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Coherent dynamics of excitons in a stack of self-assembled InAs quantum dots at 1.5-μm waveband

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Abstract

We investigate the excitonic dephasing in a stack of self-assembled quantum dots (SAQDs) by using a four-wave-mixing (FWM) technique performed in the optical telecomm-fiber wavelength region at 6 K. A sample used in our experiment is a 150-layer stack of InAs SAQDs embedded in InGaAlAs grown on InP(3 1 1)B substrate fabricated by molecular beam epitaxy. By using a novel strain-controlled technique, the resonant wavelength of the exciton ground state (GS) ranges from 1.25 to 1.5 μm which is much longer than that in typical In(Ga)As SAQDs. In the weak excitation region, the intrinsic dephasing time of excitons at the excitation wavelength of 1.43 μm reaches 770 ps which is much longer than that in most SAQDs with the resonant wavelength of <1 μm. We also find a strong anisotoropy of the signal intensity with respect to the crystal axis attributed to the orientation of InP(3 1 1)B substrate and the elongated shape of QDs.

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Keywords: Quantum dot; Dephasing; Four-wave mixing; Optical anisotropy

1. Introduction

Semiconductor quantum dots (QDs) have recently been a subject of great interest because of their many novel device applications to photonics and quantum information technologies [1,2]. In these applications, the dephasing of optical polarization plays a decisive role, because it limits the number of possible quantum operations and determines the homogeneous broadening of QD laser active media. Epitaxially grown self-assembled QDs (SAQDs) are particularly interesting, since they show very high quality of crystalline and optical properties, ultralong dephasing time of the exciton

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ground-state (GS) transition in the nanosecond range [3–5].

In spite of extensive studies about the excitonic dephasing in SAQDs and quantum islands in a quantum well, previous experiments have been performed at the wavelength below 1.2 μm. One reason is that large strain arising from lattice mismatches at the boundaries of QD, barrier medium and GaAs substrate increases the energy of an exciton. In addition, widely used Ti:sapphire lasers and Si photodetectors are available only below 1 μm. A weak signal from QDs makes it difficult to detect a dephasing process by using a coherent spectroscopy such as four-wave mixing (FWM).

Recently, new techniques to compensate the strain have been developed [6] that allow us to control an excitonic energy in SAQDs. A single photon emission [1] and QD lasing [2] have been demonstrated at optical telecommunication wavelengths in the past several years, which have made major impacts with a great potential of SAQDs. Nevertheless, there is no report on the excitonic dephasing in SAQDs in this wavelength region. In this paper, we report the first measurements of excitonic dephasing in a stack of InAs SAQDs by using the FWM technique performed at the wavelength of over 1.4 μm.

2. Samples and experiments

The investigated sample consists of 150 layers of vertically stacked InAs SAQDs separated by 20-nm-thick $In_{0.46}Ga_{0.11}Al_{0.43}As$ spacers on InP(3 1 1)B substrate. Molecular beam epitaxy was used for the fabrication. Its details are shown in Ref. [6]. The backside of our sample was antireflection coated to prevent multiple reflections. The composition of spacer layers was precisely controlled to compensate strains caused by a tensile stress. Fig. 1 shows photoluminescence spectra measured at 12 K and room temperature. The exciton GS energy at 12 K ranges from 0.827 (1.5 μm) to 0.992 eV (1.25 μm) which are much lower than that of ordinary In(Ga)As/GaAs SAQDs on GaAs(0 0 1) [3–5]. This fact supports that the strain in our sample is considerably smaller compared with previously reported samples [3–5]. Moreover, the reduction of strain enables us to stack over 100-layers of QDs as well as to improve their size uniformity. This allows us to measure the weak signal from QDs with high signal-to-noise ratio.

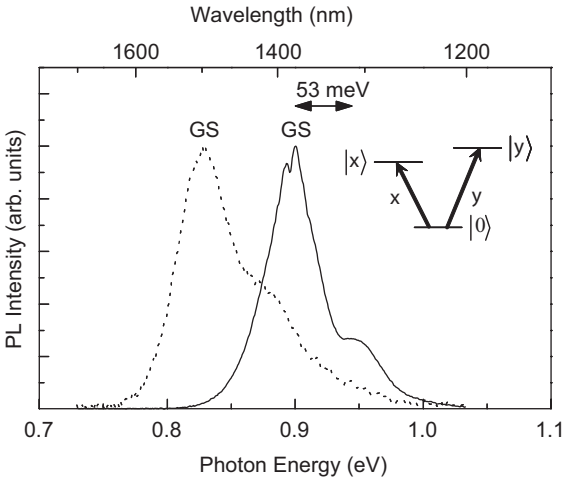

Fig. 1. Photoluminescence spectra at 12 K (solid line) and room temperature (dotted line) under nonresonant CW excitation. PL peaks at 0.900 and 0.953 eV correspond to a ground and a first-excited states of excitons, respectively. Inset: energy level diagram for an asymmetric QD.

