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Femtosecond photodeflection spectroscopy in thin monocrystalline plates of germanium*

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A highly sensitive photodeflection spectroscopy technique with femtosecond time resolution has been developed. Using this technique, we have measured accurately the shape of acoustic pulses, which were generated from the ultrafast phonon emission in a germanium plate. Supersonic expansion of photoexcited electron-hole plasma was observed. The characteristic velocity of plasma diffusion was evaluated. It exceeded the longitudinal sound velocity in germanium by a factor of 4.0.

Keywords: ultrashort acoustic pulse, femtosecond laser pulse, photodeflection technique
PACC: 7847, 4280W, 4335, 6860, 6320

1. Introduction

Generation and application of ultrashort acoustic pulses have played a very important role in in-line monitoring of semiconductor processing and in the optimization of materials for ultrafast electronic switches and devices. With the development of ultrafast laser technology, short laser pulses are now available for the excitation of ultrashort acoustic pulses in condensed matter. With time-resolved picosecond or femtosecond optical pump and probe methods,[1−4] acoustic or optical phonons have been detected through photoelastic or electro-optic effects, respectively. In 1988 Rothenberg[5] observed the picosecond thermal expansion of an absorbing surface after a short heating pulse. The measurement was performed on Si by adopting the method of photothermal deflection to a pump-probe technique using 1 ps laser pulses. In their measurement, the spatial inhomogeneity of the laser-generated elastic strain along the irradiated surface induced gradients in the phase of the probe laser beam, and then caused its deflection. But this work presumably contained an error in sign, because ample justification was given for this in the work of Wright and Gusev in 1995.[6] They observed the volume contraction in bulk silicon crystals on excitation with femtosecond visible optical pulses. The surface deformation and the generated acoustic strain were detected by measuring the reflectivity changes of probe beam. They suspected that a sign error or sample contamination might be responsible for the opposite polarity of the signals reported in Ref.[5]. Wright and Kawashima[7] in their work used an optical technique and observed directly the ultrafast vibrations of the surface of thin opaque films in the 0.1THz range in the time domain. Motivated by the success of photothermal deflection measurements of transient thermal expansion and surface waves in the MHz to GHz range,[5] they extended such measurements up to ∼0.2THz with polycrystalline films of the group-VI transition metals, molybdenum and chromium.

The pump beam induced change in the optical characteristics of the sample can cause a deflection signal from the photoexcited area without any surface motion. Such an optical contribution to the deflection signal, or an optical deflection signal, was observed by probing the photoexcited surface,[5] and it is determined by the transverse gradient in the phase of the pump beam induced reflectivity. In Ref.[8] laser picosecond acoustic measurements were carried out for the amorphous sputtered thin films of *a*-Ge and

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a-InSb on sapphire substrates using the pump–probe technique. They extended this technique to a geometry in which the pump beam was incident on the substrate side of a thin film deposited on a transparent substrate. This allowed background signals from non-acoustic sources to be greatly reduced. The resolution of measurement of small change in reflectance was $\delta R/R \approx 10^{-7}$rms. Back surface probing enables us to measure a pure sound-induced deflection signal, or an acoustical deflection signal. So the time-resolved pump–probe deflection technique (pump and probe beams are incident at the opposite surfaces of a sample) is a necessary method to study the shape and other important parameters of ultrafast photoexcited acoustic pulses. In previous work[9] we developed a picosecond deflection technique and observed coherent sub-nanosecond acoustic pulses in a germanium single crystal. The picosecond system was based on the lock-in amplification technique with double modulation at high (f_0=6.2MHz) and low (f_1=800Hz) frequencies (the photodetection system operated at the fixed frequency f_0). Using femtosecond laser pulses, almost all of the reported experiments concerning generation and detection of ultrafast acoustic pulses were based on measurements of the reflection or transmission changes of the probe beams affected by the presence of the strain wave.[10]

