Coherent Nonlinear Optical Properties in Quantum Dots

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Abstract

This paper discusses optical nonlinear functions in semiconductor quantum dots (QDs) and their underlying physics and possible applications. The optical functions involve controlling light with no energy dissipation. The applications include conventional devices as well as future devices based on quantum phenomena, so they will be very useful in various technological areas. Theoretical results explain the physical principle of the nonlinear functions and the concept of the coherent slowdown of light. Experiments with actual QDs show that their optical responses agree well with the theoretical results. Light absorption was successfully controlled in a QD. These results clearly reveal that QDs provide intriguing optical functions and possible innovative applications.

1. Introduction

Semiconductor quantum dots (QDs) have been attracting considerable attention because they may have valuable applications in both conventional and future devices [1]. A QD is a semiconductor nanostructure, as shown in **Fig. 1(a)**. Here, an electron is confined in the InGaAs quantum dot because InGaAs has a smaller band gap than AlGaAs. QDs can confine electrons inside their structures and behave like atoms, but unlike atoms, their properties can be artificially designed. They can greatly improve the performance of many optical and electronic devices [1] and have been predicted to be good candidates for the fundamental elements in quantum information processes [2]. They could potentially lead to many useful technological innovations in many application areas.

An exciton, which is composite particle consisting of an electron bound to a hole, can be created in a QD by laser light irradiation. The processes for creating a single exciton (X) and a biexciton, which is a pair of excitons (XX), in a QD are shown in **Fig. 1(b)**. X and XX exist stably in the QD and contribute to the quantum information process as fundamental quantum bits (qubits). These fundamental bits can be represented as in **Fig. 1(c)**, where the ground state g is $|0\rangle = |00\rangle$, X is $|1\rangle = |01\rangle$, and XX is $|11\rangle$. X and XX in a QD provide a one-qubit rotation gate and a two-qubit correlation gate. These two gates are necessary for quantum computing, which is expected to lead to a paradigm shift in computing and information processing [3]. QDs will contribute to the development of next-generation computers.

The qubits in QDs have another characteristic. The creation processes of X and XX can be coherent processes; that is, they can be reversible with no energy dissipation. This allows us to control the creation and erasure processes using appropriate lights. These control processes correspond to the gating processes with the qubits in computing. QDs are known to preserve coherence in the creation and erasure processes longer than other semiconductors [4]-[6]. This allows us to make many gating processes, which are indispensable when performing quantum algorithms.

The system composed of X and XX (Fig. 1(c)) has three independent states that interact via lights. Since X and XX are created and erased coherently, this three-level system can be called a coherent three-level system. Several intriguing optical functions can be expected in a coherent three-level system such as quantum interference effects in photoabsorption, inversionless lasing, and lossless light speed control [7]. These phenomena are known as examples of

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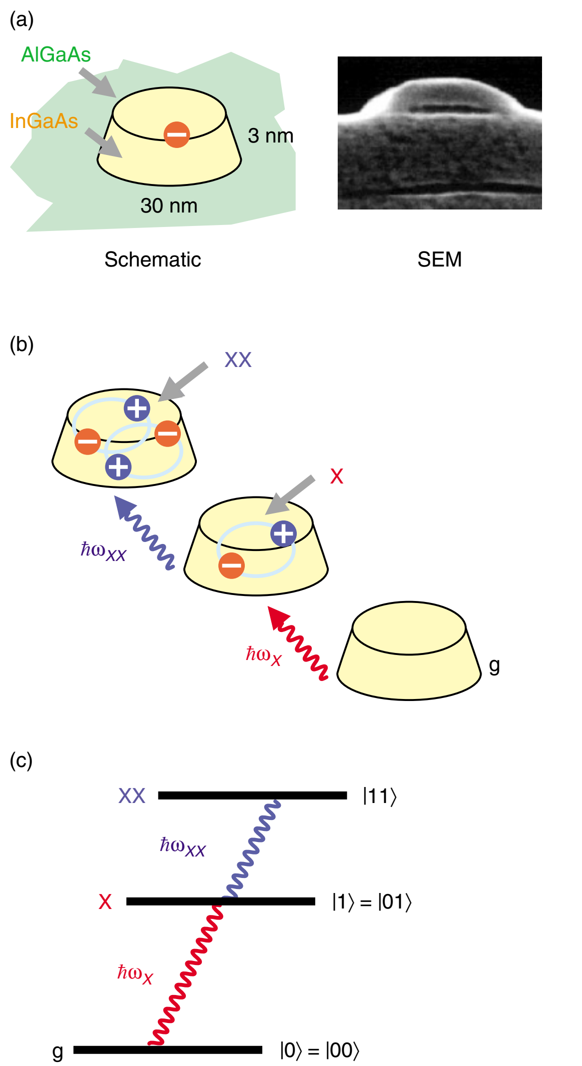


