Coherent phonon emission in the supersonic expansion of photoexcited electron-hole plasma in Ge

N. V. Chigarev, D. Yu. Paraschuk, and X. Y. Pan
Physics Department, Moscow State University, Moscow 119899, Russia

V. E. Gusev
Laboratoire de Physique de l'Etat Condensé, UPRESA-CNRS 6087, ENSIM, Université du Maine, av. O. Messiaen, 72085 Le Mans, France

(Received 23 February 2000)

Coherent subnanosecond acoustic pulses generated by a laser-excited electron-hole plasma in a germanium single crystal are detected by a time-resolved pump-probe photodeflection technique. It is found that the front duration of these hypersound pulses is controlled by the time of plasma diffusion at supersonic velocities. The characteristic velocity of plasma diffusion evaluated from the experiments exceeds the longitudinal sound velocity in germanium by a factor of 1.5. The hypersound pulse shapes provide evidence for supersonic diffusion of the electron-hole plasma in a semiconductor at room temperature on a subnanosecond time scale.

The interaction of charge carriers with acoustic waves is a classical problem of solid-state physics. An interesting aspect of this problem in general (and of the acoustoelectric effect, in particular) concerns the regimes where the directional motion of carriers is transonic or supersonic.[1] In the latter case the acoustic waves can be amplified by carrier drift, and the process of phonon emission by carriers can become stimulated.[1] In both cases a strong inverse influence of the excited (or amplified) acoustic waves on the carrier motion is expected. Consequently, an important question to be answered is: Can the free carriers in a solid be accelerated to supersonic velocities, i.e., can they overcome the sound barrier? In the early 1980s this question was actively studied in relation to the motion of electron-hole (*e-h*) droplets in Ge at liquid-helium temperature.[2] To the best of our knowledge all the attempts to accelerate an *e-h* droplet to a supersonic velocity under an inhomogeneous stress have been unsuccessful.[2]

To evaluate the expansion velocity of laser-excited plasmas, optical methods based on the detection of photons emitted[3] or scattered[4] by the plasma have been applied. However, use of these techniques have not given an unambiguous answer to the above question about the sound barrier. Photoacoustic spectroscopy provides a method for plasma expansion velocity measurement that is sensitive to either subsonic or supersonic plasma motion, in either the hydrodynamic or diffusive regime. In fact, any transient inhomogeneous distribution of free carriers in space is a source of acoustic waves excited owing to the carrier-phonon interaction mediated by the deformation potential. For example, the hydrodynamic expansion of a photoexcited plasma in a crystal sample involves propagation of a well-defined plasma front from the laser-irradiated surface. In the case of an 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.[5] 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. In the diffusional regime of plasma motion the arrival time of the acoustic pulse at the rear surface is not well-defined because the plasma boundary is delocalized in space [Fig. 1(a)]. However, according to the theory of sound photoexcitation via the electron-phonon potential,[6] the supersonic plasma expansion can broaden the front of the coherent acoustic signal arriving at the rear surface compared to the width predicted by a simple thermoelastic sound excitation mechanism.

In this paper the observation of coherent acoustic pulses excited during the supersonic diffusion of photoexcited *e-h* plasma at room temperature in monocrystalline Ge is reported. We begin with estimates of key physical quantities in

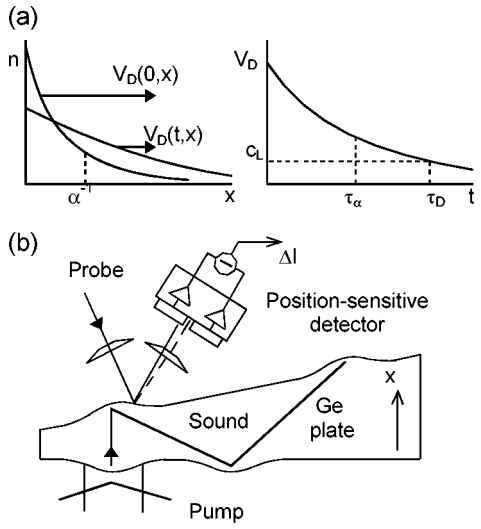

FIG. 1. (a) The *e-h* plasma density *n* and characteristic velocity V_D for the diffusional regime are plotted schematically versus the coordinate normal to the irradiated surface and the time after photoexcitation, respectively. (b) Diagram of the photodeflection technique with back surface probing.

