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Time Response of One-Dimensional Photonic Crystals with a Defect Layer Made of Semiconductor Quantum Dots

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The effective optical nonlinearity of semiconductor quantum dots can be enhanced in one-dimensional photonic crystal structures. We have fabricated a one-dimensional photonic crystal with a structural defect which is made of CdSe quantum dots. Besides the enhancement of degenerate four-wave mixing efficiency, a fast response of transient population grating in an order of a picosecond was obtained in the one-dimensional photonic crystal. This structure can be used as a future photonic device with high nonlinearity and fast response.

KEYWORDS: one-dimensional photonic crystal, localized mode, semiconductor quantum dot, CdSe, optical nonlinearity, degenerate four-wave mixing, photon echo, transient grating

For the future practical application of photonics, it is necessary to find nonlinear optical materials with high optical nonlinearity and fast response. In general, however, there is a trade-off between the magnitude of nonlinear susceptibility and the response time, and it is not easy to satisfy these two features at the same time.[1)] As a solution of this problem, we propose to use fast response nonlinear materials in photonic crystal structures which can enhance the nonlinear optical effect.

In the recent progress of microfabrication technology, much effort has been devoted to the study of mode control of radiation field by using the photonic crystal structure.[2)] We can consider various types of photonic crystal structures from one-dimensional to three-dimensional ones depending on the targets. For example, in order to control spontaneous emission[3)] or light propagation,[4)] three-dimensional photonic crystals are suitable. On the other hand, in the case of the control of laser beams which are close to plane wave, we need not to have three-dimensional photonic crystals but one-dimensional photonic crystals (1-D PCs) are sufficient. When a structural defect is introduced in the photonic crystal, photon-localized state can be created in the photonic band gap. In this case, electric field around the defect layer will be enhanced due to photon localization. Therefore, if the defect layer is made of optical nonlinear material, the enhancement of nonlinear optical effect can be expected. For the enhancement of optical nonlinearities, the 1-D PC is the best candidate since the incident light field is fully coupled to the localized defect mode only in the 1-D PC. The 1-D PC structures with a defect layer can be considered as nonlinear Fabry-Perot etalons or microcavities. However, the concept of photonic crystals has more diversity than that of nonlinear Fabry-Perot etalons or microcavities, and includes not only one-dimensional but also two- or three-dimensional structures.

In our previous work,[5)] we studied the photonic band structure of the 1-D PCs and the propagation of ultrashort light pulses through in the 1-D PCs by using a white-light Michelson interferometer.[6)] Furthermore, we observed the enhancement of nonlinear optical effect, for instance, absorption saturation and degenerate four-wave mixing (DFWM). In that experiment we used a dye-molecule doped polymer film as the nonlinear defect layer and utilized its resonant transition, i.e. absorptive nonlinearity. Dye-doped polymer films are good media for phase conjugation optics due to their large third-order nonlinearity. We generated phase-conjugated image by using the 1-D PC with a defect layer.[5)] However, in general, the excited-state lifetime of dye molecules is in an order of a nanosecond, so that dye-doped polymer films are not suitable for ultrafast optical switching in the picosecond region.

Semiconductor quantum dots are attractive because of their high optical nonlinearity and fast response. They are expected as the nonlinear material for the future photonics devices. Much effort has been devoted to the study of semiconductor quantum dots,[7)] especially on quantum confinement effect. The most characterized and well-understood confinement effect is blue-shifting of the ground state excitonic absorption with decreasing the size of quantum dots. If the size of the quantum dots decreases, the surface-to-volume ratio increases, and electronic and optical properties become sensitive to the surface condition. Therefore, the excited-state lifetime becomes shorter due to the surface recombination effect. This is one of the important quantum confinement effects.

In the present study, we fabricated a 1-D PC with a defect layer made of semiconductor quantum dots. The defect layer was a silica glass doped with CdSe quantum dots made by co-sputtering CdSe and SiO_2, and it was sandwiched between 5-period quarter-wave stacks of SiO_2 and TiO_2 bilayers as shown in Fig. 1. The optical thickness of each layer of SiO_2 and TiO_2 on both sides of the defect layer was 488 nm/4. The diameter of the CdSe quantum dots is about 4 nm. The transmission spectrum of the 1-D PC thus made and the absorption spectrum of the CdSe quantum dots in glass are shown in Fig. 2 as a solid curve and a dashed curve, respectively. Several defect modes are created in the photonic band gap region, since the optical thickness of the defect layer is substantially larger than half-wavelength, 488 nm/2.

