Oscillatory effects in photostimulated surface reconstruction

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Illumination of the surface of a semiconductor can stimulate structural transitions there, e.g., can alter the reconstruction of the surface. Under certain conditions, oscillations consisting of alternate transitions from one phase to another may be excited. An experiment is proposed for detecting such oscillations.

Kapaev *et al.*[1] have recently predicted the possibility that a semiconductor can undergo a Peierls transition into a metallic state when illuminated with intense light. Suslov[2] subsequently showed that relative oscillations of two sublattices of a crystal, with an amplitude on the order of the interatomic distance, may be excited under these conditions.[1)] Observing this effect would require meeting some extremely severe conditions, since it would be necessary to avoid overheating the lattice due to the high excitation levels of the electron system. In this note we wish to note that by switching from a three-dimensional system to a two-dimensional system one could considerably simplify the observation of these oscillations.

In a metal which exhibits a Peierls instability, the potential energy V is written as a function of the relative displacement x of the two sublattices of the crystal as follows[4]:

$$V(x)=-\alpha\frac{E_g{}^2}{U}\ln\frac{U}{E_g}+\beta x^2,$$

$$E_g\backsim|x|,\quad \alpha,\beta>0 \tag{1}$$

(curve 1 in Fig. 1). The first term here describes a decrease in the energy of the electron gas due to the opening of an energy gap E_g at the Fermi level; the second describes the energy increase due to elastic strain; and U is the width of the allowed band of the metal. Illumination causes some of the electrons to move across the gap, and the energy advantage described by the first term in (1) decreases. When the electron density above the gap satisifes $n > n_c \sim n_0 E_g^0/U$, the dependence $V(x)$ has the form shown by curve 2 in Fig. 1 (n_0 is the total electron density, and E_g^0 is the size of the energy gap at the point x_0). Illumination can therefore cause a system to undergo a transition from a Peierls insulator state ($x = x_0$, $E_g = E_g^0$) to a metallic state[1] ($x = 0$, $K_g = 0$).

If the rate $\partial n/\partial t$ at which electron-hole pairs are generated satisfies the condition

$$\tau_E \lessdot n_c(\partial n/\partial t)^{-1} \lessdot \tau_{\rm rec}, \tag{2}$$

where $\tau_E \sim \omega_D^{-1}(E_g^0/\hbar\omega_D)$ is the energy relaxation time of the electrons in the metallic phase (ω_D is the Debye frequency), and $\tau_{\rm rec}$ is the recombination time of the electron-hole pairs in the insulating phase, determined primarily by Auger processes, then self-excited oscillations will arise in the system.[2] The oscillatory situation most convenient for experimental study arises when the pair generation rate $\partial n/\partial t$ is just slightly above the threshold for the insulator-metal transition. As the density n of the electrons above the gap increases, the system goes from point A to point B in a time $\sim\tau_{\rm rec}$ and begins to oscillate between B and B' with a frequency $\sim\omega_D$. These oscillations are damped on the time scale of the phonon-phonon relaxation $\tau_1 \sim \omega_D^{-1}(U_b/T)$ (U_b is the binding energy of the lattice). On the other hand, the rapid recombination near the point $x = 0$, where the recombination results primarily from single-phonon processes, causes the electron density n in the conduction band to decrease with a characteristic time $\tau_2 \sim \tau_E E_g^0/\hbar\omega_D$. The system goes from curve 2 to curve 1 (Fig. 1), and after a time $\sim\tau_1$ it returns to point A. The entire cycle is then repeated from the beginning. At higher generation rates $\partial n/\partial t$, the conditions studied in Ref. 2 arise. As was pointed out in Ref. 2, the effect described there can be observed in semiconductors with small values of E_g^0, but it would be considerably simpler to achieve by making use of the surface states of a semiconductor.

Semiconductors have Tamm states localized near their surfaces. The spectrum of Tamm states forms a two-dimensional band which is partially filled with electrons liberated through the breaking of covalent bonds near the surface. In a two-dimensional system of this sort, a Peierls instability can occur and give rise to a lattice constant at the surface which is larger than that in the interior of the semiconductor. It can also lead to so-called reconstruction of the surface.[5] Conse-

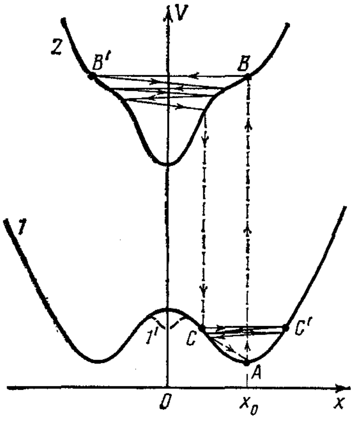

FIG. 1.

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quently, by illuminating a surface one could induce insulator-metal structural transitions there or excite surface oscillations of the atoms according to the mechanism outlined above. For changing the reconstruction of a surface it would be convenient to use irreversible phase transitions for which the metallic state is metastable (curve 1′ in Fig. 1) for the excitation of the self-excited oscillations: reversible transitions with an absolute instability of the metallic phase (curve 1).

