

Magnetic properties of $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ alloys

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The magnetic field dependences of the magnetization ($B \leq 9$ T, $T = 2-70$ K) of the samples from a single crystal $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ ($y = 0.01$) ingot, synthesized by the Bridgman method, are studied. It is established that, in accordance with the generally accepted model of the rearrangement of the electronic structure of alloys during doping, there is no paramagnetic contribution of single scandium ions located in the nodes of the metal sublattice in the studied samples. The magnetization of the samples contains several contributions: superparamagnetism of scandium clusters, linear in field diamagnetism of the crystal lattice and the paramagnetism of free electrons, as well as the oscillating contribution of the de Haas-van Alphen effect. The field dependences of the main contribution of scandium clusters are successfully approximated using the Langevin function. The average concentration, magnetic moment and the total magnetic moment of the clusters per unit volume of the sample were determined with an increase in the impurity concentration along the ingot.

Keywords: PbTe-based alloys, 3d-transition metal impurities, rearrangement of electronic structure, field dependences of magnetization, superparamagnetism of scandium clusters.

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1. Introduction

Impurity ions in dilute magnetic semiconductors (DMSs) based on $\text{A}^{\text{IV}}\text{B}^{\text{VI}}$ semiconductors with admixed 3d transition metals substitute atoms in the metal sublattice and may assume two charge states: Im^{2+} and Im^{3+} . The magnetic moments of impurity ions are defined by their spin moments only, and the orbital moments are considered to be „quenched“ [1–3]. The magnetic properties of these DMSs, the majority of which are paramagnetics, are ultimately defined by the concentration of impurity ions in states 2+ and 3+, which have different magnetic moments, and the position of the Fermi level relative to the edges of energy bands and impurity levels [1–9].

Impurity ions $\text{Sc}^{2+}(3d^1)$ in alloys doped with scandium should have the minimum spin ($S = 1/2$) and, consequently, magnetic moments and be magnetically neutral in the $\text{Sc}^{3+}(3d^0)$ state. It is known that the resonance impurity scandium level (E_{Sc}) in $\text{Pb}_{1-x-y}\text{Sn}_x\text{Sc}_y\text{Te}$ ($x = 0-0.045$) alloys is positioned high within the conduction band, much higher than the Fermi level in undoped samples ($E_{\text{Sc}} \approx E_c + 280$ meV at $T = 4.2$ K) [10,11]. Therefore, when the donor Sc impurity concentration increases in the course of doping, the processes of self-ionization of impurity ions ($\text{Sc}^{2+} \rightarrow \text{Sc}^{3+} + e_{\text{band}}$) induce filling of the unoccupied states below the scandium level (first in the valence band, then in the conduction band), $p-n$ conductivity type inversion, and a

monotonic increase in the electron density from 10^{16} to almost 10^{20} cm^{-3} . In this case, Sc ions at the nodes of the metal sublattice should initially be in the electrically active, but nonmagnetic state 3+. Impurity paramagnetism, which is associated with the emergence of magnetically active scandium ions in state 2+, may be observed only when the conduction band fills up to the resonance scandium level, the Fermi level is pinned by this level, and the electron density reaches saturation ($n_{\text{sat}} \approx 10^{20} \text{ cm}^{-3}$).

At the same time, it was demonstrated experimentally that $n\text{-Pb}_{1-y}\text{Sc}_y\text{Te}$ [12] and even $p\text{-Pb}_{1-x-y}\text{Sn}_x\text{Sc}_y\text{Te}$ with the Fermi level located in the valence band [13] feature a considerable „Curie-Weiss“ paramagnetic contribution to magnetization, which is apparently attributable to the existence of ions or clusters of $\text{Sc}^{2+}(3d^1)$ ions in crystals, at low temperatures. These results contradict the above model of rearrangement of the electronic structure of alloys in the process of doping with scandium and warrant further study of the magnetic properties of these DMSs. In view of this, the focus of the present study is on the field dependences of magnetization ($B \leq 9$ T, $2.0 \leq T \leq 70$ K) of samples from a single-crystal $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ ($y = 0.01$) ingot with the scandium concentration varying along the ingot. The study was aimed at identifying the key contributions to magnetization, verifying the above model of rearrangement of the electronic structure in the process of doping, and determining the magnetic moments

of paramagnetic centers and the dependence of their concentration on the impurity scandium concentration in alloys.

2. Samples and experimental procedure

Samples from a single-crystal $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ ingot with nominal scandium concentration $y = 0.01$, which was synthesized by the vertical Bridgman method from a mixture of PbTe and Sc_5Te_8 , were examined. The initial compounds were synthesized in advance from high-purity components (Pb — 99.9999%, Sc — 99.9%, Te — 99.9997%). The method of preparation of the initial components and synthesis of single crystals was discussed in detail in [10,11,14]. The ingot was cut perpendicularly to the growth axis, which was aligned with crystallographic direction $\langle 111 \rangle$ to within several angular degrees, by string cutting into 28 disks ~ 1.5 mm in thickness. These disks were numbered, and the sample numbers used below correspond to this numeration.

The homogeneity of samples and the distribution of impurity scandium along the ingot were determined by X-ray fluorescence microanalysis using a LEO SUPRA 50VP (LEO Carl Zeiss SMT Ltd, Germany) scanning electron microscope with an INCA Energy+ (Oxford Instruments, England) microanalysis system. All samples were fairly homogeneous within the limits of experimental error. In contrast to $\text{Pb}_{1-x-y}\text{Sn}_x\text{Cr}_y\text{Te}$, $\text{Pb}_{1-x-y}\text{Sn}_x\text{V}_y\text{Te}$, and $\text{Pb}_{1-y}\text{Fe}_y\text{Te}$ alloys where microscopic inclusions of compounds formed by impurity atoms and tellurium and regions enriched with impurity atoms were found [5,14–16], all $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ samples are free from such regions and noticeable inclusions of the minor phase. This implies that the limit of solubility of impurity scandium in the ingot was not exceeded (possibly, due to the specifics of doping with scandium: dissolution of scandium from Sc_5Te_8 in lead telluride in the process of growth).