The 1-D PC with a defect layer can be considered as a microcavity where the light field of the resonant mode, or the defect mode, is localized around the defect layer. If all the layers are transparent, the light intensity at the defect layer is enhanced from the input intensity by a factor of $G = (n_B/n_A)^{2N}/2$, where n_A and n_B are the refractive indices of the layer A and B, respectively, and N is the number of pe-

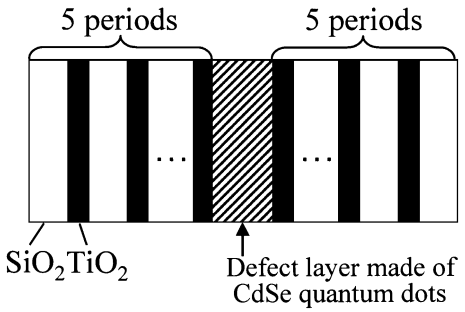

Fig. 1. Model of 1-D PC structures with a defect layer made of CdSe quantum dots.

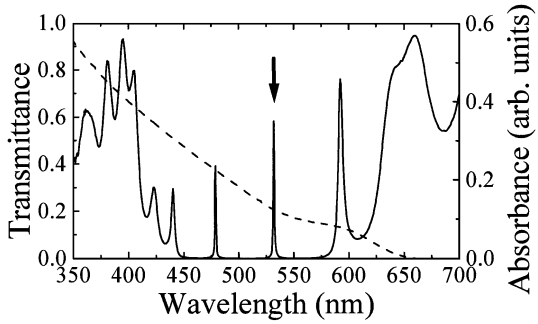

Fig. 2. Transmission spectrum of 1-D PC with a defect layer (solid curve), where the arrow shows the excitation wavelength, 532 nm. The defect layer is made of CdSe quantum dots in glass. The dashed curve shows the absorption spectrum of CdSe quantum dots in glass.

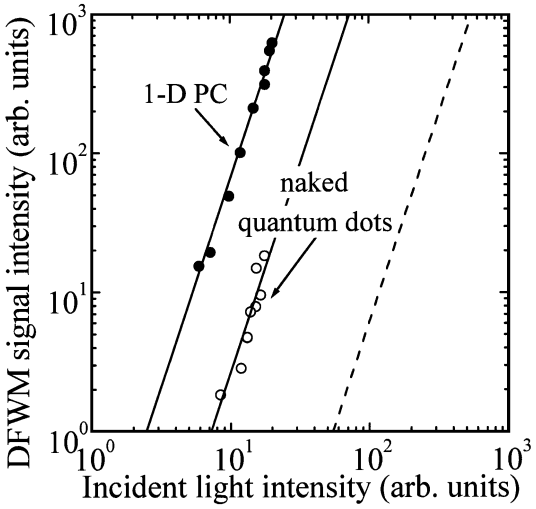

Fig. 3. Dependence of generated DFWM intensity on the incident light intensity. The closed circles are for the 1-D PC whose transmittance at 532 nm is 60%, and the open circles are for the naked CdSe quantum dots in glass whose transmittance is 60%. The dashed line is the simulation for the naked CdSe quantum dots in glass whose transmittance is 99%. The solid and dashed lines indicate the cubic-power low dependence on the incident light intensity.

riods of A and B bilayers on each side of the defect layer.[8] Since $n_A = 1.46$ and $n_B = 2.35$, and $n_B/n_A = 1.6$ for the stacks of SiO_2 and TiO_2, we can obtain the intensity enhancement of the order of 10^2 and 10^4 for $N = 5$ and $N = 10$, respectively.

