6-3 Ultrafast electron control of optical device

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The many-body effect of the excitons in the semiconductor quantum dots is investigated in the frequency and time regions. The Rabi oscillation of the exciton in the semiconductor quantum dots is observed by the stimulated photon echo method. This is the first direct observation of the Rabi oscillation of the excitons. The signal intensity changes periodically with the excitation intensity. The sharp spectrum from the single quantum dot is also observed by the micro-photoluminescence. The existence of phonon bottleneck is clarified from the temporal shape of the photoluminescence signal. These results show that the quantum dots is one of the good candidates for the device of the quantum computation.

Keywords
Rabi oscillation, Quantum dots, Photon echo, Phonon bottleneck, Micro-photoluminescence

1 Introduction

As part of efforts to develop quantum information communication, a considerable number of theoretical proposals and basic experiments have been carried out in recent years. In connection with these efforts, we determined that to realize "q-bit" of quantum computation, it is first necessary to find a material with a long coherence time and in which superposition of wave functions and the phase state can be controlled. Specifically, our group paid particular attention to the excited states of semiconductor quantum dots and tried to control the wave function thereof with an ultrashort pulse in order to evaluate feasibility of q bit computation in a solid. For a semiconductor single quantum dot, an extremely sharp exciton luminescence spectrum was observed; it is known that the luminescent properties of this spectrum are similar to those of an atom or molecule. However, many uncertainties remain relating to optical response in the time region. In this report, we present the experimental results obtained by the stimulated photon echo method with respect to basic problems such as: (1) whether the optical properties in the time region are similar to those of atoms or molecules, (2) whether the semiconductor exciton has a long coherence time, and (3) whether the exciton can be controlled by an ultrashort laser pulse, among others; to the best of our knowledge, this represents the first such research of its kind.

2 Background

First, we will briefly discuss exciton in the semiconductors, its relaxation process, and its physical environment. In the case of optical excitation, an electron in the valence band in a compound semiconductor is excited to the conduction band; the electron and the hole generated in the valence band exert a force against each other via Coulomb interaction to create a stable energy state. At this time, the electron/hole pair creates a state similar to that of a hydrogen atom, with the hole taking the role of the proton. This state is called the exciton state. Because the exciton has an extremely large overlap integral of the elec-

tron and the hole, it acts as an excellent luminescence center in a semiconductor, and has historically been a target of many investigations relating to phenomena in the time region, nonlinear optical effects, and so on. There are generally two processes of relaxation of the exciton state. One is called "energy relaxation" or "population relaxation," where the exciton itself disappears and where the relaxation time normally lasts a few nanoseconds (or slightly longer). The other process is called "phase relaxation." In this case the phase of the exciton by excitation light is relaxed through collision with the phonon or memorized excitons. Commonly the phase relaxation time of this exciton has ranged from a few picoseconds to approximately 10 ps (in the longest case). If this short relaxation time were to be positively employed, various applications would develop, such as an ultrafast switch using photon echoes, for example (a brief explanation of the photon echoes will be provided below).

When discussing the phase relaxation of the exciton in a semiconductor, the dimensions of the system become crucial. In bulk crystals, the exciton-polariton state in which the exciton is strongly coupled with light is generated, and hence the phase relaxation time cannot be measured by conventional photon echo methods. Further, since in the semiconductor quantum well (a one-dimensional confinement system) there are potential barriers in the perpendicular direction to the layer and the movement of the exciton is limited, the energy level is quantized in this direction and it becomes possible to measure phase relaxation by the photon echo[1][2]. Up until now, phase relaxation time has been measured in samples of this semiconductor quantum well structure, or in layered compound semiconductors, each of which has a very similar structure to the above samples. However, the majority of the measured samples have a rapid phase relaxation time—as fast as about a few picoseconds. The main causes of this rapid phase relaxation are attributed to relaxation through exciton-phonon collision[2][3], exciton-exciton collision,[1][4], and so on. Since in a one-dimensional confinement system there remains a degree of freedom in a layer direction, interaction with the phonon is comparatively strong. On the contrary, in a three-dimensional confinement system (such as the quantum dot system discussed later) this interaction with the phonon is restricted, and it is thus predicted that the phase relaxation time will be longer. Incidentally, one example of what one may find in a two-dimensional confinement system is what is called a quantum wire. However, in the collection of samples, research on phase relaxation of the exciton in a semiconductor has mainly been carried out in respect of one-dimensional confinement systems.