In this paper we present a femtosecond photodeflection technique with high sensitivity, which combines the advantages of precise cw photodetection and femtosecond time resolution. The femtosecond deflection system is quite simple in comparison with our previous picosecond system and with much higher time resolution (probably 10^3 times). There has been no report of such experiments, which directly measured the shape of ultrafast acoustic pulses using a deflection technique with femtosecond time resolution when pump and probe beams are incident on the opposite surfaces of a sample. The shape of acoustic pulses was obtained in a thin Ge plate with femtosecond laser pump and probe beams at 830nm wavelength. The travelling of ultrafast acoustic pulse within the sample enables us to measure accurately the sample thickness. Moreover, the interaction of charge carriers with acoustic waves is a classical problem of solid-state physics. An interesting aspect of this problem concerns the regimes where the directional motion of carriers is transonic or supersonic.[11] Recently Wright and his collaborators[12] investigated the generation and detection of picosecond acoustic-phonon pulses in a thin slab of GaAs using ultrashort optical pulses. They compared the optical phase variation with a simple theory for ambipolar diffusion and reached a conclusion that carrier diffusion has a significant effect on the shape of the phonon pulses generated. The phonon pulse duration was measured to be ~25ps, four times longer than that expected from optical-absorption considerations alone, indicating that hot carriers penetrate more than 100nm into the sample during the phonon pulse generation process. In our femtosecond optoacoustic experiment the characteristic velocity of the plasma diffusion evaluated from this experiment exceeded the longitudinal sound velocity by a factor of 4.0, which confirmed the supersonic diffusion of carriers. In our picosecond measurement[9] the characteristic velocity of plasma diffusion exceeded the longitudinal sound velocity by a factor of 1.5.

Our photodeflection technique may have interesting application in acoustic microscopy. Using ultrashort acoustic pulses, it is possible to achieve much higher spatial resolution than it was in the case of traditional nanosecond optoacoustics.[13]

2. Experiment

Our femtosecond optoacoustic experiment was based on a conventional pump and probe setup using a Ti:sapphire laser system (Mira 900F, Coherent, USA) pumped by Verdi-10 (Coherent, USA) at 10W. The laser wavelength was centred at 830nm. The repetition rate of laser pulses is 76MHz and the pulse width is 120fs, with an average power of about 1.5W. The laser beam was split into a probe beam and a pump beam (Fig.1). The average power of pump beam was about 230mW, and that of probe beam was about 80mW. The pump beam passed through a delay line and was modulated by a mechanical chopper at 1200Hz and focused onto the front surface of the Ge plate to generate an acoustic pulse by a lens with a focal length of 10cm, while the probe beam was focused by a lens with a focal length of 6cm onto the rear surface of the sample. The diameters of the pump and probe beams were 100μm and 30μm, respectively. The acoustic pulse was photoexcited near the front surface of the sample and shortly afterwards the pulse arrived at the rear surface to cause a displacement and curvature of the surface. Reflected from the rear surface, the probe beam was deflected by an angle $\delta\theta$, which was then transformed by a lens collimating the probe beam into its transverse displacement. The latter was measured with a position-sensitive detector

(PSD). Finally, the signal from PSD was transmitted to a lock-in amplifier (SR830, Stanford Research, USA), which was connected to a personal computer. To separate the optical and the acoustical deflection signals, we use the fact that a photogenerated acoustical disturbance will go away from the photoexcited volume with sound velocity. To avoid the optical contribution of pump beams to the deflection signal, it is necessary that the penetration depth of the photoexcitation should be less than the thickness of the sample. In our case an optically polished plate of undoped Ge with a thickness of $\sim 100\mu$m was used as the sample. The plate was oriented along [111] and had the form of a wedge with an angle 3°. The depth of photoexcitation was only about 0.25μm (interband absorption index $\alpha = 4\times 10^4 \mathrm{cm}^{-1}$ at wavelength 830nm). In our previous experiments with plane-parallel plates of Ge, the deflection signal had quite a complicated shape owing to the multiple reflections of the acoustical pulse at the front and back sample surfaces. To avoid such interference effects, in this work we used the samples in the form of a wedge.

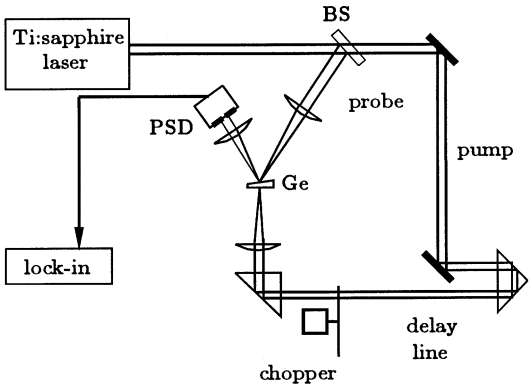


Fig.1. Schematic diagram of the experimental setup.

However, an optical contribution to the deflection signal from the back surface in general is possible via the photoelastic effect because the stress pulse can also induce a transverse gradient in the phase of the reflectivity. While, under our experimental conditions the photoelastic contribution to the deflection signals is negligible. This follows from our test experiments in which the acoustically induced reflection signal from the rear surface was not observed.

