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Intermetallide-based magnetic materials

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1. Introduction

Intermetallic compounds of transition f- and d-metals are interesting in that they can possess two systems of electrons of different natures, namely localized and itinerant, which can be responsible for magnetism. Rare-earth atoms have their 4f electrons localized both in space and in energy. The interaction of an orbital momentum with the lattice crystal field results in a large magnetic anisotropy and a large magnetostriction. At the same time, the indirect exchange interaction between the 4f electrons is relatively weak, and the magnetic ordering temperatures of rare-earth metals are low. Unlike 4f metals, the energy spectra of transition 3d metals form quasicontinuous bands. 3d (or iron group) metals typically have a high magnetic ordering temperature but a low anisotropy. Rare-earth intermetallic compounds often combine the magnetic characteristics of pure 4f and 3d metals to yield materials with a high ordering temperature and immense anisotropy (or magnetostriction) simultaneously — which enables their use as permanent magnets, as magnetostrictive or magnetoresistive materials, and for magnetic cooling.

The magnetic properties of rare-earth intermetallides have been the subject of intense study since the pioneering work by K P Belov and his team at Moscow State University in the late 1960s. In Ekaterinburg, the initiators of research into the field were S V Vonsovskii, Academician, and Ya S Shur, Corresponding Member of the USSR Academy of Sciences, both at the Institute of Metal Physics (IMP) of the Ural Branch of the USSR Academy of Sciences, and A V Deryagin, Corresponding Member of the USSR Academy of Sciences, at Ural State University. With a host of exciting results obtained in recent years, it is beyond the ambition of this review to cover the entirety of what has been done and achieved in this vast area of the physics of magnetic phenomena. The discussion will be limited to the work carried out at the UrB RAS IMP in close cooperation with other research centers on magnetism of intermetallides, both in Russia and abroad.

2. Permanent-magnet materials

From the middle 1970's onwards, the highest values of specific magnetic energy have been achieved in permanent magnets made of highly anisotropic intermetallide phases such as $SmCo_5$, Sm_2Co_{17}, and $Nd_2Fe_{14}B$ [1]. From the application point of view, of most importance is the segment of high energy-consuming permanent magnets based on Nd–Fe–B [2], whose global production is about 100,000 tons a year. Russia is lagging far behind developed countries both in production volume (about 140 tons a year) and in the property level of rare-earth permanent magnets. In particular, Russian-produced permanent magnets typically have a maximum energy product $(BH)_{max}$ of 40.0 MG Oe — in contrast to the specific magnetic energy of 50.0 MG Oe accessible on the world market. Recently, the joint research efforts of the IMP, Ural State University, the Ural Electromechanical Plant, and the Moscow Steel and Alloys Institute has resulted in developing a low-oxygen technology for fabricating sintered permanent Nd–Fe–B magnets using strip-cast alloys. With the basic technological processes optimized, a maximum energy product $(BH)_{max} \geqslant$ 50 MG Oe was obtained industrially, which matches the level of the best magnet manufacturers and is 25% superior to the domestic commercial analogs [3]. Importantly, though, the maximum theoretical value for the $Nd_2Fe_{14}B$ phase is 68.0 MG Oe, leaving significant room for improvement. The maximum theoretical value of the energy product for a permanent magnet is determined by magnetization. The material showing the highest magnetization at absolute zero temperature is holmium, for which the theoretical value of $(BH)_{max}$ is 350.0 MG Oe. Thus, the potential for magnetic materials based on intermetallic compounds between rare-earth metals and iron group metals is far from exhausted.

High-technology industries including computer hardware, robotics, and automobile engineering are unimaginable without permanent magnets. In the rotors of high-speed centrifuge-driving hysteresis motors, only mechanically strong hard magnetic materials can be used. The IMP Laboratory of Ferromagnetic Alloys developed a high-strength hard magnetic alloy based on nanocrystallized Fe–Cr–Co, whose combination of functional properties makes it unique in the world. Its mechanical properties are close to those of high-strength steels, whereas its magnetic characteristics are optimum for hysteresis motors [4].

