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Experimental evidence of thermoelectric amplification of acoustic waves in nickel

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We present experimental results on the detection of an exponentially growing acoustic signal in nickel as a function of the temperature gradient applied to the sample. These results are interpreted as resulting from the onset of the thermoelectric amplification of acoustic waves in the sample.

The interaction of carriers with acoustic waves gives rise to a number of acoustic instabilities in semiconductors and metals. Probably the best known and studied of these instabilities is the amplification of sound in the presence of an external dc electric field.^{1,2} Nevertheless, several other mechanisms are also known to lead to amplification of acoustic waves in solids. In particular, the amplification of acoustic waves by a temperature gradient (the so-called thermoelectric amplification), originally suggested by Bonch-Bruevich and Gulyaev,³ has been the subject of several theoretical works.³⁻⁶ The basic idea of the thermoelectric amplification may be summarized as follows. Under the action of a temperature gradient a semiconductor or metal exhibits an electric current such that the carrier drift velocity v_d is given by⁵

$$v_d = \chi \tau k_B |\nabla T| / m, \quad (1)$$

where τ is the carrier relaxation time, ∇T is the temperature gradient in the sample, and χ is a factor which depends on the electron scattering mechanism and whether the electrons are nondegenerate or not.⁵ Assuming the scattering mechanism to be due to acoustic phonons, for a nondegenerate electron gas χ reduces to $\chi = 9\pi/8$, whereas for the degenerate case $\chi = 3/4 (\pi \epsilon_F / k_B T)^{1/2}$, where ϵ_F is the Fermi energy; i.e., Eq. (1) tells us that there is a thermoelectric field applied to the sample. This thermoelectric field will cause the space charge to drift; if the drift velocity is supersonic (i.e., $v_d > v_s$, v_s being the sound velocity in the medium), the bunches of carriers will emit phonons in complete analogy with the Cherenkov effect.

In this communication we report on the detection of the thermoelectric amplification of acoustic waves in nickel. To our knowledge, the results presented here are the first experimental evidences of this phenomenon reported so far. The temperature gradient was established by means of CO₂ laser pulses focused in one of the nickel sample faces. This active

face was previously oxidized in a flame to minimize the reflection losses of the CO₂ laser radiation. The detection system consisted of a 28- μ m-thick PVF₂ piezoelectric film in intimate contact with the back (nonilluminated) face of the nickel sample. The output signal from the PVF₂ film after being amplified was fed into a programmable digitizer attached to an oscilloscope (Tektronix 7D20). The whole sample detector system as well as the focusing optics were placed in a vacuum chamber (10⁻¹ Torr) in order to prevent dielectric breakdown in air and to optimize the thermal and the acoustic coupling of the CO₂ laser to the sample. In Fig. 1 we show schematically our experimental arrangement. The sample length l , the laser pulse energy E_p , and pulse duration τ_p were chosen such that (i) the heat deposited at the sample surface does not diffuse appreciably during the pulse duration, and (ii) the optical penetration depth is much smaller than the sample length. Define the following parameters: k the thermal conductivity of the sample; ρ the density; c the specific heat; $\alpha = k/\rho c$ the thermal diffusivity; $\tau_d = \pi l^2/\alpha$, the thermal diffusion time; β the optical absorption coefficient of the sample; n the electron density; and σ the electrical conductivity. Condition (i), entailing that the heat diffusion during the pulse duration is negligible, is satisfied provided $\tau_d \gg \tau_p$ or $l \gg (\alpha \tau_p / \pi)^{1/2}$, whereas condition

FIG. 1. Schematic arrangement of the experiment set up for studying acoustic wave instability due to a temperature gradient.

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(ii), ensuring the optical deposition of heat at the surface, is satisfied if $\beta l \gg 1$. Thus, to get a well defined temperature gradient during the pulse duration, one should work with thin samples and short pulses. To get a feeling for the figures involved, let us consider a 500- μm -thick Ni sample ($\rho = 8.9 \text{ g/cm}^3$, $c = 3.94 \text{ J/cm}^3 \text{ }^\circ\text{C}$, $k = 0.9 \text{ W/cm } ^\circ\text{C}$, $\alpha = 0.23 \text{ cm}^2/\text{s}$, $n = 5.4 \times 10^{22} \text{ cm}^{-3}$, $v_s = 3 \times 10^5 \text{ cm/s}$, $\sigma = 1.1 \times 10^{17} \text{ s}^{-1}$). For a 100-ns laser pulse the thermal diffusion length $(\alpha \tau_p / \pi)^{1/2}$ is 0.86 μm and condition (i) is well satisfied. As for the threshold condition for the onset of the thermoelectric amplification one needs a temperature rise in the active face ΔT given by [cf. Eq. (1)]