Fig. 1. Characteristics of quantum dots. (a) Schematic drawing and scanning electron micrograph of a quantum dot. (b) Creation processes of an exciton (X) and a biexciton (XX) in a quantum dot. (c) Energy level structure of (b).

electromagnetically induced transparency (EIT) in atoms and ions. All these functions are optical nonlinear processes that require two kinds of independent lights such as $\hbar\omega_X$ and $\hbar\omega_{XX}$ in Fig. 1(c). In particular, the function that allows us to control the speed of light is very useful in optical switching or routing as a temporary optical buffer memory and in quantum computing for tuning many gating pulses. The optical nonlinear properties in a coherent three-level system in a QD yield valuable applications in conventional and future devices.

EIT processes were proposed in a theoretical analysis of atoms interacting with lights [7]. This concept has been experimentally demonstrated in several actual ions and atoms [8]. The demonstration utilized several states of atoms; lights corresponding to $\hbar\omega_X$ and $\hbar\omega_{XX}$ in Fig. 1(c) were chosen so that their energies were resonant with the atomic states. Since the resonant energies were determined by atoms and ions, the optical functions were not controllable in terms of light wavelengths and corresponding energies. In contrast, QD state energies change according to the semiconductors used to make them and the QD size. These properties allow us to achieve the optical functions with controllable light wavelengths, which makes them very useful for actual applications.

In this paper, we present the fundamental physics of and corresponding experimental results for the optical nonlinear properties of QDs. The coherent control of light is highlighted in several optical processes. Possible controllability is discussed based on the results of a theoretical analysis. We present the results of an experimental demonstration of the nonlinear properties of actual QDs and discuss the present experimental status and future prospects. These experimental results are an important step towards coherent control of light.

Section 2 describes optical responses such as the absorption and refractive index spectra and presents the results of a theoretical calculation for a coherent three-level system. Characteristic shapes appear in the spectra as a result of coherent quantum interference effects. Processes for slowing light are induced by using the large derivative value in the refractive index spectrum. Section 3 describes experiments on actual QDs. It also describes a method for evaluating the nonlinear coherent process. Experimentally measured absorption spectra were similar to theoretically predicted ones. Preliminary results for controlling the absorption properties of a light (probe light) by using another light (pump light) are also reported.

2. Coherent control of light by using optical nonlinear effects

Let us consider a coherent three-level system with two lights called the pump and probe in **Fig. 2(a)**. The intensity of the probe light is assumed to be much weaker than that of the pump beam. The pump beam energy is fixed so that it is resonant with the energy difference between states 2 and 3. The optical responses of this system can be analyzed using a semi-classical theory with the density matrix method [9]. The density matrix method can include coherent effects between three levels and yields important