In the pump–probe deflection measurement the deflection signal is a relative change of the PSD photocurrent $\delta i/i$, which is proportional to the surface displacement δu. The minimal detectable PSD signal depends on intensity and pointing stability of the probe, PSD noise,[14] etc. The ultimate sensitivity of $\delta i/i$ is determined by the noise of the PSD photocurrent. We have achieved this noise-limited sensitivity with a stable and intense femtosecond laser by using a lock-in detection technique and obtained $\delta i/i \approx 1\times 10^{-6}$.

3. Results and discussion

In this section we discuss the experimental results and simulations of the acoustic pulse photoexcited in Ge. In most of the optoacoustic experiments,[1–4] sound wave was generated in metals or semiconductors by photoexcited thermal expansion of the sample. Besides the thermal mechanism, there exist nonthermal mechanisms of sound photoexcitation in semiconductors. In non-piezoelectric semiconductors the primary way of influence of the photoexcited free carriers on the lattice is via electron–phonon coupling, which can be characterized by deformation potential. For the typical material Ge or Si, the acoustic wave excited via the electronic mechanism is larger than via the thermal mechanism. The pump laser pulse absorbed in a semiconductor generates a temperature and also free carriers moving in the space and time. We describe both thermal $T(z,t)$ and carrier density $N(z,t)$ plane waves using one-dimensional approximation. The pump photon with energy $\hbar\omega_{\mathrm{L}}$ creates a free electron–hole pair and its excess energy $(1-E_{\mathrm{g}}/\hbar\omega_{\mathrm{L}})$ transfers to the lattice immediately at a time resolution of $\sim$0.1 ns. We can find $T(z,t)$ and $N(z,t)$ from the diffusion equations:[13]

$$\frac{\partial T}{\partial t} = \chi\frac{\partial^2 T}{\partial z^2} + (1-E_{\mathrm{g}}/\hbar\omega_{\mathrm{L}})\frac{(1-R)If(t)\alpha\exp(-\alpha z)}{C}, \tag{1}$$

$$\frac{\partial N}{\partial t} = D\frac{\partial^2 N}{\partial z^2} + \frac{(1-R)If(t)\alpha\exp(-\alpha z)}{\hbar\omega_{\mathrm{L}}}, \tag{2}$$

where χ and D are the thermal and ambipolar diffusivity respectively; R is the reflectivity of the pump pulse with peak intensity I and envelope $f(t)$; α the interband absorption index at ω_{L}; C the specific heat and E_{g} the band gap. In Eqs.(1) and (2) we neglect the carrier recombination due to the fact that for our case of Ge, there exists no bimolecular recombination and in the time scale of interest ($\sim$1ns) linear volume recombination can be neglected. In our case at the photoinduced free carrier density ($\sim 10^{18}\mathrm{cm}^{-3}$) and temperature increase (<1K), the linear model is justified and thus Auger recombination is insignificant in the time scale[15,16] and other parameters D, α, etc are assumed to be constants.

The electronic mechanism of sound photoexcitation should dominate over the thermal mechanism in Ge.[13] It is noted that this is true in the case of other semiconductors for photoexcitation by an ultrashort laser pulse.[6,13] In the case of sound excitation via the electronic mechanism the spectrum of displacement is[13]

$$\tilde{u}_{\mathrm{e}}(\omega) = -\frac{(1-R)d}{\hbar\omega_{\mathrm{L}}\rho c_{\mathrm{s}}^2} \times \frac{M\omega_{\mathrm{D}}}{\omega_{\mathrm{D}} + \mathrm{i}\omega} \left[\frac{\omega_{\mathrm{D}}}{\omega^2 + M^2\omega_{\mathrm{D}}^2} + \frac{1}{(\sqrt{-\mathrm{i}\omega} + \sqrt{\omega_{\mathrm{s}}})(M\sqrt{\omega_{\mathrm{D}}} + \sqrt{-\mathrm{i}\omega})}\right] I\tilde{f}(\omega), \tag{3}$$

where $\omega_{\mathrm{D}} = c_{\mathrm{s}}^2/D$ is the characteristic frequency at which the wavevectors of acoustical and carrier density waves are equal, $M = \alpha D/c_{\mathrm{s}}$ is the ratio of the sound transit time to the time of free carrier diffusion through the pump absorption region, and $\omega_{\mathrm{s}} = S^2/D$ is the characteristic frequency at which the velocity of the carrier density wave and the surface recombination rate are equal. Here we review the effects of sound propagation on the shape of photoexcited acoustic pulse. The Gaussian distribution of the pump beam intensity at the photoexcited surface (z=0) with radius a induces a corresponding distribution of mechanical displacement $u(r) = u(0)\exp(-r^2/a^2)$. For an acoustic signal with initial spectrum $\tilde{u}(\omega, r, 0)$ propagating in an isotropic medium, we have the spectrum at the distance z in the quasioptical approximation

$$\tilde{u}(\omega, r, z) = \frac{\tilde{u}(\omega, r, 0)}{1 + \mathrm{i}z/z_{\mathrm{d}}} \exp\left\{-\frac{r^2}{a^2(1 + \mathrm{i}z/z_{\mathrm{d}})}\right\}, \tag{4}$$

where $z_{\mathrm{d}} = \omega a^2/2c_{\mathrm{s}}$ is the characteristic diffraction length for each spectral component of the signal.

