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Features of the formation of magnetic properties of $3d$-transition metals in films of solid solutions of R-T ($R =$ Y, La, Gd; $T =$ Co, Ni)

© A.N. Nizaev, E.V. Kudyukov, A.N. Gorkovenko, E.A. Step-nova, N.V. Selezneva,
A.V. Svalov, V.N. Lepalovskiy, V.O. Vaskovskiy

Ural Federal University after the first President of Russia B.N. Yeltsin,
Yekaterinburg, Russia

E-mail: azat.nizaev@urfu.ru

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The magnetic properties of films of binary systems R_xT_{100-x} ($R =$ Y, La, Gd; $T =$ Co, Ni) obtained by magnetron sputtering in a range enriched in $3d$ metals ($x < 50$) have been studied. The concentration and temperature dependences of spontaneous magnetization, Curie temperature, and average magnetic moment per transition metal atom are obtained. The differences in the magnetism of R-T systems containing different $3d$-elements are interpreted.

Keywords: thin films, rare earth metals, transition metals, magnetic moment, ferrimagnetism

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1. Introduction

Classical $3d$-transition metals such as Fe, Co, Ni, as well as their binary alloys, have been studied for a long time due to the presence of unique magnetic properties due to the band nature of the magnetism of these metals. Their distinctive properties include high values of spontaneous magnetization and low hysteresis, which persist over a wide temperature range. Of particular interest are planar media based on them (single-layer films, composites, sandwich structures), which have certain prospects when used as functional media for magnetodynamics and spintronics devices [1–3] — key practical applications of modern condensed matter physics.

At the same time, interest has recently increased in the study of magnetically ordered media with functionally new properties, which include non-collinear magnetic structures (aspero-, spero-, and spermagnetic). Their existence is usually associated with a specific structural state of objects, leading to an inhomogeneous exchange interaction and dispersion of magnetic anisotropy. The media in which such structures are observed include solid solutions of rare earth and transition metals in the film state, which have a relatively high temperature stability of the magnetic state over a wide concentration range. The vast majority of works devoted to non-collinear magnetic structures relate to nanoscale films of binary systems R_xT_{100-x} ($R =$ Gd, Tb, Dy, Ho; $T =$ Fe, Co, Ni) [4–15]. Amorphous films with $x < 50$ are of particular interest. In this range of compositions, the $3d$-element has a spontaneous contribution to the total magnetization, whereas the R-component may represent a spatially disoriented, but „rigid“ in the exchangeable sense, magnetic structure. As a result, the combination of two subsystems — rare earth (R) and transition (T) — can lead to a spermagnetic structure.

An experimental confirmation of the formation of non-collinearity of magnetization in the R-subsystem is the effective magnetic moment per atom of a rare-earth element determined according to magnetometry data. The difference between this value and the tabular value suggests that in reality the system of R magnetic moments is non-collinear [16–18]. However, isolating the contribution of the R-component is a separate task, since the partial contribution of the $3d$-subsystem remains unknown, changing due to modification of its band structure under the influence of the electronic subsystem of R-elements. To solve this problem, non-magnetic La or Y [19,20] are used as R, which have a close atomic radius with R-elements, as well as a similar configuration of the outer part of the electron shell. Thus, in (Y, La)-T systems, the contribution to spontaneous magnetization remains only from the T-element, and it is possible to determine its effective magnetic moment depending on the composition of the R-T system.

The purpose of this paper is to systematically study the magnetism of $3d$-transition metals (Co, Ni) in the systems Y-T, La-T, Gd-T and its quantitative characterization in the form of concentration dependences of the effective magnetic moment on T-metal atoms.

2. Samples and research methodology

All film samples of the Y-Co, Y-Ni, La-Co, Gd-Co, and Gd-Ni systems were obtained by magnetron sputtering in an argon atmosphere in the co-sputtering mode of single-component R and T-targets. The samples were deposited on Corning glass substrates in the presence of a permanent magnetic field (process field) with a strength of 250 Oe. The composition of the films was varied by changing the ratio of deposition rates of R- and T-metals. The nominal thickness of the films was 100 nm. All samples had a protective coating in the form of a 5 nm thick Ta layer. Structural certification of the samples was carried out on a Bruker D8

