Spin relaxation of excitons in zero-dimensional InGaAs quantum disks

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We report the observation of spin relaxation of excitons in zero-dimensional semiconductor nanostructures. The spin relaxation is measured in InGaAs quantum disks by using a polarization dependent time-resolved photoluminescence method. The spin relaxation time in a zero-dimensional quantum disk is as long as 0.9 ns at 4 K, which is almost twice as long as the radiative recombination lifetime and is considerably longer than that in quantum wells. The temperature dependence of the spin relaxation time suggests the importance of exciton–acoustic phonon interaction. © *1998 American Institute of Physics*. [S0003-6951(98)02111-1]

In recent years, a number of studies has been made on spin relaxation in semiconductors by using polarization dependent photoluminescence (PL) for circularly polarized excitation light,[1–6] time-resolved PL,[3,4] and pump–probe techniques.[6] Spin relaxation of carriers in bulk and quantum wells is believed to be caused by momentum-dependent spin flip interactions such as the k^3 term in the zinc-blende Hamiltonian[7] or the electron-hole exchange interaction[8] which can be viewed as an effective magnetic field. Elastic scattering of carriers, which is responsible for dephasing processes such as momentum relaxation, plays an important role in these spin relaxation mechanisms.[7,8] The spin relaxation of carriers is in general a fast process because there is a continuum of states available for spin relaxation. Contrary to this, the full quantization of the energy spectrum in the zero-dimensional (0D) structure makes all the elastic processes inefficient; these processes are usually invoked to explain the rapid spin relaxation in bulk materials[1,9,10] or in two-dimensional (2D) systems.[3–5,11] Because of the discrete density-of-states and the absence of energy versus momentum dispersion, no elastic process to flip the electron spin can occur because there is no state available between the levels. The complete lack of continuous band states allows only a fundamental process between the discrete levels. A study on the spin relaxation in the 0D nanostructure therefore gives insight into how the spin relaxation proceeds in the most simple energy level scheme, and will also give important information on the physics of the spin relaxation in semiconductors.

The quantum confinement achieved by adding a lateral confinement onto a 2D structure gives a disk-shaped structure. In this structure, there is a well-defined spin excitation condition involving pure spin exciton states. Also, the large heavy-light valence band splitting is a consequence of the strong confinement along the growth direction which is much tighter than the lateral confinement. The structure is grown via a unique spontaneous reorganization of an InGaAs/AlGaAs heterostructure grown on a GaAs (311) *B* substrate.[12–14] This spontaneous reorganization results in arrays of well-ordered InGaAs disk-shaped structures with diameters of 30–100 nm and thickness of 3–5 nm automatically embedded in AlGaAs barriers. Such quantum disk structures also have the unique feature that the effective lateral extent can be tuned between 2D and 0D simply by controlling the disk's lateral size. We measured the spin relaxation time of excitons in undoped InGaAs quantum disks by using a polarization dependent time-resolved PL method. We show that the spin relaxation time of excitons in 0D quantum disks is extremely long compared with that in the 2D quantum well and that the spin relaxation rate increases linearly with temperature. We discuss possible mechanisms for the spin relaxation of excitons in terms of the comb-shaped density-of-states in quantum disks and of scattering by thermal phonons.

The $In_xGa_{1-x}As$ quantum disk samples were grown by low-pressure metalorganic vapor phase epitaxy (MOCVD) on (311) *B* GaAs substrates. Figure 1(a) shows a schematic representation of the quantum disk structures. The quantum disks were spontaneously formed via a strain-driven self-organized process which occurs during interruptions in the growth of a sequence of AlGaAs and $In_xGa_{1-x}As$ layers.[12–14] The $In_xGa_{1-x}As$ quantum disks are completely buried in the AlGaAs barriers via dynamic mass transportation as depicted in Fig. 1(b), which shows a scanning electron microscope photograph of a stain-etched cleaved (110) facet. We used two quantum disk samples whose indium

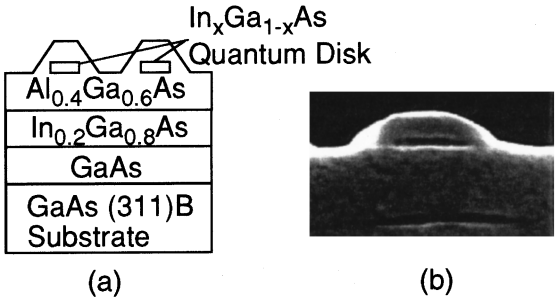

FIG. 1. (a) Schematic drawing of the sample structure of the quantum disk used in the measurement. (b) Cross section of the InGaAs quantum disk. This photograph was taken by using a scanning electron microscope (SEM).