$$\Delta T > (m v_s l / k_B \tau \chi). \quad (2)$$

The temperature rise due to the absorption of the CO_2 laser radiation is expressed in terms of the laser pulse energy as

$$\Delta T = \beta E_p / c A, \quad (3)$$

where A is the area of the illuminated region. Substituting Eq. (3) into Eq. (2) one gets the threshold laser fluence for the onset of the thermoelectric amplification, namely

$$(E_p / A)_{\text{th}} > (c m v_s l / \beta k_B \tau \chi). \quad (4)$$

Now, for a metal irradiated by long-wavelength radiation such that $\sigma \gg \omega$, the optical absorption coefficient is given by⁷ $\beta = (8 \pi \omega \sigma / c^2)^{1/2}$, which for our Ni sample reduces to $\beta = 7.39 \times 10^5 \text{ cm}^{-1}$. The electron relaxation time is estimated using Drude's relation ($\tau = m \sigma / n e^2$) to be $\tau = 0.8 \times 10^{-14} \text{ s}$. As for the value of χ we have estimated it using the corresponding expression for the degenerate case [$\chi = 3 (\pi \epsilon_F / k_B T)^{1/2} / 4$] to be $\chi = 18.78$. Substituting

these values in Eq. (4) one finally gets the threshold pulse fluence

$$(E_p / A)_{\text{th}} \geq 3.53 \text{ J/cm}^2.$$

In Fig. 2 we show the output signal of the PVF_2 piezoelectric film as a function of the CO_2 laser pulse fluence (100-ns duration), for a 500- μm -thick nickel sample. The laser beam was focused on the blackened face of the Ni sample by means of a Ge lens producing a spot size of 0.4-mm diameter. Each point in Fig. 2 corresponds to an average over 16 pulses at a repetition rate of 1 Hz. The pulse energy was varied by means of a Mylar attenuator of variable thickness. The signal from a pyroelectric energy meter (Molelectron J3-05) placed outside the chamber (Fig. 1), calibrated against a Joulemeter (Gen. Tec. ED-500) placed inside the vacuum chamber after each set of measurements, gives us the value of pulse energy incident into the sample. The full and dashed lines in Fig. 2 correspond to the best fitting of our data. Up to a pulse fluence (J_p) of 5 J/cm^2 our data is best fitted by a power law curve $S = 1.43 J_p^{2.18}$, represented by the full line, whereas for fluences greater than 5 J/cm^2 the data are best fit by an exponential law, $S = 10.0 \exp(0.34 J_p)$, represented by the dashed line. The transition from the power law region into the instability (exponential) region is experimentally estimated to occur at 5.2 J/cm^2 , which is reasonably close to the theoretically predicted value of 3.53 J/cm^2 . Actually, this discrepancy between the theoretical and experimental values of the threshold fluence can be accounted for by relaxing the implicit assumption in Eq. (3) that all the light absorbed goes into heat. Introducing a light to heat conversion efficiency η , one should replace β in Eq. (3) by $\eta \beta$. If one does this the threshold pulse fluence reads $(E_p / A)_{\text{th}} = 3.53 / \eta (\text{J/cm}^2)$; equating this expression to the experimental value of 5.2 J/cm^2 one finds $\eta = 0.68$ for the conversion efficiency. In fact, the data shown in Fig. 2 may be interpreted as reflecting the occurrence of acoustic wave instability as follows. Assume a crude model whereby (i) the detected acoustic signal is proportional to the number of phonons excited in the sample by the drifting carriers during a pulse duration; (ii) the phonons are described in terms of a kinetic equation involving stimulated emission of phonons by the carriers.² The energy delivered by the thermoelectric field to the electrons is dissipated mainly by emission of phonons so that the phonon population reaches an equilibrium in a very short time; from the energy balance, the electron energy loss rate to the phonons is $(n e v_d)^2 / \sigma$, so that the phonon equilibrium population $\bar{N}_k$ may be written as $\bar{N}_k \sim (n e v_d)^2$.

