phase boundaries produced in orthoferrities in the case of spin reorientation, $H=0$. The corresponding equations are Lorentz-invariant, and the characteristic velocity coincides with the minimal phase velocity of the spin waves c. In our problem there must of necessity be present a strong magnetic field that upsets the Lorentz invariants of the equations and leads to a nontrivial dependence of the DW structure on its velocity.

An important singularity of the problem of the motion of 90° DW is that when the external field deviates from H_{tr}, induced motion of the DW must arise. Thus, it is possible to investigate experimentally the dynamics of nonlinear waves in AFM, and this can be of interest for the physics of antiferromagnetism. Such an experiment can be carried out, e.g., in accordance with the scheme proposed by Chetkin *et al.*,[2] the only difference being that the AFM sample should be placed in a constant magnetic field close to H_{tr} (e.g., 6 kOe for $CuCl_2 \cdot 2H_2O$ or 94 kOe for MnF_2). The DW velocity is linear in ΔH as $\Delta H \to 0$, but when ΔH is increased the velocity $v(H)$ saturates, i.e., nonlinear motion of the DW should set in. The transition to the most interesting nonlinear regime occurs at sufficiently low value of $(H-H_{tr}) \sim \pi\lambda M_0$ [see (22)], e.g., at $H-H_{tr} \sim 10$ Oe, if $\lambda \sim 10^{-2}$ and $M_0 \sim 10^3$ G.

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[1)] $[\mathbf{mh}] \equiv \mathbf{m} \times \mathbf{h}$, etc.

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Investigation of mictomagnetic MnBi alloys

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MnBi alloys in the mictomagnetic state were obtained and subjected to x-ray-diffraction, microstructural, and differential-thermal analysis. The electric, magnetic, and elastic properties were investigated, as well as the effect of high hydrostatic pressure (up to 10 kbar) on the temperature dependences of the magntization. It was established that the mictomagnetism arises in two-phase system consisting of the high-temperature MnBi phase and Bi. New data were obtained on the magnetic phase transitions and on the influence of hydrostatic compression on the mictomagnetic and superparamagnetic properties of MnBi. Exchange interactions in the investigated alloys are discussed on the basis of these new data.

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INTRODUCTION

The physics of magnetic phenomena has recently been enriched with new concepts (spin glass, mictomagnetism,[1] speromagnetism, asperomagnetism[2]) that describe various disordered states of a spin system. Despite the intensive theoretical and experimental investigations, many aspects of the magnetic behavior of these systems remain unclear, to a considerable degree because of insufficient knowledge of the mechanism of the exchange spin coupling in each concrete case.

We report in this paper the production and investigathat of MnBi alloys whose magnetic properties differ radically from those hitherto known (for MnBi) and recall the mictomagnetic Cu-Ni and Au-Fe systems that were extensively investigated by Beck.[1] According to his ideas, mictomagnetism is determined by the presence of ferro- and antiferromagnetic clusters that are randomly frozen in a nonmagnetic spin-glass matrix. When the temperature is lowered in the region of T_0 (the freezing temperature of spin glass) a transition from the superparamagnetic to the mictomagnetic state takes place. The characteristic symptoms of this state are: a) the absence of hysteresis or saturation of the magnetization; b) very large changes in the magnetic parameters, depending on the magnetic annealing of

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the alloy. This combination of properties was observed by us in the MnBi alloys, which are known[3,4] to be high-coercivity magnetic materials.

Much interest has been paid in recent years to thin ferromagnetic MnBi films, which can be used as magneto-optical media for information recording.[5] A major shortcoming of manganese-bismuth alloys as magnetic materials, however, is the instability of their magnetic characteristics, due to the presence of magnetic phase transitions that are accompanied by changes in the crystal structure. We shall show presently that an investigation of these alloys in the mictomagnetic state yields new data on the exchange interactions and phase transitions in MnBi.

We describe in this paper a procedure for the preparation of mictomagnetic Mn-Bi alloys and the results of their x-ray diffraction, microstructural, and differential thermal (DTA) analysis, as well as of the measurement of the temperature dependence of the electric resistance, of the magnetization isotherms, and of the elastic moduli. We investigated the influence of high hydrostatic pressure (up to 10 kbar) on the magnetization and on the transition temperature T_0. We note that the mictomagnetic and superparamagnetic properties of alloys at high hydrostatic pressures have not been investigated before by anyone.