The FWM measurements were performed using optical pulses of ~170 fs duration at 76 MHz repetition rate from an optical parametric oscillator pumped by a mode-locked Ti:sapphire laser. The time-integrated FWM signal in the direction of $2\boldsymbol{k}_1-\boldsymbol{k}_2$ was selectively detected using an InGaAs photodetector in the transmission geometry. The intensity of $\boldsymbol{k}_1$ pulse coincides with that of $\boldsymbol{k}_2$ pulse. It is well known that the measurement of time-integrated FWM signals as a function of time delay gives us direct information on the dephasing time of microscopic polarization. The central energy of excitation pulses was tuned to 0.868 eV which is 31 meV below the center of the inhomogeneously broadened GS energy of excitons in order to avoid the contribution of the first excited state of excitons (corresponding to the PL peak at 0.953 eV). All measurements were performed at 6 K.

3. Results and discussion

Fig. 2 illustrates time-integrated FWM intensity versus time delay τ for various excitation intensities. Both of the first- and second- excitation pulses were linearly polarized along the $[\bar{2}\,3\,3]$ direction. In all cases, the FWM signals are dominated by long-lived signals showing a single exponential decay. A beat signal with a period of 1.3 ps around $\tau \sim 0$ is observed and followed by pronounced nonexponential signals with faster decay time at $\tau < 50$ ps (see the inset of Fig. 2). In this paper, we focus on the slow component to determine the intrinsic dephasing time of our sample, though the beat signal and the fast component also give us important information on the initial dephasing process as discussed in previous papers [3–5]. We fit the slow component to a single exponential decay function to obtain the dephasing time and the FWM signal intensity.

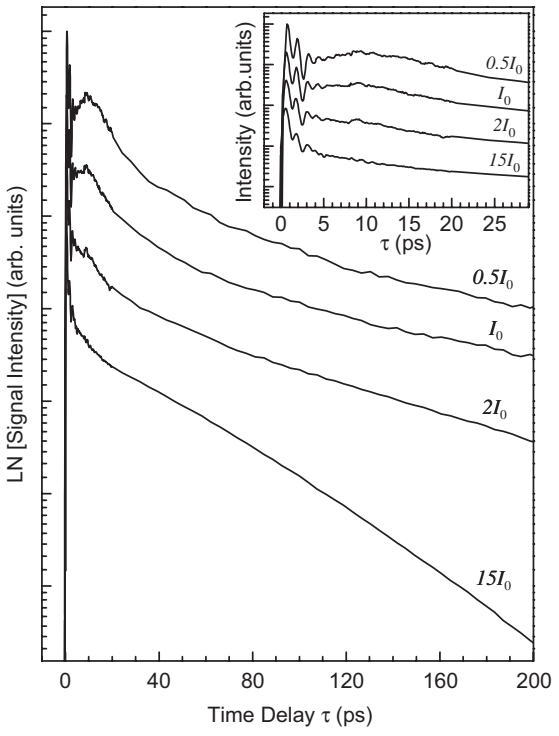

Fig. 2. Time-integrated FWM intensity versus time delay τ at 6 K for various excitation intensities. I_0 corresponds to approximately 3 MW/cm^2. Inset is a magnified view for $\tau < 30$ ps.

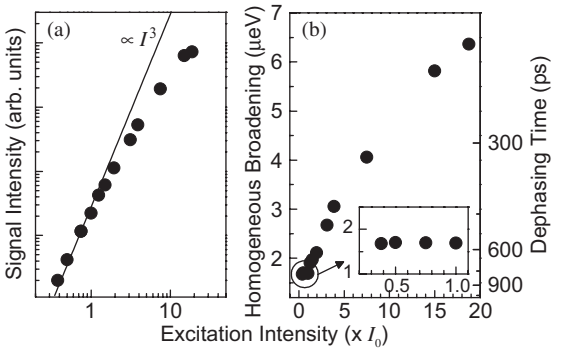

Fig. 3. Excitation intensity dependence of: (a) the FWM signal intensity and (b) the homogeneous broadening (dephasing time) estimated from the results of Fig. 2. Below I_0, signal intensity increases in proportion to a cube of excitation intensity I and the homogeneous broadening is almost independent of I. Above I_0, signal intensity saturates and the homogeneous broadening rapidly increases.

The fitting parameters are shown in Fig. 3. I_0 corresponds to the excitation energy of $\sim$3 MW/cm^2. The signal intensity increases perfectly proportional to the third order of the excitation intensity in the weak excitation region and saturates above I_0. In the excitation region below I_0, the so-called $\chi^{(3)}$ region, the dephasing time is almost independent of the excitation intensity (see the inset of Fig. 3(b)). This constant dephasing time indicates that the dephasing in this region is caused not by an exciton–exciton scattering but by an exciton–phonon interaction and population decay. This implies that only a single exciton is excited per dot in the $\chi^{(3)}$ region.

