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Transient grating studies of phase and spin relaxations of excitons in GaAs single quantum wells

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ABSTRACT

Exciton spin and phase relaxations at low temperatures in GaAs/AlGaAs single quantum wells were investigated by using transient grating technique. The technique allows us to obtain the exciton lifetime, spin relaxation, and phase relaxation in the same setup. In combination with a series of single quantum wells grown on the same substrate, the well width dependence of exciton spin relaxation was studied. The obtained spin relaxation results were analyzed with their phase relaxation times in a framework of MAS mechanism, and were in good agreement with the calculated results. Especially, the motional narrowing character of the exciton spin relaxation was well demonstrated.

1. Introduction

The spin relaxation processes of excitons and electrons in semiconductor nanostructures have been a central topic of intense experimental and theoretical investigations. Spin-based applications such as quantum information processing boost up the studies of spin coherence, especially, in quantum dots since quantitative information on spin relaxation or spin lifetime is essential to the design and successful implementation of devices.

As in bulk semiconductors, in the quantum well (QW), a conduction band electron and a valence band hole can bind into an exciton due to Coulomb attraction. However, the exciton states are strongly modified by the confinement of electrons and holes in one direction. Because of the increased wavefunction overlap and the ability of exciton to acquire appreciable momentum during its long lifetime, the long-range exchange interaction has larger importance for spin relaxation process in type I QW than in bulk materials. Based on the studies since around 1990, exciton spin relaxation mechanism effective for GaAs QWs got settled; for room temperature, the scattering during the spin precession in the effective magnetic field originating from the spin splitting in conduction band, and for low temperatures, the exchange interaction between electron and hole in an exciton. The former is the D'yakonov-Perel'(DP) mechanism [1] and the latter is the QW version (Maialle–Andrada–Sham (MAS) mechanism [2]) of the Bir–Aronov–Pikus (BAP) mechanism [1] for electron spin relaxation in bulk semiconductors, respectively. However, there exists only a few reports on the well width dependence of spin relaxation, which corresponds to change the wavefunction overlap and allows one to deduce the dominant spin relaxation mechanism. Furthermore, in most of their reports, phase relaxation has been assumed as a constant regardless of the well width [3–6].

In the present work, the exciton spin and phase relaxations at low temperatures in GaAs/AlGaAs single quantum wells (SQWs) were studied by using transient grating technique. This technique allows us to obtain the exciton lifetime, spin relaxation, and phase relaxation for resonant condition in the same setup. The character that spin coherence depends strongly on the scattering events gives rise to a sample-to-sample distribution of the experimental spin relaxation times. The extent of confinement also should affect significantly to the spin coherence through the change of scattering. Therefore, the used technique gives a complete set of the information about spin relaxation dynamics.

2. Sample and experimental setup

The sample used here is GaAs/$Al_{0.3}Ga_{0.7}As$ SQWs. A series of SQWs with different well width separated by 200-Å-thick AlGaAs layers were grown on a (0 0 1) GaAs substrate by molecular beam epitaxy. The sample was mounted in a cold finger cryostat and was kept at 10 K through the experiments. Important basic values

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Table 1
Important values of the used SQWs.

QW sample	A	B	C	D
L_W (Å)	120	72	48	36
E_{XH} (eV)	1.5368	1.5669	1.6069	1.6448
ΔE_{XL-XH} (meV)	6.8	15.4	23.7	29.4
E^{b}_{XH} (meV)	9.0	10.8	12.2	13.0
τ_m (ps)	7.9	1.8	3.6	6.2
τ_S (ps)	25.0	68.2	23.0	17.0
τ_R (ps)	730	230	380	270

L_W, E_{XH}, ΔE_{XL-XH}, and E^{b}_{XH} represent the well thickness, heavy hole exciton energy (10 K), splitting energy between heavy hole and light hole excitons (10 K), and binding energy of heavy hole exciton, respectively. Also, the obtained values of phase relaxation time τ_m, exciton spin relaxation time τ_S, and exciton lifetime τ_R at 10 K in the present experiments are shown in the lower part.

obtained by the steady state photoluminescence (PL) and PL excitation (PLE) measurements are listed in the upper part of Table 1.

