Mössbauer spectra of ^{119}Sn in the gap-free state of a semiconductor

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A decrease in the probability for the Mössbauer effect by $\sim 40\%$ has been detected near the band-inversion point in the system of narrow-gap semiconductors $(SnTe)_x (PbSe)_{1-x}$. This result is attributed to a structural change in the phonon spectrum due to a strong electron–phonon interaction.

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The $A^{IV}B^{VI}$ narrow-gap semiconductors and the solid solutions which they form have recently been the object of many studies. Among these compounds are some in which, upon a change in the stoichiometric composition with the temperature, there is an inversion of the conduction band and the valence band. In this case the compound or solid solution goes through a gap-free state. Volkov and Kopaev[1] have suggested that a strong electron–photon interaction occurs in the system of solid solutions $Pb_{1-x}Sn_xTe$. If so, a change in the electron spectrum in the gap-free state should cause a change in the phonon spectrum. In an effort to detect this change, Nikolaev *et al.*[2] studied this system by Mössbauer spectroscopy, and they indeed noticed a pronounced decrease in the probability for the Mössbauer effect near the band-inversion point. Nikolaev *et al.*[2] labeled this decrease a "softening" of the phonon spectrum of the crystal.

In the present experiments we have attempted to determine whether this softening of the phonon spectrum is peculiar to the $Pb_{1-x}Sn_xTe$ system or is a more general effect. For these experiments we selected the system $(SnTe)_x(PbSe)_{1-x}$, in which a band inversion occurs at $x \sim 0.6$ and in which anomalous features are observed in the behavior of the carrier concentration, the thermal-emf, and the electrical conductivity.[3]

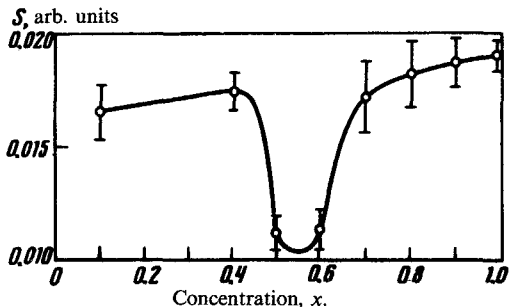The procedure used to prepare the samples can be outlined a follows: First, single

FIG. 1. Composition (x) dependence of the area under the Mössbauer-spectrum curve, S, for $(SnTe)_x$ $(PbSe)_{1-x}$ solid solutions.

crystals are grown by the Bridgman method.[4] These crystals are then crushed and deposited on an aluminum foil in an alcohol solution of BF-2 cement. The thickness of all the absorbers was 5 mg/cm^2 in terms of natural tin. The γ source was a 5-μCi Pd ^{119}Sn source. The source and absorber were at room temperature. Each sample was measured 7–10 times. The measurements were carried out in the moving-absorber mode. The Mössbauer spectra of all the samples contain single, unbroadened lines. Computer-assisted least-squares calculations yielded the line width Γ, the isomer shift δ, and the magnitude of effect, α. The area (S) under the spectral curve, which is directly proportional to the probability for the Mössbauer effect, is calculated from

$$S = \pi a \Gamma /2.$$

Figure 1 shows the behavior of the area under the Mössbauer spectrum. We see that the area S decreases by nearly 40% at $x \sim 0.55$. A similar change was observed by Nikolaev et al.[2] Such large changes in the Mössbauer-effect probability are usually observed only at first-order phase transitions. The $(SnTe)_x(PbSe)_{1-x}$ system has a cubic structure of the NaCl type; x-ray diffraction measurements have revealed no phase transitions anywhere in the concentration range studied.[3] The lattice constant also varies linearly. Under the assumption of a strong electron–phonon interaction, this result can be explained on the basis that the phonon spectrum softens because of a
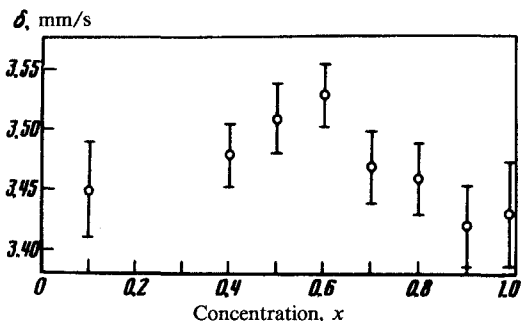

FIG. 2. Change in the isomer shift δ in the $(SnTe)_x(PbSe)_{1-x}$ system as a function of x. The results are expressed with respect to a BaSnO$_3$ source.

change in the electron spectrum in the gap-free state, since the decrease in the area under the curve implies a weakening of the chemical bond of the tin atoms and an increase in their vibration amplitude. This result also shows that there is a decrease in the frequencies of certain optical vibrational modes of the tin atoms. The results of these experiments thus confirm the conclusion of Nikolaev et al.[2] and show that there is a change in the phonon spectrum of the crystal in the gap-free state.

Figure 2 shows our results on the isomer shift as a function of x. In contradiction of the results of Ref. 2, there is an increase in the isomer shift (by about 0.1 mm/s) in the gap-free state. This result shows that the density of s electrons at the tin nucleus increases.

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