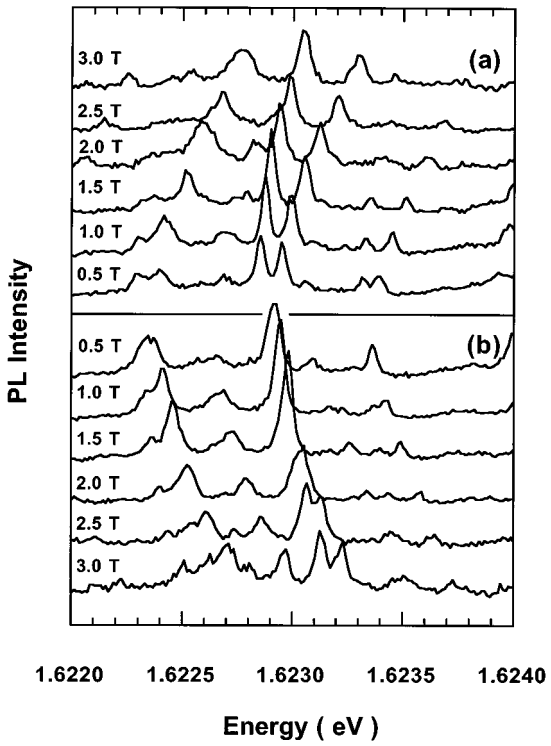

FIG. 2. Polarization dependence of excitonic splittings from single quantum dots in an external magnetic field. (a) $\sigma+$ excitation; $B_{\text{total}}=B_0+B_N$. (b) $\sigma-$ excitation; $B_{\text{total}}=B_0-B_N$.

levels increases. In addition, a shift in the energy of both levels to higher energy with increasing magnetic field is observed. A different field dependence of the splitting of the line is observed with $\sigma-$ excitation [Fig. 2(b)]. In this case, resolvable splitting of the luminescence is not observed until magnetic fields greater than 2.0 T are applied, while the shift in the energy of the luminescence is similar to that observed for $\sigma+$ excitation.

These experimental observations can be explained using a simplified Hamiltonian,

$$\hat{H}=g^*\mu_B B_0 S_z+\alpha B_0^2+\langle A\cdot I\rangle_z S_z, \tag{1}$$

which neglects the exchange interaction, but includes a hyperfine term.[19] In this expression, g^* is the exciton g value in the z direction, μ_B the Bohr magneton, S_Z the excitonic spin projection along the quantization axis, z, and α a constant which depends on the in-plane spatial extent of the exciton. B_0 is the external magnetic field, oriented along the z direction. $\langle A\cdot I\rangle_Z$ is the ensemble average of the z component of the hyperfine interaction, reflecting the degree of orientation of the nuclear moments. The first term in the Hamiltonian in Eq. (1), linear in applied magnetic field, is the spin-dependent Zeeman interaction; the second term describes the spin-independent diamagnetic shift of the exciton energy levels; the final term describes the hyperfine interaction. Eq. (1) can be written

$$\hat{H}=g^*\mu_B(B_0+B_N)S_z+\alpha B_0^2, \tag{2}$$

where the hyperfine interaction is now expressed as an effective nuclear field B_N:

$$B_N\equiv\langle A\cdot I\rangle_z/g^*\mu_B. \tag{3}$$

The orientation of the nuclear magnetic moments is determined by the electronic spin polarization, which in turn depends on the optical selection rules. For GaAs quantum dots, changing the excitation from $\sigma+$ to $\sigma-$ flips the orientation of the electron spin polarization. The nuclear moments can therefore be aligned either parallel or antiparallel to the external field, depending on the polarization of the incident light with respect to the direction of the external magnetic field. The effective internal magnetic field will then add to or subtract from the external field, leading to differences in the total effective magnetic field in the spin-dependent term in the Hamiltonian in Eq. (2), which depend on the polarization of the light.

In comparing the experimental results in Fig. 2 with the Hamiltonian in Eq. (2), the diamagnetic shift of the exciton energy levels, spin-dependent splittings, and polarization-dependent Overhauser shifts in the splittings are clearly identifiable. Consider again the magnetic field dependence of the exciton with a zero-field energy of ~1.623 eV. In Fig. 3(a), the energies of excitonic recombination from the Zeeman-split pair are plotted as a function of applied magnetic field for $\sigma+$ excitation. The average values are then fit to a quadratic magnetic field dependence, giving a value for the diamagnetic shift α of 26 μeV/T^2 for this exciton. An analysis of the diamagnetic shift of several other excitons localized in quantum dots in this quantum well gave values of α ranging from 21–34 μeV/T^2. Effective g values are also readily obtained from an analysis of the data. In Fig. 3(b), the excitonic Zeeman splitting for $\sigma+$ excitation is fit to a linear field dependence, giving a slope of 64 ± 5 μeV/T, or a g^* value of 1.1. Similar g^* values were obtained from an analysis of other dots in this quantum well.

