Polarized photoluminescence spectroscopy of single self-assembled InAs quantum dots

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(Received 17 June 1998; revised manuscript received 21 August 1998)

We have investigated the spin relaxation of carriers in single InAs/GaAs self-assembled quantum dots using near-field optical spectroscopy. Polarized photoluminescence spectroscopy of Zeeman split peaks in an external magnetic field allows us to investigate spin relaxation in the excited states. The excitation energy dependence of the polarization of each spin component shows the suppression of spin relaxation to be quenched at higher relaxation energies. This effect suggests a change in relaxation processes in the self-assembled dot systems. [S0163-1829(98)51740-7]

Atomiclike zero-dimensional (0D) systems, i.e., quantum dots, introduce new phenomena to semiconductor physics.[1] Among several fabrication techniques, self-assembled quantum dots (SADs) fabricated by the Stranski-Krastanow (SK) growth mode are promising 0D structures and are expected to possess real 0D character.[2,3] In 0D structures, an important subject of discussion has been a relaxation mechanism of excited carriers due to their discrete states with large energy differences. However, optical studies of SAD structures have been based on measurements of large ensembles of dots, so the results include the effect of inhomogeneous broadening which prevents accurate measurement of the optical properties of dots.

Investigation of single-dot properties has been accomplished by optical probing techniques with high spatial resolution. Because of the absence of the inhomogeneous broadening effect in a single dot, a narrower linewidth allows for precise measurements of luminescence character.[2–9] In addition, the application of magnetic field provides information about spin-related effects such as Zeeman splittings,[6–8] Overhauser shifts,[6] and spin-flip transitions in single dots.[8]

In this work, we consider relaxation processes in single InAs/GaAs SAD's by using polarized photoluminescence (PL) spectroscopy in an external magnetic field. The optical spectroscopic studies from a single dot have been performed with a near-field scanning optical microscope (NSOM).[9,10] As discussed in a previous report, luminescence features from single InAs SAD's showed Zeeman spin splittings as a function of applied magnetic field.[10] By monitoring the polarization of the pairs of luminescence lines for different excitation polarizations, it is possible to observe spin relaxation in the SAD structures, and thus to gain new information about carrier relaxation. The results clearly show a difference in the polarization properties of the split lines to be quenched at higher relaxation energies.

The InAs/GaAs SAD structures are grown by molecular beam epitaxy. The dots are capped with 50 nm of GaAs. Atomic force microscope images show the dots to have lateral size ~ 20 nm, height ~ 2 nm, and density $\sim 10^{10}$ cm^{-2}. A Ti:sapphire laser is used as a light source. The carriers are excited through an etched fiber probe with a submicrometer aperture, and the luminescence is collected by the same fiber (i.e., *illumination-collection mode* NSOM).[10] The experiments are performed in a He-gas flow cryostat at ~ 7 K. Luminescence is dispersed by a 250 mm single or a 1 m double monochromator and detected by a charge-coupled-device camera.

A typical magneto-optical luminescence spectrum from a single dot is shown in Fig. 1. In this measurement, the luminescence is collected in the Faraday configuration with fields $B=0-8$ T, and dispersed by the 1m double monochromator. The luminescence line shape at 0 T is fitted well as a Lorentzian function with a linewidth less than 100 μeV. The application of the magnetic field makes it possible to observe splittings due to the different spin states of the carriers. The splitting changes linearly as a function of applied magnetic field. The average value of the energies of the two split peaks shows a diamagnetic blueshift. The shift of the energy is proportional to B^2. Quantitative analysis of the energy shift shows the parabolic coefficient to be 3.7 μeV/T^2. This value

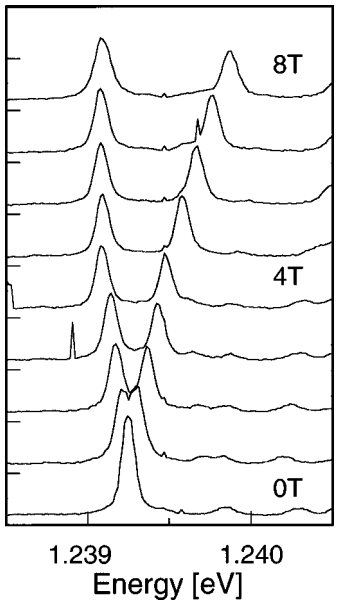

FIG. 1. Typical unpolarized photoluminescence spectra from a single dot at various magnetic field. The spectra are shifted vertically for clarity. The carriers are excited at 1.476 eV and collected luminescence is dispersed by a 1-m-long double monochromator. A splitting coefficient of 100 μeV T^{-1} and a diamagnetic shift of 3.6 μeV T^{-2} are observed in this dot.