ALLOY PREPARATON AND EXPERIMENTAL PROCEDURE

The Mn-Bi alloys were obtained in the following manner: finely ground and well mixed powders of chemically pure Mn (99.8%) and Bi (99.999%) in a ratio 45 at.% Mn and 65 at.% Bi were presented into a bar, which was inserted in an alundum crucible placed in a quartz ampoule. The ampoule was evacuated to 5×10^{-3} Torr and then filled with argon and sealed. The alloys were synthesized in two stages: 1) melting in an induction furnace, in which the temperature was raised in 1.5 hours to 1000 °C and kept at this temperature for 20 minutes; 2) annealing at 440 ± 10 °C in a "selite" furnace for two hours and then slowly cooling the alloy to room temperature (in the turned-off furnace).

The x-ray photographs of the powdered samples were obtained with an RKD-57.3 camera by an asymmetrical method in chromium and iron radiation. The lattice periods were determined by graphically extrapolating to zero the functions

$$a,\ c=f(\varphi),\quad \varphi=\cos^2\theta/\sin\theta+\cos^2\theta/\theta.$$

The thermograms were obtained by the DTA method, using an optical derivatograph DO-102, in an argon atmosphere, with calcined aluminum oxide as the standard. The apparatus made it possible to record, besides the thermal effects, the change in the weight of the investigated sample as it was heated. The error in the determination of the phase-transition temperature was ±10°.

The magnetic measurements were made with a Domenicali pendulum magnetometer in fields up to 16 kOe. At high temperatures, to prevent oxidation, the sample was placed in an argon-filled glass ampoule. The magnetization at high hydrostatic pressure (up to 10 kbar) was measured by a procedure similar to that described by us earlier.[6] The elastic properties (the Young and shear moduli) were measured by a resonance method using a compound vibrator and piezo-quartz excitation at frequencies of the order of 100 kHz. The resistivity of the Mn-Bi alloys was measured by the usual potentiometer method with direct current.

MEASUREMENT RESULTS

The microstructural and x-ray diffraction phase analyses, as well as the DTA thermograms, have shown that the Mn-Bi alloys synthesized by us consist of two phases, Bi and MnBi, in approximately equal amounts. On the x-ray diffraction patterns obtained at room temperatures, all the MnBi-phase interference lines were identified on the basis of a hexagonal lattice of the NiAs type (space group Pb_3/mmc); the obtained unit cell parameters were $a=4.340\pm0.005$ Å and $c=5.980\pm0.007$ Å, in good agreement with the data published by Heikes[7] for the high-temperature (HT) modification of MnBi.

On the DTA thermograms, thermal effects were found at the temperatures 483, 543 (melting of eutectic MnBi + Bi), 630, and 790 K (decomposition of MnBi into Mn and liquid Bi). These temperatures correspond to the phase diagrams of two-phase systems consisting of Bi and of the HT phase of MnBi (see, e.g., Ref. 8). A new result obtained in the present paper is the observation, at 483 K, of a phase transition connected with heat release.

Figure 1 shows the magnetization isotherms $\sigma(H)$ measured in fields up to 16 kOe at various temperatures from 77 to 547 K. It is seen from the plots that no saturation of the magnetization appears in the entire temperature and field interval investigated. We note that measurements in strong pulsed magnetic fields, which will be separately reported, have shown that $\sigma(H)$ remains nonlinear at room temperatures up to 250 kOe. The variation of σ with the field is fully reversible, as is seen from the results of the measurement of the hysteresis loops at 83 K (curve 1) and 293 K (curve 2) shown in Fig. 2. It is seen from the presented magnetization-reversal curves that the rema-

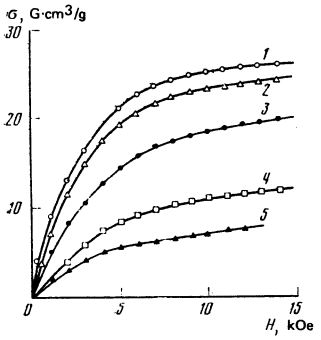

FIG. 1. Isotherms of the magnetization of the mictomagnetic manganese-bismuth alloys at the temperatures: 1—143 K, 2—173 K, 3—283 K, 4—535 K, 5—547 K.