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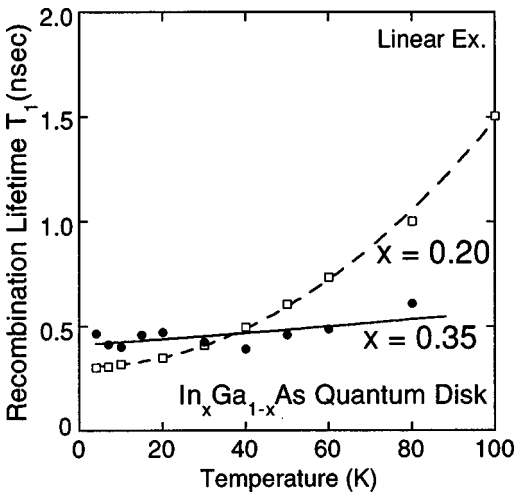

FIG. 2. Temperature dependence of recombination lifetime for large ($x=0.20$) and small ($x=0.35$) quantum disks. The diameters of the disks are 100 nm ($x=0.20$) and 30 nm ($x=0.35$). The solid and broken lines are guidelines for the eye. The exciting light is linearly polarized.

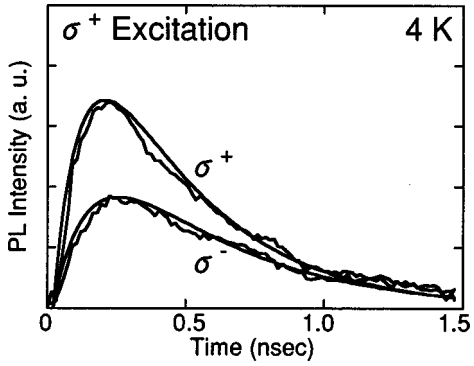

FIG. 3. Time evolution of σ^+ and σ^- PL signals at 4 K for 0D quantum disks (30 nm quantum disk). The excitation is σ^+ polarization. The energy difference between the excitation and the PL is about 100 meV. The excitation intensity is about 1×10^8 cm^{-2}. The solid curves are fitted curves of a combination of two exponential decay functions.

contents x are 0.20 and 0.35. The lateral sizes of the quantum disks were estimated to be about 100 and about 30 nm, respectively, for $x=0.20$ and for $x=0.35$. We have confirmed that the PL is due to radiative recombination of excitons confined in the quantum disks.[15,16] The temporal evolution of the exciton luminescence was measured using a cw mode-locked Ti–sapphire laser (repetition rate: 82 MHz, duration: 1.5 ps) as the excitation source and a synchroscan streak camera. The excitation light was circular polarized (σ^+) by means of a λ/4 plate, and the PL was analyzed into its σ^+ and σ^- components using a second λ/4 plate coupled to a polarizer placed in front of the entrance slit of the monochromator. The spin relaxation time can be evaluated from the σ^+ and σ^- components of PL. The excitation density was kept low (about 1×10^8 cm^{-2}) to avoid many-body effects. The excitation energy was set in the nonresonant condition; the energy difference between excitation and detection was about 100 meV in all measurements.

First, to check the effect of lateral confinement we measured time-resolved PL for a variety of temperatures for linearly polarized excitation which excites excitons of both spin orientations. Here, the decay time directly corresponds to the radiative recombination lifetime of the exciton, since at this low temperature nonradiative processes should be negligible. Figure 2 shows the temperature dependence of the recombination lifetime for two quantum disk samples. The recombination lifetime for the $x=0.20$ disk increases almost in proportion to temperature, because the excitons begin populating nonradiative (optically inhibited) exciton states. In contrast, the recombination lifetime of the $x=0.35$ disk stays almost constant against temperature. This is essentially a consequence of the 0D discrete density-of-states as was already confirmed in Refs. 15 and 16 and prevents excitons from thermalizing into higher nonradiative excited states.

The time evolution of σ^+ and σ^- PL signals at 4 K for the 0D disk ($x=0.35$) is shown in Fig. 3. Exciting light is σ^+ polarized. The solid curves are fitted by assuming a single exponential rise followed by a single exponential decay. A large difference in the signal intensity between σ^+ and $\sigma-$ signals is clearly seen. Such features were also observed in 2D quantum wells but only under resonant excitation.[3–5] The time dependent degree of circular polarization $P(t)$ is defined by $P(t)=[I^+(t)-I^-(t)]/[I^+(t)+I^-(t)]$, where $I(t)$ is the temporal intensity of circularly polarized PL. When t is much larger than the build-up time of $I^+(t)+I^-(t)$, $P(t)$ is proportional to $\exp(-2t/\tau_s)$, where τ_s is the spin relaxation time. Therefore, we can obtain the spin relaxation time from the decaying portion of $P(t)$. The spin relaxation time τ_s is evaluated to be 0.9 ns in Fig. 3. This relaxation time is substantially longer than those reported for 2D quantum wells[3–6] and it is even longer than the recombination lifetime, 0.4–0.5 ns. Although the excitation is in the nonresonant condition and spin relaxation may proceed simultaneously with intraband energy relaxation, most of the spin relaxation will occur after the energy relaxation into the exciton ground state. The intraband energy relaxation time (less than 100 ps) is 10 times smaller than the spin relaxation time and thus the former contribution is negligible. In distinct contrast, for the larger 2D disk of $x=0.20$, the difference between σ^+ and σ^- PL signals was negligible within the experimental resolution. This suggests a spin relaxation time as short as 50–100 ps which is the time resolution of our measurement system. A straightforward conclusion is therefore that the stronger lateral confinement results in a substantial decrease in the spin flip rate in the disk-shaped 0D structure. Due to the absence of energy versus momentum dispersion in all directions, no elastic process giving rise to spin flip transitions can occur because there is no state available between levels. Therefore the essential cause of such in inefficient spin relaxation rate is the 0D discrete density-of-states.