The average of the estimated dephasing time in the $\chi^{(3)}$ region is approximately $T_2 = 770$ ps, corresponding to the homogeneous broadening of 1.7 eV. Such ultralong dephasing time of over several hundreds picosecond at the wavelength $>1.4\,\mu$m have never been reported to our knowledge. Moreover, the obtained dephasing time is much longer than that of typical In(Ga)As SAQDs previously measured in the time and frequency domain [3,5]. Universally, the dephasing of an exciton is attributed to the population decay as well as the pure dephasing without changing the exciton occupation. In our sample, the population lifetime is estimated to be over 1 ns from the result of transient-grating measurements performed at the

same temperature: thus, the dephasing time can be over 2 ns if the pure dephasing is negligible. The measured dephasing time in our sample is much shorter than the upper limit of dephasing time. This result clearly indicates that a pure dephasing process considerably contributes to the dephasing in our sample at measured temperature. In the weak excitation region, such pure dephasing is dominated by elastic interactions between an exciton and acoustic phonons within the same dot. In order to clarify that point, more detailed experiments will be needed.

On the other hand, the homogeneous broadening rapidly increases with the excitation intensity above I_0. In the intermediate excitation intensity, the broadening shows linear dependence on the excitation intensity. Such a linear increase of the homogeneous broadening has been observed in a GaAs quantum well [7] and has been explained by the excitation induced dephasing arising from an exciton–exciton scattering. In QDs, the similar behavior has been theoretically predicted by Schneider et al. in 2004 [8]. The detailed discussion will be published elsewhere.

Let us next discuss the polarization dependence of the FWM signals with respect to the crystal axes. For a circularly shaped QD, the excitonic GSs consist of two degenerate, opposite circular polarized states which can be excited by right- and left-circularly polarized light. However, if the symmetry of the confinement potential is reduced, the mixing of these exciton states via the exchange interaction lifts the degeneracy, resulting in two linearly polarized states whose polarizations are aligned along the orthogonal in-plane axes of the dot structure (see the inset of Fig. 1). From atomic force microscope images of our QD layer, most of QDs are found to be elongated along the $[\bar{2}\,3\,3]$ direction of InP(3 1 1)B substrate, confirming a lower symmetry of our QDs. Typical lateral extent is estimated to be ~65 nm along the $[\bar{2}\,3\,3]$ direction and ~51 nm along the $[0\,1\,\bar{1}]$ direction, respectively. Fig. 4 shows FWM signals measured in various polarization configurations for the excitation intensity of I_0. When polarizations of $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ pulse align along the $[\bar{2}\,3\,3]$ and $[0\,1\,\bar{1}]$ directions, the slow component of the FWM signal completely vanishes as shown in Fig. 4(a). When the polarizations are rotated 45° keeping the relative angle of polarization to 90°, the slow components appears again. We thus conclude that for the $[\bar{2}\,3\,3]$ and the $[0\,1\,\bar{1}]$ polarization only one of the states of $|x\rangle$ and $|y\rangle$ is significantly excited. Fig. 4(b) indicates the FWM signals for the parallel polarization along the $[\bar{2}\,3\,3]$ and the $[0\,1\,\bar{1}]$ directions, respectively. The time profile is very similar to each other, while the signal intensity significantly depends on the crystal axis. Qualitatively, such an optical anisotropy is attributed to the orientation of InP(3 1 1)B substrate [9] as well as the asymmetric shape of QDs.

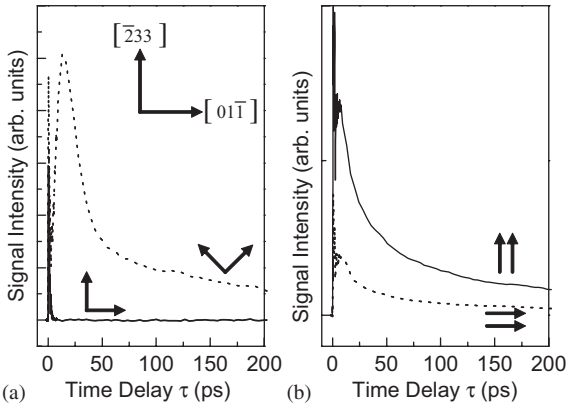

Fig. 4. FWM traces for different polarizations of excitation pulses. (a) Two excitation pulses are linearly crosspolarized. The polarization of $\boldsymbol{k}_1$ pulse is along the $[0\,1\,\bar{1}]$ direction (upper) and (lower) rotated 45° with respect to the $[0\,1\,\bar{1}]$ direction and (b) two excitation pulses are linearly copolarized. The polarization directions are aligned along the $[\bar{2}\,3\,3]$ (upper) and $[0\,1\,\bar{1}]$ (lower) directions.

4. Conclusion

We performed the FWM measurements at the excitation wavelength of over 1.4 μm and could observe an ultralong coherent signal from a 150-layer stack of strain-reduced InAs SAQDs at 6 K. Due to the larger confinement energy of our sample, the dephasing time of excitations in the weak excitons region reaches 770 ps which is much longer than that of most of SAQDs with the resonant wavelength to exciton GS of <1 μm. The polarization-sensitive measurements show a strong optical anisotoropy depending on the orientation of substrate.

The excitonic GS resonant energy and the dephasing characterictics of our SAQDs can be controlled by slightly changing fabrication parameters. Further systematic studies would open potential applications to various photonic devices using different samples.

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