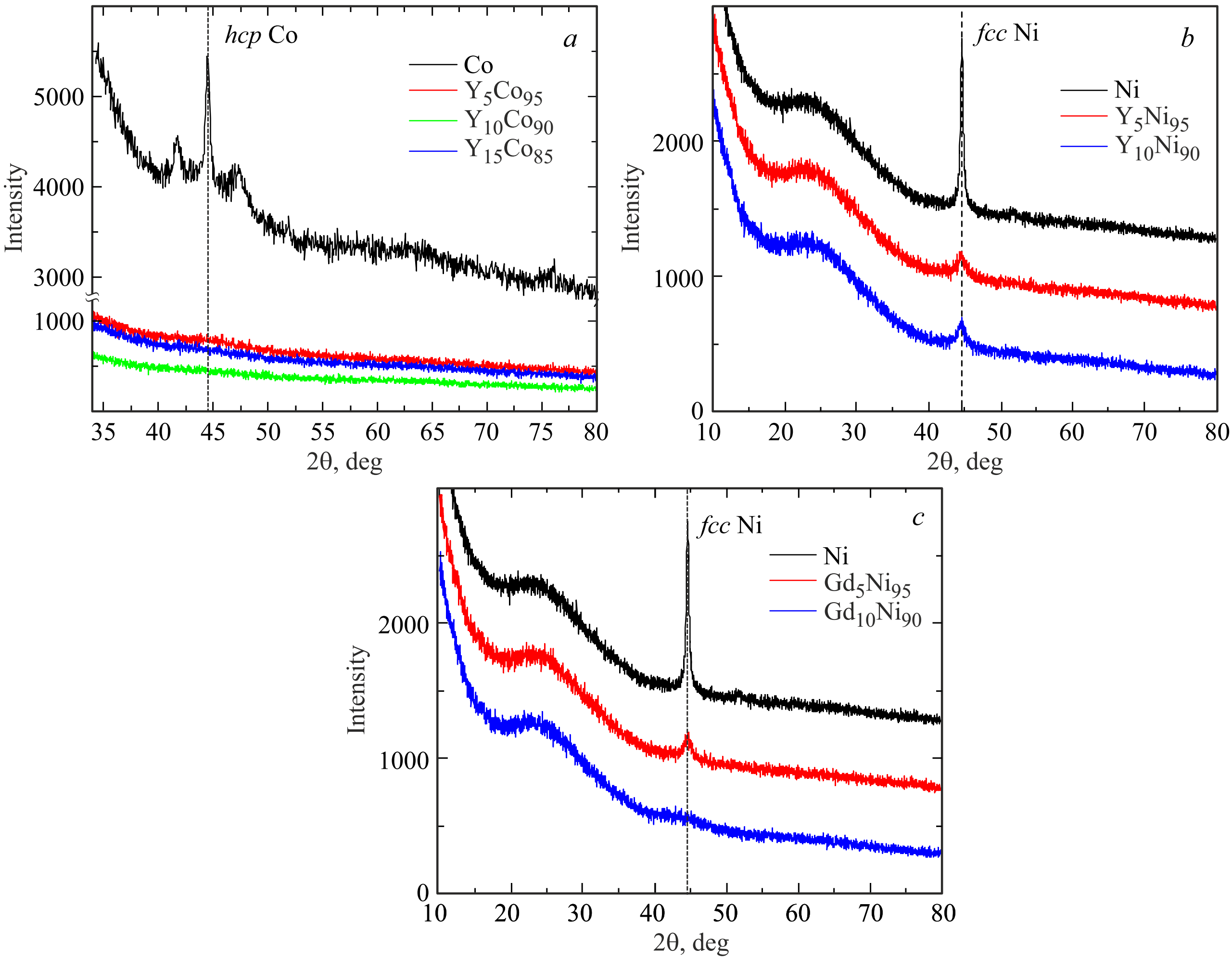

Figure 1. Diffractograms of film systems Y-Co (*a*), Y-Ni (*b*), Gd-Ni (*c*).

diffractometer, the magnetic properties were determined using MPMS EverCool and PPMS DynaCool measuring complexes (optional „vibration magnetometer“), as well as a LakeShore vibration magnetometer in the temperature range from 5 to 700 K in a magnetic field up to 70 kOe, which was oriented in the plane of the film samples.