In contrast to PbTe -based alloys doped with Cr, V, and Fe, the distribution of Sc along the single-crystal ingot is inverse in nature: the impurity concentration increases from the end to the origin of the ingot (Fig. 1). This anomalous impurity distribution in $\text{A}^{\text{IV}}\text{B}^{\text{VI}}$ semiconductors has been observed earlier in $\text{Ge}_{1-x}\text{Sn}_x\text{Te}$ alloys doped with Mn and has been attributed to the fact that impurity atoms are pushed back to the origin of the ingot in the process of growth of a single crystal due to the formation of heavy MnTe complexes in the liquid phase [10,17,18]. Therefore, the exponential $y(L)$ dependence typical of impurity Mn was used to approximate the distribution of Sc in the studied $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ samples [10]:

$$y = y_0 + A_1 \exp\left(-\frac{L}{t_1}\right) + A_2 \exp\left(-\frac{L}{t_2}\right), \quad (1)$$

where $L = h/h_0$ is the relative coordinate of a disk, h is the distance from the ingot origin to the disk center, h_0 is

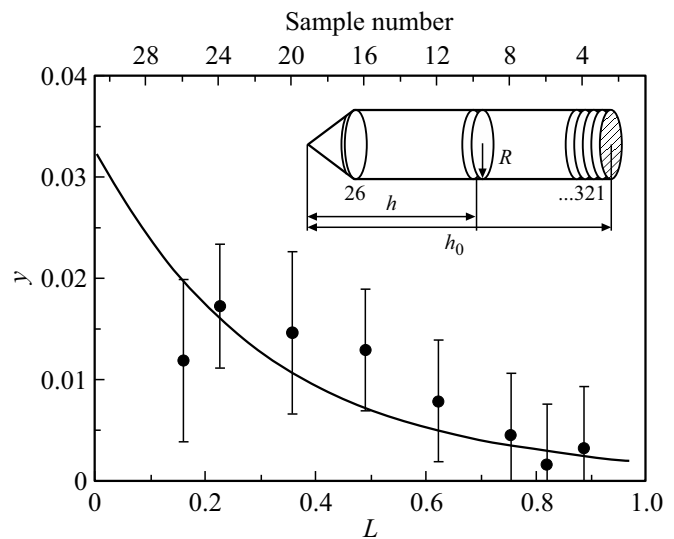


Figure 1. Distribution of impurity scandium along the length of the single-crystal $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ ($y = 0.01$) ingot (dots correspond to X-ray fluorescence microanalysis data, and the curve is the result of approximation in accordance with formula (1) [10]).

the total length of the ingot, and A_1, A_2, t_1, t_2 are fitting parameters.

The spatial distribution of scandium along the ingot was estimated using the least-squares method and Eq. (1) with account for the fact that the area under the curve should correspond to the overall concentration of scandium in the ingot ($y = 0.01$). In what follows, we assume that the concentration of impurity scandium increases monotonically from the end of the ingot to its origin in accordance with the approximation curve plotted in Fig. 1.

Samples similar to a rectangular parallelepiped in shape and with a mass of 40–80 mg were cleaved out of disks at liquid nitrogen temperature to examine their magnetic properties. The field dependences of magnetization of $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ alloys were studied at a temperature of 2.0–70 K in magnetic fields up to 9 T using a vibration magnetometer of a PPMS-9 (Quantum Design, USA) setup.

3. Field dependences of magnetization

It was found that field dependences of magnetization $M(B)$ of all the studied samples at low temperatures contain a contribution linear in field, which is dominant in high magnetic fields ($B > 2$ –4 T), and a paramagnetic contribution of the „Brillouin“ type that is nonlinear in field and reaches saturation in high fields of the indicated intensity (Fig. 2, a). In addition, certain samples (in particular, samples 10 and 20) feature an oscillating magnetization dependence, which is apparently related to the de Haas–van Alphen (DHVA) effect, in the linear sections of their $M(B)$ dependences.

The contribution linear in field may have both negative (primarily in samples from the end part of the ingot