In the present study, however, we discuss about the 1-D PC structures with a defect layer where the defect layer is absorptive. In this case, the intensity enhancement factor G at the defect layer is reduced by the absorption, and it is expressed as[8]

$$G = 2(n_B/n_A)^{2N}[2 + \pi\kappa\,(n_B/n_A)^{2N}/n_x]^{-2}, \qquad (1)$$

where κ and n_x are the extinction coefficient and the refractive index of the defect layer. The pure absorptive nonlinearity can be justified if the defect-mode wavelength falls well inside the absorption band. We chose absorptive nonlinearity because it can be induced by much smaller light intensity than the case of dispersive nonlinearity.

In order to see this enhancement effect, we have performed the DFWM and compared the signal intensity from the 1-D PC with that from a naked quantum dots in glass. Since the wavelength of the transmission peak of the 1-D PC depends on the angle of incidence, we used box-CARS (coherent anti-Stokes Raman scattering) configuration[9] of DFWM where the angles of incidence of all the three excitation beams and the output beam were the same, 2 degrees.

In the present experiment, the second harmonic of a Q switched Nd:YAG laser with a pulse width of 5 ns was used for excitation. One of the transmission peaks of the 1-D PC was controlled to be at the excitation wavelength, 532 nm, as shown by an arrow in Fig. 2. The wavelength is within the inhomogeneously broadened absorption band of CdSe quantum dots in glass. The transmittance of the transmission peak at this wavelength is 60%. From this transmittance, the single-pass transmittance of the defect layer at 532 nm is calculated to be 99%.[8] The multiple reflection in the 1-D PC resulted in the total transmittance of 60% at the transmission peak. As a reference sample, we prepared naked CdSe quantum dots in glass whose transmittance at 532 nm is the same as that of the 1-D PC, 60%. The experimental result is shown in Fig. 3, where the DFWM signal intensities are plotted against the incident light intensity. We observed a 20-time enhancement of DFWM generation efficiency of the 1-D PC compared with that of the naked quantum dots in glass. This value of the enhancement factor is much smaller than the theoretically obtained value of 3,500 for 60% transmittance.[8] We cannot tell the definite reason for this discrepancy at present, but the residual optical imperfection especially in the defect layer might be the main reason. The light scattering by the imperfection is greatly enhanced by the cavity effect of the present 1-D PC with a defect layer.

The single-pass transmittance of the defect in the present 1-D PC is 99%. The DFWM signal from the defect layer of the transmittance 99% without the PC structure was very small and could not be detected. However, the signal intensity can be calculated from the result of the naked sample with transmittance 60% and it is shown by a dashed line in Fig. 3. If the DFWM generation efficiency is compared for a same CdSe quantum dots doped glass of transmittance 99% with and without the PC structure, the enhancement factor would be 8,000.

Fast response is required for the future nonlinear optical devices. We performed time-resolved DFWM, or 3-pulse photon echo, experiments to study the time response of the 1-D PC. In the 3-pulse photon echo, the intensity of the echo signal is expressed as

$$I(\tau,\ T) = I_0 \exp(-4\tau/T_2 - 2T/T_1), \qquad (2)$$

where τ and T are the time separations between the 1st and

the 2nd pulses, and the 2nd and the 3rd pulses, respectively, and T_1 and T_2 are the excited-state population lifetime and the polarization dephasing time, respectively. By setting $\tau = 0$, the T dependence of $I(0, T)$ gives the information about the excited-state lifetime or population grating decay time T_1. On the other hand, by setting $T = 0$, τ dependence of $I(\tau, 0)$ gives the information about the polarization dephasing time T_2.

The box-CARS beam configuration was used also in the 3-pulse photon echo experiments. The excitation laser system was based on a cw mode-locked Ti:Sapphire laser (Spectra Physics, Tsunami). Its output pulses were amplified at a 1 kHz repetition rate by a regenerative amplifier (Spectra Physics, Spitfire) and seeded into an optical parametric amplifier (Spectra Physics, OPA-800) to produce the tunable light pulse whose pulse width is about 160 fs. The wavelength of the output pulses was tuned to one of the transmission peak of the 1-D PC. The 3-pulse photon echo experiments were performed at room temperature.

