Thermalization of nonequilibrium phonons in a-Si:H

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The transmission of heat pulses through a silicon single crystal has been studied. The pulses were produced by the optical excitation of the silicon or of an a–Si:film grown on its surface. Another effective mechanism for the thermalization of phonons has been found to occur in a film of amorphous silicon.

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Nearly the only way to obtain information on the kinetic properties of high-frequency (superthermal) phonons—the phonon–phonon interaction and the scattering by defects—is to study the relaxation and propagation of nonequilibrium phonons. Essentially no such data have been reported for the a–Si-H system.

In the present experiments we used the standard heat-pulse method in a transmission geometry (Fig. 1). The sample a silicon single crystal, was immersed in a bath of liquid helium at 1.8 K. The beam from a nitrogen laser was attenuated by filters and focused by a lens onto the front face of the sample. The size of the focal spot was varied by moving the lens in the axial direction. The energy incident on the sample ranged from 5×10^{-3} to 5 μJ, and the corresponding energy density range from 0.1 to 0.5 J/cm^2.

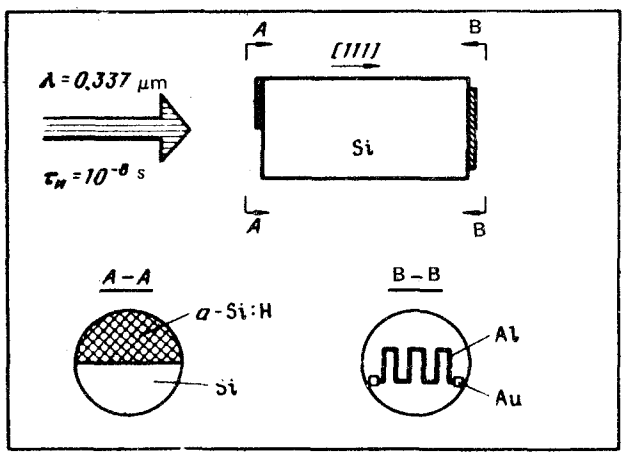

FIG. 1. Experimental arrangement. Si sample ($\rho\sim8$ k Ω cm, $L = 1$ cm, diameter = 0.6 cm). A-A—Front surface of the sample; hatched region—the a-Si-H film; near surface of the sample, with the deposited aluminum bolometer. The dimensions are 2×2 mm, and the thickness is 350 Å.

An a–Si-H film 0.6 μm thick was synthesized in a capacitive rf (5.28-MHz) glow discharge in a mixture of 4% monosilane and argon at a pressure of 0.1 Torr in the reaction vessel and at a substrate temperature of 280 °C. The electrical and optical properties of this film were similar to those customarily cited (see Ref. 1, for example): $\rho \sim 10^{10}$ Ω cm, a photoresponse $\sim 10^5$, an optical gap width of 1.88 eV (Ref. 2), and a hydrogen concentration ~ 16 at.%. The reflection coefficients for light at 0.337 μm of the a–Si:H and of c–Si were approximately the same according to our measurements (~ 0.32).

The phonon detector was a superconducting bolometer consisting of granulated aluminum, whose output signal was amplified, processed by a strobe-integration technique, and fed to an x, y chart recorder.

Figure 2 shows experimental curves for two values of the absorbed energy density. We can draw some conclusions from this figure.

1. At low energy densities the signals obtained during the absorption of the laser pulse in the a–Si-H film and in the c-Si have sharply different shapes (when the film is present, there are some additional narrow signals of a ballistic type; the tails of the signals nearly coincide).

2. At high energy densities the difference between the signals in the two cases is not as large.

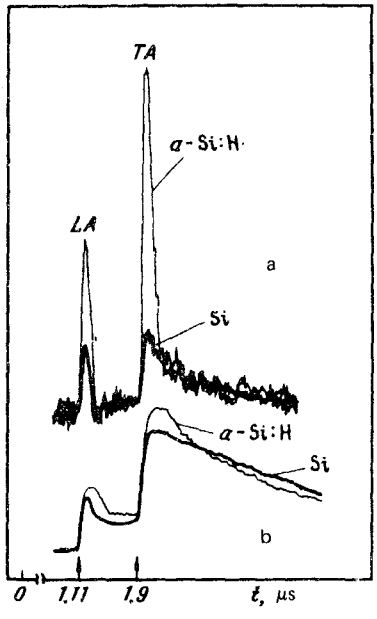

FIG. 2. Heat pulses which arise during the absorption of a laser beam in an a-Si:H film and at the surface of a silicon crystal, for two values of the absorbed energy density: a—2 μ J/cm^2; b—3×10^4 μ J/cm^2. The LA and TA peaks are the responses of the bolometer to the arrival of longitudinal and transverse acoustic phonons. The two signals in each part of the figure, a and b, were recorded at the same gain level.

3. The presence of signals of the ballistic type for the a–Si-H implies that the a–Si-H/c-Si interface is of good quality.