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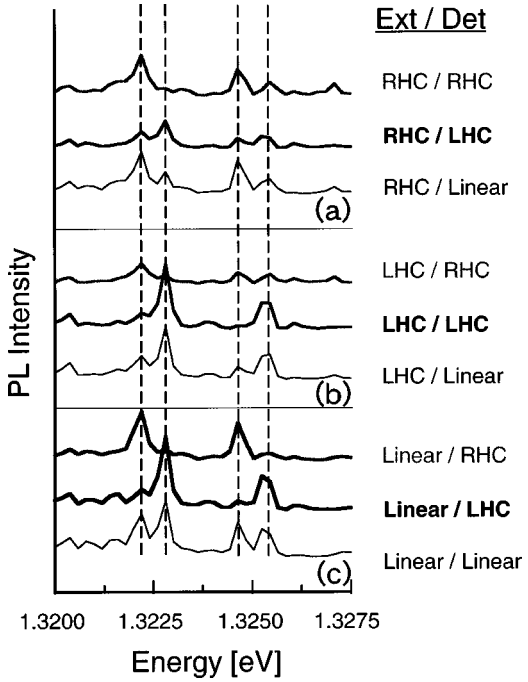

FIG. 2. Luminescence spectra for all polarization geometries at 8 T. Luminescence is dispersed by a single 250 mm monochromator. Two pairs of Zeeman split line pairs can be seen. The excitation energy is 1.409 eV. The spectra are shifted vertically for clarity.

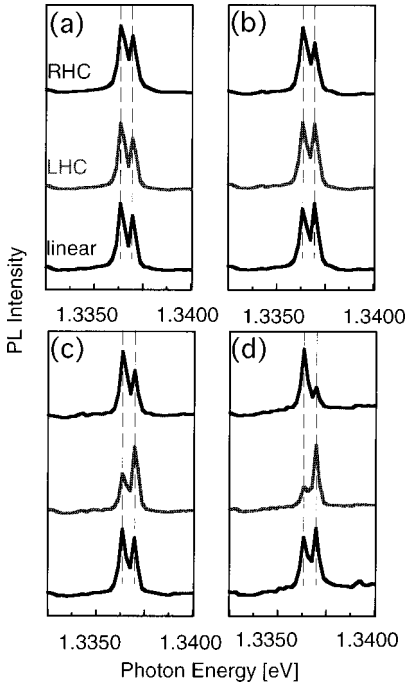

FIG. 3. Comparison of the split lines at 8 T in different excitation polarizations configuration (top: RHC, middle: LHC, bottom: linear) and various excitation energies E_{ex}= (a) 1.476, (b) 1.442, (c) 1.425, and (d) 1.409 eV. The collected luminescence is detected with linear polarization and dispersed by a single 250 mm monochromator.

is consistent with that observed in far-field magneto-PL spectra.[11,12] The observed diamagnetic shift indicates that strong confinement in the lateral direction is achieved in our SAD structures.

Considering the optical transitions between the conduction band ($|\pm 1/2\rangle$) and the heavy-hole valance bands ($|\pm 3/2\rangle$), for an optically excited electron-hole pair there are two allowed states: $|e,\mathrm{hh}\rangle = |+1/2, -3/2\rangle$ and $|-1/2, +3/2\rangle$. Recombination of $|+1/2, -3/2\rangle$ and $|-1/2, +3/2\rangle$ generates left-hand circular (LHC) light and right-hand circular (RHC) light, respectively. Therefore spectroscopy with polarization discrimination provides information about the spin states. Figure 2 show luminescence spectra for different polarization geometries at 8 T. The carriers are excited with circularly [Figs. 2(a), and 2(b)] or linearly polarized light [Fig. 2(c)] and the resulting luminescence polarizations are measured by turning a variable wave plate. The excitation energy is set to be 1.4089 eV and the detection energy is about 80 meV lower. When carriers are excited with linearly polarized light, the opposite polarities of the split lines are clearly shown—see Fig. 2(c). As discussed below, the excitation energy of 1.409 eV results in carriers being excited in dot states. When the excitation and detection polarizations are crossed, the intensities of the split peaks are small compared to when polarizations are the same. There still remain small peaks in crossed polarization due to imperfect polarization selectivity of the experimental setup or to spin relaxation within the recombination.

Figures 3(a)–3(d) show PL at 8 T with different excitation polarizations at various excitation energies. The splitting energy is estimated to be about 0.6 meV, where spin relaxation within a recombination is sufficiently suppressed. In Fig. 3(a), the split peaks show the same relative intensities in each excitation configuration. In contrast, when the laser energy is tuned to lower energies, for each circular excitation polarization one of the two peaks is suppressed, due to selective excitation. In Fig. 3(d), this effect is clearly seen. This indicates that at this high magnetic field, spin relaxation is suppressed because of the large exchange energy involved.[13] Because the dots reside on a wetting layer (WL) in SAD structures, the observed PLE spectra show a 2D-like continuum of states with a large nonresonant region. The absorption band edge of the WL is around 1.42 eV (see below). In Fig. 3(d), where the excitation energy is below 1.42 eV, the carriers are excited in the dot states. Therefore, suppressed spin relaxation is observed. On the other hand, in Fig. 3(a) the carriers are excited in the WL. The spin relaxation of carriers is mainly caused by dephasing processes such as momentum relaxation. In higher-dimensional structures (i.e., bulk, quantum well), spin relaxation is observed to be as fast as several tens of picoseconds because of the continuum of energy states.[14] The spin relaxation observed in the WL [Fig. 3(a)] may be explained by such fast spin-relaxation processes. Another explanation is that the scatterings of carriers may relax their spin polarities; carriers may be scattered from each other in the WL before being captured in each dot state.