Unlike a one-dimensional confinement system, in cases where the electron is confined in the semiconductor quantum dot (a three-dimensional confinement system), the energy of the electron becomes completely discrete. Therefore, the interaction between the electron and the phonon differs largely from that of a bulk case where the electrons can move freely in a crystal, and it is predicted that the collision probability between the electron and the phonon decreases drastically (resulting in a phonon bottleneck effect)[5][6]. It is considered that as a result of the significant decrease in the collision probability, the non-radiative process of the excited state becomes negligibly small and the life time of the excited state is determined by the radiative process[7][8]. This means that the excited state is maintained for a long time in the semiconductor quantum dot, and it is thus considered that the population inversion necessary for laser radiation can easily be realized. Therefore, the semiconductor quantum dot is regarded as a highly promising material for a low-threshold, high-efficiency laser. Further, the phase information of the polarization formed coherently in the semiconductor quantum dot by laser light is naturally maintained for a long period, as the process of collision with the phonon has been frozen[9][10]. A number of researchers find that the possibility of controlling the phase of

the exciton wave functions in this manner renders the quantum dot an unprecedentedly suitable material on which to perform quantum computation.

3 Luminescent property of a single quantum dot

Here we will discuss how the line width of the electronic level (especially of the exciton level) in the GaAs quantum dot—a semiconductor quantum dot—is sufficiently narrow, creating a phonon bottleneck.

First we investigated the luminescence (micro-photoluminescence; μPL) from a micro region (size: not more than 1 μm) comprised of a number of GaAs/$Al_{0.4}Ga_{0.6}As$ quantum dots (dot size: about 15 nm, density: about $10^{19}cm^{-2}$) formed by liquid-drop epitaxy method. Fig.1 shows the luminescence spectrum for this sample. The horizontal axis denotes the energy of excitation light, and the vertical axis shows the intensity of the emitted light at that energy (or wavelength of light). The left-hand graph in Fig.1 shows a normal PL spectrum of collected luminescence from a wide spatial region. The spectra on the high-energy side (beyond 1.9 eV) represent luminescence from the AlGaAs barrier layer; the broad luminescence band with its center in the vicinity of 1.7 eV represents the luminescence from the excitons in the quantum dots. It is assumed that, since it has been observed that the fabricated quantum dots are not of uniform size and all luminescence from excitons whose confinement energies vary only a little from dot to dot, the spectral width of the luminescence from the quantum dots becomes considerably wider than the luminescence line width of a single quantum dot (inhomogeneous broadening).

One method for observing the luminescence from a single quantum dot in a system where the luminescence's spectral width is broadened by superposition of luminescence from a significant number of quantum dots (a system of inhomogeneous broadening) is to narrow the area to be observed. In doing so, the quantum dots in the region decrease in number and the superposition of luminescence is dissolved, and accordingly the luminescence of an individual quantum dot becomes observable. Since the density of quantum dots in the sample used in this measurement is about 10^9 cm^{-2} and a region with an area of 1 μm^2 contains only about 10 quantum dots, the luminescence from an area on this order of size may be observed virtually in terms of a single quantum dot. In this study, to observe the luminescence from the micro region only, a microscope was used to collect the luminescence from a region having a size of 1 μm. The results are shown in the right-hand figure. In the case of an luminescence from an area having a size of 1 μm, the spectrum shows a set of sharp peaks, each of which reflects the luminescence from an individual quantum dot. This data, showing a set of sharp lumines-