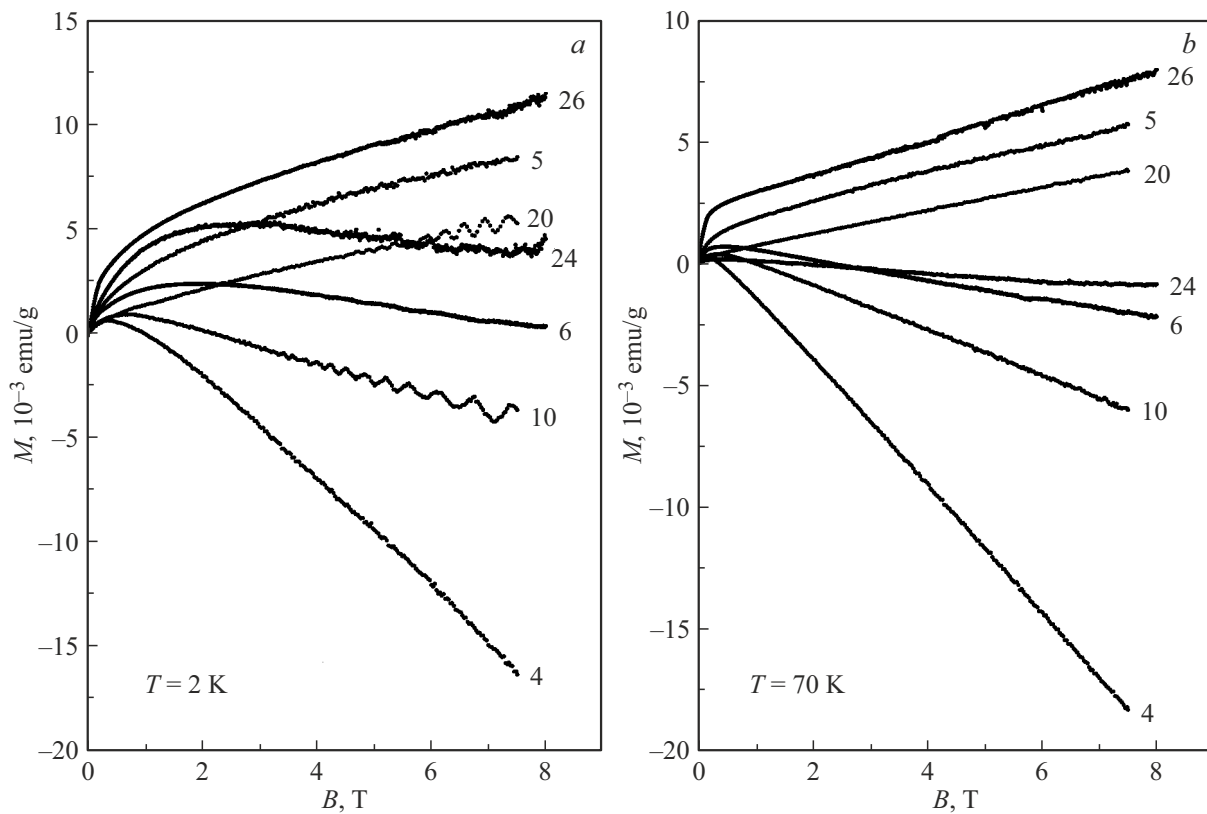


Figure 2. Field dependences of magnetization of $Pb_{1-y}Sc_yTe$ alloys at temperatures $T = 2$ K (a) and $T = 70$ K (b) (the numbers next to curves correspond to the sample numbers).

with the lowest electron densities) and positive (primarily in samples with the highest electron densities that were positioned closer to the origin of the ingot) slopes. Magnetic susceptibility χ corresponding to this contribution increases nonmonotonically with scandium concentration along the ingot length from approximately $-2.5 \cdot 10^{-7}$ to $+0.8 \cdot 10^{-7}$ $\text{emu} \cdot \text{g}^{-1} \cdot \text{Oe}^{-1}$. Since the theoretical and experimental values of the diamagnetic susceptibility of the crystal lattice for undoped PbTe and PbTe doped with $3d$ transition metals fall within the range from $-3.0 \cdot 10^{-7}$ to $-3.5 \cdot 10^{-7}$ $\text{emu} \cdot \text{g}^{-1} \cdot \text{Oe}^{-1}$ [1,4,19–21], it is fair to assume that the magnetization contribution linear in field consists primarily of the diamagnetic contribution of the crystal lattice and the generally paramagnetic contribution of free electrons, which increases in magnitude with concentration of impurity scandium along the ingot length. It is unclear why the contribution of free electrons varies irregularly with concentration of impurity scandium along the ingot length. However, it is likely that the reason for this is related to the specifics of paramagnetism of free carriers in many-valley semiconductors with a nonparabolic and highly anisotropic electron spectrum (all alloys based on lead telluride belong to this class of compounds [22]) and to the fact that the samples were oriented arbitrarily relative to the magnetic field in the measurements of field dependences of magnetization.

When the measurement temperature increases from 2 to 70 K, the linear contributions to magnetization of the studied samples remain almost unchanged, while the DHVA oscillations and the „Brillouin“ nonlinearity of the $M(B)$ dependences vanish rapidly (Fig. 2, b). At the same time, an additional feature of the field dependences of magnetization becomes visible at higher temperatures: a contribution nonlinear in field that saturates rapidly with increasing field intensity (in fields up to 1 T) and was masked by the „Brillouin“ contribution at low temperatures. A similar contribution to magnetization has been observed earlier in $Pb_{1-y}Fe_yTe$ alloys and has been associated with the existence of magnetic clusters of impurity ions, the concentration and mean magnetic moment of which increased with impurity iron concentration in the samples [6–8,23].

Assuming that this additional contribution in the present case is also related to the formation of clusters of scandium ions, we used the technique proposed in [7,8] to isolate it. It is assumed in this approach that the „Brillouin“ contribution to magnetization is linearized in the studied range of magnetic fields at $T = 70$ K and is combined with other contributions linear in field (diamagnetism of the crystal lattice and the contribution of free carriers). Therefore, the contribution of clusters of scandium ions in each sample may be determined by subtracting the complete linear run of magnetization from the field dependence