We can calculate the model pulses by taking into consideration the propagation effects (absorption and diffraction) from the spectrum of displacement [Eq.(3)] using inverse Fourier transformation.

Although the interference effects are suppressed by increasing the wedge angle, the acoustical pulse measured by the deflection method will be broadened for some wedge angles. This broadening appears because the acoustic pulses arrive at the probe spot in different time. The wedge angle $\gamma \approx 3°$ corresponds to the broadening which can be estimated as $\gamma d_{\mathrm{p}}/c_{\mathrm{s}} \approx$280ps, where d_{p} is the diameter of the probe beam and c_{s} is the sound velocity in Ge. To make it clear, we did the following simulation. We calculated three acoustic pulses using Eq.(3) with different moment appearance at the rear surface over the probe spot. Then we took the average of the three curves and compared it with the initial one. In Fig.2 the dotted line shows the average profile (with the wedge effect), the other three solid lines show the acoustic pulses which appear at the moment $t = -t_0$, t=0, $t = t_0$, respectively (t_0=80ps). Actually, such research was performed for the Ph.D. thesis of Pan.[17] It is shown that this effect does not have any significant influence on the shape of the acoustic pulse. Therefore, ignoring the wedge effect does not affect the calculation and the fitting procedure.

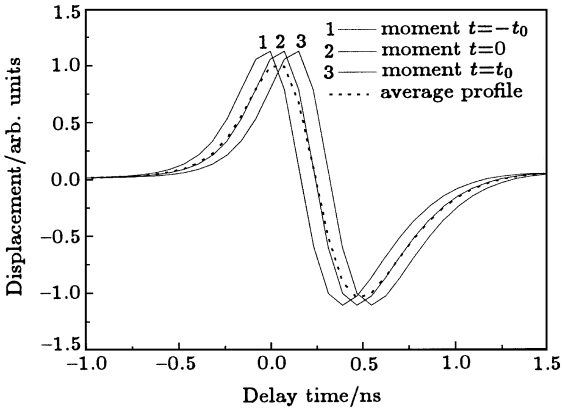


Fig.2. The influence of the wedge effect on the shape of the acoustic pulse.

The typical shape of simulated deflection signal is shown in Fig.3 (dotted line). The risetime of the acoustic pulse is estimated to be 230ps and the full width of the acoustic pulse at 1/e-maximum is about 480ps. For fitting the finite duration of both the pump and the probe laser pulses has been taken into account. The following parameters were selected for the theoretical calculation:[13,18] D=55cm^2/s, z=110μm. The theoretical shape of deflection signal is also shown in Fig.3 (solid line). The fitting of theoretical calculation to the experimental results leads to the conclusion that the dominant mechanism of photoexcitation acoustic pulses is electronic mechanism. The fitting ambipolar diffusivity $D \approx 55 \pm 10$cm^2/s is consistent with known data D=65cm^2/s for Ge.[13] We found that the sound absorption index is proportional to the square of the frequency $\alpha_{\mathrm{s}} \approx \beta f^2$, where $\beta \approx 3.5\mathrm{cm}^{-1}\cdot\mathrm{GHz}^{-2}$. And the estimated attenuation is in good agreement with the known data for Ge.[18] It has been discussed in our previous picosecond optoacoustic experiment[9] that absorption attenuates the high-frequency components, while diffraction suppresses the low-frequency ones. More details can be found in Ref.[17].

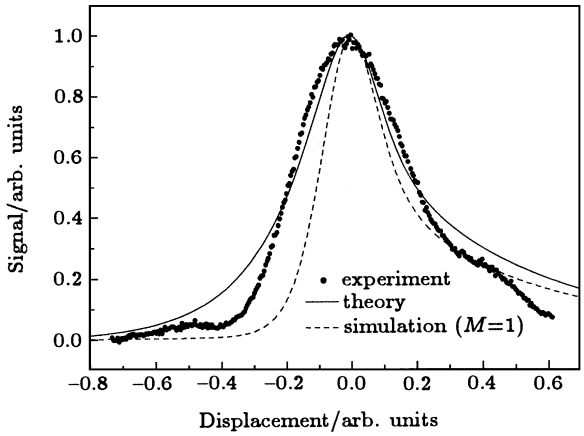


Fig.3. Deflection signal of acoustic pulse (propagation length z=110μm) and theoretical calculation.

