Study of intensity-dependent nonlinear optical coefficients of GaP optical crystal at 800 nm by femtosecond pump-probe experiment

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The intensity-dependent two-photon absorption and nonlinear refraction coefficients of GaP optical crystal at 800 nm were measured with time-resolved femtosecond pump-probe technique. A nonlinear refraction coefficient of 1.7×10^{-17} m^2/W and a two-photon absorption coefficient of 1.5×10^{-12} m/W of GaP crystal were obtained at a pump intensity of 3.5×10^{12} W/m^2. The nonlinear refraction coefficient saturates at 3.5×10^{12} W/m^2, while the two-photon absorption coefficient keeps linear increase at 6×10^{12} W/m^2. Furthermore, fifth-order nonlinear refraction of the GaP optical crystal was revealed to occur above pump intensity of 3.5×10^{12} W/m^2.

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Nonlinear optical effects are of current interest because of their potential applications to optical switching and optical phase conjugation[1]. Nonlinear optical coefficients and saturation effects have been investigated in semiconductors such as ultraviolet-grade fused silica, CdS[2], ZnSe quantum dots[3], molecular quantum wires[4], and semiconductor-doped glass[5−8].

Gallium phosphate (GaP) is a typical semiconductor. The nonlinear susceptibility tensor element d_{36} of GaP crystal has been measured by Levine and Bethea at 1.318 μm using the fringe-interference method[9]. The two-photon absorption (TPA) cross section of GaP crystal was also detected at 3.56 eV using TPA normalization[10]. Later, Rychnovsky *et al.*[11] carried out time-resolved picosecond pump-probe measurements and a Z-Scan technique to observe the nonlinear refraction and TPA coefficients of GaP crystal at 532 nm.

In the present work, we measured the intensity-dependent TPA and third-order nonlinear refraction coefficients of GaP optical crystal at 800 nm by time-resolved femtosecond pump-probe method. Moreover, the saturation of the third-order nonlinear refraction and the fifth-order nonlinear refraction of GaP crystal at 800 nm were investigated.

The sample used in our experiment is an orange-colored and transparent GaP single crystal, which is parallelogram with dimensions of 3×2 (mm) and thickness of 300 μm. The surface is coated with transmission film. The carrier density is 10^{18} cm^{-3}. The crystal structure is zinc blende and the orientation of crystal surface is $\langle 110\rangle$. A direct energy band gap at 2.78 eV and an indirect band gap at 2.27 eV near the X point[11] exist.

The experimental setup is shown in Fig. 1. A Ti:sapphire femtosecond pulsed laser, operating at a wavelength of 800 nm with a pulse width of 80 fs and a repetition frequency of 82 MHz, was used in our time-resolved pump-probe experiment. The laser beam was separated into pump and probe beams by a 10:1 beam splitter. The probe beam was delayed by an optical time delay line and then propagated parallel to the pump beam. The optical time delay line was drived by a step motor and the precision of movement was 0.01 μm, so the time resolution of delay line was 0.6 fs. Finally, both the pump and probe beams were split by a 7:3 beam splitter, then the transmissive part was impinged upon the GaP crystal by a focus lens ($f = 10$ cm) and superposed over with each other, while the reflective part was incident upon the BBO crystal. The reflective part was used to determine the zero point of the delay time between the pump and probe beams, which was the peak position of second harmonic frequency signal.

The average power of the pump and probe beams were first tuned to 10 and 1 mW, and the diameters of the focused pump and probe beams inside the sample were 40 and 36 μm, respectively. Consequently, the pump beam

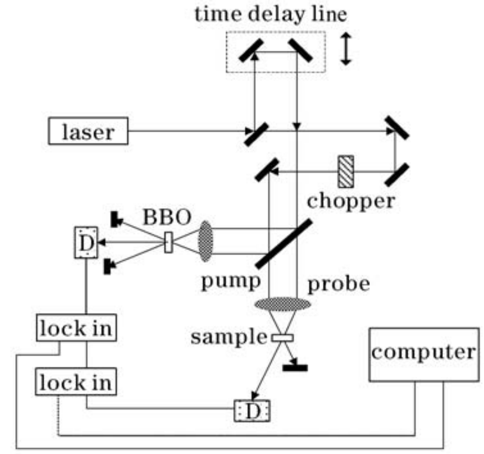

Fig. 1. Time-resolved pump-probe experimental setup.

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intensity in the sample was 1.3×10^{8} W/cm^{2}. The transmissive probe beam was detected by a Si detector, and then input into a lock-in amplifier. A chopper was put in the path of the pump beam, and set at a frequency of 400 Hz, which was locked with the phase of lock-in amplifier. Finally, a computer recorded the data from the lock-in amplifier and plotted the differential transmission curve of probe beam as a function of delay time.

We measured the time-resolved pump-probe signals while the pump intensity was set as 1.3×10^{8}, 1.5×10^{8}, 1.8×10^{8}, 2.3×10^{8}, 3.5×10^{8}, 4.6×10^{8}, and 6.7×10^{8} W/cm^{2}. The typical experimental data are depicted in Fig. 2.