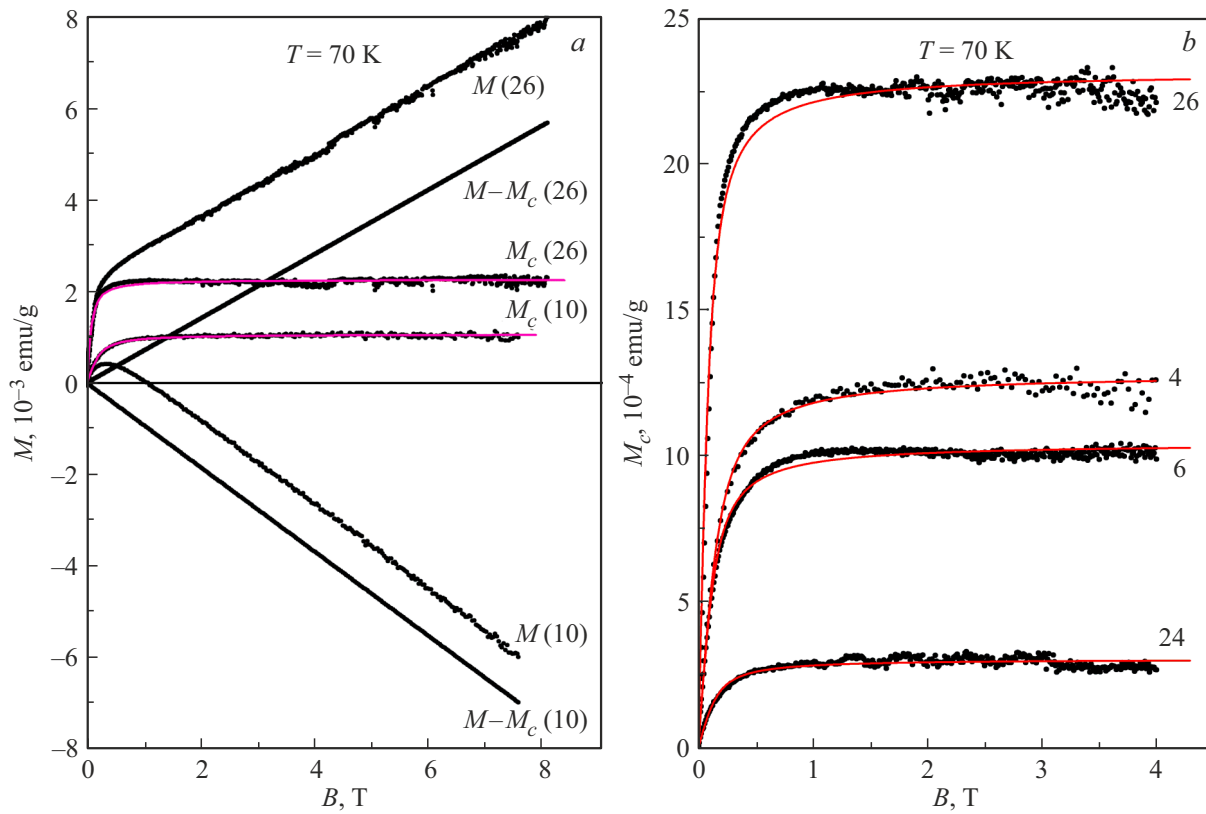


Figure 3. Examples of isolation of the contribution of magnetic clusters (M_c) to the magnetization of samples 10 and 26 (*a*) and field dependences of this contribution $M_c(B)$ for a series of samples (*b*) (the numbers next to curves correspond to the sample numbers).

of magnetization M at $T = 70$ K. Figure 3, *a* illustrates the results of application of this method for isolating the contribution of magnetic clusters (M_c) to samples 10 and 26, and Fig. 3, *b* shows the field dependences of the obtained cluster contribution for samples 4, 6, 24, and 26.

Assuming that the examined samples are similar to $Pb_{1-y}Fe_yTe$ alloys studied earlier in that the $M_c(B)$ contribution of clusters of scandium ions remains unchanged at lower temperatures, we then subtracted it from the initial $M(B)$ dependences measured at $T = 2$ K for all samples (Fig. 4, *a*). Finally, we determined the $M_0(B)$ contribution linear in field, which contains the contributions of the crystal lattice and free carriers at $T = 2$ K, using the sections of $(M - M_c)(B)$ dependences corresponding to high fields ($B > 5$ T) and subtracted it from the $(M - M_c)(B)$ dependences for each sample. The $(M - M_c - M_0)(B)$ (Fig. 4, *b*) dependences obtained this way should contain only the paramagnetic contribution to magnetization of the studied $Pb_{1-y}Sc_yTe$ alloys and may be used to determine the parameters of paramagnetic centers in samples.

4. Parameters of clusters and paramagnetic centers

The experimental $M_c(B)$ dependences from Figs. 3, *a* and *b* were used to determine the parameters of scandium

clusters, which, just as in earlier studies [7,8,23], were represented as small particles containing a large number of impurity atoms or ions. It was assumed that the magnetic moments of clusters, which are oriented arbitrarily in zero magnetic field, become ordered rapidly as the magnetic field intensity increases. Therefore, the classical Langevin function was used to approximate the field dependences of cluster contribution $M_c(B)$ [24]:

$$M_c(B) = \frac{1}{\rho} N_c m_c \left[\coth\left(\frac{m_c B}{k_B T}\right) - \left(\frac{k_B T}{m_c B}\right) \right], \quad (2)$$

where $\rho \approx 8.2 \text{ g/cm}^3$ is the density of $PbTe$, N_c is the mean concentration of clusters, m_c is the mean magnetic moment of clusters, and k_B is the Boltzmann constant.