Parameter D is the coefficient of ambipolar diffusion. In our experiments it is determined by the diffusion coefficients for electron (D_e) and hole (D_h) in undoped crystalline Ge:

$$D = \frac{D_e D_h}{D_e + D_h}. \tag{5}$$

The characteristic diffusion depth ($\sqrt{D\tau_a}$ $\approx$2.4μm) of the electrons within the phonon generation time (τ_a $\approx$1ns) when D=55cm^2/s.

One progress of the femtosecond deflection experiments is that we can use the quasioptical approximation to explain the observed acoustic pulses. In picosecond experiments[9] the observed pulse at z=110μm had a bipolar shape with a short negative phase of high amplitude. Therefore, for short propagation lengths only the shape of the front can be well reproduced by the model. We associate this difference with an appreciable contribution of low frequencies to the observed pulse shapes at $\sim$100μm that was not properly taken into account in the quasioptical approximation. The criterion for use of the quasioptical approximation is $\lambda/d \ll 1$, where d is the diameter of the acoustic wave and λ is the wavelength. We have $\lambda/d \approx 1.27$mm this case, while it was about 1.27mm our previous picosecond experiment. In addition, we can estimate the magnitude of the surface displacement μ $\approx$3pm when the typical peak-to-peak photocurrent change is $\delta i/i \approx 1.5\times10^{-5}$, whereas μ is estimated to be 10pm from Eq.(3).

In the case of instantaneous photoexcitation of the sample, whose thickness exceeds the light absorption depth of exciting laser pulse, the time of the acoustic pulse arrival at the rear surface of the crystal depends on the plasma front velocity.[9] If the latter is supersonic, the acoustic pulse associated with the moving front arrives at the rear surface earlier than that directly generated near the photoexcited surface. Parameter D=55cm^2/s corresponds to a Mach number $M \equiv V_D/c_s \approx 4.0$, where $V_D = \alpha D$ is the diffusional velocity of photoexcited electron–hole plasma.[9] From the analysis we also found that the parameter D determines the risetime of the acoustic pulses. In Fig.3 the dashed line shows the model pulse front in the case of sonic diffusion (M=1). In our experiments the photoexcited electron–hole plasma diffused with supersonic velocities V_D. Moreover, a series of acoustic signals with different powers of pump beams were experimentally investigated. The amplitude of acoustic signals was proven to depend linearly on the power of pump beams. This is in agreement with Eq.(3).

Our technique provides a highly precise method to measure the thickness of very thin film or the thickness difference of samples. In Fig.4 the two acoustic pulses were obtained with the propagation length z=114.5 and 114 μm. The acoustic pulse arrived at the rear surface at different moments when the thickness of sample changed. The corresponding difference of delay time $\Delta T = \Delta z/c_s$= 0.089ns, and the experimental difference of delay time is 0.095ns. The estimated value is in good agreement with the experimental result. Scanning the surface of the sample, we can detect such a small difference of thickness. By this highly precise technique we can perform a contactless measurement of the sample thickness with small spatial resolution, or even smaller when a shorter laser pulse is applied.

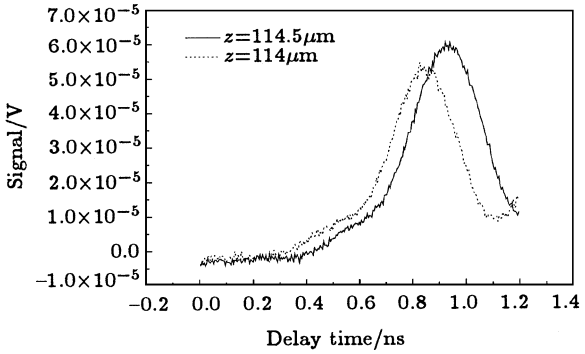


Fig.4. Precise measurement of the thickness of sample.

4. Conclusion

In summary, we have presented the experimental results concerning the photodeflection measurement

on the shape of ultrafast acoustic pulses with femtosecond time resolution. The generation of acoustic pulses by using femtosecond laser was explained with a theory of the excitation and relaxation of low-density electron–hole plasma. Supersonic expansion of photoexcited electron–hole plasma was observed. The characteristic velocity of plasma diffusion evaluated from the experiments exceeded the longitudinal sound velocity in germanium by a factor of 4.0.

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