Structural transformations in InSb induced by a strong electromagnetic radiation

G. G. Gromov, V. V. Kapaev, Yu. V. Kopaev, and K. V. Rudenko
P. N. Lebedev Physics Institute, Academy of Sciences of the USSR

(Submitted 25 June 1987)
Pis'ma Zh. Eksp. Teor. Fiz. **46**, No. 3, 119–121 (10 August 1987)

Nonequilibrium phase transitions in indium antimonide are studied experimentally. A model for the phase transition is proposed.

It has been previously established theoretically[1] that a semiconductor can undergo a nonequilibrium phase transition as a result of bombardment with a strong electromagnetic (laser) radiation. This prediction requires experimental verification.

A InSb semiconductor was chosen because its nonequilibrium (metallic) phases can be stabilized by fast quenching to low temperatures.[2] Accordingly, the samples immersed into a liquid nitrogen were bombarded with laser light ($\lambda = 1.064$ μm, $\tau = 10$ ns). As samples we used single-crystalline InSb substrates and polycrystalline films.

Experimental studies showed that a layer of a highly reflecting phase is formed at the pulse energy $P = 0.06$ J/cm^2. The plot of the reflection coefficient $R(\lambda = 0.63$ μm) versus P in this case has a characteristic shape (Fig. 1), which is governed by the change in the thickness h of the layer of the phase. The value of h which was determined by means of controlled etching and subsequent measurement of the surface profile depends linearly on P (Fig. 2a). The highly reflecting phase is formed at energies much lower than the rated melting point in liquid nitrogen ($P_{\rm mp}^{\rm rated} = 0.095$ J/cm^2).

Holding the irradiated samples for a time under normal conditions brings about an inverse transition which is accompanied by a decrease in R to the original value (46%). This causes the appearance of a strong temperature dependence ($T = 288$–333 K) of the phase relaxation time, $\tau_{\rm rel}$, which has a shape typical of the thermally activated processes, $\ln \tau_{\rm rel} = -52.6 + 1.7 \times 10^4/T$ ($\tau_{\rm rel}$ is in min.).

An x-ray structural analysis (based on Debye's method) of polycrystalline films showed that the metallic InSb phase (presumably of the "β-Sn" type) is the highly reflecting metastable phase, consistent with the agreement between the interplanar distances ($d = 2.92$, 2.56, and 2.04 Å) and the known data.[2] An increase in the density ($\Delta\rho = 20\%$; Ref. 3), characteristic of a "sphalerite-metallic phase" phase transition in InSb has been found to occur in this case. The fact that a smooth surface which "collapses" by Δh (Fig. 2b) as a result of irradiation can be restored to its original state is evidence that such a process occurs. A comparison of Δh with the thickness of the phase that forms (Fig. 2) allows us to calculate its density, $\rho = 6.9 \pm 0.2$ g/cm^3.

An important feature here is that the phase transition in InSb occurs through the intermediate state. When a train of pulses (N) affects the behavior of the curve R

 © 1987 American Institute of Physics

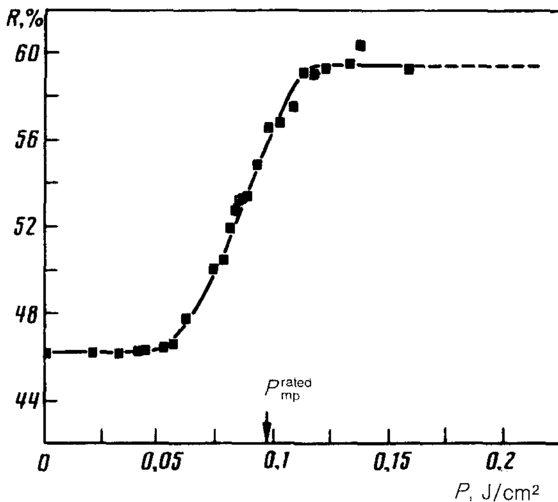

FIG. 1. The reflection coefficient $R(\lambda = 0.63\ \mu m)$ of the irradiated InSb surface versus the energy of the pulse p.

versus N (along the discrete N scale), the formation of the highly reflecting phase ($N > 1$) is always preceded ($N = 1$) by a state with a lower R relative to the initial value ($\Delta R = 4$–5%). The formation of the wurtzite modification of InSb in this case can be predicted on the basis of the data on the x-ray structural analysis from the presence of a strong reflection, $d = 3.46$ Å.