In tungsten- and gallium-doped Fe–Cr–Co alloys, by using hardening, annealing, and intense plastic rolling deformation, a multiphase nanocomposite state was formed instead of a conventional modulated structure, resulting in a high-strength high-plasticity material (conditional yield limit of up to 1500 MPa, and relative elongation of up to 6%). Structure studies [5, 6] using nuclear gamma resonance (NGR), X-ray diffraction and transmission electron microscopy established that the hardening of the alloy and the formation of a highly coercive state are due to the decomposition of a homogeneous α-Fe-based solid solution combined with the evolvement of nano-sized particles of the tungsten-enriched paramagnetic intermetallic phase.

3. Materials for magnetothermal applications

The employment of the magnetocaloric effect in cooling machine design [7] is believed in the last few years to be one of the most promising applications of magnetic materials. Unlike the mechanical compressors currently used in refrigerators, magnetic cooling devices are noiseless, consume less electric power, and are ecologically safe. While most of their working prototypes utilize gadolinium as a working medium, a number of rare-earth intermetallides, such as $La(Fe,Si)_{13}H_x$ and RCo_2 (where R is a rare-earth element), also show considerable promise for application. Because the temperature change due to the demagnetization of a material is determined by magnetic entropy, it follows that compounds whose magnetization is strongly temperature-dependent near room temperature hold promise as magnetic cooling materials. This understanding stimulated recent interest of research-

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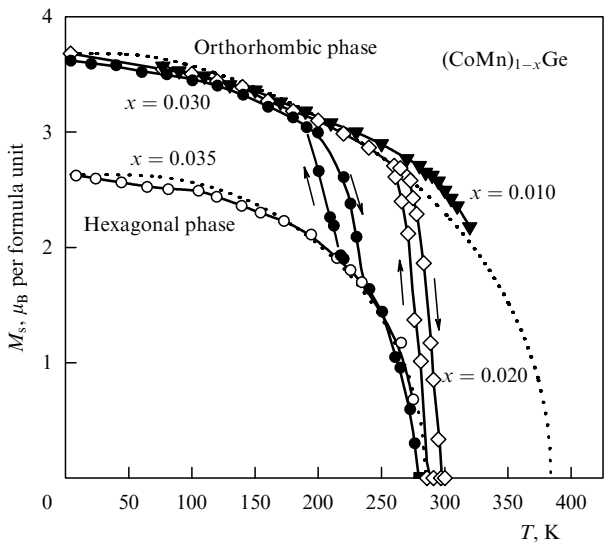

Figure 1. Spontaneous magnetization of $(CoMn)_{1-x}Ge$ compounds as a function of temperature for different x. Dashed lines fit calculated results for the hexagonal and orthorhombic phases.

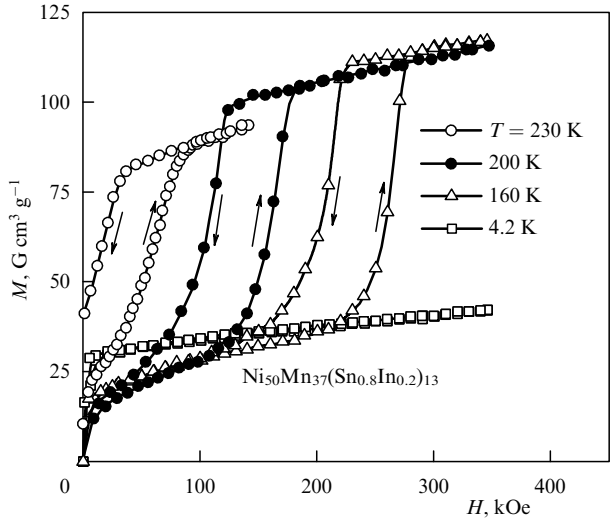

Figure 2. Magnetization curves of the $Ni_{50}Mn_{37}(Sn_{0.8}In_{0.2})$ alloy illustrating a martensite transformation induced by a pulsed magnetic field.

ers in compounds with structural phase transitions near room temperature, with various structural components having widely different magnetizations. The most widely known examples are Heusler Ni_2MnGa alloys [8] and alloys based on $Gd_5(Si,Ge)_4$ [7]. The IMP is currently searching for effective magnetocaloric materials based on CoMnGe, Ni–Mn–Sn–In, etc.