Finally, the large polarization-dependent differences in excitonic splittings shown in Fig. 2 are explained by changes in the orientation of the internal effective nuclear field relative to the external magnetic field. Quite simply, in Fig. 2(a), the effective nuclear field B_N is aligned parallel to the external magnetic field and the total effective magnetic field in the spin-dependent term in Eq. 2 is equal to B_0+B_N. In Fig. 2(b), the nuclear field is aligned antiparallel to the applied field, giving a total effective magnetic field $B_T=B_0-B_N$. At 2.5 T, this difference is equal to 157 μeV, as shown in Fig. 3(b). Given an excitonic splitting of 64 μeV/T, a shift of 157 μeV corresponds to a change in the total magnetic field of ~2.5 T, or an effective nuclear field B_N of 1.3 T.

Evidence of the hyperfine origin of the polarization-dependent shift in excitonic splittings is given by the strong dependence of the magnitude of the splitting on excitation intensity. As shown in Fig. 4(a), for $\sigma+$ excitation, the magnitude of the observed splitting increases from 143 to 192 μeV as the excitation power goes from 1 to 20 mW. In contrast, for $\sigma-$ excitation, the magnitude of the splitting decreases with increasing excitation power from 87 to 12 μeV.

This behavior is readily explained by the intensity dependence of the strength of the hyperfine term $\langle A\cdot I\rangle_z$, which is

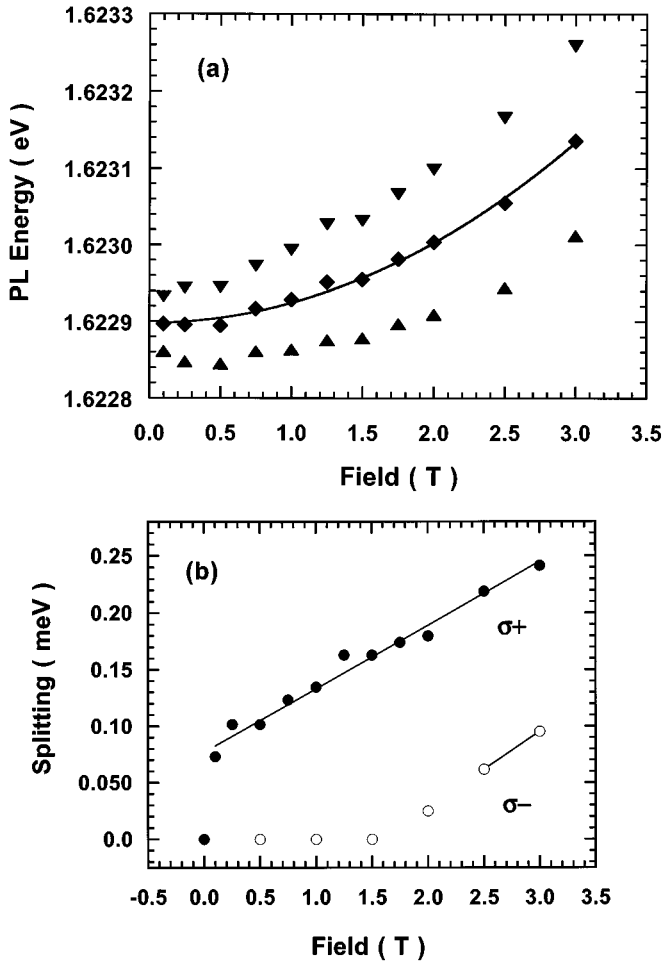

FIG. 3. (a) Magnetic field dependence of excitonic recombination from a single quantum dot for $\sigma+$ excitation. Solid triangles are higher-energy and lower-energy peaks, while closed diamonds are average recombination energies. The solide line is a quadratic fit to the average values, giving a diamagnetic shift of 26 μeV/T^2. (b) Magnetic field dependence of excitonic splittings for (●) $\sigma+$ excitation and (○) $\sigma-$ excitation. The solid line is a linear fit to the data, giving a field-dependent splitting of 64±5 μeV/T for $\sigma+$ excitation.

