Quasidiffusion and the Localized Phonon Source in Photoexcited Si

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Previous observations of heat pulses produced by localized photoexcitation of silicon do not support the predictions of phonon "quasidiffusion" via anharmonic decay and elastic scattering. Our experiments, with controlled boundary conditions, verify that quasidiffusive theory is relevant in Si under very weak photoexcitation. Beyond this domain a transition to a localized source of low frequency phonons is attributed to excited carrier interactions. Photoluminescence experiments confirm the presence of electron-hole droplets coincident with this localized phonon source.

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In a perfectly harmonic lattice, vibrational waves or phonons of a given frequency are constrained to remain at that frequency. Anharmonicity of the lattice allows a phonon to split into lower frequency phonons, conserving energy and momentum. This down-conversion process (and the corresponding up-conversion) makes the propagation of thermal energy in a solid quite different from that of particles in a gas. If anharmonic decay—a.k.a., inelastic scattering—is combined with a more rapid elastic scattering of phonons from defects in the lattice (such as that due to impurities or isotopic randomness of atoms), then the motion of nonequilibrium phonons from a point source in an otherwise cold crystal is by "quasidiffusive" expansion [1]. In this case, whenever an acoustic phonon undergoes anharmonic decay to a lower frequency ($\tau_a^{-1} \propto \nu^5$), a large increase in the effective diffusion constant (typically a factor of 2^4) occurs due to the rapid frequency dependence of the elastic scattering rate ($\tau_e^{-1} \propto \nu^4$) [2].

It is predicted [1,3] that quasidiffusion produces a broad distribution in the arrival times of acoustic phonons crossing a crystal. Surprisingly, localized photoexcitation of a Si crystal immersed in a liquid-helium bath produces temporally sharp heat pulses and sharp phonon-focusing caustics [4], characteristic of low frequency (subterahertz) phonons traveling ballistically from the excitation region to the detector. Since the phonons produced by the thermalization of photoexcited carriers should be rich in frequencies above 1 THz, it appears that either the anharmonic decay rate is much faster than calculated or some other process is bypassing this relaxation mechanism.

In search of the predicted quasidiffusive behavior, we have (1) removed the contact with liquid helium, and (2) greatly reduced the excitation density. Under these conditions a broad temporal distribution of phonons is indeed observed. Figure 1(a) shows the distribution of phonons traversing a 5.5 mm thick [100]-oriented Si crystal following defocused excitation with a 10 ns Ar^+ laser pulse. The heat pulse is detected with a granular aluminum bolometer on the face opposite the excitation, as depicted in the inset of Fig. 1(a). Except for the excitation surface, which is in vacuum, the sample is immersed in liquid He at 1.7 K. The experimental heat pulse displays a sharp onset at the ballistic arrival time of transverse phonons and a long tail which decays approximately ex-

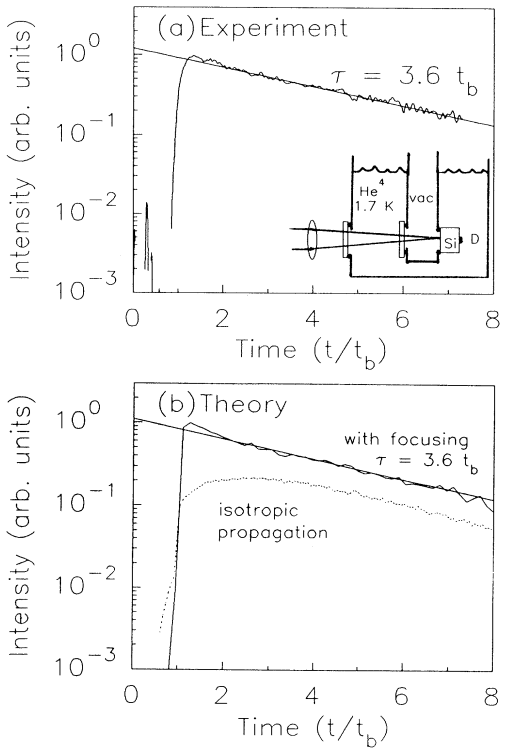

FIG. 1. (a) Intensity of the phonon signal vs time along the [100] propagation direction in photoexcited Si of 5.5 mm thickness. The intensity of longitudinal phonons in this propagation direction is small due to phonon focusing. The inset shows the experimental configuration used to isolate the excitation surface from the liquid He bath. (b) Monte Carlo simulation of the phonon signal vs time for two different models described in the text. Both models include three phonon branches; the solid curve includes phonon focusing.

