Measurement of the third order nonlinear optical coefficients of GaP and chirp parameter of laser pulse

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The spectral resolved femtosecond pump-probe experiment is applied to measure the third order nonlinear refractive index coefficient, two-photon absorption coefficient of GaP crystal, and the chirp parameter of the input laser pulses. The results show that nonlinear refractive index coefficient is 2×10^{-18} m^2/W, two-photon absorption coefficient is 2×10^{-11} m/W at wavelength of 783.5 nm, and the chirp parameter of laser pulse is 1×10^{25} s^{-2}. Furthermore, the mechanism of nonlinear refraction due to two-photon absorption in GaP crystal and experimental results are discussed.

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As high-speed optical fiber communication has been rapidly developing, all-optical communication becomes of the great research interest. It is important to study all-optical modulation, all-optical switching, and all-optical transistor. For this purpose optical nonlinearity of the material has attracted much interest. Gallium phosphate (GaP) is a kind of typical nonlinear optical semiconductor material, which was studied in detail as a luminance diode and an acousto-optic modulator in the past[1]. Levine and Bethea[2] measured its nonlinear susceptibility tensor element d_{36} at 1.318 μm using fringe-interference method with Nd:YAG laser. Later Rychnovsky *et al.*[3] measured its nonlinear refraction change and two-photon absorption coefficient at 532 nm by picosecond (ps) pump-probe technique and Z-scan techniques. In this letter we report the measurement for nonlinear refraction coefficient and two-photon absorption coefficient of GaP at 783.5 nm by spectrally resolved femtosecond (fs) pump-probe technique, and the chirp parameter of laser pulse is also determined at the same time.

We performed spectrally-resolved pump-probe experiment in GaP crystal by fs Ti:sapphire laser. Central wavelength of laser was tuned to 788.5 nm and output power was 200 mW. The laser beam was separated into two beams by a splitter (10:1), with the stronger beam as pump and the weak one as probe. The probe beam was delayed by optical delay line. Both pump and probe beams were focused and overlapped in GaP crystal by a focusing lens (f = 10 cm). Before the sample average powers of the pump and probe beams were 10 and 1 mW, respectively. The radius of focusing spot was 20 μm, thus the intensity of pump beam was about 1×10^{12} W/m^2. The transmitted probe beam was focused into monochromator and then detected at different wavelengths by a photomultiplier. The output signals from the photomultiplier were recorded by lock-in amplifier. A chopper was placed in the path of pump beam before the sample and its phase was locked with that of the lock-in amplifier. Output data from lock-in amplifier were sent into a computer. The figure of experimental setup was presented in Ref. [4].

We measured transmitted signals of probe beam at the wavelengths of 779.5, 783.5, 788.5, 796.5, and 804.5 nm. Experimental data were shown in Fig. 1 as dashed lines. Two-beam coupling equation was derived by Kang *et al.*[5] and further developed by Wang *et al.*[6]. We used analytical solution derived by Wang *et al.*[6] to fit our experimental results. In nonlinear coupled-wave propagation equation for probe field, Wang assumed linearly chirped pulses as $E(t) = E^0 \exp(-t^2/\tau^2 + ibt^2 + i\omega_0 t)$, where b is chirp parameter of laser pulse, and obtained the spectrally resolved differential transmittance for the probe field as

$$\frac{\Delta T}{T}(\Delta t, \delta) = 2\left[\frac{3 + b^2\tau^4 + 4b^2\tau^4}{(9 + b^2\tau^4)^2}\right]^{1/4} \times \exp[g(b, \delta, \tau)] \times \exp\left\{\frac{[\Delta t - T_s(b, \delta, \tau)]^2}{[\Gamma(b, \tau)]^2}\right\} \times (2\Delta\Phi \sin\Theta - q\cos\Theta), \tag{1}$$

with $\Delta\Phi = kn_2 I^0_{\text{pu}} L$, $q = \beta I^0_{\text{pu}} L$, $T_s = \frac{-\delta b\tau^4}{2(3+b^2\tau^4)}$, $\Gamma = \tau\left[\frac{9+b^2\tau^4}{2(3+b^2\tau^4)}\right]^{1/2}$, $g = \frac{\delta^2\tau^2}{2(1+b^2\tau^4)(3+b^2\tau^4)}$, $\Theta = \frac{4b}{9+b^2\tau^4}\left(\Delta t + \frac{3\delta}{4b}\right)^2 - \frac{\delta^2}{4b(1+b^2\tau^4)} - \frac{1}{2}\tan^{-1}\left(\frac{2b\tau^2}{3+b^2\tau^4}\right)$.