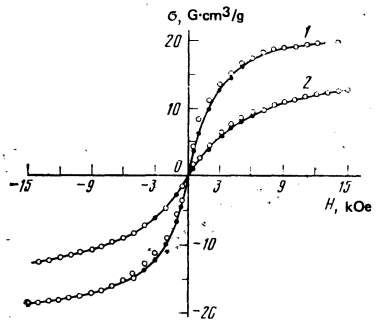

FIG. 2. Magnetization-reversal curves of mictomagnetic manganese-bismuth alloys at temperatures: 1—83, 2—293 K.

nent magnetism and the coercive force of the Mn-Bi alloys investigated by us are close to zero.

Figure 3 shows plots of the temperature dependence of the magnetization, measured under different conditions of prior heat treatment: curve 1 was obtained in a field 0.8 kOe after cooling the sample to 4.2 K without a magnetic field. In this case a sharp maximum is observed on the $\sigma(T)$ curve at $T_0 = 95$ K. In stronger fields, e.g., $H = 9.5$ kOe (curve 2), the maximum spreads out. Prior cooling to 4.2 K in a field 9.5 kOe and measurements at $H = 9.5$ kOe lead to the vanishing of the maximum of the $\sigma(T)$ curve, and the magnetization is in this case independent of temperature in the entire interval $T < T_0$. At high temperatures $T > T_0$, both $\sigma(T)$ curves, which were obtained under different magnetic annealing conditions, merge (curves 1 and 3 of Fig. 3).

It must be noted that measurements of the temperature dependence of the magnetization in weak fields, ~0.1 kOe, and in the region 220–250 K, have revealed a strong variation of the magnetization with temperature, as well as irreversibility of the curves obtained in the course of heating and cooling. These results confirm the previously observed[9,10] presence of a magnetic phase transition in the MnBi + Bi eutectic.

Magnetic annealing and the conditions of the measurement of σ manifest themselves very strongly in the temperature dependence of the magnetic susceptibility. In measurements of the magnetization in sufficiently

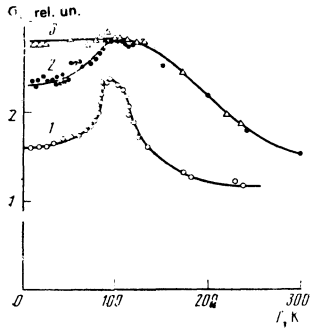

FIG. 3. Temperature dependence of the specific magnetization of a manganese-bismuth alloy in various magnetic fields: 1—0.8 kOe, 2—9.5 kOe (1, 2—cooling of the sample in the absence of a magnetic field, 3—cooling and measurement in a magnetic field 9.5 kOe.

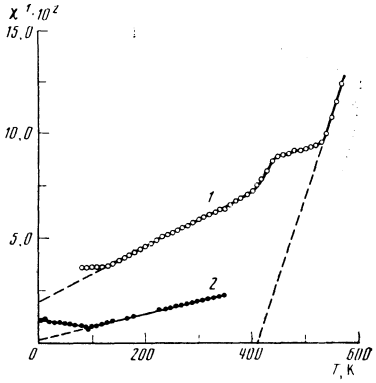

FIG. 4. Temperature dependence of the reciprocal susceptibility (cm^3/G) of the manganese-bismuth alloy in various magnetic fields: 1—8 kOe, 2—0.8 kOe.

strong fields ($H = 8$–10 kOe) the magnetic susceptibility $\chi = \sigma/H$ (where H is the measurement field) follows the Curie-Weiss law with a negative asymptotic Curie point $\Theta = -150$ to -170 K. In this case, if H is weak (~0.8 kOe), the temperature dependence of the susceptibility obeys the Curie-Weiss law and $\Theta = 0$ K, as can be readily seen from the $\chi^{-1}(T)$ plots shown in Fig. 4. Thus, it is possible to observe in the same sample, depending on the measurement conditions, different plots of the temperature dependence of the magnetization, as shown in Fig. 3.