The results of approximation for the studied samples are represented by solid curves in Figs. 3, *a* and *b*, and the corresponding cluster parameters are listed in Table 1. It is evident that, just as in $Pb_{1-y}Fe_yTe$ alloys studied earlier, the mean number of impurity atoms or ions in clusters is at the level of $(1-2) \cdot 10^3$. At the same time, in contrast to $Pb_{1-y}Fe_yTe$ alloys, the mean magnetic moment of clusters (m_c) and, consequently, the mean size of clusters vary only weakly and irregularly along the ingot length. The mean values of cluster concentration N_c and total magnetic moment of clusters in unit volume $N_c m_c$ also vary nonmonotonically along the ingot length and are

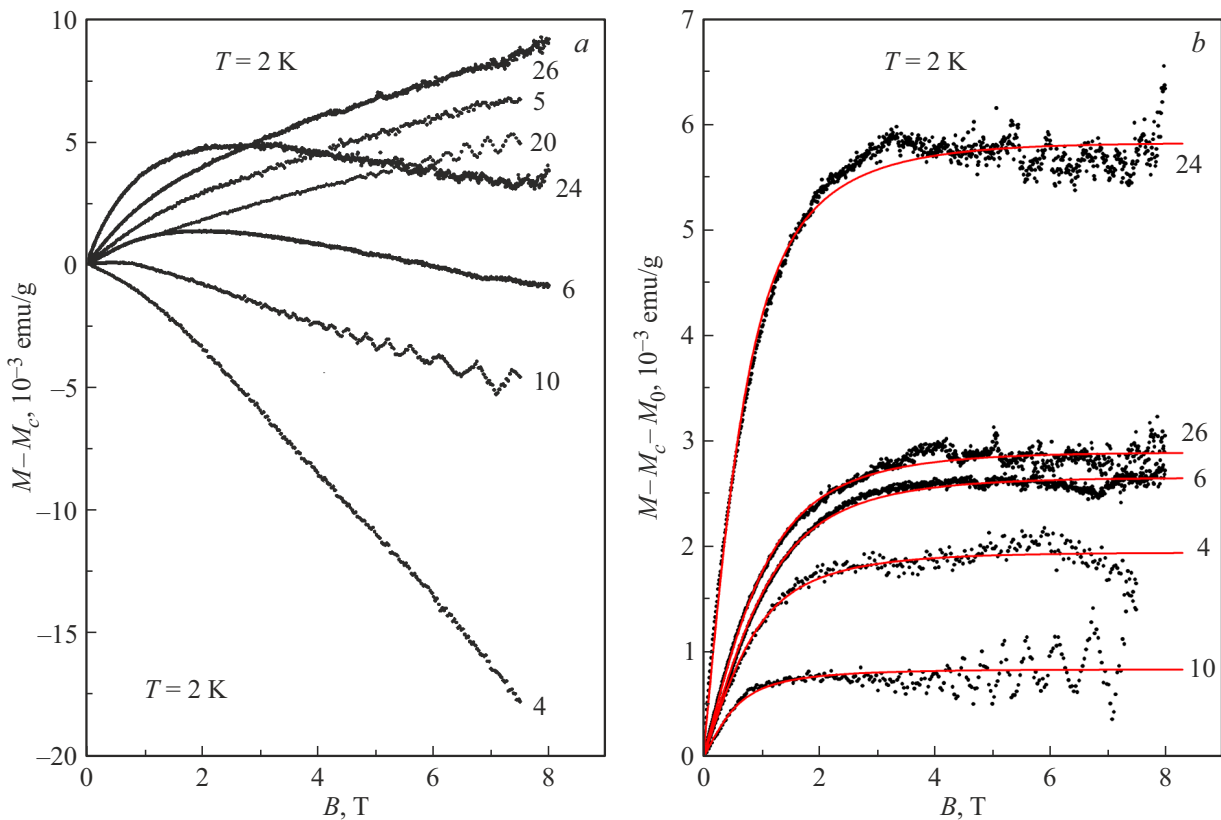


Figure 4. Field dependences of magnetization of $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ alloys at temperature $T = 2$ K after the subtraction of the contribution of magnetic clusters (a) and subsequent subtraction of the contribution linear in field (b) (the numbers next to curves correspond to the sample numbers).

Table 1. Parameters of scandium clusters determined based on the field dependences of magnetization M_c of the $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ samples at $T = 70$ K

Sample number	y	m_c, μ_B	$N_c, 10^{14} \text{ cm}^{-3}$	$N_c m_c, 10^{17} \mu_B/\text{cm}^3$
4	0.0024	1318	8.6	11.3
5	0.0027	684	21.0	14.4
6	0.0029	1618	5.7	9.2
10	0.0041	1054	8.9	9.4
20	0.0106	1128	2.3	2.6
24	0.0160	1455	1.9	2.7
26	0.0197	2333	8.8	20.4

approximately an order of magnitude lower than those in $\text{Pb}_{1-y}\text{Fe}_y\text{Te}$ alloys.

It was taken into account in the determination of the key parameters of paramagnetic centers in the studied $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ alloys that scandium ions are, in contrast to iron and vanadium impurity ions that are magnetically active in both charge states ($2+$ and $3+$) and may produce additive contributions to magnetization, magnetically active only in the $\text{Sc}^{2+}(3d^1)$ state with spin $S = 1/2$. Therefore, the experimental $(M - M_c - M_0)(B)$ dependences (Figs. 4, b

and 5) for each sample were approximated with the modified Brillouin function [7,9,25,26]:

$$M(B) = \frac{1}{\rho} N g S \mu_B \left(\frac{2S+1}{2S} \coth \left(\frac{2S+1}{2S} \frac{g S \mu_B B}{k_B (T - T_0)} \right) - \frac{1}{2S} \coth \left(\frac{1}{2S} \frac{g S \mu_B B}{k_B (T - T_0)} \right) \right), \quad (3)$$

where $\rho \approx 8.2 \text{ g/cm}^3$ is the density of PbTe , N and S are the concentration and spin of scandium ions in the $\text{Sc}^{2+}(3d^1)$ state, the g -factor is equal to 2, μ_B is the Bohr magneton, k_B is the Boltzmann constant, and T_0 is a fitting parameter that covers the probable weak interaction of impurity ions.