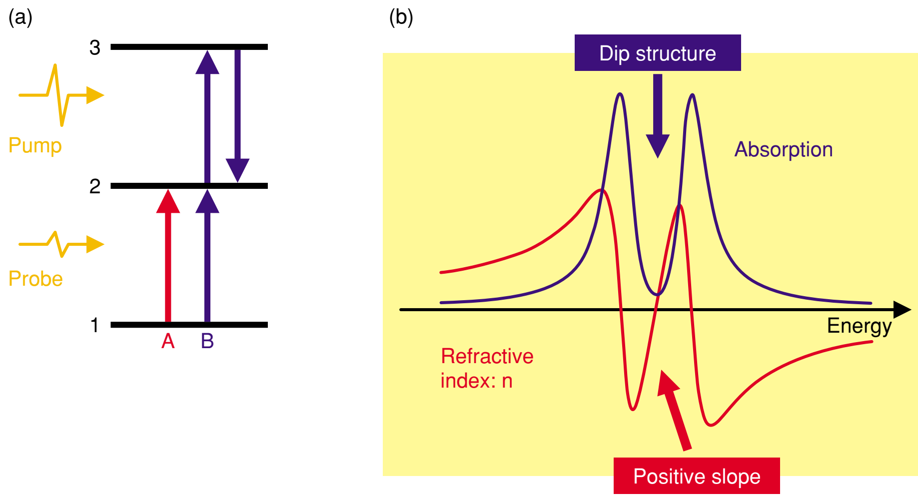


Fig. 2. (a) Coherent three-level system used in a theoretical analysis.
(b) Calculated optical responses for the three-level system.

optical properties such as the absorption and refractive index spectra. These optical properties play a crucial role in determining the performance of various optical devices. The rate equation method is often used to analyze the characteristics of laser diodes. Although it can simulate the main features of these devices, it assumes that the coherent effects are negligible, so it is unsuitable for analyzing the system shown in Fig. 2(a).

Examples of calculated absorption and refractive index spectra for the three level-system are shown in **Fig. 2(b)** as a function of probe light energy. These spectra have several characteristic features. A dip-shaped structure can be seen in the absorption spectrum. This results from coherent interference effects. In Fig. 2(a), there are two possible ways for the probe beam to be absorbed. One involves a simple optical absorption process from state 1 directly to state 2 (process A). The other involves optical absorption from state 1 to state 2 via state 3 mediated by the pump beam (process B). The latter process occurs when the pump beam intensity is strong, as we assumed for our calculation. Since the optical absorption of process B interferes destructively with that of process A, the absorption disappears when the probe beam energy is resonant with the energy difference between states 1 and 2. On the other hand, when the probe beam energy is not in a resonant condition, the destructive interference effect decreases and strong photoabsorption can be seen. As a result, a dip-shaped structure is seen in the absorption spectrum. Since a region that is transparent to the probe light is created by another light (pump light), which is an electromagnetic wave, the phenomenon is called electromagnetically induced transparency (EIT).

The dip-structure in the absorption spectrum produces a region in the refractive index spectrum that has a steep positive slope, as seen in Fig. 2(b). In the theoretical analysis, the density matrix method yields these two spectra (absorption and refractive index). Alternatively, a spectrum can be calculated from the other spectra using the Kramers-Kronig relation. According to this relation, the dip structure in the absorption spectrum leads to a positive-slope region in the refractive index, which yields a function for slowing light. When the probe light is a pulsed light, two kinds of velocity can be determined: the phase velocity and the group velocity. The group velocity is the effective speed of the pulse envelope and it determines the transmission time of information carried by several optical pulses. The group velocity v_g may be expressed as

$$v_g = \frac{c}{n(\hbar\omega) + \hbar\omega \dfrac{\partial n(\hbar\omega)}{\partial \hbar\omega}}, \tag{1}$$

where c is the velocity of light in a vacuum and $n(\hbar\omega)$ is the refractive index of the medium. The large positive slope such as that seen in Fig. 2(b) results in the large denominator in Eq. (1). This in turn results in a small v_g value and a reduction in the speed of the probe light induced by quantum interference effects. This slowdown effect depends strongly on the slope of the refractive index and the shape of the absorption spectrum. If the pump light can control the shapes of these spectra, then the slowdown function is controllable.