The foregoing regularities differ strongly from the known magnetic properties of the *HT* phase of MnBi (Ref. 8) and are perfectly analogous to the behavior of mictomagnetic systems.[1] Figure 5 shows for comparison the temperature dependences of the magnetizations of our two-phase alloy, which is made up of the *HT* phase of MnBi and Bi (curve 1) and of the *HT* phase of the MnBi single crystal (curve 2) according to the data of Chen and Stutius.[8]

The principal distinguishing features of $\sigma(T)$ of the two-phase alloy of the *HT* phase of MnBi with Bi (curve 1, Fig. 5) is the following. At low temperatures, σ does not vary with temperature. With increasing T, a change of the magnetic state takes place in the region $T_0 \simeq 100$–110 K, after which σ gradually decreases with

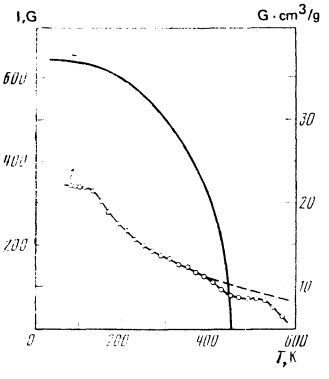

FIG. 5. Temperature dependence of the specific magnetization σ of the mictomagnetic manganese-bismuth alloy in a field $H = 0.8$ kOe (curve 1) and the magnetization I of a single crystal of the HT phase of MnBi according to Ref. 8 (curve 2).

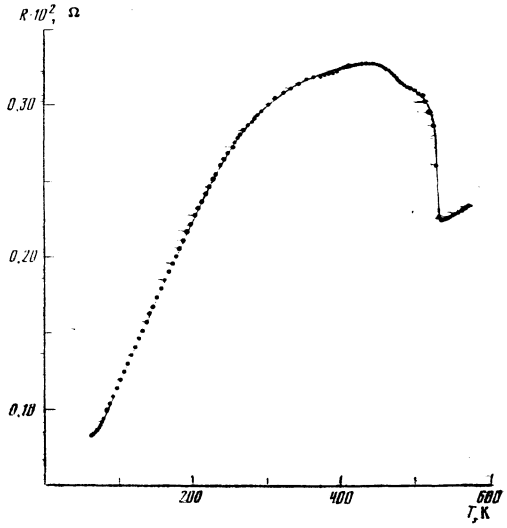

FIG. 6. Temperature dependence of the electric resistance of the manganese-bismuth alloy.

increasing T. At high temperatures, near the Curie point of the HT phase of MnBi (440 K), as well as at the melting point of the eutectic (538 K), strong changes of σ are observed. It is seen from the $\chi^{-1}(T)$ curve of Fig. 4 that in the interval $440 < T < 538$ K the susceptibility obeys the Curie-Weiss law $\chi = C/(T-\Theta)$ with a large negative constant Θ. Above the eutectic melting point, $T > 538$ K, the Curie-Weiss law is also satisfied, but the constant Θ is in this case positive and equal to 420 K.

Additional information on the magnetic phase transitions in MnBi alloys were obtained by us by investigating the temperature dependence of the resistance, as shown in Fig. 6. It is seen from the $R(T)$ plot in the region of 440 K, i.e., at the Curie point of the HT MnBi phase, the sign of the temperature coefficient of the resistivity is reversed and the resistivity changes from metallic to semiconducting; at 383 K there is distinct bend in the $R(T)$ curve, and at the eutectic melting point 538 K the temperature coefficient of the resistivity again becomes positive. As already noted, our phase transitions at 483 and 538 K were observed also on the DTA thermograms. No anomalous changes of R were observed at low temperatures, in the vicinity of T_0.

Figure 7 shows the temperature dependences of

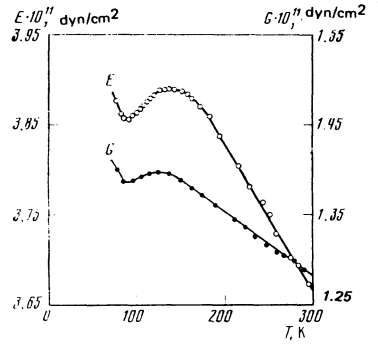

FIG. 7. Temperature dependence of the Young and shear moduli E and G of the manganese-bismuth alloy.

