Nonlinear optical properties of Phosphorous-doped Si nanocrystals embedded in phosphosilicate glass thin films

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Abstract: Nonlinear optical properties of phosphorus (P) -doped silicon (Si) nanocrystals are studied by z-scan technique in femtosecond regime at around 1.6 eV. The nonlinear refractive index (n_2) and nonlinear absorption coefficient (β) of Si-ncs are significantly enhanced by P-doping. The enhancement of n_2 is accompanied by the increase of the linear absorption in the same energy region, suggesting that impurity-related energy states are responsible for the enhancement of the nonlinear optical response.

OCIS codes: (190.4720) Optical nonlinearities of condensed matter; (190.0190) Nonlinear optics

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1. Introduction

Silicon nanocrystal (Si-nc) is a topic of great interests in the field of optelectronics because of its high quantum efficiency of photoluminescence (PL) and relatively large nonlinear optical responses[1, 2, 3, 4, 5, 6]. The large nonlinear optical response has been reported in various forms of Si-ncs such as porous Si prepared by electrochemical etching[7, 8], Si-ncs

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doped SiO_xN_y deposited by plasma enhanced chemical vapor deposition (PECVD)[10, 9], Si-ncs doped SiO_2 prepared by cosputtering[11], laser ablated Si-ncs deposited on quartz substrate[12, 13], and so on. In these literatures, dependence of the nonlinear optical response on the size and volume fraction of Si-ncs has been studied in detail. Although the origin of the large nonlinear optical response is still not fully clarified, the quantum confinement effects are often believed to be responsible[9, 11].

It has been demonstrated experimentally[15, 16, 17, 18, 19] and theoretically [20, 21, 22] that the electronic band structure, and the resultant optical and electrical transport properties of Si-ncs are significantly modified by impurity doping. Experimentally, PL properties of Si-ncs were found to be very sensitive to the impurity doping[15, 16, 17]. The doping of either n- or p-type impurities results in strong quenching of the PL, due to efficient Auger process between photo-excited electron-hole pairs and impurity-supplied carriers[17]. The quenching can be suppressed by doping n- and p-type impurities simultaneously because of the compensation of carriers within Si-ncs. The PL of the codoped and compensated Si-ncs appears at very low energy; the PL peak reaches 0.9 eV in heavily-doped an compensated Si-ncs [15, 16]. These observed phenomena are successfully reproduced, at least qualitatively, by first principles calculations [20, 23].

In this paper, we study the effect of impurity doping on the nonlinear optical properties of Si-ncs by using the samples of phosphorus(P)-doped Si-ncs embedded in phosphosilicate glass (PSG) thin films. We show that P-doping further enhances the large nonlinear optical responses of Si-ncs and is thus an effective way to control the nonlinear optical properties of Si-ncs.

2. Experimental procedure

P-doped Si-ncs embedded in PSG thin films were prepared by a cosputtering method. Si, SiO_2 and PSG were simultaneously sputter-deposited in Ar gas on a quartz substrate. Then the deposited films were annealed in a N_2 gas (99.999 %) atmosphere for 30 min at 1150 $^\circ$C to grow nanocrystals in the films. The size of Si-ncs was estimated by cross-sectional transmission electron microscopy (TEM) observations[24]. The average diameter (D) was about 4.0 nm, and the standard deviation was about 1.0 nm. The concentration of excess Si (C_{exSi}) and P_2O_5 (C_P) were obtained by electron probe micro analysis (EPMA). C_{exSi} was about 6.7 vol% and P_2O_5 was changed from 0 to 1.2 mol% . The linear refractive indices were estimated from the volume ratio of Si and SiO_2 with the application of the Bruggeman effective medium theory [25] and was about 1.54 at 800 nm.

The thickness of the samples was estimated by physical-contact-type surface roughness measurement and was about 12μm. The evidence that P atoms are doped in substitutional sites of Si-ncs and are electrically active was shown in our previous work by infrared absorption and electron spin resonance (ESR) spectroscopy [19]. The nucleation of phosphorous particles can be ruled out because they are thermodynamically unstable in SiO_2 matrix.