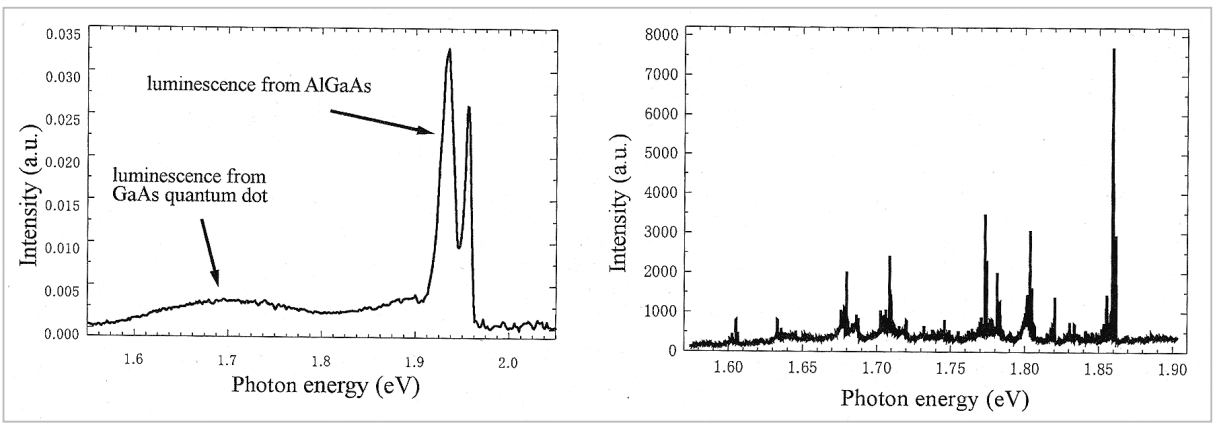

Fig.1 *Luminescence spectrum (right) and micro-photo luminescence spectrum (left) of GaAs/$Al_{0.4}Ga_{0.6}As$ quantum dot*

cence lines on the high-energy side that are much higher than the energy of the exciton (1.515 eV) in a GaAs bulk crystal, suggest that GaAs quantum dots are actually generated in this sample. The spectral width of an individual luminescence line was below the resolution (0.2 meV) of the measurement system; however, it turned out that the width of the exciton level in the GaAs quantum dot was sufficiently narrow, as expected.

When increasing the intensity of the laser light used for excitation, the luminescence spectrum from the quantum dot changes drastically. One example is shown in Fig.2. With weak excitation, a luminescence line was observed in the vicinity of 1.8112 eV, and as the excitation intensity increases, a new peak emerges at the lower energy of 0.8 meV, which develops into a main peak surpassing the luminescence line at the high energy level. When the excitation intensity is further increased, a new group of luminescence lines emerges at an approximately 25 meV higher energy level. This behavior can be interpreted to mean that the electronic structure of the quantum dot assumes a shell structure and that there are many-body interactions among the excitons generated in a single dot[11]. In the example of Fig.2, an exciton is present consisting of the electron and the hole, both residing in the lowest-energy S shell, and the luminescence emitted when the electron and the hole recombine with each other presents a luminescence peak in the vicinity of 1.8112 eV. Under conditions where the strength of the excitation light is sufficiently weak, on average, one or fewer electron/hole pairs is injected into the quantum dot, which permits only the 1.8112 eV luminescence line to emerge. When increasing the excitation light gradually, a state where two carriers exist in a single dot is allowed. The lowest energy state in this situation is that in which two excitons occupy the S shell, in which a stabilized bi-exciton energy state is created due to the many-body effect. Since light luminescence from this state is regarded as an optical transition from the stabilized bi-exciton state to the one-exciton state, the luminescence line shifts from one exciton luminescence line to a low energy level, as seen by the change in the amount of the stabilization energy. The luminescence peak at the lower energy of 0.8 meV reflects such an optical transition from the bi-exciton state. When further increasing the excitation intensity to effect an increase in the number of carriers in the quantum dot, a new luminescence line emerges at an approximately 25 meV higher energy level. From this phenomenon, it is apparent that the P shell of the holes, the next shell, resides at an energy level approximately 25 meV higher than that of the S shell. Based upon an experiment where the excitation intensity was increased even further, it was determined that the P shell of the electrons is located at an energy level approximately 80 meV higher than that of the S shell.