Two-dimensional systems have several advantages over three-dimensional systems for observing these oscillations. In the oscillation stage *B–B′–C–C′–A*, most of the energy stored in stage *A–B* ($\sim E_g^0 n_c \sim E_g^{02} n_0/U$ per unit volume) is converted in a time $\sim \min(\tau_1, \tau_2) \lesssim 10^{-11}$ s into heat, which is evolved in an essentially local fashion, since the heat does not escape from the two-dimensional layer over a time shorter than the phonon-phonon collision time. The resultant heating must be small in comparison with the melting point T_m:

$$E_g^{02}/U \ll T_m. \qquad (3)$$

In three dimensions, the typical values are $E_g^0 \sim 1$ eV and $U \sim 10$ eV, and the left side of (3) would be ~ 1000 K, or comparable to T_m. For two-dimensional Tamm bands, the energies are typically an order of magnitude smaller ($U \sim 1$ eV, $E_g^0 \sim 0.1$ eV), and the left side of (3) would be ~ 100 K, much smaller than the typical melting temperatures.

2. In stage *A–B*, the heat evolution is insignificant, and the energy evolved in the stage *B–B′–C–C′–A* propagates a diffusion length during a oscillation cycle ($\sim \tau_{\rm rec}$):

$$L_D \sim (sl\tau_{\rm rec})^{1/2} \sim (sa\tau_{\rm rec} U_b/T)^{1/2} \sim 10^{-6} - 10^{-5}\ \text{cm}, \qquad (4)$$

where l is the meanfree path of a Debye phonon, a is the distance between atoms, s is the sound velocity, and $\tau_{\rm rec} \sim 10^{-9}$–$10^{-11}$ s. In the two-dimensional case, the energy evolved in a single atomic layer propagates a distance of 10^2–10^3 atomic layers, and the average heating over a time $\sim \tau_{\rm rec}$ is ~ 1 K. For one-dimensional diffusion, the surface temperature would increase with the time t in proportion to $t^{1/2}$, and one could observe $\sim 10^6$ oscillation cycles before the melting point is reached. If special heat sinking were arranged, the oscillations could also occur in a steady state.

In the three-dimensional case, L_D is short in comparison with the damping length for the light wave, $\alpha^{-1} \sim 10^{-5}$–10^{-4} cm, and heat sinking would not be effective. The maximum observable number of oscillation periods would then be $\sim UT_m/E_g^0$, which could hardly be raised above 10^2.

3. The onset of a Peierls instability is facilitated as the dimensionality of the space is reduced, so that in two dimensions it is simpler to choose a material with an absolute instability of its metallic phase (curve 1 in Fig. 1)—it will quite often be metastable (curve 1′). Evidence for this assertion comes from the abundance of reversible structural transitions at the surfaces of semiconductors and metals [e.g., the transition from a 7×7 structure to a 1×1 structure on the silicon (111) face].

The optical absorption coefficient γ of a two-dimensional layer of electrons can be calculated by analogy with the approach used in the three-dimensional case[6]:

$$\gamma = \frac{8\pi^2}{q}\frac{e^2}{m^2c^2}\sum_{\mathbf{k}} (\hat{a}\mathbf{p}_{\mathbf{k}}^{vc})^2 \delta(\varepsilon_{\mathbf{k}}^c - \varepsilon_{\mathbf{k}}^v - \hbar\omega),$$

where q and $\hat{a}$ are the wave vector and polarization vector of the light wave, $\varepsilon_{\mathbf{k}}^v$ and $\varepsilon_{\mathbf{k}}^c$ are the dispersion laws of the valence band and of the conduction band, and $p_{\mathbf{k}}^{vc}$ is the matrix element of the momentum operator between states with quasimomentum **k** in the valence band and the conduction band. Near the absorption edge we have $\gamma \sim \alpha\theta(\hbar\omega - E_g^0)$ (α is the fine-structure constant); i.e., $\sim 1\%$ of the energy of the light wave is absorbed in the two-dimensional layer. The rest of the energy passes unhindered across the thickness of the crystal, so that the radiation frequencies corresponding to transitions between Tamm states ($\hbar\omega$ must be several times larger than the gap, $E_g^0 \sim 0.1$ eV, in the two-dimensional spectrum) reach the region of transparency of the bulk material. To excite oscillations, we would need light with an energy flux density

$$J \sim \frac{\hbar\omega}{\gamma}\frac{n_c}{\tau_{\rm rec}} \sim 10^5 - 10^8\ \text{W/cm}^2.$$

The oscillations can be detected from oscillations of the reflection coefficient, which should increase by some tenths in stage *B–B′*, where the oscillations occur near the metallic state.

In order to observe this effect one should select materials with Auger recombination times as short as possible. The most suitable materials by this criteria would be semiconductors for which the ordinary three-particle Auger process is forbidden by the particular features of the energy spectrum. The maximum velocity in the band, $V_{\max}$, would have to be low in comparison with $(E_g^0/m^*)^{1/2}$ here, where m^* is a characteristic effective mass. In this case, the Auger recombination would involve a phonon, and its time could be estimated from

$$1/\tau_{\rm rec} \sim \omega_D (n/n^*)^2, \quad n^* \sim 1/r_B^2$$

(r_B is the first Bohr radius, and n and n^* are two-dimensional densities). With $n/n^* \sim 10^{-1}$–10^{-2} we find $\tau_{\rm rec} \sim 10^{-9}$–$10^{-11}$ s as suggested above.

The coherent lattice oscillations discussed above were used in Ref. 2 to propose a nonthermal mechanism for laser annealing. It follows from the present study that this mechanism could be implemented not only for a light source with a phonon energy $\hbar\omega \sim 1$ eV but also for a lower-frequency source with $\hbar\omega \sim 0.1$ eV.

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[1)] For a further discussion of this question, see the recent paper by Kopaev *et al.*[3]

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