Concentration of magnetic ions N and temperature T_0 were used as fitting parameters in plotting the theoretical field dependences of magnetization. The results of calculations for two of the studied samples are represented by dashed curves in Fig. 5. It turned out that an acceptable fit between the theoretical and experimental curves is impossible to obtain in any sample at a fixed spin of scandium ions ($S = 1/2$), since the experimental magnetization values increase much faster with B (and tend to saturation in significantly weaker magnetic fields) than the theoretical ones. A closer fit may be obtained if one

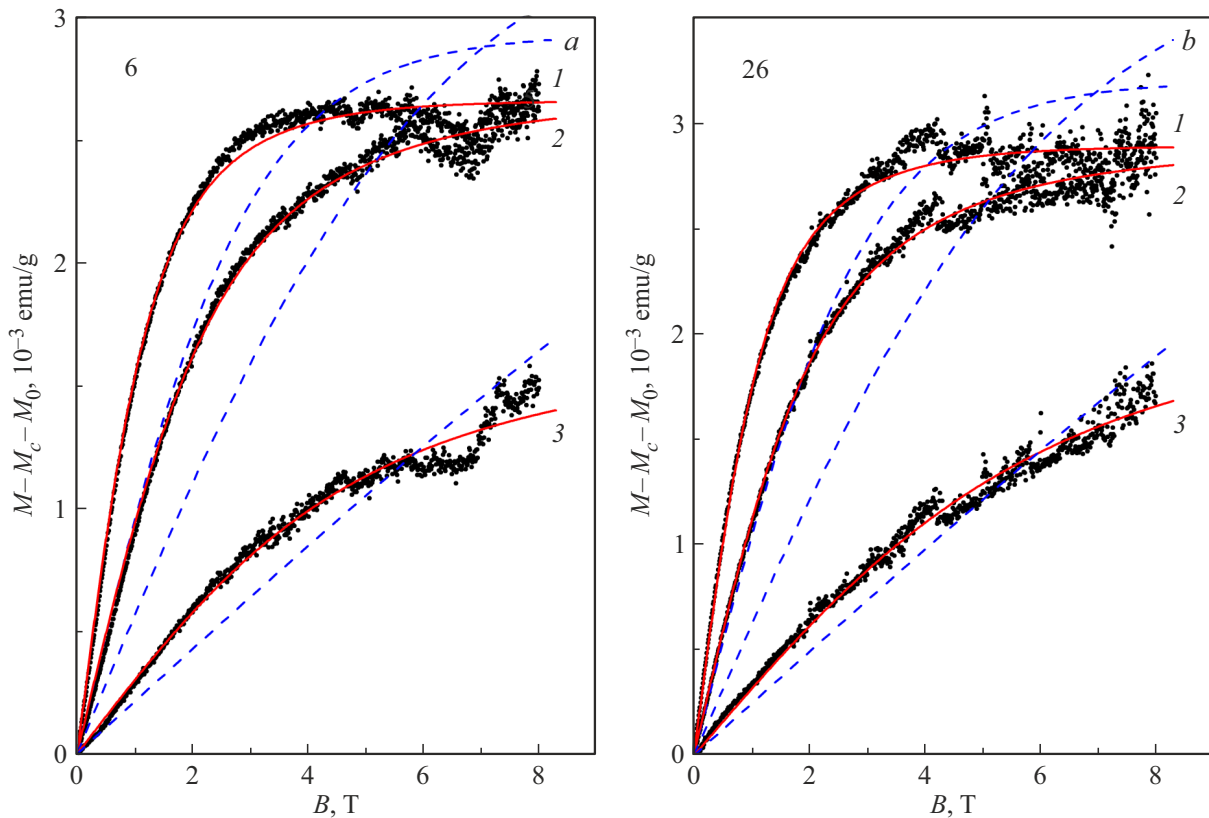


Figure 5. Field dependences of magnetization $(M - M_c - M_0)(B)$ of samples 6 and 26 approximated with the modified Brillouin function (dashed curves) and the Langevin function (solid curves) at different temperatures T , K: 1 — 2, 2 — 4.2, 3 — 15 K.

assumes that clusters of impurity atoms or ions serve as magnetic centers both at $T = 70$ K and at low temperatures and introduces the third fitting parameter (magnetic moment of clusters).

In order to verify this assumption, we approximated the $(M - M_c - M_0)(B)$ dependences from Fig. 5 with Langevin function (2) regarding mean magnetic moment m_c and cluster concentration N_c as variable parameters (solid curves in Fig. 5). Using the same approach, we also plotted the theoretical curves in Fig. 4, *b* for a series of samples studied at $T = 2$ K. The key parameters of clusters in all samples are listed in Tables 2 and 3. It is evident that the fit between the theoretical and experimental curves improved appreciably. At low temperatures, the mean magnetic moment of clusters is ~ 250 times lower (5–6 Bohr magnetons), while the mean concentration of clusters is 3 orders of magnitude higher than the corresponding values at $T = 70$ K (see Tables 1, 2, and 3). Therefore, since the mean contribution of clusters with small magnetic moments to overall magnetization $M(B)$ of samples may be characterized by the $N_c m_c$ product, it is approximately two times higher than the contribution of clusters with large magnetic moments.