Figure 4(a) shows the echo decay curve $I(0, T)$ for naked CdSe quantum dots deposited on a glass plate by co-sputtering CdSe and SiO_2. The size of the dots is about 4 nm, and there are a lot of surface trapped states around the dots, therefore the excited-state lifetime is very short less than a picosecond.[10,11] On the other hand, Fig. 4(b) shows the decay curve $I(0, T)$ for the present 1-D PC with a defect layer made of quantum dots. In this case the decay time is a little enlarged compared with that for the naked quantum dots, but it is still in an order of a picosecond. This is because the cavity lifetime of the present 1-D PC structure is about 100 fs as can be seen from the width of the transmission peak shown in Fig. 2.

Figures 5(a) and 5(b) show the echo decay curve $I(\tau, 0)$ for the naked quantum dots and that for the 1-D PC, respectively. These echo decay curves give the information about the polarization dephasing time T_2 of the quantum dots. In the case of the naked quantum dots, since the polarization dephasing time of quantum dots at room temperature is considered to be less than the excitation pulse width,[7] the decay curve reflects the third order correlation of the laser pulse. However, in the case of the 1-D PC, the decay curve is broadened by the cavity lifetime of the 1-D PC as shown in Fig. 5(b).

These 3-pulse photon echo experiments show that, regardless of the nonlinearities originating from the excited-state population or the polarization of CdSe quantum dots in the present 1-D PC, we can obtain a very fast time response in an order of a picosecond. The thin film structure of the 1-D PC facilitates the use of this structure for phase conjugation optics.[5]

In conclusion, we obtained the 20-time enhancement of the DFWM generation efficiency in the 1-D PC against the naked quantum dots. By reducing the optical imperfection of the defect layer and optimizing the 1-D PC structure, we can expect much larger enhancement of the nonlinear effect. We also demonstrated by the 3-pulse photon echo experiments, that the ultrafast response time of the DFWM in an order of a picosecond can be obtained in the 1-D PC. The 1-D PC structures may be one of the key devices for the future optical processing.

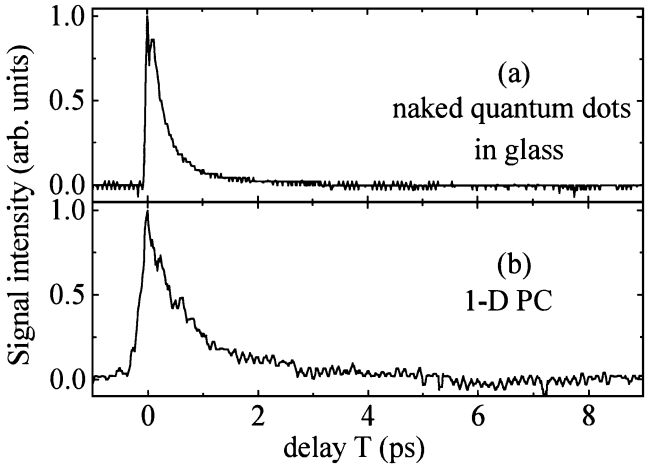

Fig. 4. 3-pulse photon echo decay curve $I(0, T)$ for the naked CdSe quantum dots in glass (a) and for the 1-D PC (b).

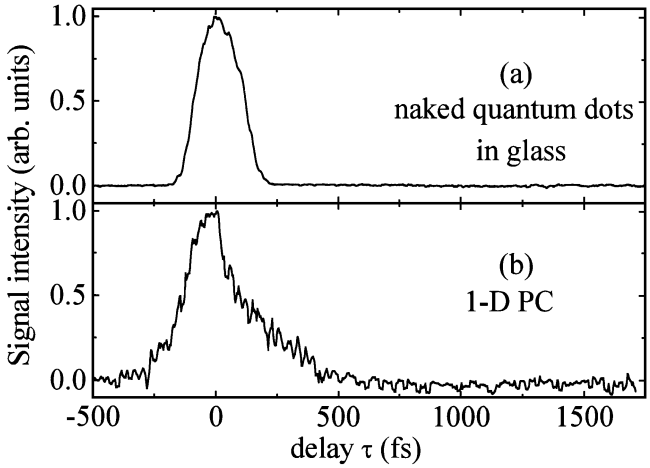

Fig. 5. 3-pulse photon echo decay curve $I(\tau, 0)$ for the naked CdSe quantum dots in glass (a) and for the 1-D PC (b).

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