Decoherence of exciton complexes in single InAlAs quantum dots measured by Fourier spectroscopy

S. Adachi, a N. Yatsu, R. Kaji, and S. Muto b
Department of Applied Physics, Hokkaido University, Sapporo 060-8628, Japan

H. Sasakura
CREST, Japan Science and Technology Agency, Kawaguchi 332-0012, Japan

(Received 28 August 2007; accepted 28 September 2007; published online 17 October 2007)

We report the single-photon Fourier spectroscopy of exciton and exciton complexes in single self-assembled InAlAs/AlGaAs quantum dots. A Michelson interferometer was inserted in the photoluminescence path for measuring the coherence length of the time-averaged emission of neutral excitons and their complexes. The effects of exchange interactions and fluctuations in the surrounding excess charges were estimated by comparing the coherence times and the excitation energies of the excitons and their complexes. © 2007 American Institute of Physics. [DOI: 10.1063/1.2800373]

Single quantum dot (QD) devices attract considerable attention in the field of semiconductor nanostructures because of their remarkable potential for single-photon emission, single-electron storage, and manipulation of single qubits for quantum information processing, which rely on discrete energy level of the QD due to its three-dimensional (3D) quantum confinement. However, a semiconductor QD can interact with its environment through carrier-phonon, carrier-carrier, and electron-nuclei interactions. In particular, electron-hole exchange interaction and electron-nuclei interactions are supposed to be enhanced due to the nanoscale 3D confinement. The suppression or positive utilization of the above interactions to realize an ideal isolated artificial atom is still a major challenge in the field of the semiconductor nanotechnology. The coherence time which is deduced from the coherence length or linewidth is an important parameter for the characterization of the single-photon wavepacket and emission and, therefore, to provide single-photon sources for quantum information processing. Currently, single-photon sources are an active research area, and the production of single photons on demand has been demonstrated using a variety of QDs with and without cavities in the visible to near-infrared spectral range.

The samples contain layers of In0.75Al0.25As QDs embedded in an Al0.5Ga0.7As barrier layer grown on a (100) GaAs substrate by molecular beam epitaxy. The QDs are formed in the Stranski-Krastanow growth mode. For single-QD spectroscopy, small mesa structures with a typical top lateral size of ~150 nm are fabricated. A cw-Ti:sapphire laser beam traveling along the QD growth direction is focused on the sample surface by a microscope objective. A Michelson interferometer inserted in the photoluminescence path is used to measure the coherence length of the time-averaged emission; this is a type of time-domain spectroscopy called single-photon Fourier spectroscopy and was first demonstrated by Kammerer et al.9 The obtained data show the variation of the interference fringe contrast (visibility) with the path-length difference between the two arms of the interferometer and give the magnitude of the Fourier transform of the intensity spectra. Fourier spectroscopy is an interesting method to explore the spectral dynamics of a single transition by both high temporal and high spectral resolutions with very low photon losses.10-12 The PL passing through the interferometer was dispersed by a triple-grating spectrometer (f=0.64 m) and was detected by a liquid-nitrogen-cooled Si-charge-coupled device camera with a typical accumulation time of 500 ms.

Figure 1(a) shows the contour plot of the PL intensity as a function of linear polarization angle at 0 T; the PL intensity was obtained by rotating a half-wave plate before a fixed polarizer in front of the spectrometer. The sample was excited at the wetting layer (WL) of ~1.698 eV by the depolarized light. The figure shows almost all the spectral lines from a single InAlAs QD target for WL-excitation condition. Various measurements, including those of the quantities depicted in Fig. 1, indicate that the PL lines in the figure originate from the same single QD, and the PL lines are attributed to X0 (spin-triplet positive trion), XX0 (neutral biexciton), XX+ (positively charged biexciton), X0 (neutral exciton), and X+ (spin-singlet positive trion) from the low-energy side. Each line of the X0 and XX0 PLs has a clear fine structure and the dependence of linear polarization is anticorrelated; the high/(low)-energy line of X0 and the low/(high)-energy line of XX0 have the same x(y) polarization. While the XX0 state consists of spin-paired electrons and holes and is free from exchange interactions, the neutral exciton state splits due to anisotropic exchange interactions (AEI) resulting from the...
the polarization of the PL component. The solid lines are the fitting curves.