On the basis of the pseudopotential method[1] it was shown that a nonthermal mechanism for the phase transition[4] in InSb from the semiconducting ground state with a sphalerite structure (phase I) to the "β-Sn"-type metallic state (phase III)

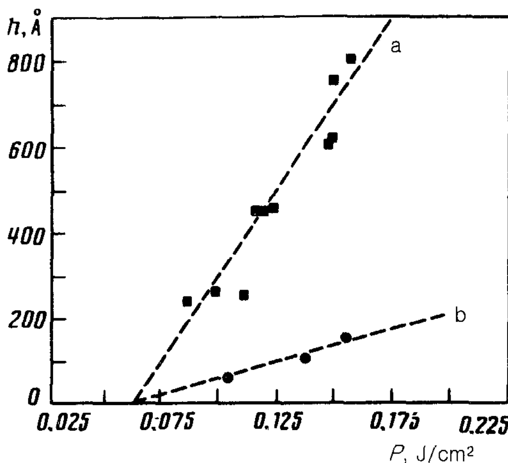

FIG. 2. The results of profile measurements of the irradiated surface. (a) The thickness of the highly reflecting phase, h; (b) "collapse" of the irradiated region of the surface, Δh.

requires the carrier density to be $n_{cr} \cong (2\text{–}3)\times 10^{21}$ cm^{-3}, which is larger by a factor of 5–10 than the value obtained experimentally. Under these conditions the phase transition may be brought about by increasing the concentration of excitations in the individual regions up to n_{cr} in comparison with the average concentration n, by analogy with the situation in the electron-hole drops.[5,6] Under our experimental conditions ($n \sim 10^{20}$ cm^{-3}) the excitations can be assumed to be the ideal gas, and the change in the width of the band gap E_g, which depends on n $[E_g = E_g^0(1-\gamma n)]$, and in the reduced mass m accounts for the condensation of these excitations into the new phase. Such a transition can be described thermodynamically if the lifetime of the excitations in each phase is greater than the phase-transition time. This is not the case for the metallic phase (phase III). As was shown above, however, the phase transition proceeds through the intermediate semiconducting wurtzite phase (phase II). For each of the two phases ($i = \mathrm{I}$, II), the excitation energy E_i is $E_i = A(n^{5/3}/m_i) + BnE_{gi}^0(1-\gamma_i n)$. The quantity $\delta E = \Delta E_T + (E_{\mathrm{II}} - E_{\mathrm{I}})$, where ΔE_T is the equilibrium difference in the energies of phases II and I, characterizes the possibility of the phase transition. If $E_{g\mathrm{II}}^0 < E_{g\mathrm{I}}^0$, the difference in the energies, as in the case of the electron-hole drops, is minimum. In InSb the situation apparently is the reverse ($E_{g\mathrm{II}}^0 > E_{g\mathrm{I}}^0$). With increase in n, δE increases, goes through a maximum, and then decreases monotonically. Choosing the values of E_g, m, and γ (which are so far not known experimentally) for phase II in a reasonable range, we find $n_{\max} \approx 10^{19}$ cm^{-3}. If the average concentration is $n > n_{\max}$, the system tends to condense the excitations in the band of phase II. These excitations are periodically spaced in the maxima of the electromagnetic field. An increase of their concentration is limited by a transition at $n > n_{cr}$ to the metastable (at $n = 0$) metallic state (phase III). In δE the values of n for phases I and II are different for the inhomogeneous state.

To clarify the question of the further evolution of the system, we solved the electrodynamic problem of the penetration of laser light into the periodic structure consisting of layers of metal of width a at the semiconductor surface. The maximum of the flux, $P_{\max}$, which penetrates into the structure, is situated in the semiconductor region near the metal-semiconductor interface. At $a/d < 0.4$ $P_{\max}$ is greater than the value of P_s for a homogeneous semiconductor surface ($P_{\max}/P_s$ may be as high as 1.5), which accounts for the increase in the thickness of the metal layer. Because of the metastable nature of the metallic state, the value of a will increase. The metallic state becomes stabilized because the radiation flux P_m which penetrates into the metallic layers is much higher than P_0 for a homogeneous metallic surface (for small a we have $P_m/P_0 \cong 2$).

At $a > 0.4$ additional flux maxima appear at the center of the semiconductor region, giving rise to the formation of the metallic phase even in that region.

The mechanism which we have examined here thus accounts for the fact that the volume of the metallic phase is much greater than the value of the ratio of the average concentration of the carriers in the sample to the critical concentration for the transition to the metallic state.

[1]Yu. V. Kopaev, V. V. Menyaĭlenko, and S. N. Molotkov, Fiz. Tverd. Tela 27, 3288 (1985) [Sov. Phys. Solid State 27, 1979 (1985)].

[2]V. F. Degtyareva, I. T. Belash, G. V. Chipenko, E. G. Ponyatovskiĭ, and V. I. Rashchupkin, Fiz. Tverd. Tela **25**, 2968 (1983) [Sov. Phys. Solid State **25**, 1712 (1983)].
[3]V. I. Petrosyan, O. I. Vasin, S. I. Stenin, N. F. Bondarenko, and P. A. Skripkin, Pis'ma Zh. Eksp. Teor. Fiz. **26**, 10 (1977) [JETP Lett. **26**, 8 (1977)].
[4]V. V. Kapaev, Yu. V. Kopaev, and S. N. Molotkov, Mikroélektronika **12**, 499 (1983).
[5]S. G. Tikhodeev, Usp. Fiz. Nauk **145**, 3 (1985) [Sov. Fiz. Usp. **28**, 1 (1985)].
[6]L. V. Keldysh, in Proceedings of the Ninth International Conf. on Physics of Semiconductors, Nauka, Leningrad, 1968, p. 1307.

Translated by S. J. Amoretty