Thus, it turned out that only the Langevin function with the assumed formation of magnetic clusters of scandium atoms or ions provides a satisfactory approximation of

Table 2. Parameters of scandium clusters determined based on the field dependences of magnetization $M - M_c - M_0$ of two $Pb_{1-y}Sc_yTe$ samples at different temperatures

Sample number	T , K	m_c , μ_B	N_c , 10^{17} cm^{-3}	$N_c m_c$, $10^{17} \mu_B / \text{cm}^3$
6	2.0	4.2	5.6	23.7
	4.2	5.0	4.7	23.6
	15	10.0	1.5	15.1
26	2.0	4.6	5.6	25.9
	4.2	5.8	4.4	25.3
	15	7.8	2.5	19.3

Table 3. Parameters of scandium clusters determined based on the field dependences of magnetization $M - M_c - M_0$ for $Pb_{1-y}Sc_yTe$ at $T = 2$ K

Sample number	y	m_c , μ_B	N_c , 10^{17} cm^{-3}	$N_c m_c$, $10^{17} \mu_B / \text{cm}^3$
4	0.0024	5.6	3.1	17.2
5	0.0027	3.6	4.7	16.9
6	0.0029	4.2	5.6	23.7
10	0.0041	9.3	0.80	7.4
20	0.0106	4.4	1.6	7.2
24	0.0160	7.0	7.4	51.6
26	0.0197	4.6	5.6	25.9

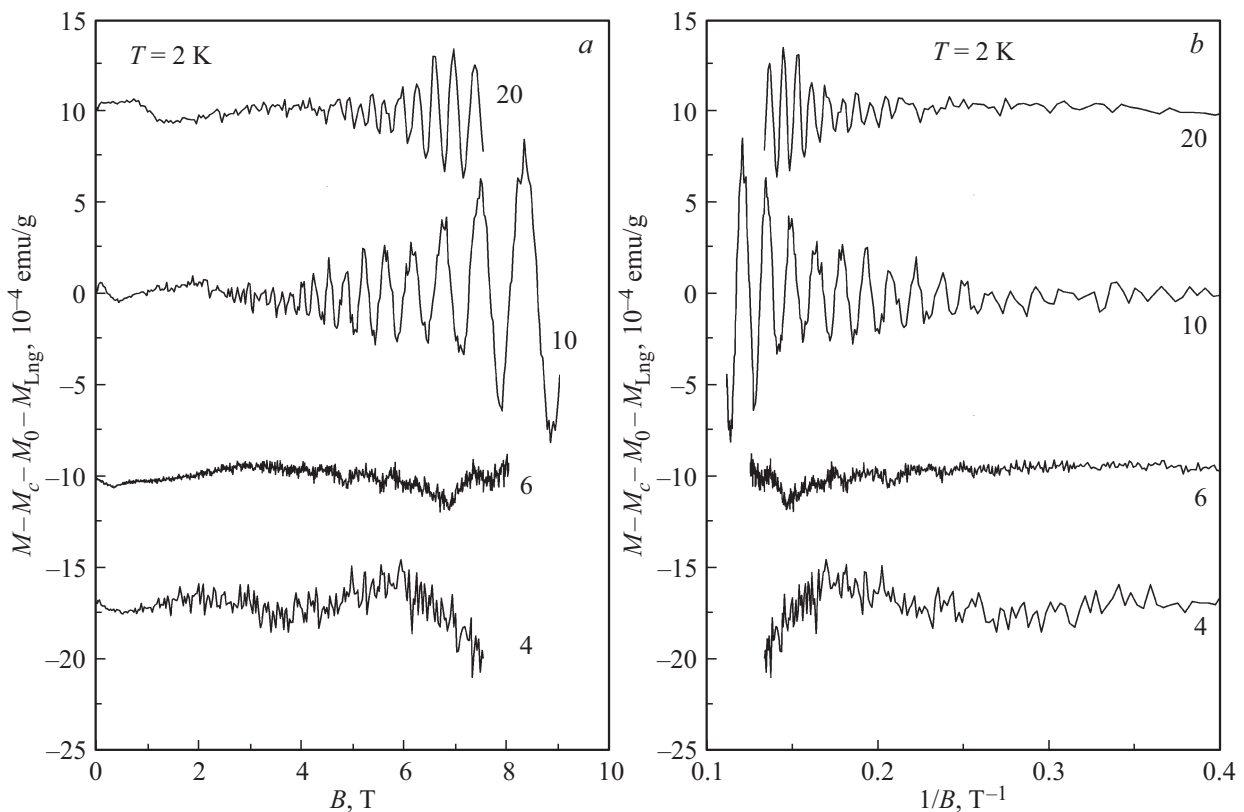


Figure 6. De Haas–van Alphen oscillations in direct and reciprocal magnetic fields at temperature $T = 2$ K for certain $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ samples.

the field dependences of magnetization both at $T = 70$ K and at low temperatures $T < 15$ K. Therefore, one can conclude that, just as was assumed in the common model of rearrangement of the electronic structure of PbTe-based alloys doped with scandium (see Introduction), scandium ions in charge state $3+$ located at the nodes of the lead sublattice do not indeed provide contribution to magnetization. At the same time, the entire paramagnetic contribution, which was referred to above as the „Curie–Weiss“ or „Brillouin“ contribution, is apparently attributable not to the paramagnetism of single scandium ions, but to the superparamagnetism of clusters of Sc^{2+} ions or scandium atoms that may be located both at the nodes and at interstitial sites of the lattice and are characterized by a wide distribution over the magnetic moment magnitude (i.e., over the cluster size), which may vary from several to several thousand Bohr magnetons.