The slowdown function can be characterized by the denominator in Eq. (1). We define this denominator as slowdown factor S; namely,

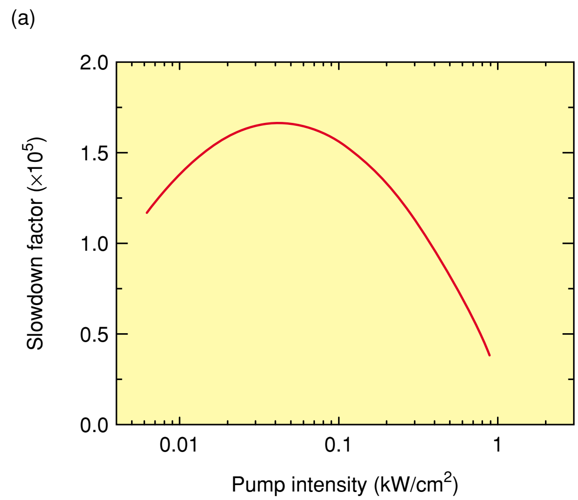
$$S = n(\hbar\omega) + \hbar\omega \frac{\partial n(\hbar\omega)}{\partial \hbar\omega}. \quad (2)$$

The slowdown factor S is shown as a function of pump intensity in **Fig. 3(a)**. This calculated result corresponds to a three-level system in actual QDs. In this calculation, we used reported physical parameters [10]. The slowdown factor changes with pump intensity, meaning that QDs can provide a controllable slowdown function. The maximum slowdown factor is about 1.7×10^5, which enables us to delay optical pulses by about 567 ns using a device 1 mm long. This delay time may be increased by modifying the physical parameters in QDs. A schematic diagram of an actual delay device is shown in **Fig. 3(c)**.

Some slowdown factors that correspond to familiar speeds are shown in **Fig. 3(b)** to provide a more intuitive understanding of the slowdown effects. The velocity of light in a vacuum corresponds to S = 1, i.e., no slowdown. The walking speed of a human, which is about 1 m/s, corresponds to $S = 3 \times 10^8$. The present slowdown factor in QDs of 1.7×10^5 falls between the speed of the space shuttle and the speed of a bullet shot from a gun. If the speed of light can be reduced to human walking pace, then this function will be very useful for optical routing, switching, and storage. Moreover, the slowdown function does not involve any energy dissipation. The ability to slow light will enable us to develop valuable and innovative optical devices that will find many applications.

3. Fundamental experiments in actual QDs and discussion

We examined the optical nonlinearity induced by an exciton and a biexciton in a single QD [11], [12]. This requires a well-defined exciton-biexciton system and large dipole moments, which characterize the coupling strength between exciton and lights. Our QD sample met both these requirements. A demonstration of the slowing of light requires many QDs where the sum of their cross-sections is larger than the cross-section of the probe beam. However, the fundamental properties of the slowing processes are determined by optical nonlinearities in a single QD, so results of experiments on a single QD will provide important reference data for slow light demonstrations with many QDs. We observed strong optical nonlinearity in the exciton absorption of an InGaAs QD by using single-dot photoluminescence excitation (PLE) spectroscopy. We measured the absorption spectrum of an exciton interacting with a biexciton in an isolated QD. In a QD with a weak confinement



(b)

S	
1	Velocity of light in vacuum
4×10^4	Velocity of space shuttle
1.7×10^5	Velocity of light in QDs
6×10^5	Speed of bullet from a gun
3×10^8	Walking speed of human

