Low-temperature anomaly of the velocity of ultrasound in $Pb_{0.75}Sn_{0.25}Te(In)$

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A new method of studying the acoustic properties of semiconductors, which is based on direct conversion of electromagnetic and ultrasonic waves, is proposed. At $T \sim 20$ K an indium-doped lead-tin telluride exhibits an anomalous feature in the velocity of longitudinal ultrasound. This anomaly is caused by the transition of the crystal to the ferroelectric phase.

The practical application of lead-tin tellurides as radiation sources or radiation detectors in the far-infrared region, in high-resolution molecular spectroscopy, in gas analysis, etc., targets these semiconductor compounds as objects of extensive experimental and theoretical research. The terms that determine the band gap in PbTe and SnTe are mutually inverted, so that a change in the composition $Pb_{1-x}Sn_xTe$ causes the width of the band gap to smoothly decrease from several tenths of eV to zero at $x \sim 0.35$ ($T = 4$ K). It is assumed that at low temperatures and large x the solid solutions $Pb_{1-x}Sn_xTe$ undergo a ferroelectric phase transition accompanied by a rhombohedral distortion of the cubic lattice of these solid solutions. The transition temperature tends toward zero at $x \sim 0.3$ and at lower x there is a tendency for the lattice to be unstable.[1] The transition to the ferroelectric phase in SnTe, on the other hand, has been confirmed by x-ray-diffraction studies.[2] There is only indirect evidence of a phase transition in $Pb_{1-x}Sn_xTe$. Anomalous features on the temperature-resistance curves (the mobility minimum at T_c), which shift upward with the temperature as the value of x is increased, have been reported, for example, in Refs. 3 and 4. The dielectric constant has been seen in Ref. 5 to exhibit anomalous features.

An n-type impurity doping, in particular, indium doping, of lead-tin tellurides has a significant effect on the properties of these compounds. At $T \sim 25$ K $Pb_{1-x}Sn_xTe(In)$ has been found to exhibit in the composition range $0.22 \leqslant x \leqslant 0.28$ an unusually high photosensitivity, a long relaxation time of the photoresponse and of the conductivity,[6] a spontaneous polarization,[7] and a switching and delayed dielectric breakdown.[8] The role of impurity centers in lead-tin tellurides is not completely clear.

In some models[9,10] the anomalous properties $Pb_{1-x}Sn_xTe$ are explained in terms of the "local phase transition"—the rearrangement of the impurity centers (defects) in lead-tin tellurides, rather than in terms of the ferroelectric phase transition in these materials. To determine the nature of the observable anomalies, it is necessary to carry out additional experiments which would yield information primarily on the lattice properties of crystals.

In this letter we study the temperature dependence of the propagation velocity of the transverse ultrasound S_t and longitudinal ultrasound S_l in a $Pb_{0.75}Sn_{0.25}Te$(0.4% In) single crystal. The sample is a 0.12-cm-thick plane parallel plate ~2 cm in diameter. The normal to the sample's plane and the direction of propagation of the ultrasound coincide with the [001] fourfold symmetry axis. A direct conversion of electromagnetic and ultrasonic waves in a highly conducting medium was exploited to generate the ultrasound.[11] For this purpose, a thin aluminum film (~1 μm) was deposited on both surfaces of the plate. The striking of the metal surface by an electromagnetic wave in a static magnetic field H_0 caused the metal surface to generate an ultrasound which subsequently propagated into the interior of the semiconductor. The excitation is caused by the Lorentz interaction of the current in the metallic film with the field H_0. The generation of both the longitudinal and transverse ultrasound of a given polarization is one of the many advantages of this method. The longitudinal ultrasonic waves were generated in a field H_0 parallel to the surface of the plate and the transverse ultrasonic waves were generated in a field normal to the plate. Two coils wound around the sample were used to transmit the ultrasonic waves through the sample and to receive them. In the experiment we studied the resonant features of the surface impedance of the plate which were detected upon the appearance of ultrasonic standing waves in the plate. The acoustic-resonance frequencies $f_{l,t} = nS_{l,t}/2d, n = 1,3,\ldots$ were measured within $\Delta f = 10^{-5} f_{l,t}$. This value defines the relative measurement accuracy of the velocity of ultrasound. The measurements were carried out at frequencies $f \sim 10^6$ Hz over a temperature interval 4–80 K in a field $H_0 = 50$ kOe. The temperature dependences of S_l and S_t are shown in Fig. 1. The velocity of longitudinal ultrasound changes anomalously near $T \sim 20$ K. The relative

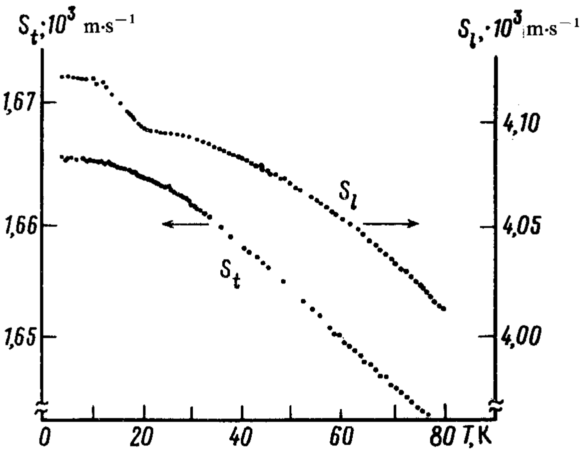

FIG. 1. Temperature dependence of the velocity of transverse ultrasound S_t and longitudinal ultrasound S_l.

magnitude of the effect is $\Delta S/S_l \sim 0.5\%$. The velocity of transverse ultrasound has no anomalies of any sort in the same temperature interval.

Let us estimate the anomaly of the elastic moduli which can be observed upon rearrangement of the impurity centers. A change in the charge state of indium atoms upon lowering the temperature below 20 K is accompanied, as follows from Ref. 9, by a change in the location of the impurity in the crystal cell. In a $Pb_{1-x}Sn_xTe$ lattice the stable states of In vary by 2×10^{-2} eV on the energy scale. This value is comparable to the elastic modulus of lead-tin telluride (~ 10 eV). In the sample under study the impurity concentration is 0.4%. Consequently, the anomalous feature of ultrasonic velocity which is associated with the rearrangement of the impurity centers is $\Delta S/S \sim 10^{-5}$; i.e., it exceeds the observable effect by no more than 10^{-2}.

The anomalous velocity of longitudinal ultrasound observed at $T\sim 20$ K can be interpreted, in our view, as a manifestation of the ferroelectric phase transition in lead-tin telluride. Indium is used to reduce the band-carrier density, since the impurity level falls into the energy gap within the composition range $0.22 \leqslant x \leqslant 0.28$. A low density of free charge carriers [$n \lesssim 10^{10}$ cm^{-3}] (Ref. 6)] sharply reduces the Coulomb screening of ions, thereby facilitating the formation of ferroelectric dipoles in the crystal lattice. The absence of particular features on the $S_t(T)$ curve is attributable, according to Ref. 12, to an appreciably lower sensitivity (by two or three orders of magnitude) of the shear modulus to compressibility fluctuations near the phase transition.

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