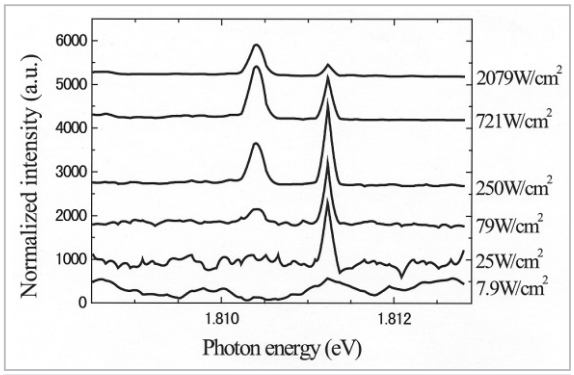

Fig.2 *Excitation dependence of microphoto luminescence spectrum of $GaAs/Al_{0.4}Ga_{0.6}As$ quantum dots*

That the GaAs quantum dot system will display a phonon bottleneck can be predicted from the fact that the luminescence spectral line of the single quantum dot is extremely narrow, and it can be directly observed by measuring the temporal evolution of the luminescence. The temporal evolution of the luminescence from the GaAs/AlGaAs quantum dot (with a dot diameter of 15 nm and a density of about 10^{10} cm^{-2}) is shown in Fig.3. The upper figure in Fig.3 shows the relationship between the energy of the pulsed laser used for excitation and energies used to measure the temporal evolution of the luminescence. Specifical-

ly, the excitation energy was set at about 1.75 eV, and the temporal evolution of luminescence was measured for energies lower than the energy by the following amounts: (a) the energy difference between the S shell and the P shell of the hole; (b) the energy of the LO phonon; and (c) the energy difference between the S shell and the P shell of the electron. The lower figure in Fig.3 shows the temporal evolution of luminescence for the respective energy positions (a), (b), and (c) specified above. In the case of (b), corresponding to the resonant Raman process, the signal rises in resolution of time (on the order of approximately 10 ps) and thereafter decays, whereas in the cases of (a) and (c) (each of which corresponds to a transition process from the P shell to the S shell), the signal rise time is slow (a few tens of picoseconds). This indicates that when the P shell exciton is excited, it cannot be relaxed to the S shell exciton, as no phonon is present with the appropriate energy to relax the P shell energy to the S shell energy; other, more complicated processes that take more time are required to relax the energy. Therefore, these results provide first-hand evidence that a phonon bottleneck exists within this system.

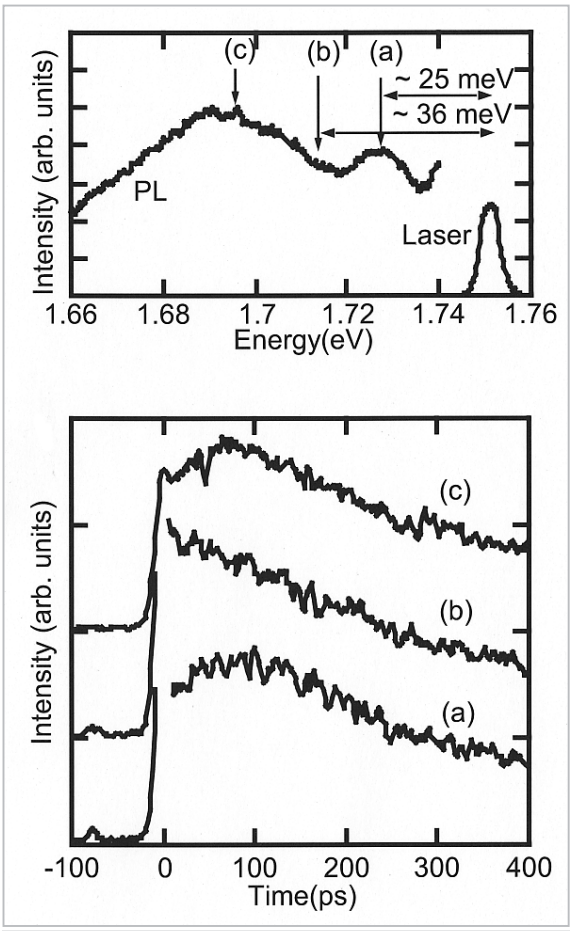

Fig.3 *Luminescence spectrum (upper) and time development of luminescence (lower) of GaAs/AlGaAs quantum dots*