PL spectra were measured by using a single grating monochromator and an InGaAs near-Infrared diode array. The spectral response of the detection system was calibrated with the aid of a reference spectrum of a standard tungsten lamp. For the measurement of the nonlinear optical properties, a z-scan method was used. Details of the z-scan method is found elsewhere[14]. Briefly, in the z-scan method, the tight focusing gaussian beam is vertically irradiated onto a sample and the sample is moved along the direction of the beam propagation (z axis). The transmitted light intensity is recorded as a function of the distance from the focal point (z). When all of the transmitted light is detected (open aperture), the transmittance ($T_{op}(z)$) is determined by the nonlinear absorption coefficient (β), and its dependence on z is

$$T_{op}(z) = 1 + \frac{\beta I_0 L}{1 + (z/z_0)^2}, \tag{1}$$

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where I_0, L, and z_0 are the peak intensity of the beam, sample thickness and the diffraction length of the beam, respectively. Note that T_{op} doesn't depend on the nonlinear refractive index (n_2) but only on β, thus open aperture measurement provides the information on β.

When a small aperture is placed in front of the detector to cut peripheral regions of the transmitted light (closed aperture), the transmittance (T_{cl}) depends both on n_2 and β . The information on n_2 is extracted by the division of T_{cl} by T_{op},

$$T_{cl}/T_{op}(z) = 1 + \frac{4\Delta\phi}{((z/z_0)^2+9)((z/z_0)^2+1)}, \tag{2}$$

where $\Delta\phi$ is the nonlinear phase change. n_2 is obtained from $\Delta\phi$ as,

$$n_2 = \frac{\lambda\alpha\Delta\phi}{2\pi I_0(1-e^{-\alpha L})}, \tag{3}$$

where α and λ are the linear absorption coefficient and the wavelength of the beam, respectively.

For the gaussian beam, we used the mode-locked Ti:shaphire femtosecond laser with the pulse width of 70 fsec and the repetition frequency of 82 MHz. The photon energy was changed from 1.48 to 1.65 eV. The incident beam was focused on a sample by a lens with the focus length of 100 mm. The beam waist and diffraction length determined by a knife edge method were 18 μm and 1.1 mm, respectively. The peak intensity of the beam was typically 10 GW/cm^2. No notable change of nonlinear optical properties was observed in the intensity range of 0.5-20 GW/cm^2, suggesting that thermal effect was negligible in this measurement condition[10, 9]. The validity of the obtained data was checked by measuring a fused quartz plate as a reference.

3. Results and discussion

Figure 1 summarizes the PL and absorption properties of P-doped Si-ncs. Details of these properties are found in the literatures[17, 19]. In Figure 1(a), the absorption spectra of P-doped (C_P=0.8mol%) and pure Si-ncs are shown. In pure Si-ncs, absorption due to the valence-to-conduction-band transition starts at around 1.5 eV. In P-doped Si-ncs, in addition to the interband transition, intra-valence-band transitions appear below 1.3 eV. The observation of the intraband transitions evidences the doping of electrically active shallow impurities in Si-ncs[17, 19]. In Fig. 1(a), we also notice that interband transition of P-doped Si-ncs near the band edge is larger than that of pure Si-ncs. This absorption enhancement is very similar to that observed in P-doped bulk Si[29, 30], and is probably due to the impurity-related states predicted by some theoretical calculations[31]; P-doping causes the formation of the energy state just below the conduction band of Si-ncs, and enhances the absorption near the band edge. It should be noted here that each Si-nc should have discrete impurity-related states, because the number of impurity atoms in Si-ncs is very small and hence the formation of an impurity band is not expected. Apparently, this seems to contradict with the observed very broad enhancement near the band edge. The broadness can be attributed to the inhomogeneities of the number- and position-distributions of P atoms within a Si-nc and the size- and shape-distributions of Si-ncs. It should be stressed here that the size of pure and P-doped Si-ncs is similar and thus the difference of the spectral shape cannot be attributed to the size difference.