From the fitting in Fig. 1 that is narrower than that of similar clear anticorrelated patterns. Here, the interference fringe contrast served for the coherence of the electron spin.

The change on the excitation energy from the WL to the state. From the fitting in Fig. 1(b), the bright exciton splitting \( \delta_x \) is found to be \( 115 \pm 5 \, \text{meV} \) and the binding energy of \( XX^0 \) is \( +3.77 \, \text{meV} \). \( X^0 \) consists of two holes with the paired spin and one electron; therefore, exchange interaction is ineffective and \( X^0 \) has no fine structure. Hence, \( X^0 \) has a linewidth that is narrower than that of \( X^0 \), which suggests a long coherence of the electron spin. \( X^0 \) has a negative binding energy \( (-0.89 \, \text{meV}) \), which implies that a repulsive force acts between an exciton and a hole of \( X^0 \). The \( XX^0 \) and \( XX^0 \) PLs are weak as compared to the others in this QD but show similar clear anticorrelated patterns. Here, \( XX^0 \) PL implies the transition from \( XX^0(J_z = \pm 3/2) \) to \( XX^0(J_z = \pm 1/2, \pm 5/2) \). Moreover, \( XX^0 \) PL arises from the transition from \( XX^0(J_z = \pm 1/2, \pm 5/2) \) to the hole state \( h(J_z = \pm 3/2) \). \( XX^0 \) and \( XX^0 \) contain excited holes that occupy the \( p \)-shell and exchange interactions between the \( s \)-shell and the \( p \)-shell carriers cause the state mixing and splitting, leading to a fine structure.

Figure 1(c) shows a typical visibility plot of the single-QD PL at the WL excitation. By varying the temporal delay \( \tau \), the detected signal is given by \( I(\tau) = I_0(1 + 4 \sin^2 \phi(\tau)/h + \sin^2 \phi(\tau)/h) \), where \( I_0 \) is the average signal intensity, \( E_0 \) the central detection energy, \( V(\tau) \) the interference fringe contrast (visibility) given by the modulus of the Fourier transform of the intensity spectrum, and \( \phi(\tau) \) a slowly varying phase factor related to the line profile asymmetry. (An example of the rapid oscillation of the interference fringe is seen in the raw data in the upper panel of Fig. 3) In Fig. 1(c), \( XX^0(x+y) \) represented the \( x \)-polarized component and \( XX^0(x+y) \) represents both the \( x \)- and \( y \)-polarized components. Since the InAlAs QD target has a relatively large bright exciton splitting \( 2 \delta_x \), the visibility of the whole fine structure components of \( XX^0 \) and \( XX^0 \) oscillates with a period of \( h/2 \delta_x \sim 36 \, \text{ps} \) due to classical polarization interference. The visibility decay of \( XX^0(x+y) \) shows a clear Gaussian shape of \( V(\tau) \) with a coherence time of \( \sim 25 \, \text{ps} \), which indicates spectral fluctuations in the ground state energy. This is supposed to be due to the environmental charge because the effect reduces experimentally due to a near-resonant excitation such as the 1 LO resonant excitation discussed later (Fig. 2). Although the oscillatory and nonoscillatory behaviors of \( XX^0 \) are the same as those of \( X^0 \), depending on the detected polarization components, the coherence time of \( XX^0 \) is clearly different from that of \( X^0 \) and is longer \( \sim (40 \, \text{ps}) \), which is consistent with the higher-resolution PL spectra. In the case of \( X^0 \), the coherence time is long and is almost the same as that of \( XX^0 \). The long coherence times of \( X^0 \) and \( XX^0 \) as compared with \( X^0 \) are probably due to different exchange interactions between the electron and the hole in an exciton since \( XX^0 \) and \( X^0 \) contain the paired holes with antiparallel spins; therefore, the exchange effect is ineffective. Since the coherence time \( T_2 \) is generally represented as \( 1/T_2 = 1/(2\pi\Gamma + 1/T_\text{phonon}^2 + 1/T_\text{exchange}^2) \), under a particular excitation condition, it is reasonable to assume that the spectral fluctuation effect \( 1/T_\text{fluct}^2 \) and phonon scattering \( 1/T_\text{phonon}^2 \) affect all the species equally. Furthermore, the lifetime \( T_1 \) is estimated to be \( \sim 1 \, \text{ns} \) from the single-QD-PL measurements using the streak camera. Therefore, on this timescale, the difference of the coherence times between \( X^0 \) (or \( XX^0 \)) and \( X^0 \) corresponds to the electron-hole exchange effect \( 1/T_\text{exchange}^2 \), and \( T_2 \) is estimated to be \( \sim 65 \, \text{ps} \). For other QDs in the same and different mesas in the same sample, almost similar results are obtained and the value of \( T_2^\text{exchange} \) is in the range of 50–100 ps. As compared with quantum wells where \( T_2^\text{exchange} \sim 5 \, \text{ps} \), this smaller contribution of the exchange interaction in QDs indicates a low scattering rate due to the 3D confinement.