In order to investigate the changes of spin relaxation more precisely, we measured PLE spectra in the different polarization configurations. Figure 4 shows the results at 8 T with LHC excitation. The x axis used is the difference between the excitation and detection energies, which will be referred to as relaxation energy. In spectra (a) and (b), the detection energy and polarization are set to those of the higher and lower energy peaks, respectively. The magnitude of the po-

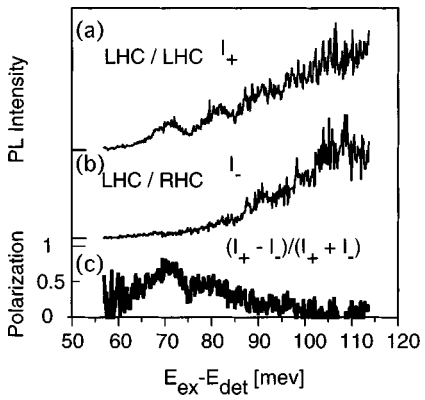

FIG. 4. (a) and (b) show corresponding photoluminescence excitation spectra which are recorded with the indicated excitation/detection polarizations. The luminescence intensity is plotted as a function of energy difference between the detection (E_{det}) and the laser (E_{ex}) energies. (c) Polarization calculated from the upper two spectra.

larization is given by $|I^+-I^-|/|I^++I^-|$, where I^+ (I^-) is the PL intensity for detection with the same (opposite) polarity. The calculated polarization is shown at the bottom in Fig. 4(c). It can be seen that when the excitation energy is decreased, the polarization is increased. The PLE peaks at 70 meV and 80 meV observed in the LHC/LHC configuration are not seen in the LHC/RHC configuration, where the polarization of excitation and detection is opposite. The WL absorption edge is at a relaxation energy of around 100 meV in these spectra. Figure 5 shows PLE spectra for five typical single dots in this sample. The arrows in the figure mark the excitation energy of 1.42 eV, at which we can see the absorption band edge of the WL. Below a relaxation energy of 60 meV, we observe a large off-resonance region due to the 0D character. In this energy region some of the dots show reproducible PLE features as marked by the gray circles in Fig. 5. Because these features are independent of luminescence energies, we suppose that they are due to phonons.[15] The PLE spectra for relaxation energies from 70 meV to the band edge exhibit unique features which depend on the dots. The PLE data of Fig. 4 correspond to these features. In Fig. 4, it is noticeable that the degree of polarization changes continuously from the energy of the WL absorption edge to that of the dot states. The PLE structures observed at the energy beyond 70 meV occur on a continuum background, which may be attributed to the dots not being perfectly 0D, and this may obscure the abrupt change of polarization. Another possibility is that there exists an energy region where the density of states changes gradually from discrete to continuum. As the upper discrete states of the quantum dots become close in energy to the continuum of 2D, strong interaction between these states will occur, thus increasing the probability of scattering.

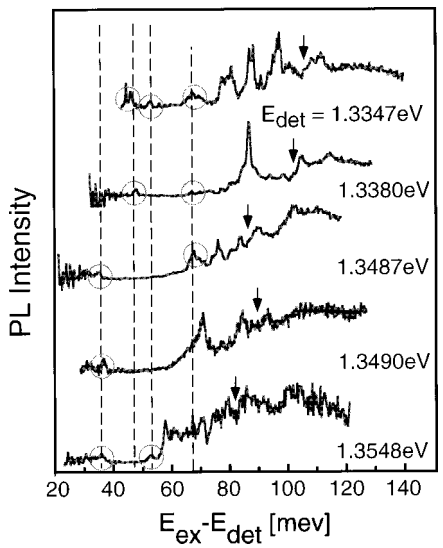

FIG. 5. Typical PLE spectra taken from five different single dots. The luminescence energies are displayed in the figure. The arrows indicate the corresponding WL absorption band edge around $E_{\mathrm{ex}}=1.42$ eV. The reproducible features are marked by gray circles. The luminescence intensity is plotted as a function of energy difference between the detection (E_{det}) and the laser (E_{ex}) energies.

In summary, single dot polarized PL measurements have been performed to study relaxation processes in InAs SAD structures. Using *illumination-collection mode* NSOM, we have investigated Zeeman spin splittings in single dots. By monitoring the polarity of fine Zeeman splittings in an external magnetic field, we observe changes of spin polarity to be suppressed during relaxation in the 0D states. The observed quenching effect allows for the study of the relaxation process in the SAD structures.

The authors thank S. Mononobe and M. Ohtsu, Kanagawa Academy of Science and Technology, for their kind arrangement of the fiber probe. They also express their thanks to Dr. J. Harris for useful discussions on relaxation processes in quantum dots. This work was supported in part by the Research for the Future Program of the Japan Society for the Promotion of Science (Project No. JSPS-RFTF96P00201), Grant-in-Aid of Priority Area by the Ministry of Education, Science and Culture, and University-Industry Joint Project on Quantum Nanostructures.

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