Nonequilibrium phonons are generated in the course of the relaxation of hot carriers, whose initial energy is $\varepsilon_0 \sim 1.2$ eV. The first step in the relaxation is the emission of high-frequency (acoutic and optical) phonons with an energy on the order of the Debye energy ($\hbar\omega_D \sim 56$ meV). After emitting about 20 such phonons, the carriers fall into the region $\varepsilon < \hbar\omega_D$, where the second step of the relaxation occurs as a result of an interaction with low-frequency acoustic phonons, with $\hbar\bar{\omega} \sim 4$ meV $= 40$ K.

Assuming that the characteristic time for the emission of a phonon $\hbar\omega_D$ is 1 ps, we find that the carriers diffuse to a depth $d \sim 1$ μm in the first step of the relaxation. The second step lasts about 10 ns; the corresponding diffusion depth is determined by the carrier lifetime. This ends the "electron" stage of the relaxation.

In the "phonon" stage of the relaxation, the absorbed energy density is very influential. The nonequilibrium phonon temperature, i.e., the Planckian distribution of phonons, is established only when the mean free path with respect to phonon-phonon interactions, l, is smaller than the dimensions of the "heated" region, d. This condition is not met at low energy densities. An energy $P \sim 1$ μJ/cm^2 evolved at a depth $d \sim 1$ μm would heat the crystal to $T \sim 20$ K. At this temperature we would have $l \sim 1$ mm $\gg d$. The circumstance that the heated region may expand by heat conduction to a depth d_T during the pulse (10 ns) would make it even more difficult to reach a steady-state temperature. Estimates indicate that we could expect a steady-state temperature only at $P \gtrsim 10^3$ μJ/cm^2; at this critical energy we would have $T = 50$ K and $d_T \sim l \sim 10$ μm.

The factors which primarily determine the phonon propagation are the decay of the phonons and their scattering by isotopes. At $\omega \sim 35$ K ~ 0.75 THz, the mean free paths with respect to both processes can be estimated to be on the order of the length of the sample, $L = 1$ cm. At higher frequencies, the decay is predominant (unless it is forbidden by conservation laws).

The phonons produced in the decay process in the first step of the relaxation accumulate on the nondecay TA branch.[3] The length of the TA signal cannot be explained on the basis of the dispersion of the group velocity of the TA phonons in their ballistic propagation, since the dispersion amounts to only 3% at those frequencies at which ballistic propagation is possible. On the basis of geometric considerations we may also estimate reflection from the sidewalls of the samples. We are thus left with the assumption that the broad tail results from a quasidiffusive propagation mechanism.[4,5] However, we still lack an explanation for the short delay in the peak of the TA signal.

In the presence of an amorphous film of the thickness in these experiments, a significant number of carriers can apparently diffuse into the crystalline substrate. The phonons produced by these carriers create that part of the signal which varies only slightly upon the deposition of the film. The phonons produced in the film become thermalized, so that their average frequency decreases, and they provide the observed ballistic TA signal and the additional ballistic LA signal (Fig. 2a). This picture, we might note, is qualitatively reminescent of the signals found during the laser heating of

a gold film on germanium.[6]

The thermalization of phonons is an amorphous film may be associated with two-level systems,[7] although no direct indications of two-level systems in a–Si:H have been found. It is also assumed[8] that the high coordination number 4, prevents the formation of such systems. Two-level systems may, however, form near ruptured bonds, and even if their concentration in a–Si-H is three orders of magnitude below the concentrations typical of glasses they would provide a thermalization of phonons, $T = 20$ K, over a distance of 1 μm.

It can be seen from these estimates that at high energy densities (Fig. 2b) a thermalization of the phonons occurs in crystalline silicon, so that the additional thermalization mechanism that operates in the amorphous film has only a slight effect on the shape of the signal.

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[1]W. E. Spear, Adv. Phys. **26**, 811 (1977).
[2]I. P. Akimchenko, V. S. Vavilov, N. N. Dymova, V. V. Krasnopevtsev, A. A. Rodina, and D. P. Utkin-Édin, Fiz. Tekh. Poluprovodn. **16**, 656 (1982) [Sov. Phys. Semicond. **16**, 422 (1982)].
[3]R. G. Ulbrich, V. Narayanamurti, and M. A. Chin, Phys. Rev. Lett. **45**, 1432 (1980).
[4]D. V. Kazakovtsev and I. B. Levinson, Pis'ma Zh. Eksp. Teor. Fiz. **27**, 194 (1978) [JETP Lett. **27**, 181 (1978)].
[5]N. M. Guseinov and Y. B. Levinson, Solid State Commun. **45**, 371 (1983).
[6]V. S. Bagaev, G. Bel'skaya-Levandovskaya, M. M. Bonch-Osmolovskiĭ, T. I. Galkina, S. Yu. Levandovskiĭ, G. N. Mikhaĭlova, A. G. Poyarkov, and G. Yung, Zh. Eksp. Teor. Fiz. **77**, 2117 (1979) [Sov. Phys. JETP **50**, 1013 (1979)].
[7]I. B. Levinson, Pis'ma Zh. Eksp. Teor. Fiz. **37**, 157 (1983) [JETP Lett. **37**, 190 (1983)].
[8]N. F. Mott and E. A. Davis, electronic Processes in Noncrystalline Materials, Clarendon, Oxford, 1971 (Russ. transl., Mir, Moscow, 1982).

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