The CoMnGe compound undergoes a diffusionless structural martensite type transition at 470 K from the high-temperature hexagonal Ni_2In type structure to a low-temperature orthorhombic TiNiSi type phase [9]. Although both are ferromagnetic, these modifications differ significantly in magnetization and in the Curie temperature T_C. By varying the composition of the system $(CoMn)_{1-x}Ge$, we were able to obtain alloys for which the structural transition near the ferromagnetic ordering temperature is one from the paramagnetic hexagonal to the ferromagnetic orthorhombic phase [10] (see Fig. 1), which shows exactly the compositions for which magnetothermal effects are expected to be strongest. For $x = 0.02$, the transition occurs in the room temperature range and involves an entropy change of 33.8 J (kg K)$^{-1}$. Applying a magnetic field in excess of the threshold value of 20 kOe in the vicinity of the structural transition increases the relative volume of the higher-magnetization orthorhombic phase.

The magnetic shape memory effect is even more pronounced in Ni–Mn–In and Ni–Mn–Sn alloys [11], in which the austenite and martensite phases differ widely in their magnetization. The $Ni_{50}Mn_{37}(Sn_{0.8}In_{0.2})_{13}$ alloy subjected to an 8-ms magnetic field pulse exhibits a field-induced martensite transformation (Fig. 2) [12]. The critical transition field decreases as the measurement temperature approaches the temperature of spontaneous martensite transformation. Both the spontaneous and the field-induced martensite transformation involve a considerable resistance change: $\Delta R/R \approx -45\%$.

4. Mixed valence compounds and band metamagnets

The situation of practical interest is when two magnetic states of an intermetallide are close in energy, in which case even a slight action on the system (such as an external field, pressure, temperature change, hydrogenation, or doping) can change drastically the magnetic and other physical properties of the system.

A free Yb^{3+} ion has one unfilled 4f electronic state and carries a magnetic moment. When in a compound, the 4f vacancy is often filled by conduction electrons, resulting in the formation of the diamagnetic Yb^{2+} state. In the $YbInCu_4$ compound, the change in valence occurs via a first-order isostructural phase transition in a narrow temperature range [13]. Although the valence of Yb decreases by a mere 0.1, the magnetic susceptibility and electrical resistance show sharp changes, and the specific heat and the sample volume undergo an anomaly (Fig. 3). At low temperatures, applying a magnetic field can destroy the low-magnetization Fermi-liquid state.

Our laboratory study of pressure and doping effects on the transition temperature, transition field, magnetic susceptibility [14], specific heat [15], and magnetostriction [16] shows that the anomaly in physical properties associated with the small change in valence in a given compound is due to a fivefold increase in hybridization factor between f electrons and the conduction electrons [17].

Ferromagnetic ordering in a system of itinerant electrons emerges in the presence of high density of electronic states near the Fermi level. In a band structure of the compound with the Fermi level lying on the steep downward portion of the density-of-states curve, exchange interactions are unstable, and a magnetic field can induce ferromagnetism in a paramagnetic compound — a phenomenon known as band metamagnetism [18]. (This usually requires a strong magnetic field, according to Refs [19, 20].) The YCo_2 compound, being a Pauli paramagnet, changes to a ferromagnetic state in an external magnetic field of 700 kOe [21]. As shown in Ref. [22], doping with aluminium reduces the critical transition field. In Ref. [23], we investigated the effect of hydrostatic pressure on the critical transition field in the $Y(Co, Al)_2$ compounds under pulsed magnetic fields. By comparing the effects of doping, hydrogenation, and pressure, it proved possible to see separately how the interatomic separation and the concentration of d-band electrons contribute to changing the transition field [24].