In order to observe the transient processes, two-pulse and three-pulse four-wave-mixing (FWM) measurements in reflection geometry were performed by using a mode-locked Ti:sapphire laser with a repetition rate of 76 MHz. In the tuning range of 750–810 nm, the laser bandwidth was $\sim$8.5 meV (pulse width $\sim$150 fs). For spin relaxation and exciton lifetime measurements, two excitation pulses with orthogonally linear and parallel linear polarizations, respectively, are overlapped spatially and temporally on the sample. As a result, the respective excitation polarizations produce the exciton spin density grating (SG) and the exciton density grating (DG) in the sample [7,8]. The probe pulse with a controlled delay time is diffracted in backward direction by the created transient grating and can monitor the decay by the diffracted intensity. The decay rate of the respective transient grating $1/\tau_{DG(SG)}$ is given as $1/\tau_{DG(SG)} = 1/\tau_{R(S)} + 4\pi^2 D_{R(S)}/\Lambda^2$, which has two components: exciton recombination (spin relaxation) and exciton diffusion (exciton spin diffusion) in exciton DG (exciton SG). Here, $\tau_{R(S)}$ is exciton recombination time (exciton spin relaxation time), $D_{R(S)}$ is diffusion coefficient of exciton density (exciton spin density), and Λ is the grating fringe spacing. In this measurements, Λ is rather wide ($\sim$20 μm) due to the small crossing angle of the excitation pulse pair. Considering the diffusion coefficient in GaAs QW ($D_R \sim 16.5\,\mathrm{cm^2/s}$, $D_S \sim 3.5 D_R$) [8], contribution of diffusion effects to the grating decay is negligible in the observed time scale. The details of the spin grating technique are seen in Refs. [7–10]. For phase relaxation measurements, one of three pulses was blocked and only two of three pulses were used to generate and monitor the exciton polarization grating (PG). The self-diffracted FWM signals were spectrally resolved or spectrally integrated for the proper purpose. In all FWM measurements, the laser wavelength was tuned to the heavy hole exciton (XH) resonance.

3. Results and discussion

As an example of the decay of the respective transient grating, we show the time-integrated FWM signals in 120-Å and 48-Å SQWs at 10 K in Fig. 1. From the uppermost panel, exciton phase relaxation (two-pulse FWM with PG), exciton spin relaxation (three-pulse FWM with SG), and exciton recombination (three-pulse FWM with DG) are represented.

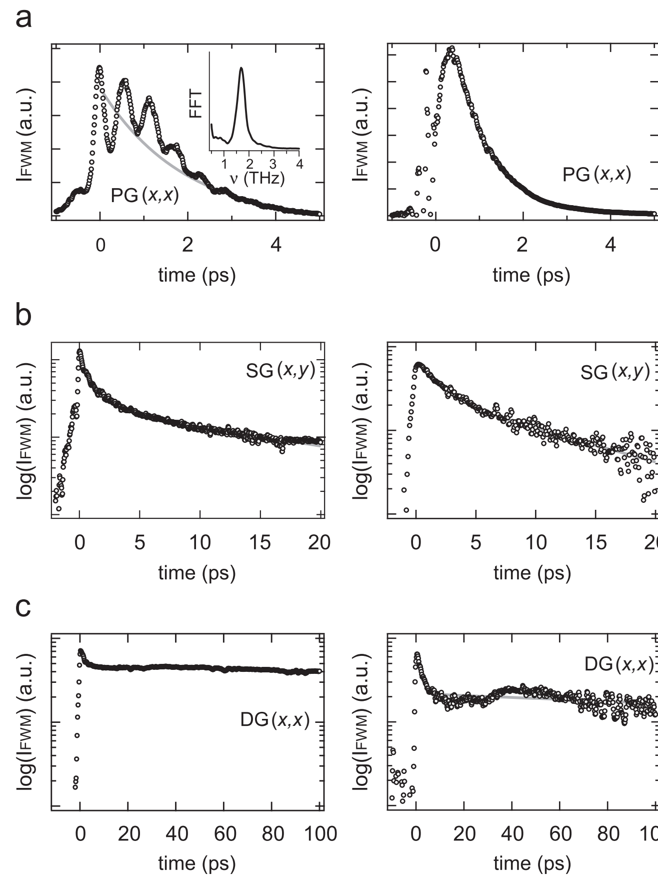

Fig. 1. Typical signals of two-pulse and three-pulse FWM measurements in 120-Å SQW (left column) and 48-Å SQW (right column) at 10 K. (a) Phase relaxation of excitons monitoring the decay of polarization grating (PG) by two-pulse FWM. Inset (left column): Fourier transformed spectrum of the signal. The peak due to the quantum beat is found to be $\sim$1.64 THz (6.8 meV). (b) Exciton spin relaxation monitoring the decay of spin density grating (SG) by three-pulse FWM. (c) Exciton lifetime monitoring the decay of exciton density grating (DG) by three-pulse FWM. In (a) of the right column, the spiky signal around the time origin is due to interference of two pulses and has no influence to obtain the phase relaxation time.

3.1. Phase relaxation

Phase relaxation of the exciton polarization occurs by the scattering mainly by phonon and other excitons in large well width, and corresponds to the momentum relaxation of the center of mass motion of excitons. It can be monitored by the decay of exciton polarization grating, which can be produced by the second pulse incoming within the time interval until the coherently created exciton polarization by the first pulse relaxes completely. Here, as an example of phase relaxation, let us consider it in a 120-Å SQW. Since 120-Å SQW has the comparable XL–XH splitting energy ΔE_{XL-XH} (see Table 1) to the laser bandwidth, the simultaneously excited XH and XL gives rise to the beating in the FWM signal. The beat period is $\sim$0.61 ps (1.64 THz) and coincides with the ΔE_{XL-XH}. This agreement and the spectrally resolved measurements (not shown here) indicate that the beat occurs quantum mechanically between XL and XH states although the biexcitonic effect is also shown in negative delay time and around the first peak. Duration of the beating and the fundamental decay are determined by the phase relaxation of XL and XH, respectively, because XL has shorter phase relaxation than XH.