this experiment. The equality of the diffusional flux of carriers to an equivalent hydrodynamic flux

$$-Dn_x = nV_D \tag{1}$$

is used as a definition of the diffusional velocity V_D,[7] where n is the free carrier density, D is the ambipolar diffusivity, x is the coordinate normal to the irradiated surface, and n_x denotes the derivative over x. Assuming an instantaneous photoexcitation for which plasma diffusion is negligible during the exciting pulse duration, we take $n \propto \exp(-\alpha x)$ in Eq. (1), where α is the light absorption index; that leads to $V_D = \alpha D$. Then the definition $V_D > C_L$ of the supersonic regime, where C_L is the longitudinal sound velocity, can be rewritten in the terms of characteristic times $D/C_L^2 > (\alpha C_L)^{-1}$. The time $\tau_\alpha \equiv (\alpha C_L)^{-1}$ corresponds to sound propagation across the light absorption depth α^{-1}. The time $\tau_D \equiv D/C_L^2$ can be interpreted as the characteristic time of plasma deceleration down to the sound velocity [Fig. 1(a)]. In fact in the case of instantaneous plasma generation at the surface the depth of plasma penetration is estimated as $(Dt)^{1/2}$ and the velocity as $V_D \simeq (D/t)^{1/2}$. Thus, $V_D \simeq C_L$ when $t \simeq \tau_D$.

The diffusing e-h plasma excites acoustic waves in which the mechanical displacement U can be found as the solution of the problem[6]

$$U_{tt} - C_L^2 U_{xx} = (d/\rho_0) n_x,$$

$$n_t - Dn_{xx} = (\alpha(1-R)I/h\nu_L)\exp(-\alpha x) f(t/\tau_L),$$

$$-C_L^2 U_x|_{x=0} = (d/\rho_0) n|_{x=0}, \quad Dn_x|_{x=0} = 0. \tag{2}$$

Here d is the e-h phonon deformation potential, ρ_0 is the equilibrium density, R is the optical reflectivity, $h\nu_L$ is the energy of the optical quantum, I is the laser intensity, and the function $f(t/\tau_L)$ describes the intensity envelope of a laser pulse of duration τ_L. The analytical solution of Eq. (2) for $\tau_L \to 0$ in the case of slow subsonic plasma diffusion ($\tau_D \ll \tau_\alpha$) gives an exponential rise of the acoustic pulse front with rise time τ_α. In the case of fast supersonic diffusion ($\tau_D \gg \tau_\alpha$) the front of the acoustic pulse is also exponential but with rise time τ_D. For a finite τ_L the condition for observation of acoustic front broadening caused by supersonic plasma diffusion is $\tau_D > \tau_\alpha, \tau_L$.[6] For a room-temperature nondegenerate e-h plasma in Ge, $D = 65$ cm^2/s and $C_L = 5.6 \times 10^5$ cm/s in the [111] direction, we have $\tau_D \simeq 200$ ps.

To measure fast acoustic transients, we have applied a pump-probe photodeflection technique[8–10] in which the pump and probe are focused on the opposite surfaces of a thin Ge plate. The optically polished plates of undoped Ge oriented along [111] (or perpendicular to it) have the form of a wedge with angle $\beta = 3°$. The acoustic pulse generated by the e-h plasma photoexcited near the front surface arrives some time later at the rear surface and causes its displacement and curvature [Fig. 1(b)]. The acoustically induced deflection angle of the probe beam is measured with a position-sensitive detector. A mode-locked Nd:YAG laser (YAG denotes yttrium aluminum garnet) producing $\tau_L \simeq 120$ ps pulses at the wavelength $\lambda_1 = 1064$ nm with a 100 MHz repetition rate was used as the optical source. The pump (pulse energy $E_1 \leqslant 1$ nJ at λ_1, $\alpha_1 \simeq 1.4 \times 10^4$ cm^{-1}) and the probe ($E_2 \sim 0.03$ nJ at $\lambda_2 = \lambda_1/2 = 532$ nm, $\alpha_2 \simeq 5.4 \times 10^5$ cm^{-1}) beams were focused to spots of area $S_1 \simeq (50\ \mu\text{m})^2$ and $S_2 \simeq (15\ \mu\text{m})^2$, respectively. To measure the signal, a high-frequency lock-in technique with double modulation was used.[11] The pump was modulated at 6.2 MHz by an electro-optical modulator and at 800 Hz by a chopper.