The inset of Fig. 1(a) shows the PL spectra of pure and P-doped Si-ncs. Both samples exhibit a broad PL band at around 1.3 eV. The PL is assigned to the recombination of electron-hole pairs within Si-ncs. This is evidenced by the temperature and the photon-energy dependence of the PL-lifetime[2], and also by the resonantly excited PL spectra[27]. In Figure 1(b), PL intensity at 1.3 eV and absorbance at 0.5 eV are plotted as a function of C_P. In the lower C_P region

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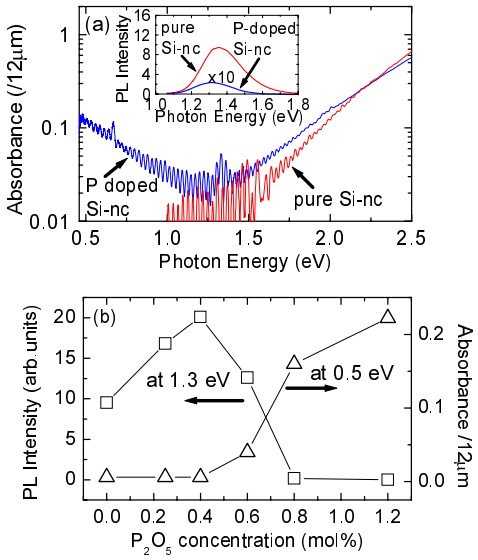

Fig. 1. (a)Absorption spectra of pure and P-doped Si-ncs (C_P=0.8mol%). The inset shows the PL spectra of the same samples. (b)P_2O_5 concentration (C_P) dependence of PL intensity at 1.3 eV (Left axis) and absorbance at 0.5 eV (Right axis).

(below 0.4mol%), no notable infrared absorption is observed, and PL intensity increases with increasing C_P. Carriers are thus not supplied within Si-ncs. The increase of the PL intensity in the C_P region is, as discussed in reference[28], considered to be due to the termination of dangling bond defects at the surface of Si-ncs by electrons supplied by doping. In the higher C_P region (above 0.6mol%), the infrared absorption increases and PL intensity decreases with increasing C_P. The PL quenching is accompanied by the shortening of the PL lifetime, and is considered to be nonradiative Auger recombination of photo-excited electron-hole pairs with the interaction with supplied carriers. It should be noted here that re-absorption by nearby clusters cannot explain the strong quenching because the samples are almost transparent in the energy range (optical transmittance $>$ 80%).

Figure 2 shows the results of z-scan measurements for (a) a closed aperture (T_{cl}), (b) an open aperture (T_{op}), and (c) the ratio (T_{cl}/T_{op}). Open squares and solid curves represent experimental data and fitted results, respectively. In Figs. 2(b) and 2(c), Eqs. (1) and (2), respectively, are used for the fittings. The solid curve in Fig. 2(a) is generated by using the parameters obtained by the fittings of Figs. 2(b) and 2(c). The agreement between the experimental data and the fitted curves is very good and the diffraction length estimated from the fitting coincides well with that measured by a knife edge method. In Fig. 2(c), all the z-scan spectra show valley to peak traces. This indicates that the sign of n_2 is positive for all the samples. We can see that the magnitude of the transmittance change depends on C_P. It increases with increasing C_P, suggesting that n_2 increases with increasing C_P.

Figure 3 shows the results of the analysis of the z-scan spectra. For the pure Si-ncs sample, i.e. C_P=0, the n_2 and β are $\sim 1.7 \times 10^{-13}$ cm^2/W and $\sim$ 1.0 cm/GW respectively. The observed n_2 is three orders of magnitudes larger than that of SiO_2, and one order of magnitude than that of bulk-Si.