At the initial stage of calculations (when the contribution of magnetic clusters determined at $T = 70$ K was subtracted from the initial field dependence of magnetization at $T = 2$ K), we obviously failed to suppress completely the contribution of clusters with small magnetic moments, since it becomes substantially linearized already at $T = 15$ K in the studied range of magnetic fields (see Fig. 5). It turned out as a result that the nonlinear magnetization part of the studied samples, which saturates rapidly with increasing B , at $T = 70$ K contains only the contribution of clusters

with large magnetic moments, while the contribution of clusters with small magnetic moments became linearized and remained unaccounted for.

5. De Haas–van Alphen oscillations

As was already noted, quantum oscillations of magnetic susceptibility (de Haas–van Alphen effect) are observed at helium temperatures in high magnetic fields ($B > 2$ T) in field dependences of magnetization of several samples against the background of the magnetization contribution linear in field (see Figs. 2, *a* and 4). In order to cancel out the monotonic magnetization contribution and isolate the oscillating one, the theoretical „Langevin“ $M_{\text{Lng}}(B)$ field dependences, which are represented by solid curves in Figs. 4, *b* and 5 were subtracted from the experimental $(M - M_c - M_0)(B)$ dependences. The results of this procedure, which was performed at $T = 2$ K for several samples, are presented in Fig. 6 (oscillating curves are shifted sequentially upward relative to the curves for sample 4). It is evident that oscillations are separated almost completely from the monotonic run of the curve after subtraction of the „Langevin“ contribution of clusters. This is indicative of a fine accuracy of the procedure of successive isolation of the primary contributions to magnetization of the studied alloys.

Oscillations are observed in a wide range of magnetic fields, and the maximum oscillation amplitude corresponds to samples 4, 10, and 20 with electron density $n \approx 1.3 \cdot 10^{17}$, $4.3 \cdot 10^{19}$, and $8.2 \cdot 10^{19} \text{ cm}^{-3}$, respectively [10]. Just as in $\text{Pb}_{1-y}\text{Fe}_y\text{Te}$ alloys studied in [7], oscillations are not monochromatic and contain a superposition of at least two close frequencies. This is attributable to the complexity of the multi-ellipsoid Fermi surface in the studied alloys and the uncertainty in orientation of samples relative the magnetic field vector in measurements of the field dependences of magnetization and complicates considerably the determination of carrier densities based on the oscillation period in the reciprocal magnetic field [7,15]. However, with the wide range of variation of electron density with scandium concentration in the studied samples taken into account, the electron densities estimated based on the period of DHVA oscillations agree sufficiently well (within 15–50%) with the data of Hall measurements [10]. This fact, the high mobilities of electrons in samples ($10^3\text{--}10^5 \text{ cm}^2/(\text{V} \cdot \text{s})$ at $T = 4.2 \text{ K}$ [10]), and the experimental observation of quantum DHVA oscillations itself suggest that scandium clusters exert only a weak influence on the homogeneity and structural perfection of samples and on the parameters of the band structure in heavily doped $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ alloys.

6. Conclusion

The field dependences of magnetization ($B \leq 9 \text{ T}$, $T = 2.0\text{--}70 \text{ K}$) of single-crystal $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ alloys with the impurity scandium concentration varying along the ingot length ($y < 0.02$) were examined. It was found that magnetization $M(B)$ includes several key contributions: the sum of diamagnetism of the crystal lattice and the generally paramagnetic free-electron contribution ($M_0(B)$) that are linear in field and temperature-independent, the oscillating contribution of the DHVA effect in quantizing magnetic fields, and the contribution that reaches saturation rapidly in stronger fields ($B < 1 \text{ T}$). The rapidly saturating contribution cannot be approximated with the modified Brillouin function, which characterizes the paramagnetic contribution of all the other impurity ions of $3d$ transition metals in PbTe -based alloys, and is apparently associated with the superparamagnetism of clusters of scandium atoms or ions. Thus, it was demonstrated that, in accordance with the common model of rearrangement of the electronic structure of PbTe -based alloys in the process of their doping with $3d$ transition metals, the studied samples do indeed lack the paramagnetic „Brillouin“ contribution of single scandium ions located at the nodes of the metal sublattice.

The contribution of scandium clusters ($M_c(B)$) to magnetization of the studied alloys was isolated and approximated fairly closely with the classical Langevin function ($M_{\text{Lng}}(B)$). The mean values of cluster concentration N_c , magnetic moment of clusters m_c , and total magnetic moment of clusters in unit volume $N_c m_c$ were determined, and

their dependences on the scandium impurity concentration were obtained. It was demonstrated that the magnetic moments of clusters in the samples vary widely (from several to several thousand Bohr magnetons), the concentration of clusters is as high as $\sim 7 \cdot 10^{17} \text{ cm}^{-3}$, and the primary contribution to the sample magnetization is produced by moderate-sized clusters.

Subtracting successively the above-mentioned primary contributions from the overall magnetization of the samples, we managed to isolate oscillating contribution $(M - M_c - M_0 - M_{\text{Lng}})(B)$ of the de Haas–van Alphen effect. The densities of free electrons in the samples were estimated based on the period of oscillations of this contribution in the reciprocal magnetic field. The obtained values are in a satisfactory agreement with the Hall measurement data. This fact and the experimental observation of quantum magnetization oscillations itself suggest that magnetic clusters exert only a weak influence on the homogeneity, physical properties, and electronic structure of heavily doped $\text{Pb}_{1-y}\text{Sc}_y\text{Te}$ alloys.

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Conflict of interest

The authors declare that they have no conflict of interest.

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