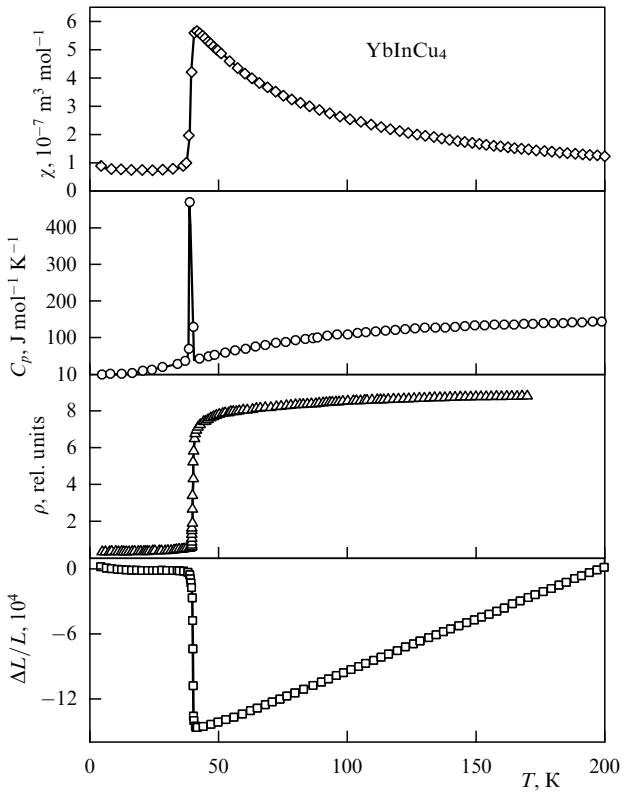

Figure 3. Magnetic susceptibility χ, heat capacity C_p, resistivity ρ, and relative change in the sample size $\Delta L/L$ as functions of temperature for $YbInCu_4$. The isostructural valence-change transition at $T = 40$ K involves a sharp change in properties.

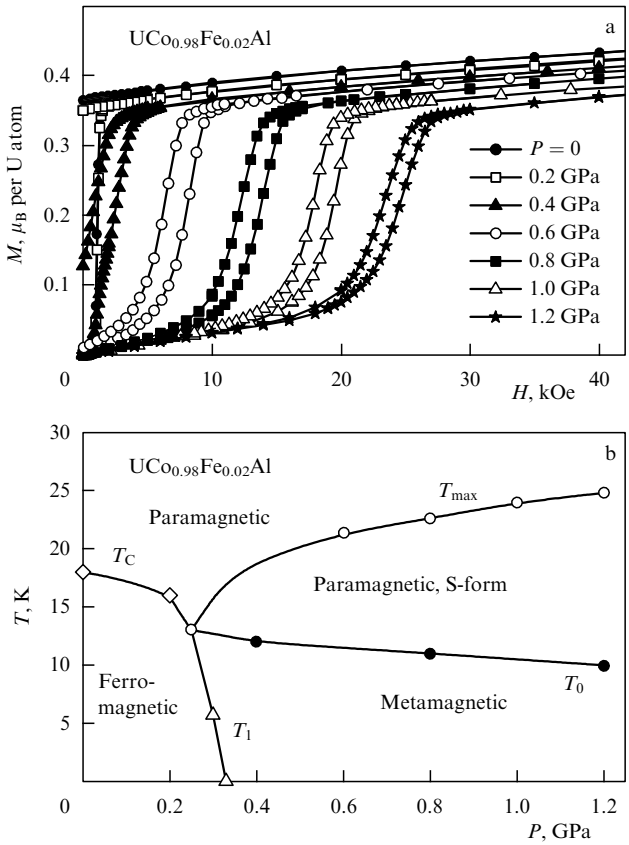

Figure 4. Hysteresis loops at 1.5 K measured for different values of external hydrostatic pressure (a), and magnetic $T-P$ phase diagram (b) for anisotropic band metamagnetic $UCo_{0.98}Fe_{0.02}Al$.

While UCoAl also shows signs of being band metamagnet [25], its properties differ strongly from those of YCo_2. The critical transition field of this compound does not exceed 6.0 kOe. The magnetic moment emerges on uranium, whereas cobalt carries no moment. Metamagnetism is observed only along the c-axis, i.e. is anisotropic, unlike the isotropic metamagnetism seen in YCo_2. The goal of this research, conducted in close cooperation with counterparts from Japan and the Czech Republic, was to reveal the general and particular aspects of an isotropic itinerant 5f-electron metamagnetism.