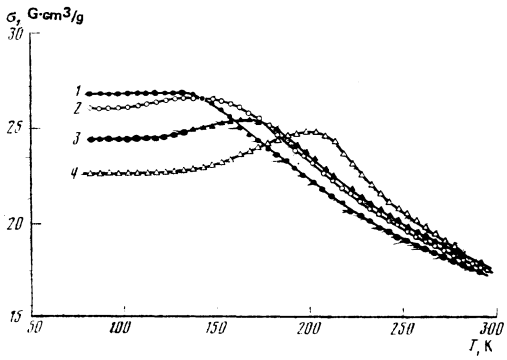

FIG. 8. Temperature dependence of the magnetization of bismuth-manganese alloys at various pressures: 1—atmospheric, 2—4.3 kbar, 3—6.3 kbar, 4—9.3 kbar.

Young's modulus (E) and of the shear modulus (G). It is seen that an anomalous change of the elastic moduli is observed only at low temperature, viz., minima on the $E(T)$ and $G(T)$ curves at the temperature $T = 95$ K near which a peak of the temperature dependence of the low-field magnetic susceptibility was observed.

In conclusion we present the results of the measurements of high hydrostatic pressure (up to 10 kbar) on the magnetic properties of the Mn-Bi alloys. Figure 8 shows the temperature dependences of the magnetization, measured at various pressures. It is seen from these curves that at low temperatures ($T < T_0$) the magnetization decreases very rapidly with increasing pressure. Conversely, at room temperature ($T > T_0$) the magnetization is increased by high pressure. On the basis of these measurements, the derivative $dT_0/dP = 8$ deg/kbar was determined from the shift of T_c.

DISCUSSION OF RESULTS

In the analysis of the obtained experimental data we consider mainly two questions: the onset of the mictomagnetism in the Mn-Bi alloys, and the magnetic phase transitions at high temperatures.

The temperature and field dependences of the alloys investigated by us indicate that two-phase systems consisting of the HT phase of MnBi and of Bi cannot be regarded as ordinary ferromagnets. The large value of the magnetization and the nonlinearity of $\sigma(H)$ can be due to the presence in the alloy of regions with short-range magnetic order (clusters) having a spontaneous magnetic moment. It is known (see, e.g., Ref. 11) that the behavior of an ensemble of noninteracting clusters is described by the superparamagnetism theory based on the Langevin equation

$$\frac{\sigma}{\sigma_\infty} = \operatorname{cth}\frac{\mu H}{kT} - \frac{kT}{\mu H}, \tag{1}$$

where σ is the magnetization of the sample at the temperature T in the field H, $\sigma_\infty = N\mu$ is the saturation magnetization, μ is the magnetic moment of the cluster, and N is the concentration of the clusters. At $\mu H \ll kT$ Eq. (1) can be transformed into

$$\frac{\sigma}{\sigma_s} = \frac{\sigma_s N V^2 H}{3kT} \tag{2}$$

where V is the volume of the cluster and σ_s is the spontaneous magnetization of the cluster and is proportional to $j_s = (\chi_0 T)^{1/2}$ where χ_0 is the initial susceptibility of the alloy. It follows from (2) that $\sigma \to 0$ as $H \to 0$, i.e., a characteristic property of a superparamagnet is the absence of magnetization hysteresis. In addition, if the magnetization isotherms are represented by the function j_s^{-1} of $j_s H/T$, then it follows from (2) that the superposition principle must be satisfied, i.e., the $\sigma(H)$ curves measured at different temperatures must coincide.