ponentially with a decay constant of $\tau = (3.6 \pm 0.3) t_b$, where t_b is the ballistic time of flight of transverse phonons in the [100] direction (for this 5.5 mm sample $t_b = 950$ ns). Removing the contact to the liquid helium is critical. Otherwise, high frequency phonons scattering diffusively near the excitation point are lost into the bath when they encounter the surface, and the remaining lower frequency phonons ($\nu < 1$ THz) travel ballistically away from the source.

The applicability of a quasidiffusive model is seen by comparing the experimental heat pulses with the Monte Carlo simulation in Fig. 1(b) (solid line). This calculation of the heat pulse resulting from injecting a nonequilibrium phonon distribution assumes the spontaneous phonon decay for silicon calculated analytically from nonlinear elasticity theory [5] (this isotropic approximation does not discriminate two transverse modes). In this theory only longitudinal phonons (L) can split into two lower energy phonons, although transverse (T) phonons can convert to L phonons by elastic scattering. (The L phonon anharmonic decay probability [2] $\tau_a^{-1} = 1.2 \times 10^{-55} [\nu/(1\ \text{Hz})]^5\ \text{s}^{-1}$, deduced from the third-order elastic constants at low temperatures, is employed.) Both the frequency and angular distributions of the decay products are determined kinematically from the energy and momentum conservation. The simulation partitions slow transverse and fast transverse phonons by their density of states and includes the isotope scattering (with scattering rate [2] $\tau_e^{-1} = 2.43 \times 10^{-42} [\nu/(1\ \text{Hz})]^4\ \text{s}^{-1}$) and anisotropy through the phonon group velocity. The model successfully reproduces the features of the experimental time trace, including the shape of the ballistic onset, which is sensitive to the anisotropy of the phonon propagation, the assumed scattering rates and the details of the allowed anharmonic processes. Because of this unique sensitivity, modeling of the quasidiffusive propagation can give insight into anharmonic decay processes which are not directly measurable by other means.

The two time traces in Fig. 1(b) illustrate the importance of the crystalline anisotropy governing the phonon propagation, which has been neglected in previous simulations. The calculation which assumes degenerate T modes and isotropy in phonon propagation (dashed curve) reproduces the decay of the quasidiffusive tail satisfactorily but the peak intensity occurs at $2.6 t_b$. The more realistic model (solid curve), including the anisotropy of the phonon propagation (i.e., noncollinearity of wave vector and group velocity directions and the resulting phonon-focusing effect) but retaining isotropic decay gives a sharp onset of ballistic intensity followed by the monotonically decaying tail. The tail of the quasidiffusive pulse can be fitted to an exponential decay, with $\tau = 3.6 t_b$. Thus, the theory of quasidiffusion based upon the above assumptions gives a late-time decay constant in good accord with our experiment. Inclusion of anharmonic decay within the T branches would tend to lower the theoretical late-time decay constant τ.