Since the GaP crystal has a direct band gap of 2.78 eV and an indirect band gap of 2.27 eV[11], the TPA and third-order nonlinear refraction are expected to occur in this system while a photon energy of 1.55 eV is absorbed. According to the previous work[11], relaxation time and recombination lifetime of photo-excited free carriers on conduction band in GaP crystal are of the order of picosecond and nanosecond, respectively. Thus, the nonlinear instantaneous response is induced by photo-generated free carriers on conduction band on the femtosecond time scale while the pump and probe femtosecond pulses with high peak power excite it. It is well known that the amplitude modulation could be induced by TPA and cross phase modulation could be induced by the nonlinear refraction.

When the pump and probe pulses overlaped with each other in the time scale, one photon from the pump beam and one photon from the probe beam will be absorbed together in the TPA process. Thus the pump-probe signal due to the TPA will show the pure absorptive pulse-shape curves as a function of delay time. When we applied a lower excitation intensity, this kind of pure absorption signal was observed, as shown in Fig. 2(a). The cross phase modulation effect induced by nonlinear refraction does not appear here due to the lower pump intensity.

With the increase of the pump intensity, the cross phase modulation effect due to the nonlinear refraction gets obvious, which contributes to the gradually increasing emission signal in the pump-probe signal curves. In Figs. 2(b) and (c), the pump-probe signals have both absorption and emission shape curves, which represent the effects of TPA and nonlinear refraction, and the effect of nonlinear refraction becomes larger and larger with increasing the pump intensity. Finally, the pump-probe signal becomes a pure emission signal at a pump intensity of 4.6×10^{12} W/m^{2}, as shown in Fig. 2(d). This means that the nonlinear refraction effect is dominant in the pump-probe signal above this pump intensity.

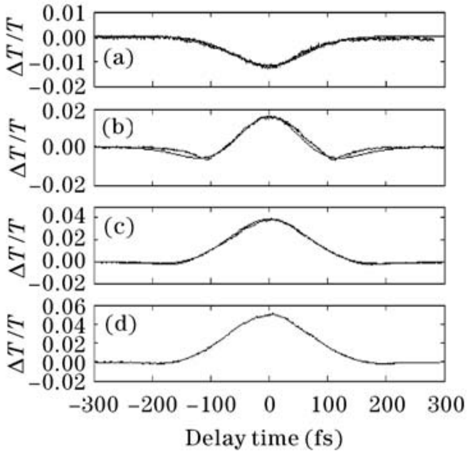

Fig. 2. Time-resolved pump-probe experimental data (dashed lines) and theoretical fitting curves (solid lines) at pump intensities of 1.3×10^{12} (a), 1.8×10^{12} (b), 3.5×10^{12} (c), 4.6×10^{12} (d) W/m^{2}. A clear correlation between the excitation intensity and the obtained pump-probe signal can be found.

These results demonstrate a clear correlation between excitation intensities and measured pump-probe signals. Therefore the intensity-dependent TPA and nonlinear refraction coefficients can be obtained from fitting the experimental data based on our previous theoretical work[12].

The nonlinear two-beam coupled-wave equation was solved in our previous work[12], in which the TPA and nonlinear refraction coefficients represented the interaction coefficients between the pump-probe beams and the GaP optical crystal, which appeared as a perturbation term in the coupling equation. In this paper, we introduce the intensity-dependent TPA and nonlinear refraction coefficients instead of the constant coefficients in the solution of the coupled-wave equation in Ref. [12] and then use this solution to fit the intensity-dependent experimental results.

The fitting results are also shown in Fig. 2. From the fitting results, we can obtain the intensity-dependent TPA and nonlinear refraction coefficients of GaP crystal at 800 nm, which are shown in Figs. 3 and 4. For example, the nonlinear refraction coefficient of 1.7×10^{-17} m^{2}/W and the TPA coefficient of 1.5×10^{-12} m/W at a pump-intensity of 3.5×10^{12} W/m^{2} are obtained. The TPA coefficient of GaP crystal at 800 nm keeps a linear increasing relation with the pump intensities, however the third-order nonlinear refraction coefficient tends to saturate at a pump intensity of 3.5×10^{12} W/m^{2}.

In the previous picosecond pump-probe experiments, due to a lower peak intensity of the picosecond pulse, the pump-probe signal showed only a pure absorption shape and a longer tail when the GaP crystal was excited at 532 nm, thus only the TPA coefficient of 7×10^{-11} m/W and the effective free carrier absorption cross section of 0.8×10^{-14} m^{2} were measured. The nonlinear refraction coefficient could not be obtained. The TPA coefficient of GaP crystal at 532 nm is one order larger than our results

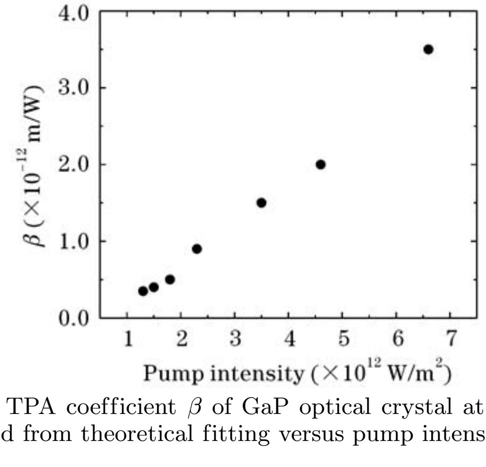

Fig. 3. TPA coefficient β of GaP optical crystal at 800 nm obtained from theoretical fitting versus pump intensity.