Structural analysis of films *R*-*T*

The X-ray diffraction analysis data of the films of the studied systems are shown in Figure 1 and in Table 1. They show that films of pure Co and Ni are in a polycrystalline state and have structures of the type *hcp* and *fcc*, respectively. Comparison of calculated values of crystal lattice parameters and X-ray density of metals with tabular values for massive samples [21,22] indicates that the structural parameters undergo a change in the film state. The hexagonal packing of Co is slightly larger in size along the directions *a* and *c*, which results in a decrease in the X-ray density of the metal. A similar situation in the framework of the FCC lattice is observed in the case of Ni.

Table 1. Lattice parameters and density values of Co and Ni

Metal	a, Å	c, Å	ρ, g/cm^3
Co	2.516 (2.507)	4.076 (4.070)	8.76 (8.84)
Ni	3.625 (3.520)	–	8.180 (8.94)

The structural state of the films of 3*d*-elements in combination with rare earths depends on the composition. An increase in the content of *R* leads to a rather rapid transition to an amorphous state in the samples of the *R*-Co system, as evidenced by the absence of diffraction lines in samples with already 5 % Y and Gd content. A slightly different situation is observed in the case of *R*-Ni films: the crystalline phase persists up to a 10 % metal content of *R*-. However, the concentration of the crystalline phase, judging by the intensity of the lines, is low. Thus, it can be argued that rare earth metals with varying degrees of efficiency act as an amorphizer in systems with transition 3*d*-metals.

3. Magnetic properties of systems $R_x\mathrm{Co}_{100-x}$

Films of rare earth metals in combination with Co demonstrate a strong dependence of magnetic properties on composition. This, in particular, can be seen from Figure 2, which shows the temperature dependences of the relative magnetization of the films of the $\mathrm{Y}_x\mathrm{Co}_{100-x}$ system. They were measured with a monotonous temperature increase in a magnetic field with a strength of 100 Oe from a state obtained by cooling the samples in a field of 70 kOe. The dependences are typical for ferromagnetic ordering. As can be seen, the efficiency of the exchange interaction in the Co subsystem decreases sharply with an increasing role of Y, which, in particular, is expressed in a significant decrease in the Curie temperature.

In addition, a change in the concentration of Y leads to a decrease in the effective magnetic moment per Co atom. Its values were determined by the values of saturation magnetization M_s of samples of the corresponding compositions. It was also assumed that the film density is a linear function of the composition. The density of the rare earth element was chosen tabular, while the Co density, taking into account X-ray diffraction data, was taken from Table 1.

The results of the obtained values of the Co moment depending on the concentration of R fare shown in Figure 3. The dots indicate the obtained moment values, the dashed lines indicate the approximation according to the quadratic law. As can be seen, the moment of pure cobalt is close to the theoretical value of $1.7\,\mu_\mathrm{B}$. As the Y or La content increases, the effective moment decreases monotonously down to compositions with $x \approx 65\,\%$. There is a slight discrepancy in the dependencies in the samples in the area of low content of „rare earth“ however, it can be noted that the dependencies $\langle\mu_\mathrm{Co}\rangle(x_Y)$ and $\langle\mu_\mathrm{Co}\rangle(x_{La})$ are quite similar in the area of $x = 50{-}60\,\%$.

This dependence of the effective moment of cobalt is a well-known attribute of amorphous films, which is associated with the filling of the $3d$-suband of cobalt which is responsible for the formation of a spontaneous magnetic moment, with electrons from the outer shells of the R-element [23]. For this reason, with a sufficiently high content of rare earth metal, the Co subsystem becomes practically non-magnetic.

Cobalt in binary systems with magnetically ordered rare earth metals in the high-cobalt region of compositions also has a ferromagnetic ordering, which, together with the negative exchange interaction between it and R-metals, leads to ferrimagnetism [24]. At the same time, the rare earth subsystem can be collinear ($R = \mathrm{Gd}$) or asperomagnetic ($R = \mathrm{Tb}$, Dy, Ho) [25]. The results obtained by us during magnetometry of the samples indicate a ferrimagnetic ordering in the Gd-Co system, which, in particular, follows from the compositional dependence of the saturation magnetization of the films $\mathrm{Gd}_x\mathrm{Co}_{100-x}$ at 5 K (Figure 4). This dependence has a non-monotonic character of variation M_s with a minimum in the area bounded by dashed lines. In fact, the nonmonotonicity $M_s(x)$ reflects a concentration change in the partial magnetizations of the Gd and Co sublattices, the values of which are compared in the region of about $x = 20\,\%$ at 5 K, that is, magnetic compensation takes place.