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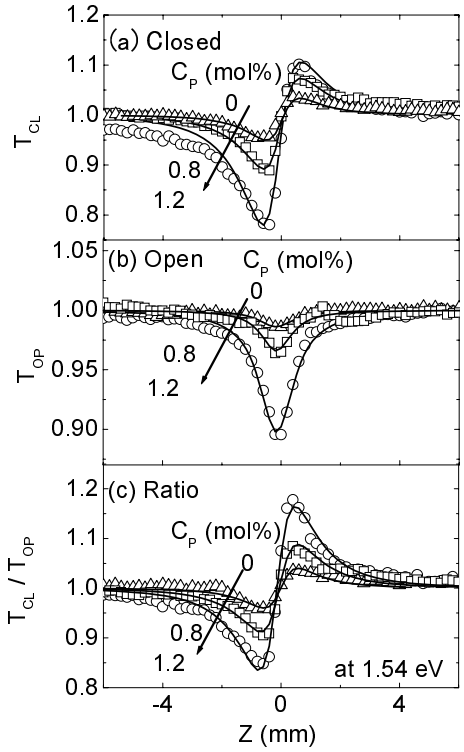

Fig. 2. z-scan measurements for (a) a closed aperture (T_{cl}), (b) an open aperture (T_{op}) and (c) the ratio of the two results (T_{cl}/T_{op}). The squares are experimental results and the solid curves are results of fittings. P_2O_5 concentration (C_P) is changed from 0 to 1.2mol% .

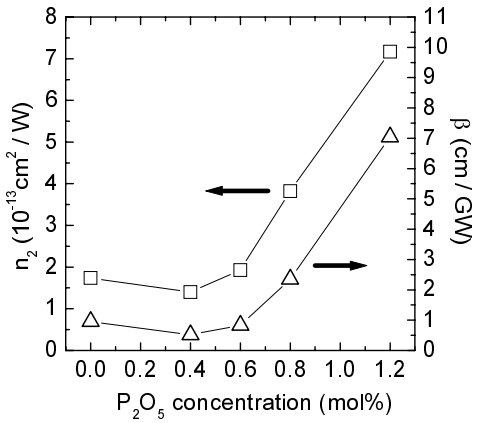

Fig. 3. P_2O_5 concentration dependence of n_2 (left axis) and β (right axis).

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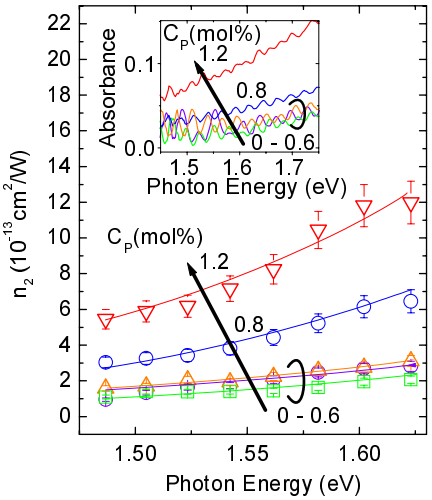

Fig. 4. n_2 spectra of samples with different P_2O_5 concentration (C_P). The inset shows the absorption spectra of the same samples.

Two different models have been proposed as the origin of the large n_2. The first one is a quantum confinement effect. This model has been proved by size-dependent n_2 enhancement[9]. The other one is that the surface state of Si-ncs is responsible for the large n_2. Klimov et al studied the transient absorption spectra of Si-ncs prepared by ion-implantation, and found a Si/SiO_2 interface state at around 1.6 eV, in addition to the size-dependent quantized states[32]. Vijayalakshumi et al showed that the surface state was responsible for the n_2 enhancement at around 1.6 eV in these samples[33, 34]. At present, no definite conclusion is obtained on the origin of the large n_2 and further intensive research is required to clarify the origin. However, since the investigation of pure Si-ncs is out of the scope of this work, we are going to focus on the effect of P-doping.

By P doping, n_2 increases from 1.7 to 7.0 $\times 10^{-13}$ cm^2/W, and β increases from 1.0 to 7.0 cm/GW. It is interesting to note that in the low C_P region, both n_2 and β are independent of C_P and are similar values to those of the pure Si-ncs sample. As shown in Figure 1, in the low C_P region, carriers are not generated within Si-ncs; supplied electrons are considered to be consumed for the termination of the dangling-bond-defects on the surface of Si-ncs. On the other hand, in the high C_P region, both n_2 and β increases with increasing C_P. One possible origin of this enhancement is the impurity-related state formed just below the conduction band of Si-ncs[31]. In the high C_P region, the states enhance optical absorption near the band edge of the Si-ncs. The enhanced absorption results in the enhancement of n_2.