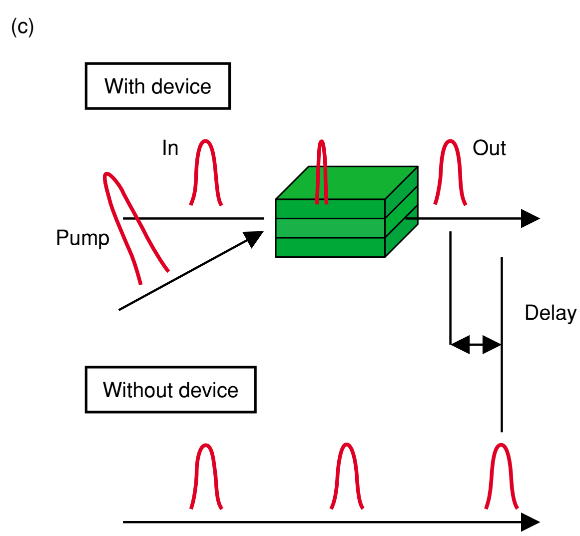


Fig. 3. (a) Calculated slowdown factor as a function of pump intensity. (b) Slowdown factors for various familiar speeds. (c) Schematic of an optical delay device.

effect, the exciton absorption spectrum had a dip-shaped structure at the exciton resonant energy. A large biexciton population induced large coherent effects between an exciton and a biexciton, which resulted in the observed absorption spectrum. A pump-probe measurement ensured that the exciton state almost disappeared as a result of the quantum interference effects in the exciton and biexciton states. These results constitute an important milestone for demonstrations of the slowing down of light.

$In_{0.4}Ga_{0.6}As$ QDs were grown by metalorganic vapor phase epitaxy (MOVPE) on a GaAs (311) B substrate [13]. The QDs were self-assembled on an $Al_{0.4}Ga_{0.6}As$ barrier layer and covered with another $Al_{0.4}Ga_{0.6}As$ layer followed by a thin GaAs capping layer. The QDs had an average lateral width of 30 nm and were 3 nm thick. This fabrication method is a kind of self-assembly. Although such methods have been reported by several groups [4]-[6], the use of a GaAs (311) B substrate is the key originality in our technique. This yields a well-defined exciton-biexciton system with large dipole moments, which are required to achieve large optical nonlinearities. The photoluminescence (PL) and PLE of the single QDs were measured with the micro-PL technique [14]. Our experimental setup is shown in **Fig. 4(a)**. The QDs were illuminated by a continuous wave Ti:sapphire laser through a microscope objective lens. The optical spot was about 2 µm in diameter. A metal mask with many holes ($0.5 \times 0.5\ \mu m^2$) was placed on top of the QD sample to detect the PL signal from a single QD. The pump-probe measurement technique with an unbalanced Mach-Zehnder interferometer was used to evaluate the optical nonlinearity of the QDs.

Measured PL and PLE spectra for a single QD are shown in **Fig. 4(b)**. The PL spectrum contains sharp clearly separated PL peaks for the lowest states of an exciton and a biexciton. The energy separation between the peaks is the biexciton binding energy. The binding energy is about 2.6 meV, which means a relatively large QD with weak confinement effects. This indicates that the dipole moments of the exciton and biexciton are much larger than those of the quantum wells and bulk.

The PLE spectra (Fig. 4(b)) for an exciton (X) and biexciton (XX) correspond to the absorption spectra. Several PLE peaks reveal the discrete density-of-states achieved in higher states. We focused on level A in the PLE spectra. Large signals can be seen in both the X and XX spectra. When the laser energy

