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Femtosecond Degenerate and Nondegenerate Pump-Probe Experiments in Bulk GaAs below the Band gap

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We perform degenerate and nondegenerate pump-probe experiments on bulk GaAs at 100 K below the band gap. We mostly observe a negative differential transmission signal both in the degenerate and nondegenerate experiments. We interpret our signal as due to two-photon absorption. This negative signal has a different origin from the normally considered band gap renormalization for resonant excitations.

I. INTRODUCTION

Ultrafast spectroscopy is a powerful tool for the investigation of nonequilibrium carrier dynamics in semiconductors. Femtosecond optical- pulse generation and measurement techniques make possible direct measurements of ultrafast relaxation processes on the time scale on which these processes occur. With the advance of femtosecond solid state lasers, femtosecond spectroscopy has become an important tool for investigation of dynamical process [1].

Most of the femtosecond experiments in semiconductors have been performed in resonant conditions: i.e., the photon energy is near the band gap energy or higher than the band gap energy. Only recently, there were some works on femtosecond nonlinearity below the band gap [2]. There have been some theoretical and experimental works on two-photon absorption when the photon energy is below the band gap [3,4]. However, because of limited tunability, these works concentrated on the comparison between different samples and not on the detailed detuning dependence measured from the band gap. In contrast to the previous studies, we investigate the two-photon absorption in GaAs using nondegenerate pump- probe experiments.

In a degenerate pump-probe experiment, the pump and the probe pulses are created from the same source. Therefore the pulse width and the center photon energy are the same for both pulses. On the other hand, the center photon energies of the pump and the probe pulses are different in a nondegenerate pump-probe experiment. To obtain different wavelengths, white-light continuum pulses are often used. By focusing amplified femtosecond laser pulses into a nonlinear medium, the white-light continuum is obtained. A part of this continuum is spectrally filtered and amplified to obtain the pump pulses and the other part is used as probe pulses with broad spectrum [5,6]. Another method is using an optical fiber and a grating pair, as in this work [7].

Optical transitions in a semiconductor are strongly influenced by the distribution of carriers in the conduction and valence bands. For direct transitions away from exciton resonances the absorption coefficient can be expressed as

$$\alpha = \alpha_o(1 - f_e - f_h),$$

where $f_{e,h}$ are the distribution functions for electrons and holes and α_o is the absorption coefficient in the absence of state filling. With a sufficiently short optical pulse a narrow band of states can be excited, creating a nonthermal distribution. As time progresses, carrier-carrier interactions lead to thermalization of the carrier distribution.

Most absorption saturation measurements were performed with very thin samples (10 - 1000 nm) in a resonant excitation condition. The reason is that experimenters have focused on the properties of real excitations. In resonant excitation, the signal itself is absorbed and the intensity is roughly proportional to $le^{-\alpha l}$ where l is the thickness of the sample and α is the absorption coefficient of the sample. In constrast, the signal due to virtual excitation is proportional to the sample thickness.

Virtual excitation is the excitation to the virtual

states, which occur in the region where the density of states is zero (below the band gap). The creation and decay of the virtual carriers are determined entirely by the exciting pulse. So the signal rises and falls faithfully with the pulse intensity.

II. EXPERIMENTAL SYSTEM

Two-color femtosecond pulses are created by a commercial Ti:sapphire laser (Coherent Mira 900). The laser is operated at 855 nm and its output is passed through a prism pair compressor for external group velocity dispersion compensation (Fig. 1). Then it is passed through a Faraday isolator and coupled into an optical fiber using a 20× microscope objective. The optical fiber has 5.5-cm-long and 3.9-μm core diameter single mode at 630 nm. In the optical fiber the spectrum is broadened from ~14 to ~130 nm full width at half maximum (FWHM) by self-phase modulation [8]. The output of the fiber is then split into two parts, which are independently compressed and spectrally filtered using two identical grating pair systems by a unity magnification telescope [9,10]. Wavelength selection of nearly Gaussian pulses is achieved by means of slits placed out of the Fourier planes [11,12]. Without slit, the output of grating is a 25 fs pulse with spectral width 150 nm (790~940 nm). The slit widths are adjusted to produce 15 nm FWHM with 80~100 fs duration time. The two beams are sent into a standard pump-probe setup.