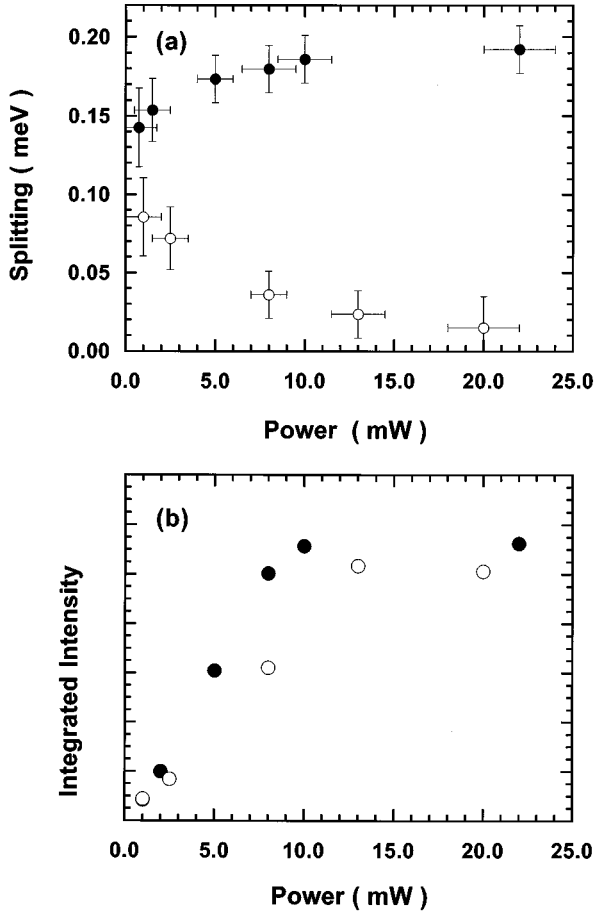

FIG. 4. (a) Dependence of observed excitonic splittings on excitation power for (●) $\sigma+$ and (○) $\sigma-$ polarizations. (b) Dependence of the integrated photoluminescence intensity from a single quantum dot on excitation power for (●) $\sigma+$ and (○) $\sigma-$ polarizations. The external magnetic field was 2.0 T.

proportional to the time-averaged probability of the existence of an exciton in the quantum dot. As the excitation power increases, the observed excitonic splitting will either increase or decrease, depending on the relative orientation of the internal effective nuclear field with respect to the external magnetic field. The strength of the hyperfine term will eventually saturate at higher powers when exciton absorption saturates. Indeed, the luminescence intensity from the dot shows evidence of saturation at the higher excitation powers, as shown in Fig. 4(b). A similar excitation power dependence of exciton luminescence from single quantum dots has recently been observed and adequately described by a simple rate equation analysis.[3] Note that the difference in the total excitonic splitting increased to ~180 μeV at the higher excitation powers, corresponding to changes in effective nuclear fields approaching 3.0 T.

There is a pronounced asymmetry in the observed splittings for fields less than 1.5 T [Fig. 3(b)] which may be related to the spin character of the exciton states. Spin-polarized electrons are necessary to dynamically polarize the nuclear system. However, in the absence of a magnetic field, the exciton states are split into two states composed of linear combinations of the two spin states.[7] The degree of nuclear polarization may therefore be limited by the character of the exciton states in low total effective fields, resulting in the observed asymmetry in the polarization dependence of the excitonic splittings.

Interestingly, the degree of nuclear orientation is apparently not directly related to the degree of polarization of the excitonic luminescence. The degree of polarization of luminescence, defined to be equal to $(\sigma+-\sigma-)/(\sigma++\sigma-)$, of the Zeeman-split pair shown in Fig. 2(a) with zero-field energy ~1.623 eV increases from ~5% to ~35% as the magnetic field increases from 0.5 to 3.0 T. In contrast, the degree of nuclear orientation, as measured by the Overhauser shift, exhibits little, if any, dependence on magnetic field in this range. Previous calculations, however, predict a strong dependence of the degree of nuclear polarization on the degree of electronic spin polarization.[8,9] The discrepancy between theoretical prediction and experimental observations may help identify details of the nuclear polarization process. For

instance, Barrett *et al.*[24] have recently postulated that the exchange-split, optically-forbidden dark exciton states are involved in the nuclear polarization process rather than the optically-allowed states.

In summary, spectrally-resolved Overhauser shifts in the luminescence of individual excitons localized by interface fluctuations in distinct $GaAs/Al_{0.3}Ga_{0.7}As$ quantum dots were reported. Based on differences in the observed excitonic splittings for nuclear magnetic moments oriented parallel and antiparallel to an external magnetic field, effective internal magnetic fields are estimated to be greater than 1.3 T in quantum dots localized in a 4.2-nm-wide quantum well. These results illustrate an important consequence of electron-nuclear interactions on the magneto-optical spectroscopy of single quantum dots.

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