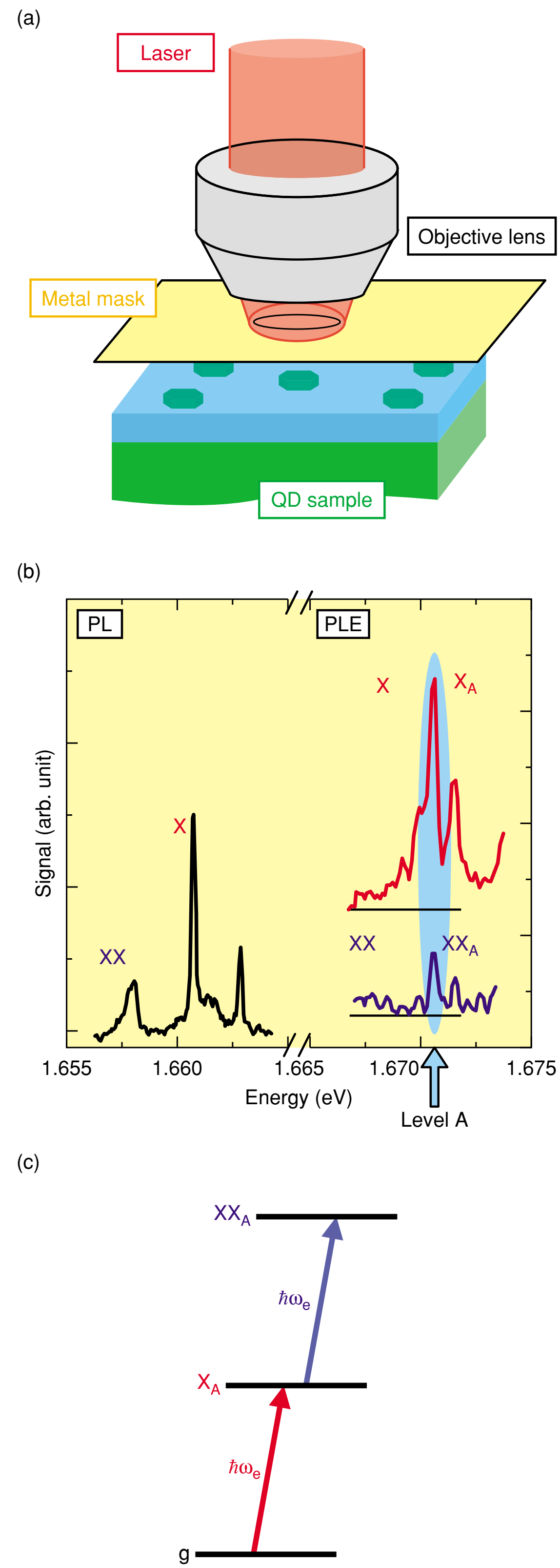


Fig. 4. (a) Schematic of measurement setup. (b) PL and PLE spectra for an exciton and a biexciton in a QD. (c) Energy level structure for X_A and XX_A.

was set to level A, an exciton and a biexciton were simultaneously created by a single excitation laser. The corresponding energy level structure is shown in **Fig. 4(c)**. It consists of three levels (g, X_A, and XX_A) that interact coherently. This energy structure is the same as that in the previous section. Therefore, we evaluated the coherent effects in X_A and XX_A.

PLE spectra of X and XX with a fine energy resolution are shown for different excitation intensities in **Fig. 5**. For the lowest excitation intensity (P_0), both X and XX have peak-shaped spectra with peaks at the same energy values. However, the spectral shapes are very different for higher excitation. For a high excitation intensity (5 P_0 and 20 P_0), the XX PLE spectrum is peak-shaped with a broader linewidth than that at P_0. In contrast, the X PLE spectrum has a completely different shape with a dip structure. The width of the dip-shaped absorption increases with excitation intensity and the absorption of X almost disappears at the exciton resonance energy of 20 P_0. This absorption spectrum is very similar to that obtained from the theoretical calculation (shown in Fig. 2(b)). The dip-shaped absorption spectrum means that there was a large coherent effect between X_A and XX_A. This spectrum leads to the steep-positive-slope region in the refractive index spectrum, resulting in the slowdown of light. Changing the dip width by means of the excitation intensity enables us to adjust the slope region and thereby gives a controllable slowdown function. The QD is a good candidate for a way to slow light. In the present QD, the slowdown function can be applied to light of 1.670 eV. This energy can be tuned by changing the QD structure. This type of controllability cannot be achieved in atom and ion systems.