The probe beam is passed through a variable delay stage and a portion of the probe beam is used as a reference. The pump beam is polarized perpendicular to the probe pulse and chopped at 1.5 kHz. The beams are focused into the sample using a lens with 50 mm focal length. The transmitted probe signal and the reference are detected using a photodiode and the difference of these two signals is amplified using a differential amplifier. Then the signal is detected by a lock-in amplifier. Data is accumulated by varying the delay between the pump and probe pulses with a computer-controlled stage moving in 0.1-μm steps while recording the lock-in output. The measurements are performed at 100 K in a 50 μm-thick intrinsic bulk GaAs sample. The sample is obtained by lapping and polishing of a commercial (Sumitomo) 350 μm-thick GaAs wafer.

III. RESULTS AND DISCUSSIONS

First we performed degenerate pump-probe experiments in the transmission geometry. Fig. 2 shows the measured differential transmission at different wavelengths and the number indicates the pump and the probe wavelength. The differential transmission ($\Delta T/T$) is the difference of the transmitted probe beam with the pump and without the pump beam, normalized by dividing the probe beam without the pump. The wavelengths of both the pump and the probe beam are changed from 810 nm to 870 nm simultaneously. The band gap energy of GaAs at 100 K is 1.501 eV corresponding to 826 nm in wavelength. So the exciting energy is mostly above the band gap at 810 nm or 820 nm and the others are below the band gap. When exciting above the band gap the transmission changed sign from negative to positive. P. Langot *et al.* observed a negative signal at short time delay followed by a slow rise and plateau when pump and probe photon energy is above the bandgap [13]. They explained this negative signal due to the band gap renormalization. Increase of the probed density of states (and thus of the absorption) by band gap renormalization predominates over reduction of the Coulomb enhancement (that reduces the absorption) resulting in

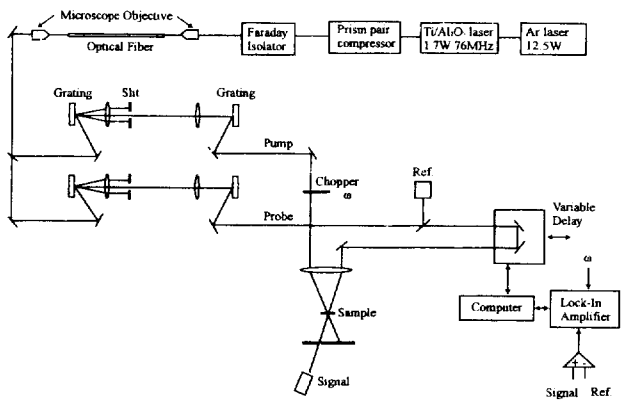

FIG. 1. The femtosecond two-color pump-probe experimental system. Pulses generated from Ti:Sapphire laser are broadened in the optical fiber and compressed in the two grating pair. The slit between two gratings selects the desired wavelength from the broad spectrum.

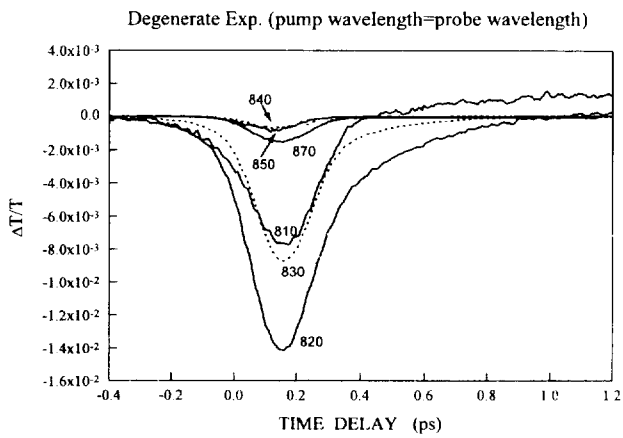

FIG. 2. Degenerate pump-probe experiments (pump photon energy = probe photon energy) in bulk GaAs when the temperature is 100K. The band gap energy of GaAs is 1.501eV (826nm).