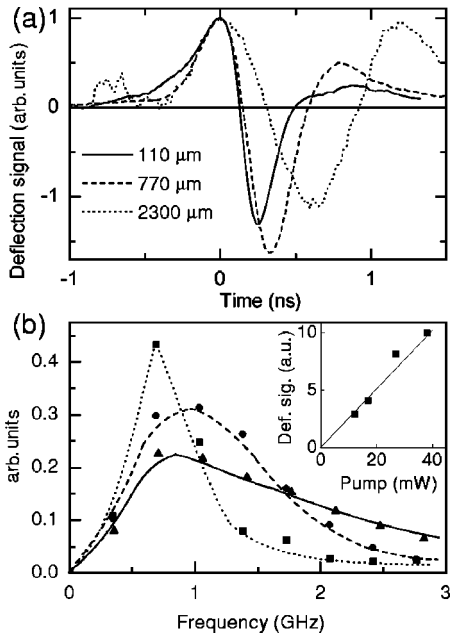

FIG. 2. (a) Normalized deflection signals at different propagation lengths for the pump at λ_1, and (b) their spectra normalized to the corresponding acoustic pulse energies (lines are guides to the eye). The inset shows the deflection signal versus average pump power.

Typical deflection signals and their spectra at different propagation paths of the acoustic pulse are shown in Fig. 2.[12] The propagation path lengths were calculated from the corresponding delays of the deflection signals. At 110 μm the hypersound pulse has nearly a bipolar shape, which transforms into a one with two maxima at longer path lengths 770 and 2300 μm [Fig. 2(a)]. The front of the hypersound pulses depends weakly on the propagation length and has a $1/e$ rise time 250 ± 50 ps.[14] The spectrum of the hypersound pulses is concentrated near 1 GHz as seen in Fig. 2(b). The maximum peak-to-peak photocurrent change was $\delta i/i \simeq 4 \times 10^{-5}$, which corresponds to a surface displacement $U \simeq 5$ pm.

Propagation effects noticeably change the pulse shapes as seen in Fig. 2(a). Diffraction and frequency-dependent attenuation control the shape of the propagating hypersound pulse. This is illustrated by the spectra in Fig. 2(b): while absorption attenuates the high-frequency components, diffraction suppresses the low-frequency ones. Consequently, diffraction results in differentiation of the initial pulse profile at $x = 0$, while attenuation leads to its broadening[15] [Fig. 2(a)]. Attenuation was calculated from the analysis of the high-frequency tails (>1 GHz) of the 770 and 2300 μm spectra in Fig. 2(b).[13] The sound absorption index was found to be proportional to the square of the frequency $\alpha_S \simeq \gamma f^2$, where $\gamma \simeq 3.0$ cm^{-1} GHz^{-2}. The calculated attenuation is in good agreement with the known data for Ge.[16] However, the

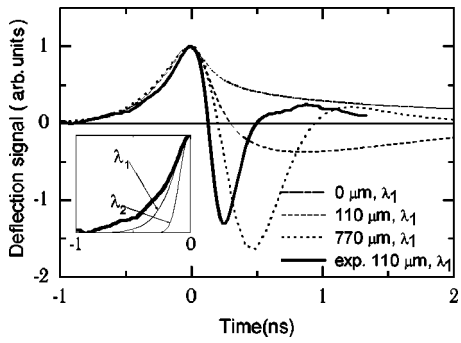

FIG. 3. Calculated pulse shapes for different propagation lengths and pump at λ_1 (broken lines, $D=60$ cm^2/s). The thick solid line depicts the measured pulse from Fig. 2(a); the inset also shows the model pulse fronts in the case of subsonic diffusion ($M=0.5$, $x=110$ μm) for the pump at λ_1 ($D=20$ cm^2/s) and λ_2 ($D=0.5$ cm^2/s).

attenuation is too small at propagation length $\lesssim 800$ μm to explain the observed broadening of the pulse front. Note that the phonon focusing effect[17] related to the elastic anisotropy of Ge can be neglected in our experiment.[13]