The change on the excitation energy from the WL to the interior of the QD allows us to observe the effect of the charge fluctuations around a QD. Figure 2(a) shows the PL excitation spectra of another InAlAs QD in the same sample at a detection energy set at the \( X^0 \) peak. Three excitation energies, denoted as 1, 2, and 3 in the figure, were selected; these energies correspond to the edge of WL, 1 LO (GaAs), and 1 LO (InAlAs) resonances, respectively. The respective visibility plots are shown in Fig. 2(b), where the case with a low-power excitation at the excitation energy of 1 is also plotted (open circles). In this QD, the coherence times are
19, 16, and 13 (18) ps at the excitation energies of 3, 2, and 1 (the case at one-fifth of the excitation power), respectively. As observed in the figure, the visibility decays with the Gaussian profile in all the cases, but the coherence time increases with a decrease in the excitation energy. In addition, the coherence time depends on the excitation power. Since the optically excited carrier density decreases according to the density of states with a decrease in the excitation energy, the coherence times of the positive trion and exciton complexes in single self-assembled InAlAs/AIGaAs quantum dots. The measurement of the coherence times of the positive trion and exciton complexes with an excited hole was demonstrated for the first time. A longer coherence time is exhibited by $XX^0$ and $X^+$ as compared with the others, and this strongly suggests that decoherence by the AEI of $s$ and $p$ shells is predominant in $X^0$, $XX^+$, and $X_T^+$; this predominance of decoherence is due to the fact that $XX^0$ and $X^+$ have spin-paired holes, which ensures that the exchange interaction is effective. All the presented data suggest that the order of decoherence in the single InAlAs QD can be understood by the exchange effect. From their excitation energy dependence and Gaussian shaped visibility curve, the effect of fluctuations in the environmental excess charges is found to be important for this type of measurements.

FIG. 3. (Color online) Visibility plot of the PLs of exciton and exciton complexes from the same single QD at WL excitation (3 K). The dashed lines are the fitting curves. The raw interferogram (upper panel) and the time-integrated spectra (right panel) are also shown.

We report the single-photon Fourier spectroscopy of excitons and exciton complexes in single self-assembled InAlAs/AIGaAs quantum dots. The measurement of the coherence times of the positive trion and exciton complexes with an excited hole was demonstrated for the first time. A longer coherence time is exhibited by $XX^0$ and $X^+$ as compared with the others, and this strongly suggests that decoherence by the AEI of $s$ and $p$ shells is predominant in $X^0$, $XX^+$, and $X_T^+$; this predominance of decoherence is due to the fact that $XX^0$ and $X^+$ have spin-paired holes, which ensures that the exchange interaction is effective. All the presented data suggest that the order of decoherence in the single InAlAs QD can be understood by the exchange effect. From their excitation energy dependence and Gaussian shaped visibility curve, the effect of fluctuations in the environmental excess charges is found to be important for this type of measurements.