The alloys investigated by us satisfy both indicated superparamagnetism criteria, as is attested by the absence of magnetization hysteresis (Fig. 2) and the superposition of the magnetization isotherms, measured at various temperatures and represented in the form of functions $(\chi_0 T)^{-1/2}\sigma$ of $(\chi_0 T)^{1/2} H/T$ on Fig. 9. It must be noted here that the superposition principle holds in a relatively limited range of fields and temperatures, and this, according to (2), can be due to the change of the cluster volume with temperature, which in turn can be due to the change of the interaction of the clusters with one another and with the Bi matrix. That the ferromagnetic clusters interact with the Bi matrix is attested to by the abrupt changes, at 538 K, of the constants Θ and C that enter in the Curie-Weiss law. In fact, for the solid and liquid states of Bi in the MnBi + Bi eutectic, the constant Θ, which characterizes the exchange interaction, turns out to be different not only in magnitude but also in sign.[1)]

Following the procedure described by Becker,[13] we have calculated the volume of the clusters V in the MnBi alloys at various temperatures. It turned out that V decreases with decreasing temperature and changes in the interval 283–143 K from 0.55×10^{-19} to 0.11×10^{-19} cm^3, so that the average diameter of clusters of spherical shape lies in the range 38–40 Å, which certainly ensures that the clusters are single-domain[14] in the indicated temperature interval.

In this case, the different magnetic states of the alloy at low ($T < T_0$) and high ($T > T_0$) temperatures can be due to the blocking of the magnetic moments of the clusters in the field of their magnetic anisotropy at $T < T_0$. To check on this assumption, we calculated the critical volumes V_p of superparamagnetic clusters at

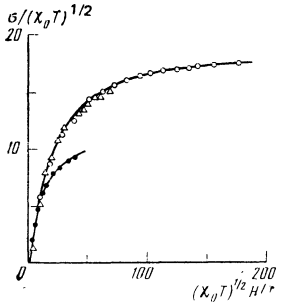

FIG. 9. Superposition of magnetization curves of manganese-bismuth alloy with allowance for the temperature dependence of the spontaneous magnetization of the clusters. Measurements at the temperatures: ○—143 K; △—283 K; ●—353 K.

77, 143, and 283 K using the formula

$$V_p = \ln(2t' f_0) kT/K, \qquad (3)$$

where t' is the measurement time (assumed to be 100 sec for statistical measurements[11]), f_0 is a frequency factor of the order of 10^9 sec^{-1}, k is the Boltzmann constant, and K is the magnetic crystallographic anisotropy constant, whose values for the HT phase of MnBi were taken from the paper of Chen and Stutius.[8]

Estimates of V_p in accord with (3) have shown that at 143 and 283 K the volume V of the clusters does not exceed the critical value V_p, in agreement with the superparamagnetic behavior of the alloys in the region $T > T_0$. At 77 K, however, the calculated value of V_p is 0.66×10^{-20} cm^3, which is less by almost one order of magnitude than the cluster volume obtained from the experimental data. In this case one should expect residual phenomena following the reversal of the alloy magnetization, since estimates of the coercive force H_c for the case of randomly oriented particles with uniaxial anisotropy $H_c \simeq K/\sigma_s$ and for expression (3),

$$H_c = \frac{\ln(2t' f_0) kT}{V_p \sigma_s}$$

lead to values of H_c on the order of 66 kOe. Nonetheless, as seen from Fig. 2, no hysteresis of the magnetization is observed at low temperatures.

It seems to us that the obtained experimental data can be explained by assuming that there exist in the two-phase manganese-bismuth alloys disordered ferro- and antiferromagnetic exchange interactions that have greatly different energies and behave differently with changing interatomic distances.

At low temperatures the magnetic system is characterized by zero magnetization, inasmuch as no special direction in the crystal has a spontaneous moment, in view of the random distribution of the exchange bonds of opposite signs. When the temperature is raised to $T_0 = T_N \approx 100$ K the regions of antiferromagnetic short-range order go over into the paramagnetic state, and the system as a whole becomes superparamagnetic, because of the preservation of the ferromagnetic clusters, in which the exchange coupling, characterized by the Curie point $T_c = 440$ K, greatly exceeds the ferromagnetic-exchange energy. All the distinguishing features of the magnetization curves of the investigated alloys, as well as the influence of the heat treatment and of the measurement conditions on $\sigma(T)$, can be attributed to the interaction between the ferro- and antiferromagnetic clusters, which produces the resultant magnetization in a magnetic field.