The metamagnetic magnetization curves of UCoAl measured for different pressures were analyzed with a phenomenological theory of metamagnetic transitions using the magnetic equation of state in the form of Landau expansion in odd powers of magnetization [26]. Matching as best as possible the theoretically predicted to experimental curves yielded the Landau coefficients for different pressures, which determine the temperature behavior of the magnetic susceptibility in the theory of spin fluctuations [27]. The numerical estimates of the maximum magnetic susceptibility and the temperature dependence of the transition critical field are in good agreement with calculated results, implying that spin fluctuation theory is good at describing the properties of anisotropic band metamagnets.

The term 'metamagnet' is also applied to ferromagnets that lose their ferromagnetism — and become metamagnetic — when subject to pressure or acted upon by thermal flux. Such a transition is of particular interest in that applying a relatively weak magnetic field drastically changes the magnetization and conductivity of the material, as well as the size of the sample. In UCoAl, ferromagnetism can be obtained by substituting nonmagnetic iron for only 2% of nonmagnetic cobalt. At a pressure above 0.4 GPa, ferromagnetism is suppressed and, as is typical of UCoAl, a metamagnetic transition occurs (Fig. 4a) and the temperature dependence of the susceptibility reaches a maximum. A study of this sample at various temperatures and pressures provided the full magnetic phase diagram in the pressure–temperature plane [28] (Fig. 4b), which agrees well with the theoretical phase diagram obtained for a spin-fluctuating band metamagnet [29]. To summarize, the results obtained place UCoAl type compounds in a new class of anisotropic itinerant 5f-electron metamagnets.

5. Layered magnets

The study of the physical properties of quasi-two-dimensional and multilayered structures is currently one of the most active areas in the physics of condensed matter. While most research is done on thin multilayered films, there are among intermetallic compounds a number of natural bulk analogs of multilayered structures, including intermetallides of the RM_2X_2 and RM_6X_6 types (R is a rare-earth metal, M is a 3d (4d) transition metal, and X is Si, Sn, or Ge). These compounds consist of intermittent layers of magnetic and nonmagnetic atoms. The strong ferromagnetic interaction of magnetic atoms within a layer results in high magnetic ordering temperatures. Therefore, the magnetic moment of each layer turns out to be a macroscopic quantity and can be treated as a classical vector. At the same time, the relatively weak and easy to control interlayer interaction turns out to be

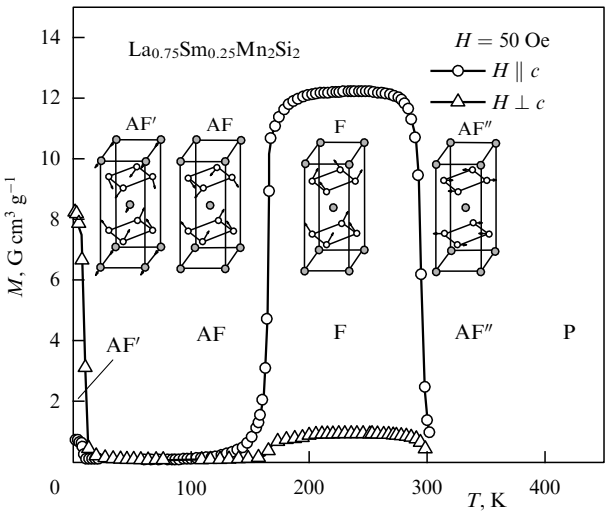

Figure 5. Temperature dependences of magnetization along different single-crystal directions at 50 Oe, and magnetic structures occurring in the $La_{0.75}Sm_{0.25}Mn_2Si_2$ intermetallide at various temperatures. (AF stands for antiferromagnetic structure, F for ferromagnetic, and P for paramagnetic.)

responsible, by and large, for the formation of the magnetic structure of the material.