In Eq. (1), Δt is relative time delay between pump and probe beams, $\delta = \omega - \omega_0$ is the frequency detuning of probe frequency with respect to its central frequency, ω_0 is the central frequency of probe pulse, $k = 2\pi/\lambda$ is circular wave number. n_2 is nonlinear refractive index coefficient and β is two-photon absorption coefficient. I^0_{pu} is the intensity of pump beam. L is propagation distance of optical field in sample, which is 200 μm. We fitted the experimental results using Eq. (1), as shown in Fig. 1 in solid line, and the values of b, n_2, and β are presented in Table 1. We obtained the values of n_2 as 6×10^{-18} m^2/W and β as 5×10^{-11} m/W at laser pulse central wavelength. The chirp parameter of the pulse is 1×10^{25} s^{-2}. Furthermore, n_2 and β showed a wavelength-dependent feature to be discussed later.

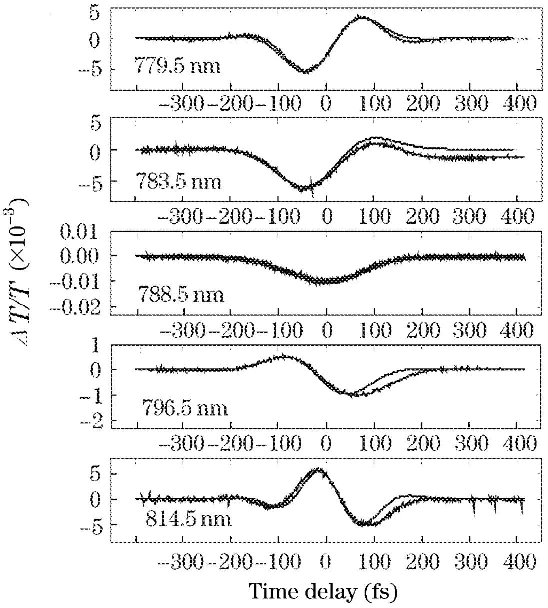

Fig. 1. Pump-probe signals (dashed lines) measured at different detection wavelengths in GaP. (a) 779.5 nm; (b) 783.5 nm; (c) 788.5 nm; (d) 796.5 nm; (e) 804.5 nm along with theoretical fitting results (solid curves).

Table 1. The Values of n_2, β, and b Obtained from the Theoretical Fitting to the Experimental Data

λ (nm)	n_2 (m^2/W)	β (m/W)	b (s^{-2})
779.5	1×10^{-18}	1×10^{-11}	1×10^{25}
783.5	2×10^{-18}	2×10^{-11}	1×10^{25}
788.5		5×10^{-11}	1×10^{25}
796.5	2×10^{-18}	2×10^{-11}	1×10^{25}
804.5	2×10^{-18}	2×10^{-11}	1×10^{28}

GaP has an indirect bandgap at 550 nm and a direct bandgap at 445 nm[1]. With the excitation at wavelength of 788.5 nm, two-photon absorption by direct and indirect bandgap transitions excites the electrons to conduction bands, which induces the nonlinear susceptibility in GaP. In Fig. 1 we got the pulse-shape absorption signal at central wavelength and dispersion-shape signals at detuning wavelengths. It can be explained that these signals were induced by two-photon absorption and cross-phase modulation produced by nonlinear refraction[7]. It is known that the nonlinear refraction coefficient and two-photon absorption coefficient are intensity and frequency dependent[8]. Rychnovsky *et al.*[3] obtained two-photon absorption coefficient of GaP of 1.9×10^{-11} m/W at 532 nm, which is similar to the result at 783.5 nm obtained in this work. Here, we show that the two-photon absorption coefficient of GaP is not influenced much by frequency. Furthermore we obtained the same nonlinear refraction coefficient and two-photon absorption coefficient at symmetric detuning wavelengths. The intensity change of probe due to phase modulation induced by pump pulse can be neglected at central wavelength[7]. This phenomenon is illustrated by experimental result in Fig. 1(c), that shows pure two-photon absorption signal at central wavelength. Thus fitting values of nonlinear refraction coefficients at central wavelength was meaningless. Sensitivity for nonlinear refraction measurement is larger with increasing detuning wavelength, as Kang *et al.*[5] presented. However, the measurement for transmission intensity of probe pulse at far detuning wavelengths, such as 779.5 and 804.5 nm, may produce more experimental errors due to their relative weak intensity and other experimental conditions. Thus we conclude that results at moderate detuning wavelengths are more reliable, for example at 783.5 and 786.5 nm, nonlinear refraction coefficient is 2×10^{-18} m^2/W, the two-photon absorption coefficient is 2×10^{-11} m/W.

In conclusion, we have measured the nonlinear refraction coefficient and two-photon absorption coefficient of GaP crystal with the pump wavelength at 788.5 nm with pump intensity of 1×10^{12} W/m^2 by spectral-resolved pump-probe technique. At the same time, the chirp parameter of laser pulse is determined. These results will provide useful parameters for GaP crystal as a candidate material of all-optical device. Further study on characteristics of two-photon absorption coefficient and nonlinear refraction coefficient of GaP crystal will be done in future work.

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