Our first conclusion is that the sound excitation by the thermoelastic mechanism is negligible in our experiment. This is predicted by the theory[6,15] and can be deduced from the following observation. We obtained nearly identical pulse shapes for pumping at wavelengths λ_1 and $\lambda_2=\lambda_1/2$. Despite the strong difference in the light absorption depths ($\alpha_1^{-1} \gg \alpha_2^{-1}$) the penetration depths of the plasma for both λ_1 and λ_2 in the time τ_L should be similar due to fast carrier diffusion. In contrast, the heated depths differ markedly. For the thermoelastic and deformational potential mechanisms of sound excitation, the acoustic pulse shape depends strongly on the penetration depth of, respectively, the lattice temperature change and the photoexcited plasma.[6,15] The similar pulse shapes measured for both pump wavelengths imply similar acoustic sources and spatial localization. Thus, we conclude that the deformation potential mechanism dominates in the hypersound excitation. The pulse shape did not depend on the pump intensity, and the deflection signal increased linearly with it as shown in Fig. 2(b), inset. This confirms that nonlinear Auger recombination is negligible on our time scales ($\sim \tau_D$) for a plasma density estimated as $n \lesssim 4\times 10^{18}$ cm^{-3}. From the experiments with chemically treated Ge surfaces[13] we conclude that surface recombination is negligible.

In order to compare the shapes of observed hypersound pulses with the model given by Eq. (2), we used its analytical solution in spectral form,[6,15] simulating the effects of sound diffraction and absorption in the hypersound spectrum. Diffraction was taken into account in the quasioptical approximation. The deflection signal was simulated as a maximum slope in the displacement profile of the rear surface of the sample. Then, using an inverse Fourier transformation, we obtained the modeled pulses (Fig. 3). The finite duration of both the pump and the probe laser pulses and the broadening caused by the slope of the wedge surfaces[14] were taken into account. As seen in Fig. 3, the main effect of propagation is the appearance of a negative phase in the signal due to sound diffraction. The pulse fronts in Fig. 3 are not sensitive to diffraction, in agreement with our experiment (Fig. 2). The measured pulse front is well reproduced by the model for the diffusivity of the *e-h* plasma $D\simeq 60\pm 10$ cm^2/s, which is consistent with known data.[18] The best correlation of the overall measured pulse shapes with the model ones was obtained for propagation lengths >500 μm. For short propagation lengths ~ 100 μm the model pulse shape has a long negative phase of small amplitude (Fig. 3) because of a relatively small diffraction effect. However, the observed pulse at $x\simeq 110$ μm has a bipolar shape with a short negative phase of high amplitude. Therefore, for short propagation lengths only the shape of the front is well reproduced by the model (Fig. 3). We associate this difference with an appreciable contribution of low frequencies ($f\sim C_L/\sqrt{S_1}$) to the observed pulse shapes at $x\sim 100$ μm that is not properly taken into account in the quasioptical approximation. For long propagation lengths $\gg 100$ μm these low frequencies are suppressed by diffraction and agreement of the model pulses (Fig. 3) with the measured ones (Fig. 2) is better. Thus, we conclude that the broadening of the pulse front is a result of hypersound excitation and is not due to its propagation.

As follows from the fits in Fig. 3, the observed broadening of the hypersound pulse front is caused by supersonic diffusion of the photoexcited *e-h* plasma. The difference between pulse fronts associated with supersonic and subsonic diffusion for both pump wavelengths is shown in Fig. 3, inset. It takes $\tau_D\simeq 200$ ps for the plasma to decelerate down to the sound velocity C_L from the initial diffusional velocity $V_D\simeq \alpha D$ [Fig. 1(a)], which for the pump at $\lambda_1=1064$ nm corresponds to a Mach number $M\equiv V_D/C_L\simeq 1.5$. This supersonic diffusion is possible because the inverse influence of emitted acoustic waves on the plasma expansion in our experiments is weak. The pressure of the acoustic field on the plasma can be estimated from our experimental data as $|d|nU_x\sim |d|nU/(C_L\tau_D)$ using the value $d\simeq 7$ eV for Ge.[2] This pressure is negligible in comparison with the internal pressure of the nondegenerate *e-h* plasma $\sim nk_BT$, where k_B is the Boltzmann constant and T is the absolute temperature.