The coexistence of opposite-sign exchange interactions and, in particular, the presence of antiferromagnetic exchange in manganese-bismuth alloys, is evidenced also by the anomalous change of the elastic properties in the region of T_0.[15] The observed change of the Young and shear moduli E and G is perfectly analogous to the anomalies of ordinary antiferromagnets at the Néel point. The simultaneous change of the Young and shear moduli near T_0 points to the existence

of anisotropic antiferromagnetic exchange in the investigated alloys. The cause of the anomalies of the elastic moduli of MnBi may be the magnetostriction phenomena connected with the strong exchange dependence of the exchange interactions, as is indicated by the large changes of T_0 under the action of high pressures.

The results of the investigation of $\sigma(T)$ at high hydrostatic pressures indicate that the existing exchange couplings of opposite sign causes the energy of the antiferromagnetic exchange to vary more strongly with interatomic distance than that of the ferromagnetic exchange. The large shifts of T_0 with change of pressure towards high temperatures, and the considerable decrease upon compression in the entire $T < T_0$ region are evidence of enhancement of the antiferromagnetic interaction when the interatomic distances decrease.

The influence of the high pressure on the ferromagnetic exchange is much weaker. This can be deduced from the weak dependence of σ on P at room temperatures. If it is recognized that the Curie point T_c of the HT phase of MnBi decreases at high pressures,[16] and $dT_0/dP = -0.7$ deg/kbar, then one should expect a small decrease of σ with increasing pressure in the interval $T > T_c$. It turns out, however, that the baric derivative of the magnetization is positive, a fact that can be attributed to enhancement of the intercluster interaction, which offsets upon compression the slight weakening of the ferromagnetic exchange with increasing interatomic distances in the cluster.

We consider in conclusion our results of the investigation of the phase transitions in MnBi. At present there are known three different magnetic-structure states of MnBi: low-temperature (LT), high-temperature, and the state with low Curie point $T_c = 240$–250 K, which was observed in the investigation of the MnBi + Bi eutectics.[9,10]

The most contradictory are the available experimental data on the phase transitions of the HT phase of MnBi. In different studies (see, e.g., Ref. 7) it was established that at the Curie point ($T_c = 470$ K) there is a simultaneous transformation of the HT phase into the LT one. It was noted in Ref. 8 that the HT–LT transition due to the change of structure occurs not at the Curie point itself, 440 K according to Ref. 8, but close to it. An investigation of MnBi films[5] has shown that the HT–LT transformation proceeds through a metastable third phase whose structure has not yet been established.

It can be concluded on the basis of our measurements of $\sigma(T)$ and $R(T)$ that at the Curie point $T_c = 440$ K no direct HT-LT transition takes place, since the magnetization decreases sharply above T_c, rather than increase as might be expected in the presence of such a transition. The results of the investigation of the resistance $R(T)$ suggest that at 440 K the ferromagnetic clusters go over into an antiferromagnetic state with a Néel point close to the eutectic melting temperature 538 K. The form of the $R(T)$ curve on Fig. 6 agrees with the theoretical premises[17] concerning the splitting of the conduction band when the metal becomes ferromagnetic, accompanied by the onset of semiconductor conductivity. Above the Néel point, which in our case is close to 538 K, the $R(T)$ dependence again becomes metallic. It can therefore be assumed that in the interval 440–538 K the manganese-bismuth alloys are in the antiferromagnetic state. It is obvious that neutron-diffraction investigations are essential to verify these assumptions.

As already mentioned, we have observed at 483 K one more phase transition that does not lead apparently to a substantial change of the magnetic state of MnBi, since no anomalies are observed in this case on the $\sigma(T)$ curve. These data, together with the discovery in the low temperature region of magnetic phase transition that depend greatly on the magnetic annealing and on the measurement conditions, are the new result of the present paper.

CONCLUSION

Two-phase alloys consisting of the HT phase of MnBi and of Bi form magnetically disordered systems that can be classified as mictomagnets.[2)]

At low temperatures ($T_0 = 95$–100 K) a transition from the mictomagnetic state into the superparamagnetic state was observed, with a temperature that is greatly increased by high pressure: $dT_0/dP = 8$ deg/kbar. The magnetization of the alloys in the mictomagnetic state is decreased by hydrostatic compression, and increases in the superparamagnetic state. Radical changes were observed in the elastic properties (the Young and shear moduli) in the region of the transition temperature T_0.