We performed a pump-and-probe measurement to examine the coherent effects as optical nonlinear effects. This measurement corresponded to controlling the absorption properties of the probe beam by using the pump beam. In the measurement, PL signals from an exciton and a biexciton were detected with two excitation beams that passed through a Mach-Zehnder interferometer as shown in **Fig. 6(a)**. The difference between the two paths was chosen to be several centimeters to make the optical interference effects between the two beams almost negligible. Although the two beams were supplied from a single laser source, they acted as independent pump and probe beams. When the X_A state is eliminated as a result of coherent effects by the pump beam, no exciton can be created by the probe beam so there is no exciton PL emission. Measurements of this type may provide the fundamental properties of an optical function using two beams.

X and XX PL intensities are shown as a function of excitation power density in **Fig. 6(b)**. The probe beam power was about 0.05 kW/cm^2. All the points indicate nonlinear portions that are the changes in PL intensity induced by the pump beam. Although the XX PL increased with pump power density and saturated above 0.3 kW/cm^2 in Fig. 6(b), the X PL increased up to 0.1 kW/cm^2 and then decreased with

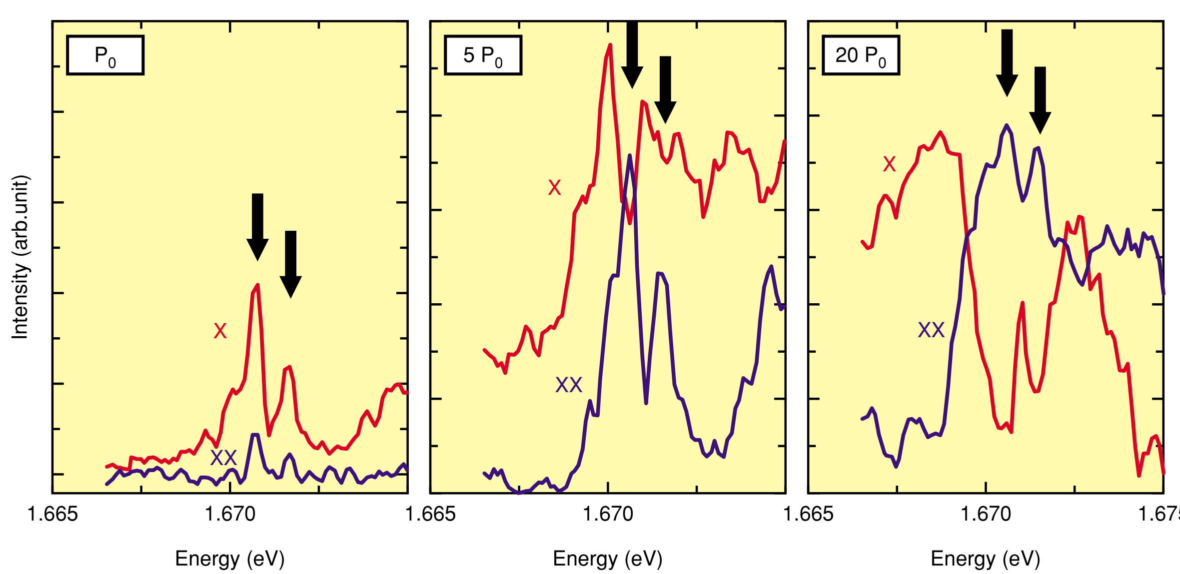


Fig. 5. PLE spectra for X and XX measured with high energy resolution. The two arrows in each panel indicate the positions of the X absorption peaks at P_0. P_0 is about 0.05 kW/cm^2.