Also, when considering the region in which the gadolinium subsystem has a collinear ordering and its effective magnetic moment is $7\,\mu_\mathrm{B}$ $(0 < x < 50)$ [25], it becomes possible to determine the contribution of cobalt to the total moment using experimental data about dependency $M_s(x)$. The corresponding effective magnetic moment of Co, depending on the concentration of Gd at a temperature of 5 K, is shown in Figure 5. As can be seen, in this case, with small additions of a rare earth element, the changes in the magnetic moment of Co are less pronounced than in films with „non-magnetic“ Y and La. There is also a difference in the concentration at which the moment vanishes: this value corresponds to 50 % in films with

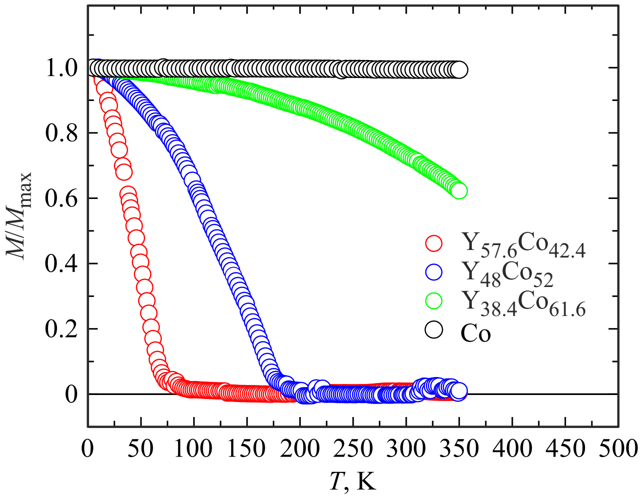

Figure 2. Temperature dependences of the reduced magnetization of films $\mathrm{Y}_x\mathrm{Co}_{100-x}$.

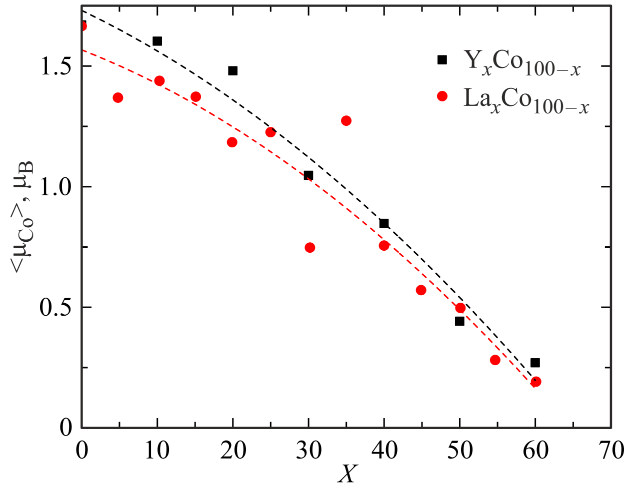

Figure 3. Concentration dependences of the effective moment of Co atoms determined on the Y-Co and La-Co systems.

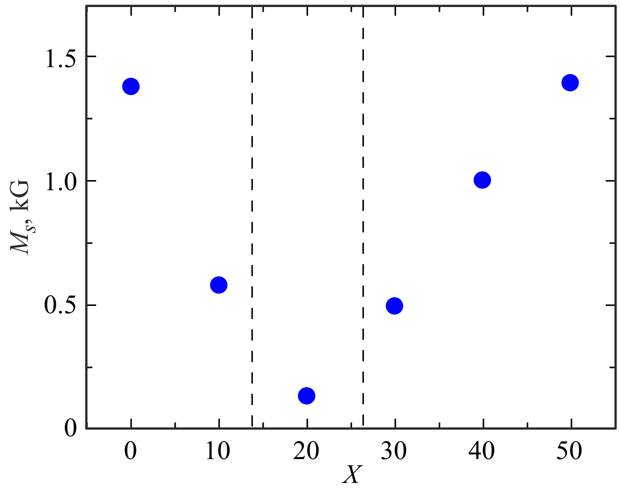

Figure 4. Concentration dependence of spontaneous magnetization at a temperature of 5 K for films of the hboxGd-Co system.