In order to validate this possibility, we investigated the relation between n_2 and absorption. Figure 4 shows the photon-energy dependence of n_2. The error bars represents the fluctuation of average laser power and beam waist and also the estimation arising from the fitting procedure. For all the samples, n_2 increases with increasing the photon energy. The inset of Figure 4 shows the absorption spectra of the same samples. We can see clear similarity between them. This suggests that the impurity-related state is responsible for the observed enhancement of the nonlinear optical response by P doping.

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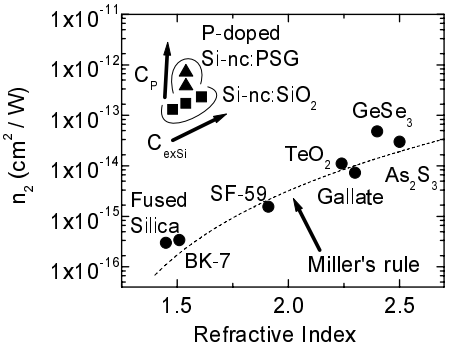

Fig. 5. n_2 is plotted as a function of linear refractive index. The dashed line is the prediction of the Miller's rule. Circles, squares and triangles are the results of several kinds of typical glasses, P-doped Si-ncs embedded in PSG (P-doped Si-nc:PSG) and pure Si-ncs embedded in SiO_2 (Si-nc:SiO_2), respectively.

In Figure 5, n_2 is plotted as a function of a linear refractive index (n_0). The dashed line is the prediction of the Miller's rule[35, 36]. The circles, squares and triangles are the data of several kinds of glasses[37, 38, 40, 39], P-doped Si-ncs embedded in PSG (P-doped Si-nc:PSG), and pure Si-ncs embedded in SiO_2 (Si-nc:SiO_2), respectively. C_{exSi} is changed from 3.3 to 10.5 vol% for Si-nc:SiO_2 and C_P is changed from 0 to 1.2mol% for P-doped Si-nc:PSG. The results of Si-nc:SiO_2 are quoted in our previous reports[11], where n_2 are obtained by the z-scan method with the same conditions as those of the present study. We can see that Si-nc:SiO_2 has relatively large n_2 and small n_0 compared to other typical glasses. For example, n_2 of Si-nc:SiO_2 is about three orders of magnitudes larger than that of a silica glass, while n_0 (=1.54) is similar to that of a silica glass. These properties are very suitable for the optical switching systems because smaller n_0 minimizes the optical coupling loss with the conventional SiO_2 fiber. As can be seen in Fig. 5, P-doping further enhances n_2 (at maximum 5 times) with no significant change of n_0. In this point, P-doped Si-ncs have great potential toward the realization of Si-based optical switching systems. It is definitely worth studying the n_2 of P-doped Si-ncs in other wavelength region, especially at around 1.5μm used in optical telecommunication industry.

4. Conclusion

Nonlinear optical properties of P-doped Si-ncs are studied by z-scan technique in femtosecond regime at around 1.6 eV. n_2 of Si-ncs is enhanced as much as 5 times by P-doping with no significant changes of n_0. This is a great improvement because generally the enhancement of n_2 is accompanied by the enhancement of n_0, which leads to the increase of the optical coupling loss with a conventional SiO_2 fiber. P-doped Si-nc is thus considered to be a promising candidate material for the realization of Si-based optical switching devices. We also showed that in the energy region of the n_2 enhancement, optical absorption is enhanced. This suggests that impurity-related energy states are responsible for the observed enhancement of the nonlinear optical response. The present results indicate that, in addition to the size and volume fraction of Si-ncs, impurity control is a parameter to control nonlinear optical responses of Si-ncs. Also, since this is, to our knowledge, the first report on the nonlinear optical properties of impurity-doped semiconductor nanocrystals, our study of P-doped Si-ncs will lead us to the better understanding of nonlinear optical properties of this kind of materials.

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