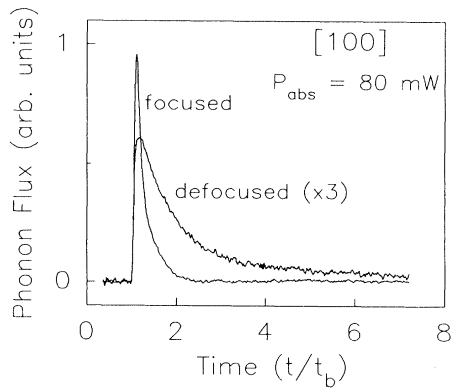

FIG. 2. Comparison of focused (power density ≈ 200 W/mm^2) and defocused (power density ≈ 2 W/mm^2) time traces. The power density is varied without changing the total power deposited in the sample by defocusing the laser spot. The spot size is estimated by measuring the sharpness of a target pattern at the sample position. The excitation area is $\pi/4$ times the full width at half maximum of the beam profile, which ranges from 20 μm to about 200 μm in the experiment of Fig. 4. The pulse broadening associated with the geometry of a larger spot size is negligible—less than 1%.

Eliminating losses into the He bath at the excitation surface is only one necessary precondition for the successful observation of quasidiffusion. The crystal must also be *very weakly* photoexcited. Figure 2 shows the remarkable changes in the heat pulses as we decrease the laser spot size at constant pulse energy, an effect discovered by Shields *et al.* [4,6]. A large increase in the intensity near the ballistic time is observed, together with a decrease in the tail signal. *Raising the excitation density appears to produce more low frequency ballistic phonons.* Consistent with this observation, phonon images obtained at tight focus show sharp caustics, which indicate that subterahertz phonons are being emitted from very near the excitation region. By subtracting a heat pulse slightly off a caustic direction (B) from one obtained along a caustic direction (A), one obtains the lifetime of the localized source of phonons. This "spatially filtered" trace [7] (A-B) isolates the pure ballistic phonons and yields a source lifetime of 160 ± 20 ns, as shown in Fig. 3 (see caption).

By continuously varying the laser spot size, we have obtained the peak intensity of the heat pulse as a function of excitation density. As seen by the solid dots in Fig. 4, a broad transition between "quasidiffusion" and "localized source" is centered about 20 W/mm^2. This excitation density is more than 3 orders of magnitude below the predicted threshold for formation of a phonon "hot spot" mediated by phonon-phonon interactions [4,8,9]. So we are faced with two questions: What is causing the phonon localization, and what has happened to the high frequency quasidiffusive phonons?

The answers appear to be associated with the nonequilibrium carriers (electron-hole pairs) which are produced by photoexcitation of silicon. At low carrier densi-

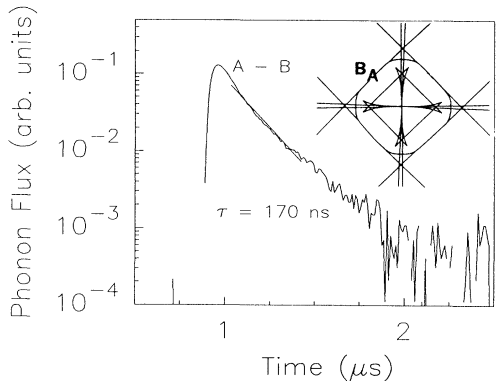

FIG. 3. Heat pulse for phonons traveling ballistically from the phonon source to the detector, obtained by subtracting time traces off (B) and on (A) a phonon-caustic direction. The inset shows the caustic pattern around the [100] direction (Ref. [4]). Accounting for a detector response of 65 ns gives an estimate of the localized source lifetime, $\tau = 160 \pm 20$ ns. This lifetime is obtained using a 20 μm (FWHM) laser spot size, over a range of incident total powers (500 mW > P > 30 mW).