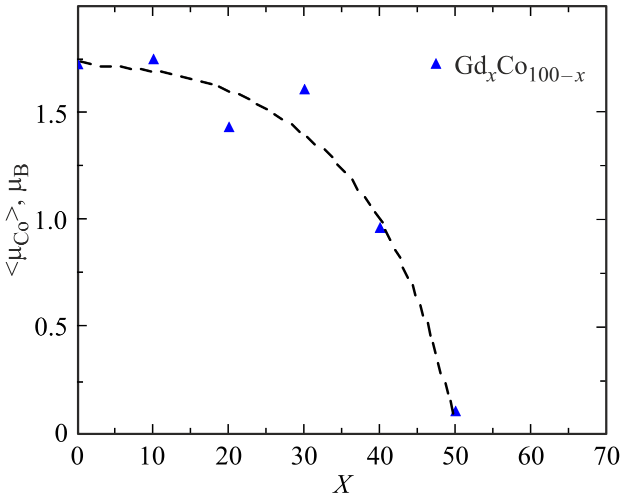

Figure 5. Concentration dependence of the effective moment of Co atoms, determined on the Gd-Co system.

Gd. These discrepancies in the course of the dependences $\langle\mu_{\mathrm{Co}}\rangle(x_R)$ $(R = \mathrm{Y, La, Gd})$ may be a consequence of the existing differences in the electronic configurations of rare-earth metal atoms. If in the case of Y and La there is a similar effect on the $3d$-subband of cobalt, then Gd is more effective in reducing the spontaneous magnetic moment Co.

4. Magnetic properties of systems $R_x\mathrm{Ni}_{100-x}$

In the case of the binary system $\mathrm{Y}_x\mathrm{Ni}_{100-x}$, the magnetism of Ni is also largely determined by the concentration of the rare earth element, as in systems with Co. Figure 6, *a* shows the dependence of the effective magnetic moment Ni on the concentration of yttrium, determined using data on the dependence $M_s(x)$ at 5 K. The film moment of the pure element $\langle\mu_{\mathrm{Ni}}\rangle(0) = 0.58\,\mu_{\mathrm{B}}$ is very close to the theoretical value of $0.6\,\mu_{\mathrm{B}}$, which once again confirms the adequacy of the chosen technique. The monotonously decreasing nature of the dependence is caused by the same modification of the $3d$-subband of nickel, which forms its spontaneous magnetic moment. However, compared with a similar dependence for Co, the achievement of a non-magnetic state here occurs at a much lower concentration of Y. A possible reason for this is a smaller splitting of Ni subbands than splitting of Co, which is leveled at a relatively lower concentration of electrons of R-elements.

The temperature stability of magnetic ordering in $\mathrm{Y}_x\mathrm{Ni}_{100-x}$ films also strongly depends on the composition. Figure 6, *b* shows the dependence of the Curie temperature of films on the concentration of yttrium. For pure Ni, the corresponding value of $T_c(x = 0) = 630\,\mathrm{K}$ coincides with the literature data for the massive sample. A rapid decrease in T_c with an increase in Y concentration indicates a strong change in the exchange bond in the Ni subsystem, depending both on the amount of splitting of the $3d$-band and on the effects of hybridization with the Y conduction band.