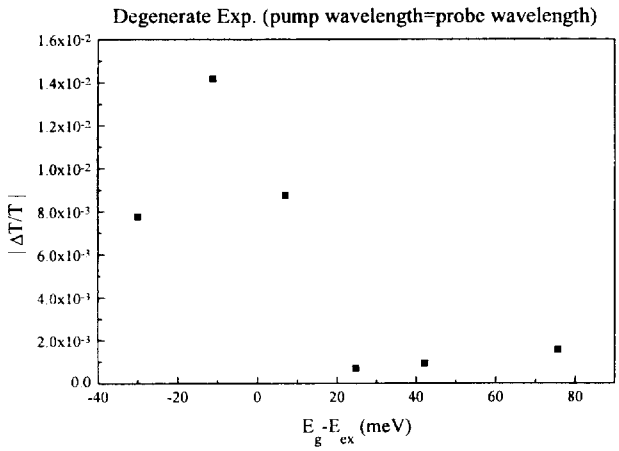

FIG. 3. Absolute value of the differential transmission as a function of the difference between the band gap energy and the excitation energy (pump photon energy) in degenerate experiment. Below the band gap, the intensity slowly increases with detuning increasing.

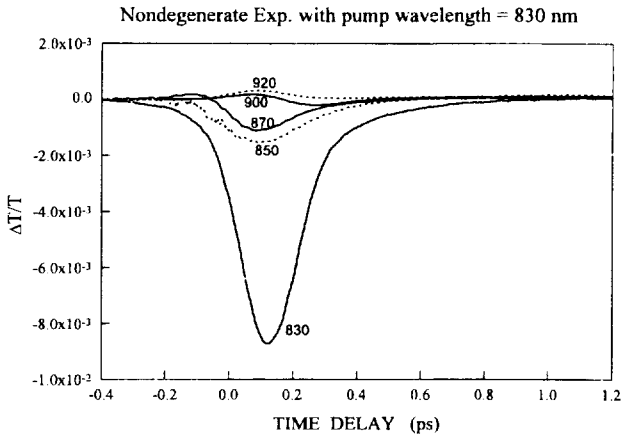

FIG. 4. Nondegenerate pump-probe experiments in bulk GaAs when the pump wavelength is 830nm. The number indicates the probe wavelength. As the probe wavelength becomes larger, the peak intensity is decreased. And there is a sign change between 870nm and 900nm.

an almost instantaneous decrease of the sample transmission.

With an excitation at below the band gap, there is still pump-induced absorption. These phenomena can not be explained by excitonic effects or band gap renormalization, because no real carrier is created. It is most likely to be caused by two-photon absorption. The two-photon absorption coefficient β_2 is given by [4]

$$\beta_2 = K E_p^{1/2} F(2h\nu/E_g)/n^2 E_g^3 \qquad (1)$$

$$F(2h\nu/E_g) = (2h\nu/E_g - 1)^{3/2}/(2h\nu/E_g)^5 \qquad (2)$$

where K is the proportionality constant, n is the linear refractive index, and E_p is related to the Kane momentum parameter [14]. The function F, whose exact form depends on the band structure, is a function only of the ratio of the photon energy ($h\nu$) to band gap (E_g), which determines the states that are optically coupled. In our case everything is constant except the photon energy. In Eq.(2), we can see that the coefficient increases rapidly until the ratio is 0.7 then slowly decrease [3]. Fig. 3 displays the absolute value of the differential transmission ($|\Delta T|/|T|$) as a function of the difference of the band gap energy and the excitation energy in degenerate experiments (Fig. 2). The absorption rapidly increases and decreases near the bandgap, then increases slowly. The slowly increasing region is consistent with the theoretical prediction. This region corresponds to the slowly decreasing region of the two-photon absorption coefficient.

Then we performed a nondegenerate pump-probe experiments. We fixed the pump energy slightly below the band gap and measured the transmission change for various probe energies (Fig. 4). In this experiment, a very interesting phenomenon is detected. As the probe energy decreases, the sign is changed. The reason for this phenomenon is not clear at the present time. The shape of the signal is delta function like. This shows that it's a virtual signal that exists only when the pump pulse overlaps the probe pulse. In Fig. 5, we plot the absolute value of the differential transmission versus the difference of the pump and the probe energies. The vertical arrow indicates the band gap energy of GaAs. As the difference of the pump and the probe energy is increased (and hence probe energy is decreased), absorption becomes weaker.