The obtained experimental data are interpreted on the basis of the assumption that there coexist ferro- and antiferromagnetic short-range regions that interact with one another as well as with the Bi matrix. At low temperatures and high pressures, the energy of the antiferromagnetic exchange increases, and at high temperatures (in the superparamagnetic state) an enhancement of the intercluster interaction appears with decrease of the interatomic distances.

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[1)]It is known[12] that the diamagnetic susceptibility of pure bismuth is decreased by almost a factor of 30 upon melting. The two-phase alloys investigated by us have high paramagnetic susceptibility, the change of which at the melting point of the MnBi + Bi eutectic exceeds substantially the indicated change of the diamagnetic susceptibility of pure bismuth upon melting.

[2)]It must be noted that ordinary mictomagnets are most frequently single-phase systems.

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Destruction of the spin glass state by indirect exchange via conduction electrons in the $x CuCr_2S_4$–$(1-x)Ga_{2/3}Cr_2S_4$ solid-solution system

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An experimental investigation was made on the magnetic and electrical properties of solid solutions in the $x CuCr_2S_4$–$(1-x)Ga_{2/3}Cr_2S_4$ system. At low values of x these solutions are insulators and exhibit properties typical of spin glasses (susceptibility peak, low-temperature magnetization hysteresis). However, when composition is varied an insulator–metal transition occurs near $x = 0.5$ and a spontaneous moment appears at the same time. A rapid rise of the magnetic ordering temperature accompanying this phase transition can be explained by the appearance of an indirect exchange via charge carriers, which suppresses the spin-glass state. Thus, in contrast to the earlier view that an indirect exchange is the main reason for the existence of a spin-glass state, the results obtained in the present study suggest a directly opposite role of this exchange in the case of insulating spin glasses. A theoretical analysis is used to show that an indirect exchange via charge carriers may destroy a spin-glass state also in spin glasses which are magnetic semiconductors. For certain values of the carrier density these materials may exhibit a heterophase state in which a crystal splits into a series of alternate spin-glass and ferromagnetic regions, with the carriers concentrated in the latter. Destruction of such a heterophase state by a magnetic field or by an increase in temperature may result in an insulator–metal phase transiton.

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The existence of spin glasses was first discovered in metallic alloys and for this reason a view has been held that a spin-glass state is a consequence of an indirect exchange (via conduction electrons), whose integral in the RKKY theory is due to an oscillatory function of the separation between localized moments. Discovery of insulating spin glasses in the $Eu_xSr_{1-x}S$ solid-solution system[1] demonstrated that in any case the RKKY indirect exchange is not necessary for their existence. Moreover, a question arises whether an indirect exchange establishes a spin-glass ordering in a disordered conducting crystal. The reasons for these doubts are as follows. Firstly, since the Fermi surface of electrons in alloys is smeared by the scattering of electrons on the random potential, the indirect exchange integral in the RKKY theory decreases exponentially with distance[2] so that in many cases it is practically constant in sign. Secondly, oscillations of the indirect exchange integral are suppressed also in the case of narrow energy bands of width comparable with or less than the s–d exchange integral,[3] which is typical of transition-element alloys.

We made an experimental observation which seemed paradoxical from the point of view of the existing ideas on the nature of spin glasses: an indirect exchange via charge carriers not only did not facilitate spin-glass ordering but destroyed a spin-glass state.

We investigated magnetic and electrical properties of polycrystalline samples of $xCuCr_2S_4-(1-x)Ga_{2/3}Cr_2S_4$ solid solutions prepared by a method described in Ref. 4. The terminal compositions of this system had very different properties: at $x = 1$ the material was a metallic ferromagnet,[5] whereas at $x = 0$ it was an insulating spin glass.[6] The solutions in the range from $x = 0$ to $x = 0.45$ were spin glasses. This was indicated by the following observations. 1) The magnetic susceptibility χ of these compositions measured in a weak static field

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