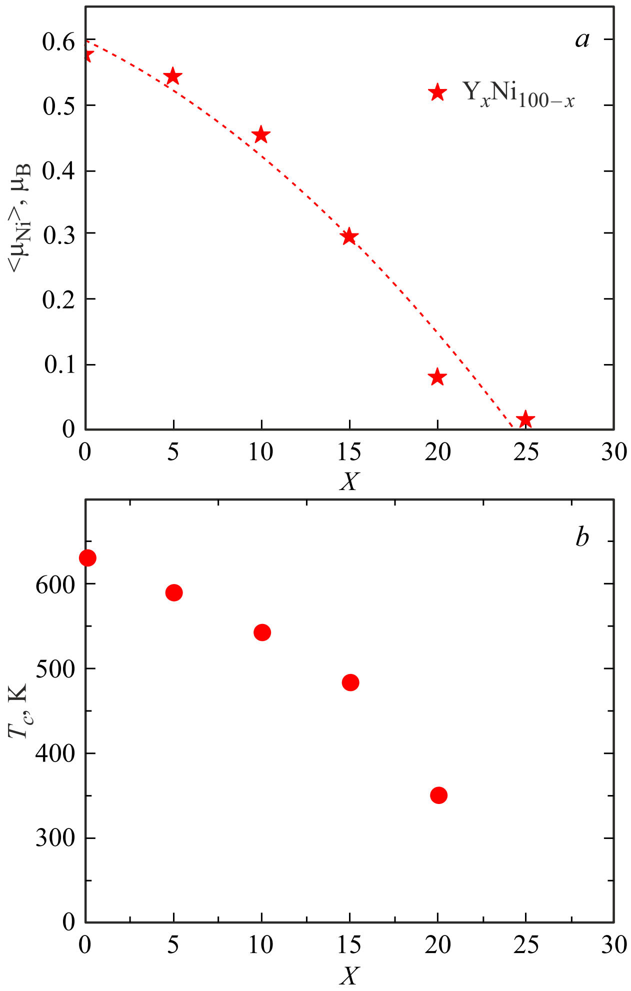

Figure 6. Concentration dependences of the effective magnetic moment of Ni (*a*) atoms and the Curie temperature of the $\mathrm{Y}_x\mathrm{Ni}_{100-x}$ (*b*) system.

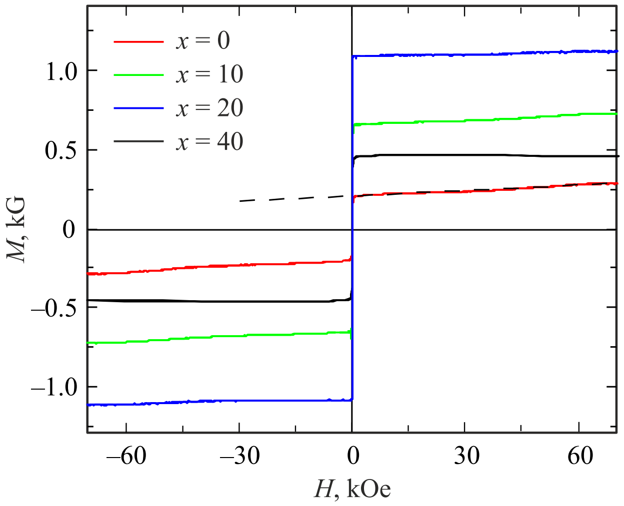

Figure 7. Field dependences of the magnetization of films of the Gd_xNi_{100-x} system, measured at a temperature of 5 K.

Figure 7 shows the field dependences of the magnetization of Gd_xNi_{100-x} films at a temperature of 5 K for different compositions, measured in the sample plane. In this case, the value M_s is determined by linear approximation of the high-field dependence $M(H)$ to the zero field (dashed line on the loop of the film $Gd_{10}Ni_{90}$). The characteristic found in this way, strictly speaking, does not coincide with the saturation magnetization of the samples and is considered by us as a quantitative assessment of the spontaneous magnetization of the ferrimagnetic phase.

The value M_s changes non-monotonously with varying composition, therefore, magnetic compensation takes place. This fact is clearly reflected in the concentration dependence $M_s(x)$ shown in Figure 8. The shaded area reflects the range of Gd concentrations in which the compensation composition is localized. In comparison with the corresponding dependence in the Gd_xCo_{100-x} system, it is found that compensation occurs at a significantly lower concentration of Gd, which is explained by the difference in the magnitude of the moments per Co and Ni atom.

As can be seen in Figure 7, the samples do not reach the saturation state even in the 70 kOe field, which indicates a possible deviation from the collinear ferromagnetic order in the Gd sublattice, which is presumably caused by the presence of a certain fraction of the frustrated magnetic phase, the formation of which is caused in nanoscale films R-T by structural heterogeneity [25,26]. The results of diffractometry show that the nanocrystalline state is preserved in these films up to the Gd content of 10–15 %. Within the framework of this structure, a magnetic order can be realized, characterized by a high dispersion of the local axes of light magnetization, which inevitably leads to frustration (uncertainty) in the location of the local magnetic moments Gd. There may also be another mechanism associated with a highly defective state of the material, in which an RKKY variant of the exchange interaction can be implemented, leading to a dispersion of both the sign and the magnitude of the exchange parameter. At the same time, the stabilizing role of Ni in terms of reducing the frustration of Gd moments is weakly manifested, as a result of which a non-collinear structure of the type of spero- or asperomagnetic remains in the gadolinium sublattice, even with a low gadolinium content. In support of this thesis, it is possible to cite the dependence of the effective magnetic moment Gd per atom, which was obtained using data on $\langle\mu_{Ni}\rangle(x)$ from the Y_xNi_{100-x} system. It is shown in Figure 9. From this dependence, it can be seen that the moment in the composition region under consideration is less important than for a free Gd atom equal to $7\,\mu_B$, which indicates the presence of a certain angle of solution of the asperomagnetic cone of magnetic moments in the Gd sublattice.

