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Determination of the Position of Antimony Impurity Atoms in PbS and PbTe Lattices by MESSBAUER EMISSION SPECTROSCOPY on ^{119}Sb (^{119}mSn) ISOTOPE

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I. INTRODUCTION

The study of the structure and other physical parameters of semiconductor materials responds to the use of these materials in manufacturing to improve the physical properties of solar cells. It was shown by the method of emission Mössbauer spectroscopy on the isotope ^{119}Sb (^{119}mSb) that the localization of impurity atoms of antimony in the PbS and PbTe lattices depends on the type of conductivity of the material. It is noted that the charge state of the ^{119}Sn antisite defect formed in the anionic sublattice of PbS and PbTe after the ^{119}Sb radioactive transformation does not depend on the position of the Fermi level. The ^{119}Sn center in the PbS cation sublattice is an electrically active substitutional impurity. The same center in the PbTe cation sublattice is electrically inactive.

In recent years, scientists have made significant progress in studying the nature of impurity states formed in lead chalcogenides by atoms of the third and fourth groups. In particular, for impurities of the third group, the effect of resonant scattering of current carriers was found [1: 201], and for impurities of the fourth group, two-electron centers with negative correlation energy were identified [2: 291]. However, there are practically no works devoted to the study of Group V impurities (As, Sb, and Bi) in lead chalcogenides. It is only known that these impurities are donors, although the fraction of electrically active atoms is much less than unity [3]. The latter fact can be explained in two ways: either a significant part of the impurity atoms form electrically inactive complexes of the (Sb_2Te_3) type in the lattice, or the impurity is distributed between the cationic (where it is a donor) and anionic (the nature of impurity states in this case is not obvious) sublattices.

The choice between alternative models is possible if the position of antimony atoms in the lattices of lead chalcogenides is determined. In the present work, such a determination was carried out for the case of impurity antimony atoms in lead sulfide and telluride (PbS, PbTe) by the method of Mössbauer emission spectroscopy on the ^{119}Sb isotope (^{119}mSb). The inset to the figure shows a diagram of the formation of the ^{119}mSn Mössbauer level using the mother isotope ^{119}Sb (half-life 38 hours). Taking into account that the recoil energy of ^{119}mSn daughter atoms due to electron capture in ^{119}Sb and neutrino emission does not exceed 1.4 eV [4: 2463], it can be concluded that radioactive transformation does not lead to displacement of tin atoms from normal lattice sites. Thus, the parameters of the ^{119}Sb (^{119}mSn) Mössbauer emission spectra should reflect the valence (charge) of ^{119}mSn atoms localized at sites occupied by antimony atoms.

The radioactive isotope ^{119}mSb was obtained by the reaction $^{120}\text{Sn} (p, 2n) ^{119}\text{mSb}$. To isolate a non-reference ^{119}mSb preparation, the target was dissolved in concentrated hydrochloric acid, extracted with SbCl_3 with isopropylene alcohol, and the preparation was purified by anion exchange. Mössbauer sources were prepared by fusing PbS or PbTe samples with a non-relative preparation $^{119}\text{SbCl}_3$, so that the estimated concentration of impurity

antimony atoms did not exceed 10^{17} at.cm⁻³. The original samples were electronic (with an excess of lead, $n \sim 10^{18}$ cm⁻³) and hole (with an excess of chalcogen, $p \sim 10^{18}$ cm⁻³) type.

The ^{119m}Sb (^{119m}Sn) Mössbauer spectra were measured on an SM-2201 industrial spectrometer at 80 K with a CaSnO₃ absorber (surface tin density 5 μg ms⁻²). The spectrum with such an absorber and a Ca¹¹⁹SnO₃ source was a single line with a FWHM of $T = 0.79 \pm 0.01$ mm s⁻¹, which was taken as the instrumental spectral line width. Typical spectra of PbS: 119Sb and PbTe: 119 Sb samples are shown in the figure; the results of their processing are summarized in the table.

Obviously, ^{119m}Sn⁰ atoms are formed from 119Sb atoms located in the anionic sublattice of PbS (PbTe). The second line (spectrum II) has an isomeric shift characteristic of stannous compounds, and it should be attributed to the ^{119m}Sn²⁺ centers in the PbS (PbTe) cation sublattice (in the nearest environment of these centers there are chalcogen atoms and the interaction of tin with them leads to an isomeric shift close to isomeric shift of the 119Sn spectrum of the SnS and SnTe compounds). Obviously, ^{119m}Sn²⁺ atoms are formed from ^{119m}Sb atoms located in the PbS (PbTe) cation sublattice.