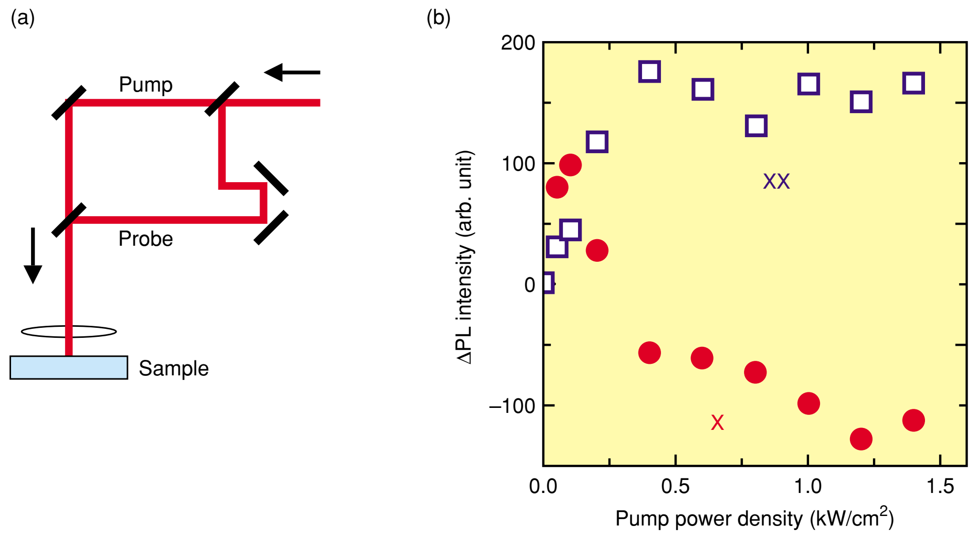


Fig. 6. (a) Schematic drawing of pump and probe measurement.
(b) Dependence of nonlinear portion of PL intensity on pump power density.

increasing pump power density. This decrease resulted from the large coherent effects that occurred between the X_A and XX_A states. The pump beam created a dip-shaped absorption spectrum as seen in Fig. 5. The coherent effects erased the X_A state and induced a large optical nonlinear effect in the X PL, as seen in Fig. 6(b). This result clearly shows that the pump beam could control the probe beam absorption. This is a fundamental requirement for the demonstration of the slowing down of light.

In this measurement, we used a conventional laser source for the excitation. In the ideal case, this nonlinear effect needs only an exciton and a biexciton. This means that only two photons are required for the optical nonlinear effects. The results clearly show that a large optical nonlinearity is feasible with several excitation photons in QDs.

Our results reveal that large optical nonlinear effects were achieved in a QD as a result of the coherent interaction between X and XX states and this could be evaluated by using another probe beam. This nonlinearity corresponds to optical control of probe beam absorption. To measure the slowing down of a probe beam, we need a technique for detecting a probe light transmitted from a QD. At the moment, there is no efficient detection method because the QD size (about 30 nm) is much smaller than the probe beam wavelength (about 750 nm). To detect the transmitted probe beam, we would have to make a sample that included many QDs. We have already fabricated such QD samples [15], [16]. They will enable us to calculate the slowdown factor in QDs from the transmission measurement results. Such measurements of the slowdown of light may be demonstrated with QD samples in the near future. Our current achievement in a QD constitutes an important step towards obtaining coherent optical device functions with many QDs.

4. Conclusion

We have described an optical nonlinear function in QDs produced by coherent processes in several quantum states such as exciton and biexciton states. These optical functions include coherent trapping, light-slowing, and inversionless laser emission as in EIT. These functions can be used in a conventional optical device to demonstrate coherent optical delay, which is very useful in the field of optical routing in optical telecommunications. Moreover, these functions may provide a temporary buffer for quantum bit coherence. Such a buffer is crucial for quantum gate control because it can adjust the timing of many optical pulses for gating processes. As a fundamental step towards achieving these optical functions, we examined the optical nonlinearity induced by the coherent process between an exciton and a biexciton in an InGaAs QD. The characteristic absorption spectra that were obtained are evidence of coherent effects mediated by quantum interference effects. The results of the pump and probe measurements reveal that probe beam absorption was controlled in a QD. These results constitute an important milestone in terms of controlling the speed of light without any dissipation. A sample with QD ensembles, which is currently in preparation, may enable us to estimate actual controllability. Appropriate designs of QDs for structures and semiconductors are also needed to clarify possible application areas. The fundamental information will enable the implementation of actual QD devices. Coherent optical nonlinearity in QDs promises valuable applications that will provide many technical innovations in various industrial fields.

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