The compositional change is also experienced by the temperature dependences of the spontaneous magnetization of the samples, measured in a field with a intensity of 100 Oe, which are shown in Figure 10. A decrease in

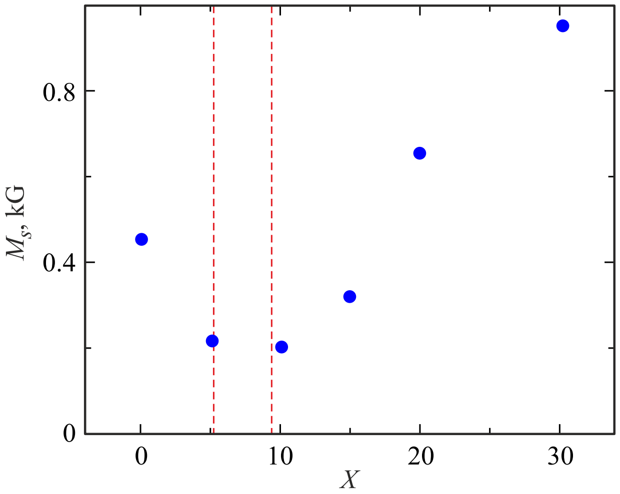

Figure 8. Concentration dependence of the spontaneous magnetization of Gd_xNi_{100-x} films, measured at a temperature of 5 K.

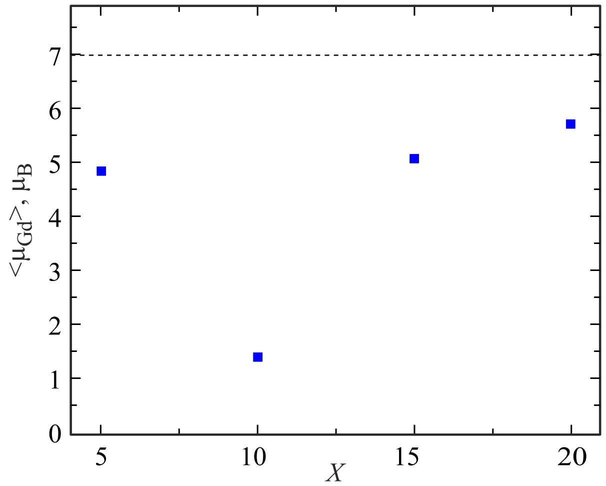

Figure 9. Concentration dependence of the effective magnetic moment of Gd atoms in the Gd_xNi_{100-x} system.

Table 2. Magnetic parameters of films of R-Co systems

x	Y_xCo_{100-x}		La_xCo_{100-x}	Gd_xCo_{100-x}	
	T_c, K	$\langle\mu_{Co}\rangle$, μ_B	$\langle\mu_{CO}\rangle$, μ_B	T_c, K	$\langle\mu_{Co}\rangle$, μ_B
0	–	1.67	1.67	–	1.72
10	–	1.60	1.44	–	1.74
20	–	1.48	1.18	215 (T_{comp})	1.42
30	–	1.04	0.75	–	1.6
40	–	0.85	0.76	–	0.95
50	179	0.44	0.50	307	0.09
60	69	0.27	0.19	216	0

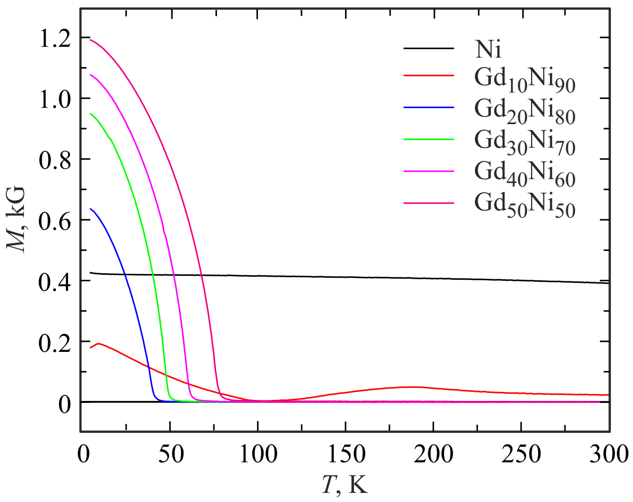

Figure 10. Temperature dependences of the reduced magnetization for films of the Gd_xNi_{100-x} system.