Thus, the location of the localization of impurity antimony atoms in the lattices of lead chalcogenides depends on the nature of the violation of the stoichiometry of the material: in samples with an excess of lead, antimony is localized predominantly in the anionic sublattice, and in samples with an excess of chalcogen, mainly in the cationic donor). The fraction of electrically active antimony atoms depends on the NT of the distribution of antimony between the sublattices, but it is always less than one (this follows from the fact that even in hole samples, a coaxial fraction of antimony atoms appears in the anion sublattice). It is worth noting that the 119Sn center in the anionic sublattice of PbS and PbTe corresponds to a type I spectrum, which is an antisite back defect and, as follows from the independence of the measured shift of spectrum I on the conductivity unit of the material, the charge state of the antisite effect does not depend on the position of the Fermi level. The 119Sn center in the cationic sublattice of PbS and PbTe (which corresponds to the type II and III spectra in PbS and the type II spectrum in PbTe) is an isoelectronic substitutional impurity. As follows from the authors' data [5], the isoelectronic impurity of tin and PbS is electrically active and plays the role of a two-electron donor with negative correlation energy. This explains the dependence of the isomeric shift of the spectrum for such centers on the type of material conductivity that we found: in electronic samples, the spectrum corresponds to the neutral state of the donor center ^{119m}Sn²⁺, and in hole ones, to the doubly ionized state (^{119m}Sn⁴⁺). In the PbTe lattice, the isoelectronic tin impurity is electrically inactive [5: 840] and this is confirmed by our data that the isomeric shift of the spectrum corresponding to these centers is independent of the position of the Fermi level.

Connection, type of conductivity	Spectrum type	Center	I.S.,mm/c	Γ,mm/c	S,%%
n – PbS	I	^{119m} Sn ⁰	2.34	1.34	78
	II	^{119m} Sn ²⁺	3.73	1.33	22
n – PbTe	I	^{119m} Sn ⁰	2.31	1.30	79
	II	^{119m} Sn ²⁺	3.41	1.32	21
n – PbS	I	^{119m} Sn ⁰	2.33	1.33	21
	II	^{119m} Sn ⁴⁺	1.23	1.32	79
n - PbTe	I	^{119m} Sn ⁰	2.33	1.32	5
	II	^{119m} Sn ²⁺	3.43	1.31	95
Inaccuracies			±0.01	±0.02	±0.02

Note: I.S. is the isomeric shift relative to CaSnO₃, H is the spectral width at half maximum, S is the area under the spectrum

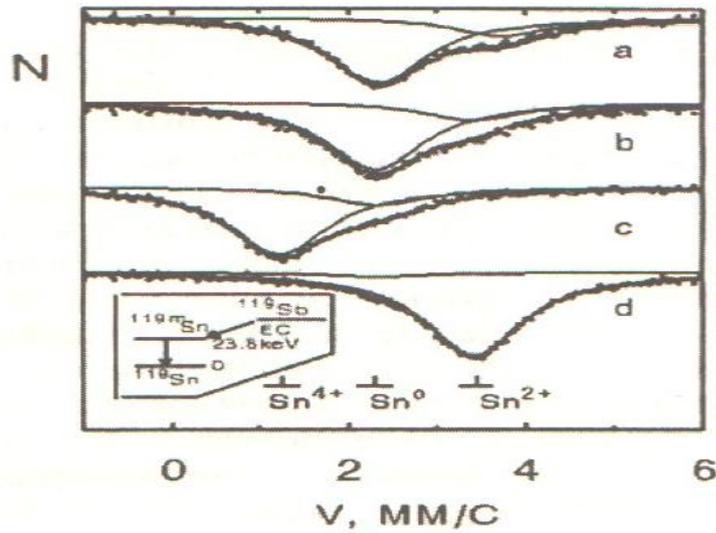


Рис.1.

Emission Mössbauer spectra of ^{119}Sb ($^{119\text{m}}\text{Sn}$) at 80 K for n-PbS (a) n-PbTe (b), p-PbS (c), and p-PbTe (d) samples. The decomposition of the experimental spectra into components corresponding to $^{119\text{m}}\text{Sn}^0$, $^{119\text{m}}\text{Sn}^{2+}$, $^{119\text{m}}\text{Sn}^{4+}$ is shown.

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