In RMn_2X_2 intermetallides, the magnitude and sign of the interlayer exchange interaction between the magnetic moments of manganese atoms in neighboring layers depend strongly on the intralayer Mn–Mn distance and hardly, if at all, on the interlayer separation [30]. In the RMn_2Si_2 family, the maximum Mn–Mn interatomic distance is observed for $R = \mathrm{La}$, and this is the only compound to exhibit ferromagnetic ordering. Replacing lanthanum by any other rare-earth metal can gradually reduce interatomic distances, thus destabilizing ferromagnetism. The $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound, with the Mn–Mn distance close to the critical value, exhibits a number of spontaneous magnetic phase transitions, due to temperature-related changes in the lattice parameters. In Fig. 5, magnetic structures determined by neutron diffraction are shown for all the phases involved [31]. From the magnetization measurements presented in Fig. 5, one determines the regions of existence of the ferromagnetic and antiferromagnetic phases. In a low-temperature antiferromagnetic state, applying a magnetic field induces a first-order phase transition to a ferromagnetic state [32], producing large changes in the parameters ($\Delta a/a = 1.5 \times 10^{-3}$) and volume of the crystal lattice, as well in the magnetic resistance ($\Delta R/R = -27\%$) [33] (see Fig. 6). Varying the relative content of the alloy components can alter the transition temperatures and critical fields of these compounds over a wide range.

If the interlayer exchange interactions are long-range, then, if there is interaction competition between the nearest neighboring and next-to-nearest layers, a spiral magnetic structure can develop, with the feature (observed in YMn_6Sn_6 [34]) that the spiral divides into two branches shifted through a certain angle. To find out the conditions for the existence of double flat spiral structures, a theory was developed — accounting for three interlayer exchange interactions — which was also employed to model magnetization processes in spiral structures [35, 36]. The results of this modeling provided an explanation for the observed abrupt change in magnetization caused by a magnetic field applied in the basal plane.

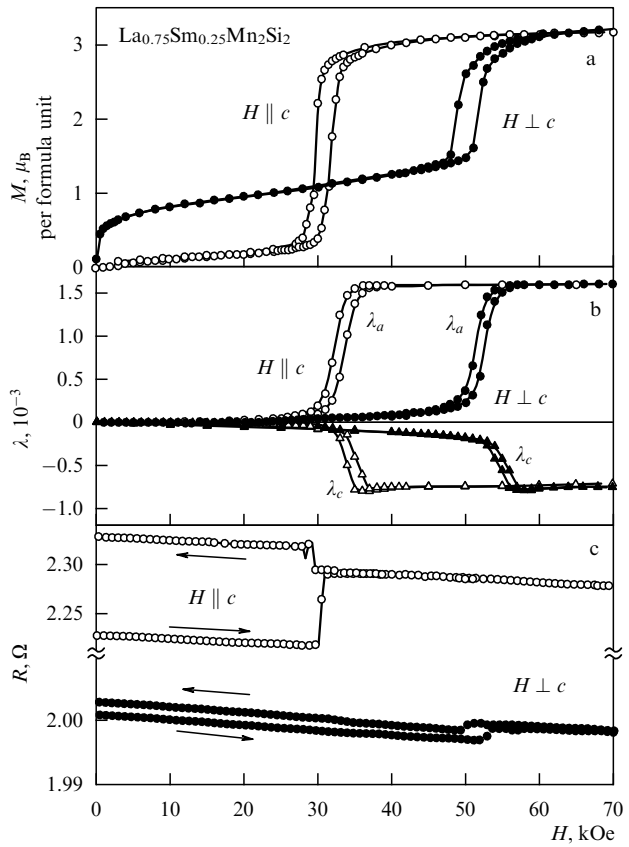

Figure 6. Field dependence of magnetization M (a), linear magnetostriction λ (b), and electrical resistance R (c) along different directions in a single-crystalline $La_{0.75}Sm_{0.25}Mn_2Si_2$ at $T = 4.2$ K.

In a highly anisotropic uniaxial ferromagnet, the application of a magnetic field along the hard magnetization axis normally causes the magnetization vector to turn smoothly in the direction of the field. However, when the magnetic energy of a sample varies nonmonotonically with angle, a field-induced spin-reorientation phase transition involving a sharp change in magnetization occurs in a certain critical field [37, 38]. Unlike most first-order phase transitions, this one exhibits a weak magnetic hysteresis, making it of interest for practical implementations.