The rate equation for the phonon population N_k may be written as²⁻⁵

$$\frac{dN_k}{dt} = \gamma_k N_k(t) - \frac{\bar{N}_k}{\tau_k}, \quad (5)$$

where N_k is the number of phonons of wave vector k , γ_k is the phonon growth rate due to its collision with the drifting carriers, τ_k is the phonon relaxation time. The growth rate γ_k may be written,³⁻⁶ quite generally, as $\gamma_k = \gamma (v_d - v_s)$, where γ is a constant dependent upon the dominant electron-phonon coupling mechanism in the material. Using

FIG. 2. Acoustic signal from the 28- μm -thick PVF_2 piezoelectric film as a function of the CO_2 laser peak pulse fluence for a 500- μm -thick nickel sample. The CO_2 laser pulse duration is 100 ns.

these expressions for $\bar{N}_k$ and γ_k , and integrating Eq. (5) over a pulse duration τ_p one gets

$$N_k(\tau_p) \sim v_d^2 \frac{\exp[\gamma(v_d - v_s)\tau_p] - 1}{\gamma(v_d - v_s)}. \quad (6)$$

For weak temperature gradient, such that $v_d \ll v_s$, the argument of the exponential is negative (i.e., one has damping) so that N_k varies as v_d^2 and therefore as J_p^2 . On the other hand, for high enough values of the temperature gradient, such that $v_d > v_s$, the argument of the exponential may become positive (i.e., amplification) with the result that N_k grows exponentially with v_d ($\sim J_p$). These features, together with the reasonable agreement between the calculated and the experimentally observed values of the threshold condition $v_d > v_s$, are, in our opinion, strong evidence in support

of our claim for interpreting the present results as due to the thermoelectric amplification of acoustic waves.

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Density of oxidation-induced stacking faults in damaged silicon

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A model for the relation between density and length of oxidation-induced stacking faults on damaged silicon surfaces is proposed, based on interactions of stacking faults with dislocations and neighboring stacking faults. The model agrees with experiments.

Thermal oxidation of diamond-lapped silicon wafers leads to the growth of extrinsic stacking faults.¹ Much research has been done on these stacking faults in an exploratory manner, but only recently, in 1984, Ishihara, Kaneko, and Matsumoto² discovered that density and length of stacking faults in diamond-lapped wafers are related to each other, independent of oxidation temperature or surface orientation. This paper explains these experimental findings and reports some experimental results obtained by ourselves. Annihilation of extrinsic stacking faults occurs, if we neglect retrogrowth, via a Hirsch reaction:

Two Shockley partial dislocations, one above and one below the (111) stacking fault plane, eliminate the stacking fault, leaving behind a perfect dislocation bounding the former stacking fault. These two Shockley dislocations must be created by an event during the oxidation. One possibility is an impingement of two stacking faults, the other is the impingement of a stacking fault and a perfect dislocation.

Stacking fault–stacking fault interactions can be divided in two categories. We call them obtuse [(111) with ($\bar{1}\bar{1}1$)] and acute [(111) with ($1\bar{1}1$) or ($\bar{1}11$)]. On oxidized (001) silicon wafers with many stacking faults, many acute impingements can be seen that apparently did not give rise to unfaulting reactions. On the other hand, on (001) planes obtuse interaction always seems to cause annihilation, as demonstrated by Hayafuji and Kawado.³ In addition, no obtuse collisions on our etched (001) samples were observed.

Next, it is known that stacking fault dislocation interaction can cause unfaulting. Attention should be paid to the fact that not all dislocations react in the same way. Some lead to an incorrect unfaulting of the stacking fault. Tan⁴ showed that a Shockley dislocation can turn a stacking fault into a four-layer-thick microtwin, i.e., leading to an incorrect unfaulting. In contrast, the same Shockley dislocation can, after dissociation into two other Shockley dislocations, lead to a correct unfaulting reaction as in reaction (1). Finally, it has to be emphasized that some dislocations do not react at all with stacking faults.

In summary:

(i) An obtuse interaction of stacking faults leads to unfaulting, leaving behind two dislocations.

(ii) An acute interaction of stacking faults does not necessarily lead to unfaulting.

(iii) Only a fraction F of the dislocations will cause unfaulting of a colliding stacking fault.

The experiments of Ishihara, Kaneko, and Matsumoto² were done with Czochralski (CZ) silicon. The high concentration of carbon and oxygen in this material favors the stacking fault formation. Dieleman and Martens⁵ showed that oxidation of (100) float-zone (FZ) silicon induces no stacking faults if the surface is not damaged. So this material is more appropriate to study effects of damage on stacking faults. Therefore, we polished (100) FZ-silicon wafers, B-doped, $1\text{--}30\ \Omega\ \text{cm}$, with $3\text{-}\mu\text{m}$ diamond powder prior to oxidation at various temperatures. To avoid stress anneal, the wafers were brought into the furnace already under (dry)