In order to gain further information on the origin of the spin relaxation, we examined temperature effects. The observed temperature dependence of the spin relaxation rate (inverse of spin relaxation time) for the same 0D quantum disk is shown in Fig. 4. The spin relaxation rate monotonically increases in proportion to temperature. For excitons in quantum wells, it has been reported that the spin relaxation time does not significantly vary with temperature below 50 K.[4] A clear temperature dependence in the present quantum disk then suggests that the dominant mechanism is different from that in quantum wells. It is known, on the other hand, that the scattering rate of excitons in 2D quantum wells due

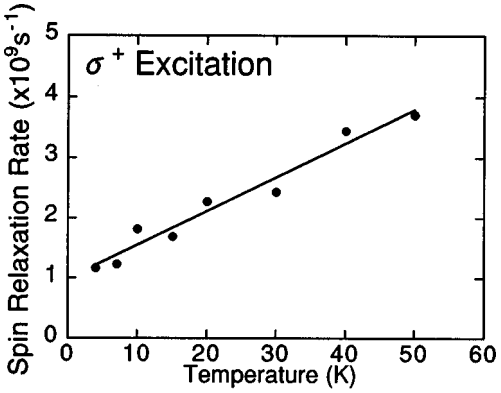

FIG. 4. Temperature dependence of spin relaxation rate for 0D quantum disks. The ordinate spin relaxation rate is the inverse of the spin relaxation time. The excitation is σ^+ polarization, the same as in Fig. 3.

to the exciton–acoustic phonon interaction increases in proportion to temperature, reflecting the Bose–Einstein statistics of the phonons.[17] The dependence proportional to temperature in Fig. 4 suggests that scattering of excitons by an acoustic phonon causes the spin relaxation in 0D quantum disks. Let us now recall mechanisms proposed to account for the spin relaxation in the bulk and in the quantum well. A direct electron-hole exchange [Bir–Aronov–Pikus (BAP)][18] yields a simultaneous flip of the electron and hole spins. Since this mechanism should have the same temperature dependence as that of the recombination lifetime, this contribution can be neglected. The D'yakonov–Perel' mechanism (DP)[7] and the Maialle–de Andrada–Sham mechanism (MAS)[8] rely on a mechanism analogous to the narrowing of the magnetic resonance linewidth in moving molecules, the so-called motional narrowing effect. In the DP, the k^3 term in the zinc-blende Hamiltonian gives an effective magnetic field, which leads to a spin splitting of the electronic states and the electron spin will precess between spin states. In the MAS, the electron-hole exchange interaction can be viewed as an effective magnetic field. In both cases, frequent random elastic scattering leads to a momentum relaxation and to the long electron spin relaxation time. In 0D quantum disks, elastic scattering events are infrequent. Moreover, it should be pointed out that spin flip mechanisms based on such motional narrowing rely on translational invariance, which is already broken in quantum dots or disks. In contrast, the Elliott–Yafet mechanism (EY)[19] is based on the mixing between two spin states; whatever the origin of the mixing is, such mixing itself can induce spin flip. Any process that leads to momentum relaxation will therefore make the spins flip, and indeed scattering by a acoustic phonon can flip the electron spin. Although the source of the mixing between two spin states is unclear at present, we tentatively attribute the phonon interaction as the source responsible for spin relaxation in quantum disks.

In summary, we have measured the spin relaxation time in InGaAs quantum disks by using a polarization dependent time-resolved PL method. The spin relaxation strongly depends on the lateral extent of the disk. It is long (0.9 ns) in small 0D quantum disks compared to that in 2D quantum wells, and is more than twice as large as the recombination lifetime. The complete lack of available states between discrete energy levels is the primary reason for inefficient spin relaxation. Suppression of any scattering process due to the 0D density-of-states makes even the spin relaxation of excitons inefficient. Its temperature dependence suggests that the scattering by thermal phonons is important. The spin relaxation occurs through transitions between two exciton states of the mixed spin components which is mediated by acoustic phonons (EY mechanism). This leads to the observed linear temperature dependence of the spin relaxation rate. We expect this work will stimulate further studies on spin relaxation processes in semiconductor nanostructures.

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