To estimate the minimum time (τ_m) for the development of the phonon instability, that is for reaching the threshold of stimulated emission of acoustic phonons, we modeled the expanding plasma by a spatially homogeneous plasma layer moving at a constant velocity $V_D=\alpha_1 D$ and neglected scattering and interaction of the emitted phonons. From the theoretical formulas[1] for a nondegenerate *e-h* plasma in Ge we have τ_m [ns] $\gtrsim (M-1)^{-1}(n[10^{18}$ cm$^{-3}])^{-1}$. Even for the peak plasma density 4×10^{18} cm^{-3} achievable in our experiment τ_m significantly exceeds the characteristic time τ_D. Thus, this process of stimulated phonon emission[19] does not prevent the supersonic expansion of the e-h plasma in our experiment.

In summary, we have used picosecond ultrasonics to observe the supersonic expansion of the photoexcited *e-h* plasma in monocrystalline Ge. Our experiments have shown that the photoexcited plasma diffuses at a supersonic velocity at room temperature during the characteristic time $\tau_D = D/C_L^2\simeq 200$ ps. This time corresponds to plasma deceleration down to the sonic velocity and is evaluated from the

profiles of the hypersound pulses excited by the expanding plasma. Our experiment proves also that supersonic diffusion of *e-h* plasma can prevent shortening of laser excited coherent acoustic pulses.[6]

Stimulating discussions with Professor O.B. Wright and his advice for the improvement of our experimental setup are gratefully acknowledged. This work was supported by INTAS, Project No. INTAS-97-OPEN-31680.

[1] J. W. Tucker and V. W. Rampton, *Microwave Ultrasonics in Solid State Physics* (North-Holland, Amsterdam, 1972).
[2] M. A. Tamor, M. Greenstein, and J. P. Wolfe, Phys. Rev. B **27**, 7353 (1983); I. V. Kukushkin and V. D. Kulakovskii, Fiz. Tverd. Tela **25**, 2360 (1983) [Sov. Phys. Solid State **25**, 1355 (1983)].
[3] A. Forchel, H. Schweizer, and G. Mahler, Phys. Rev. Lett. **51**, 501 (1983).
[4] K. T. Tsen and O. F. Sankey, Phys. Rev. B **37**, 4321 (1988).
[5] V. E. Gusev, Akust. Zh. **33**, 863 (1987) [Sov. Phys. Acoust. **33**, 501 (1987)].
[6] C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, Phys. Rev. B **34**, 4129 (1986); S. A. Akhmanov and V. E. Gusev, Usp. Fiz. Nauk **162**, 3 (1992) [Sov. Phys. Usp. **35**, 153 (1992)].
[7] G. Mahler and A. Fourikis, J. Lumin. **30**, 18 (1985).
[8] M. A. Olmstead, N. M. Amet, S. Kohmm, A. Fournier, and A. C. Boccara, Appl. Phys. A: Solids Surf. **32**, 141 (1983); M. Sontag and A. C. Tam, IEEE UFFC **33**, 500 (1986).
[9] J. E. Rothenberg, Opt. Lett. **13**, 713 (1988).
[10] O. B. Wright and K. Kawashima, Phys. Rev. Lett. **69**, 1668 (1992).
[11] D. Yu. Paraschuk, T. A. Kulakov, O. Yu. Nedosekin, N. V. Chigarev, and X. Y. Pan, Rev. Sci. Instrum. **68**, 3989 (1997).
[12] The photoelastic contribution to the deflection signals was negligible. This follows from test experiments (Ref. 13) in which the acoustically induced reflection signal from the rear surface was not observed.
[13] N. V. Chigarev, Ph.D. thesis, Moscow State University, 1998.
[14] The arrival time of the acoustic pulses at the rear surface of the wedge is broadened by $\beta\sqrt{S_2}/C_L \simeq 130$ ps.
[15] V. E. Gusev and A. A. Karabutov, *Laser Optoacoustics* (AIP, New York, 1993).
[16] D. W. Oliver and G. A. Slack, J. Appl. Phys. **37**, 1542 (1966).
[17] G. A. Northrop and J. P. Wolfe, Phys. Rev. B **22**, 6196 (1980).
[18] D. H. Auston and C. V. Shank, Phys. Rev. Lett. **32**, 1120 (1974); M. I. Gallant and H. M. van Driel, Phys. Rev. B **26**, 2133 (1982).
[19] V. E. Gusev, Fiz. Tverd. Tela **31**, 97 (1989) [Sov. Phys. Solid State **31**, 774 (1989)].