The occurrence of magnetic-field-induced first-order phase transitions is thought to be related to a special feature of the compound's magnetic anisotropy: the presence of large higher-order contributions in the expansion of the anisotropy energy in terms of the directional cosines of the magnetization vector. In accordance with the theory of single-ion anisotropy, the higher the order of the contribution, the faster its magnitude decreases with increasing temperature. Therefore, such transitions are usually observed at low temperatures in strong magnetic fields. In the $TbMn_6Sn_6$ compound, the competition among contributions to magnetic anisotropy from the Tb and Mn sublattices, combined with the presence of a spontaneous spin-reorientation transition, result in the occurrence of a magnetic-field-induced first-order phase transition in weak fields around room temperature [39]. This enabled the first neutron diffraction study of how magnetization components change in the transition process [40]. The replacement of terbium by gadolinium in a quasiternary $Tb_{1-x}Gd_xMn_6Sn_6$ system produces a disordered solid solution within the rare-earth sublattice, allowing the magnetic

anisotropy and exchange interactions to be gradually controlled to shift the critical temperature and field of a magnetic phase transition [41].

6. Nonstoichiometric intermetallides

The compounds comprising 4f and 3d metals are usually characterized by a certain stoichiometry and are therefore limited in number. All binary and many ternary intermetallides have been synthesized, and their magnetic characteristics are available in the reference and review literature. At the same time, though, some structures allow deviations from stoichiometry with the result that the properties of a compound can vary widely within a homogeneity region.

It was found recently [42] that the $R\mathrm{Ni_2Mn}$ alloys, where $R = \mathrm{Tb}$, Dy, Ho, and Er, crystallize into an fcc $\mathrm{MgCu_2}$ type structure exactly as the compounds $R\mathrm{Ni_2}$ and $R\mathrm{Mn_2}$ do, even though the number ratio of rare-earth-to-transition 3d metal atoms is 1:3. The Mn atoms occupy partly rare-earth and partly Ni sites [43], something which has never been seen before in rare-earth Laves phases. Investigations showed that the $R\mathrm{Ni_2Mn}_x$ alloys remain cubic structure over a wide Mn concentration range [44, 45]. It was found that introducing manganese sharply enhances the exchange interaction and greatly increases the magnetic ordering temperature (4–7 times more than in the original $R\mathrm{Ni_2}$). The maximum T_C is reached for $\mathrm{TbNi_2Mn_{0.75}}$ and equals 160 K.

In the $\mathrm{TbNi_2Mn}_x$ system, spontaneous magnetization monotonically decreases with increasing x. Unlike the original $\mathrm{TbNi_2}$, where the nickel sublattice barely carries a magnetic moment, introducing manganese results in the 3d sublattice acquiring a magnetic moment directed opposite to that of the terbium sublattice. The magnetization curves of ferrimagnetic samples do not saturate until magnetic fields of 15 T. The introduction of manganese results in a monotonic increase in the coercive force which is due to the pinning of narrow domain walls on structural defects. It appears that the partial replacement of terbium atoms by manganese atoms leads to local distortions in the crystal field acting on the Tb ions. As a result, local, random, uniaxial anisotropy arises, giving rise to a noncollinear magnetic structure of the terbium sublattice.

The starting $\mathrm{TbNi_2}$ compound possesses giant magnetostriction at low temperatures [46]. Adding manganese causes the anisotropic magnetostriction to decreases monotonically. Unfortunately, because local anisotropy on Tb sites in these materials is uniaxial, it is only in very strong magnetic fields that magnetostriction due to the rare-earth sublattice can be realized. However, the fact itself of the existence of nonstoichiometric rare-earth intermetallides opens wide possibilities in the search for new magnetic materials.

7. Conclusion

To summarize, the resource of utilizing magnetic materials based on the intermetallic compounds of rare-earth metals with iron group metals is far from exhausted. An important issue is to search for new materials exhibiting large magnetocaloric, magnetostrictive, and magnetoresistive effects. There is still much room for increasing the maximum energy product, extending the operating temperature range, and enhancing the stability of permanent magnets. Today, it is the development of new magnetic materials and of their fabrication technology which determines to a large measure the technological progress.

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