Fig. 6 is the measurement when the pump wavelength is fixed at 870nm, which is definitely below the band gap. We can not detect the sign change in this case. As shown in Fig. 7, absorption decreases with de-

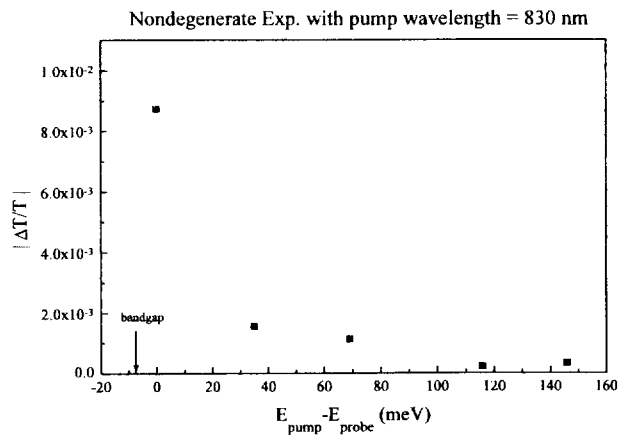

FIG. 5. Absolute value of the differential transmission versus the difference of the pump photon energy and the probe photon energy when the pump wavelength is 830nm (1.494eV). This energy corresponds to slightly below the band gap.

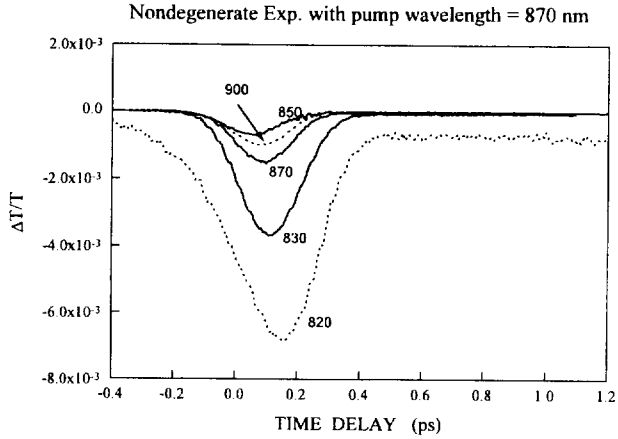

FIG. 6. Nondegenerate pump-probe experiments in bulk GaAs when the pump wavelength is 870nm.

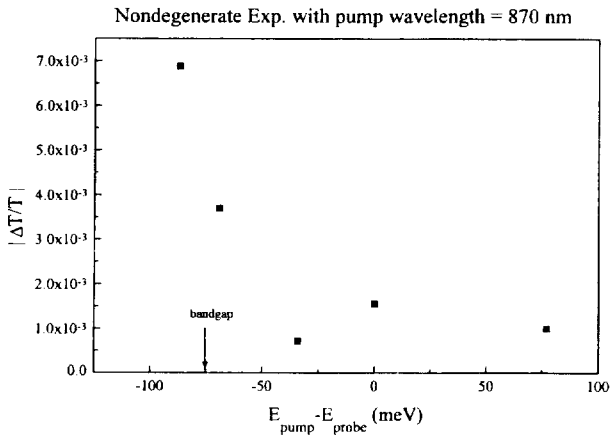

FIG. 7. Absolute value of the differential transmission versus the difference of the pump photon energy and the probe photon energy when the pump wavelength is 870nm (1.425eV), definitely below the band gap. The absorption slightly increases when the difference is zero.

tuning, but slightly increases when the pump and the probe energy are the same, and then decreases again.

IV. CONCLUSION

We performed degenerate and nondegenerate pump-probe experiments on bulk GaAs at 100K below the band gap. In degenerate experiments, the negative differential transmission (absorption) is observed when excited at both above and below the band gap. The negative signal with an excitation above the band gap is explained by band gap renormalization. We interpret the signal below the band gap as due to two-photon absorption. Mostly negative signals are also detected in nondegenerate experiments. An interesting sign change is observed when the pump energy is slightly below the band gap. Absorption increases when the probe energy is equal to the pump energy.

The dynamics of the intrinsic semiconductor below the bandgap is a very interesting topic. But there is little work about these phenomena, particularly non-degenerate experiments. Further experiments and theoretical works are certainly needed.

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