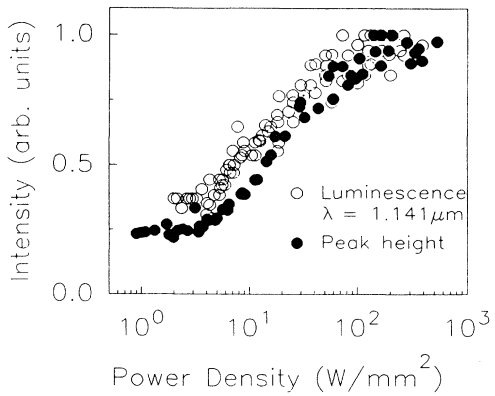

FIG. 4. Solid dots are the intensity of near-ballistic phonons arriving in the interval $t_b < 1 < 1.05t_b$, as a function of excitation-power density, obtained by decreasing the laser spot size at constant excitation power. The detected phonon signal is almost entirely ballistic at high power densities and quasidiffusive at low power densities. There is a broad transition to a low frequency phonon source around a power density of 20 W/mm^2. Open circles show the detected photoluminescence from carrier recombination in electron-hole droplets (EHD) as a function of power density, normalized at the highest power density. The rise in luminescence intensity at about 20 W/mm^2 coincides with the phonon-signal transition, providing a clear link between the EHD and the ballistic phonons. Both the phonon and the luminescence transitions are dependent upon power density rather than total power deposited in the sample. The spectra at low power displays a stronger signal from free excitons ($\lambda = 1.128$ μm) and a background due to excitons bound to impurities over a broader range including 1.141 μm. Spectral resolution = 3.5 nm.

ties hot-carrier relaxation occurs within a picosecond by emission of high frequency optical phonons. These optical phonons down-convert within a few picoseconds and provide the high frequency acoustic phonons which initiate the quasidiffusive process. However, a hot carrier created within a colder electron-hole plasma can lose its energy by either optical phonon emission or inelastic collisions with other carriers [10]. The latter process would slightly heat up the plasma as a whole, which then would emit low frequency acoustic phonons.

It is well known [11] that at a sufficient photoexcitation density of Si, electron-hole (e-h) pairs condense into droplets of electron-hole liquid, with a measured pair density of 3.3×10^{18}/cm^3. A calculation of the carrier kinetics at this density, described elsewhere [8], shows that for a small droplet ($R = 0.1$ μm) 50% of the excited carrier energy can be transmitted to other carriers in the liquid by carrier-carrier scattering. This transfer of energy to the e-h plasma as a whole raises the temperature of the droplet to about 6 K. Such a temperature would make the droplets a rich source of low frequency (subterahertz) acoustic phonons.

To test this idea, we have made photoluminescence experiments under the same conditions as the phonon experiments. We have observed the characteristic recombination luminescence emitted by electron-hole droplets and the result is shown as the open circles in Fig. 4. There is an onset of electron-hole droplet luminescence at about the same power density as the threshold for the localized phonon source.

Several previous measurements are relevant to this problem. Most important, the lifetime of electron-hole droplets in Si is measured to be 140 ns, which is in reasonable agreement with that observed for the localized phonon source (Fig. 3). The recombination within a droplet is predominantly via a nonradiative Auger process [12], providing a source of hot carriers within the colder droplets. In addition, the size and power dependence of the cloud of electron-hole droplets [13] is similar to that for the localized phonon source [4]. Based on this broad range of experimental evidence and theoretical support, we conclude that the electron-hole droplets are acting as the localized source of low frequency phonons in Si.

From this study we have learned that the frequency and spatial distribution of phonons emitted as a byproduct of carrier thermalization in semiconductors depends on boundary conditions, excitation density, and electronic properties of the semiconductor. Except for quasidiffusion observed at low excitation density, phonon distributions in other semiconductors are not likely to follow generic models because the electron-hole lifetimes and bound states vary widely. For example, GaAs is a direct-gap semiconductor having excitonic lifetimes of about 1 ns and no known electron-hole liquid. Ge, on the other hand, has an electron-hole liquid with a 40 μs lifetime and carrier interactions are expected to effect the phonon distribution. The very long recombination time in Ge

may help to disentangle the contributions of plasma heating due to nonradiative recombination and the initial thermalization processes. In both materials, the threshold for optical phonon emission is 1.5 times lower than in Si, so that a large part of the energy emitted by the carrier distribution should be in optical phonons. Finally, our photoexcitation experiments may impact the physics of phonon-based high-energy particle detectors [14], which involve much smaller deposited energies but possibly comparable local energy density. It will be interesting to view these and other situations in light of the results presented here.

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