4 Coherence time and optical response of exciton in quantum dot

It was confirmed that in the GaAs quantum dot system, the spectral width of the exciton line was very narrow due to the phonon-bottleneck effect; this suggests that the phase coherent time of the exciton polarization should be long. To confirm this implication directly in the time region, it is most effective to use the stimulated photon echo method[12], employing coherent spectroscopy with a pulsed laser. This method is performed as follows: (1) the pulsed laser light is divided into three beams, as shown in Fig.4, and the three beams are made to focus on a sample from the three directions k_1, k_2, and k_3; (2) the exciton polarizations made by the first pulse and by the second pulse interfere with each other to create a spatial interference fringe on the substance, and then the spatial interference fringe diffracts the third pulse; and (3) at this time, the sample is subjected to the first pulse and the second pulse, with the time difference between the first pulse and the second pulse being τ, and by measuring the diffracted light, it can be determined whether or not the

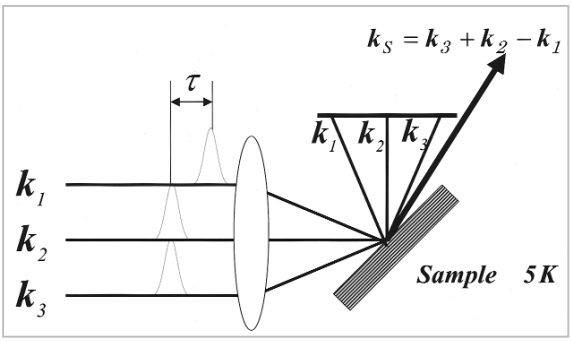

Fig.4 *Conceptual diagram of stimulated photon echo method*

exciton polarizations will interfere with each other even after the time τ has elapsed. Since the interference fringe is formed when the phase of the exciton polarization has not been destroyed, the phase relaxation information of the exciton polarization can be obtained through this method.

Fig.5 shows the results of the stimulated photon echoes method applied to the exciton in the quantum dot. Here an island structure formed by the interface fluctuation of a GaAs(11 mm)/AlGaAs quantum well was used as the GaAs quantum dot. The fact that the island structure of this sample functioned as the quantum dot was confirmed through the observation of a discrete spectrum in the measurement of μPL. Although a fast decay component of signal was observed in the vicinity of the zero point of time, after that the slow decay component followed, with a relaxation time of about 1 ns. Therefore, it can be concluded that the phase relaxation time of this system is about 1 ns. Since the phase relaxation time of the exciton in a normal quantum well is of the order of 1 ps, it was found that in comparison, the phase relaxation time of the exciton in the quantum dot is extremely long. From this attenuation time, the spectral width of the exciton level in the single quantum dot was estimated to be about 1 μeV, thus turning out to be very narrow. Regarding the rapid attenuation in the vicinity of the zero point, several mechanisms can be considered, but none has been definitively ascertained at the present stage. Similar results were obtained for the exciton captured by the island structure in a GaAs(9 nm)/AlGaAs quantum well.

In the measurement of the photon echoes shown in Fig.5, it was determined that if the time difference τ between the first pulse and the second pulse was set appropriately (where a slow delay in the photon echo was observed and the pulse intensities of the first, second, and third pulses were varied), the signal strength of the photon echo varied greatly. The results are shown in Fig.6. In the figure, the echo intensity alternately increases and decreases, in a vibrating manner, as the excitation intensity increases. This behavior can be understood in terms of the phenomenon known as the Rabi oscillation, where the exciton oscillates between two levels through interaction with laser light[13]. The results of Fig.6 are thus considered to represent Rabi oscillation of the exciton; i.e., Rabi oscillation between the ground state and the exciton state within the crystal. Normally, it is difficult to clearly observe the Rabi oscillation in a solid, because the phase relaxation time is extremely short and because under strong excitation the electron transition starts from a point k other than a point Γ in the k space[14]. However, in the quantum dot under observation, where the phase relaxation time of the exciton was as long as approximately 1 ns, the Rabi oscillation was successfully observed at a comparatively weak excitation intensity. Because the