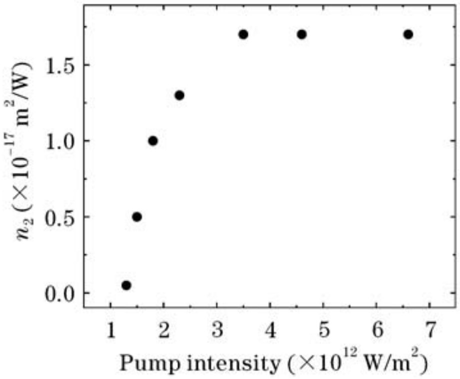

Fig. 4. Third-order nonlinear refraction coefficient n_2 of GaP optical crystal obtained from theoretical fitting versus pump intensity. A saturating nature above the pump intensity of 3×10^{12} W/m^2 is verified.

of 1.5×10^{-12} m/W. A TPA coefficient of 2×10^{-9} m/W of GaP crystal was also obtained at 348 nm using TPA normalization[10], which it is two orders larger than that at 532 nm and three orders larger than that at 800 nm. These experimental values at different excitation wavelengths are in good agreement with the theoretical prediction from the Basov model that the TPA coefficient of GaP crystal will increase as the excitation energy becomes larger[10]. These results can also be understood qualitatively as that a larger excitation energy will produce a larger free carrier density in the conduction band, so that a larger TPA coefficient is induced. This is consistent with the formation mechanism of the intensity-dependent TPA coefficient, which has a linear increasing trend with the pump intensity.

In the previous work, the Z-scan technique was used to measure the index change per photo-generated free carrier pair in GaP crystal at 532 nm[11]. The value of -3.1×10^{-16} m^3 was obtained. In our experiment, the third-order nonlinear refraction coefficients at different pump intensities were measured. With increasing pump intensity, the nonlinear refraction coefficient of GaP crystal at 800 nm increases and tends to saturate at a pump intensity of 3.5×10^{12} W/m^2.

Since GaP crystal has a direct and an indirect energy bands, the nonlinear refraction of GaP crystal mainly comes from three components: direct interband transition, indirect interband transition, and direct intraband transition[11]. Firstly, TPA in the direct interband transition and indirect interband transition will contribute to the third-order nonlinear refraction. Secondly, when the pump intensity increases, free carriers in both the direct and indirect bands saturate, and then they will be absorbed into the higher energy band due to the direct intraband transition, therefore the fifth-order nonlinear refraction is formed. The above nonlinear mechanisms of the GaP crystal can qualitatively explain the correlation of intensity-dependent TPA and nonlinear refraction coefficients in GaP crystal. When the pump intensity increases, the third-order nonlinear refraction coefficient becomes larger and larger, finally tends to saturate at 3.5×10^{12} W/m^2, however the TPA coefficient always keeps linear increase even above 3.5×10^{12} W/cm^2, as depicted in Figs. 3 and 4. This implies that the TPA in GaP crystal induces the third-order nonlinear refraction and its saturation at lower pump intensities, and then continues to contribute to the formation of the fifth-order nonlinear refraction when the pump intensity is above 3.5×10^{12} W/cm^2.

The saturation intensity of third-order nonlinear refraction coefficient of GaP optical crystal at 800 nm is estimated to be around 3.5×10^{12} W/m^2, which is of the same order as that in ZnSe quantum dots in glass-matrix thin films[5], and two orders lower than the value of 10^{14} W/m^2 in bulk CdS material[2]. The intensity-dependent pump-probe signals, such as the absorption signal at low intensity and the emission signal at high intensity, can be looked as a negative or positive logic gate, so that it could be utilized to make an all-optical switching device.

In summary, we measured the intensity-dependent TPA and third-order nonlinear refraction coefficients of GaP optical crystal at 800 nm by a time-resolved femtosecond pump-probe technique. The saturation of third-order nonlinear refraction of GaP optical crystal at 800 nm was observed at a pump intensity of 3.5×10^{12} W/m^2, however the TPA coefficient still showed a linear increasing trend at the pump intensity of 6×10^{12} W/m^2, therefore the fifth-order nonlinear refraction of GaP crystal was revealed to occur above the pump-intensity of 3.5×10^{12} W/m^2. The intensity-dependent pump-probe signals of GaP optical crystal was promising to make an all-optical switching device which could find its application in the optical communication area.

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