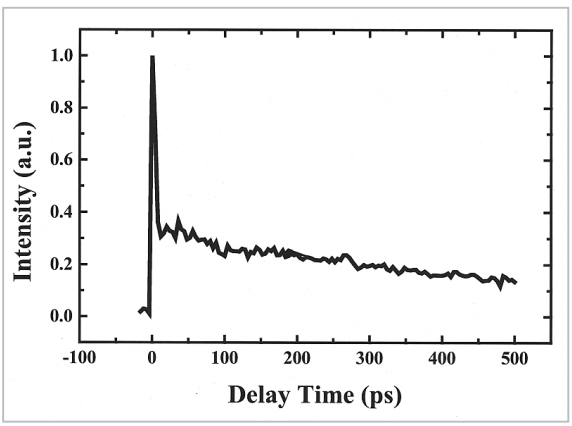

Fig.5 *Signal profile of the stimulated photon echo of GaAs/Al GaAs quantum dots*

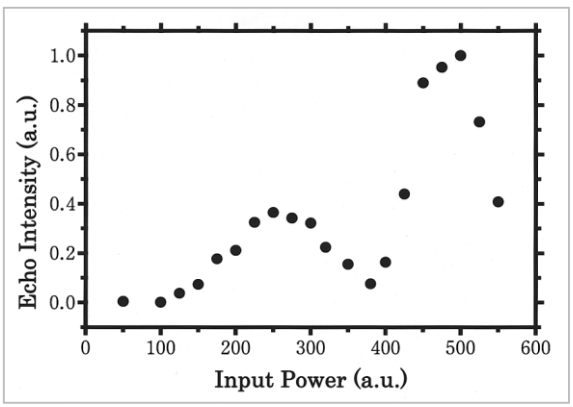

Fig.6 *Excitation dependence of photon echo intensity of GaAs/$Al_{0.4}Ga_{0.6}As$ quantum dot*

Rabi oscillation is an indispensable phenomenon in performing the phase shift of a q-bit (a basic operation of quantum computation), the data shown in Figs.5 and 6 indicate that the quantum dot is promising as a material constituting logical circuits for quantum computation.

5 Summary

We have demonstrated that in the GaAs quantum dot the line width of the electronic level (especially of the exciton level) is sufficiently narrow, the phase of the exciton polarization generated by laser light is maintained for 1 ns or more, and the Rabi oscillation of the exciton can be observed. We have shown that observation of these results may be attributed to the fact that in the GaAs quantum dot, the collision process between the exciton and the phonon is frozen by the phonon bottleneck effect and therefore the phase of the exciton is maintained for a relatively long time. From the results of this study, it has also been verified that the GaAs quantum dot is promising not only as a material for lasers having a low threshold and high efficiency but also as a material on which quantum computation may be performed.

References

1 L.Schultheis, J.Kuhl, A.Honold, and C.W. Tu, Phys. Rev. Lett. 57 (1986) 1797.
2 F. Minami, A. Hasegawa, S. Asaka, and K. Inoue, J. Lumin, 45 (1990) 409.
3 T. Takagawara, Phys. Rev. B32 (1985) 7013.
4 A.Honold, L. Schultheis, J. Kuhl, and C.W. Tu, Phys. Rev. B40 (1989) 6442.
5 U.Bocklmann and G.Bastard, Phys. Rev. B42 (1990) 8947.
6 H.Benisty, C.M.Sotomayor-Torres and C.Weisbuch, Phys. Rev.B44 (1991) 10945.
7 D.Gammon, Science 273 (1996) 87.
8 Xudong Fan, T.Takagawara, J.E.Cunningham and Hailin Wang, Solid State Commun.108 (1998) 857.
9 E.Biolatti, R.C.Iotti, P.Zanardi and F.Rossi, Phys. Rev. Lett. 85 (2000) 5647.
10 P.Chen, C.Piermarocchi and L.J.Sham, Phys. Rev. Lett. 87 (2001) 67401.
11 M.Bayer, O.Stern, P.Hawrylak, S.Fafard and A.Forchel, Nature 405 (200) 923.
12 J.Shah, Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures (Springer, Berlin, 1996).
13 L.Allen and J.H.Eberly, Optical Resonance and Two-Level Atoms (Dover, New York, 1975).
14 I. Abram, Phys. Rev.B40 (1989) 5460.

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