the spontaneous moment of the Ni sublattice with an increase in x naturally leads to an increase in the total magnetization due to the antiparallel orientation of Gd and Ni. Nevertheless, there is also a certain intermediate dependence $M(T)$, which has a nonmonotonic character with a minimum corresponding to zero magnetization at a temperature of 104 K. At this point, the magnetization of the sublattices Gd and Ni are compensated, that is, they have the same magnitude. Due to the fact that gadolinium has a lower temperature stability of the magnetic ordering compared to nickel, the Ni sublattice begins to make a predominant contribution to the total magnetization, which explains the further growth of $M(T)$ to a certain maximum value. The subsequent increase in temperature leads to a decrease in the Ni contribution and, ultimately, to the absence of spontaneous magnetization.

Magnetic compensation is also inherent in other ferrimagnetic (sperimagnetic) films R-T in the region of a higher content of T-elements. As we have already shown earlier, and in a number of other paper, the compensated state of the magnetic sublattices R and T depends both on the concentration of elements and on the temperature course of the partial dependences of the magnetization of the system [27–31], and compensation is formed in a fairly narrow range of compositions. At the same time, the existence of such a feature makes it possible to obtain in films R-T a certain analogue of antiferromagnetic ordering at certain temperatures and compositions, which may be interesting in terms of using these structures for applied purposes [32,33].

5. Quantitative characterization of film magnetism R-T

A summary of all experimental data obtained on the magnetic properties of the (Y, La, Gd)-Co and (Y, Gd)-Ni systems in the in the range of compositions enriched by 3d-metal is presented in Tables 2 and 3. The places with a dash indicate those values of the Curie temperature or the effective magnetic moment that were either not obtained during the experiment or do not relate to a specific composition.

Table 3. Magnetic parameters of films of R-Ni systems

x	Y_xNi_{100-x}		Gd_xNi_{100-x}	
	T_c, K	$\langle\mu_{Ni}\rangle$, μ_B	T_c, K	$\langle\mu_{Gd}\rangle$, μ_B
0	630	0.58	630	–
5	589	0.54	–	4.86
10	542	0.45	104 (T_{comp})	7.25
15	483	0.30	125 (T_{comp})	5.09
20	350	0.08	42	5.73
30	–	0	50	5.91
40	–	0	62	5.67
50	–	0	78	5.68

6. Conclusion

The results of a systematic study of the structural and magnetic properties of films of the binary systems hbox(Y, La, Gd)-Co and hbox(Y, Gd)-Ni over wide field and temperature ranges are presented. It is shown that films of pure $3d$-metals deposited on a glass substrate are in a polycrystalline state with structural parameters whose values differ from the corresponding characteristics of the massive state of these metals. This leads to a reduced density of the film samples, which must be taken into account when quantifying the magnetic characteristics reduced to the atomic scale. R-containing films are X-ray amorphous in the majority of the concentration range. Depending on the type of $3d$-metal, the transition of films to the amorphous state occurs at different concentrations of the R-element.

Experimental dependences of the effective magnetic moment of $\mathrm{Co}\langle\mu_{\mathrm{Co}}\rangle$ atoms on the concentration of the corresponding $R =$ Y, La, Gd have been obtained for the T-Co systems, which show acceptable comparability both among themselves and with literary sources. The $\langle\mu_{\mathrm{Co}}\rangle$ determined from different systems for a pure Co film is close to the theoretical value of $1.7\,\mu_{\mathrm{B}}$. The transition to a non-magnetically ordered state is associated with the leveling of the splitting of the $3d$-subband of Co. The compositional change in the temperature course of spontaneous magnetization of Co indicates a strong relationship between the ordering temperature and composition.

The concentration dependence of the effective magnetic moment $\mathrm{Ni}\langle\mu_{\mathrm{Ni}}\rangle(x)$ is constructed for the T-Ni system. The corresponding moment of Ni at $x = 0$ is in good agreement with the theoretical value of $0.6\,\mu_{\mathrm{B}}$. The destruction of ferromagnetism in the Ni subsystem occurs at a significantly lower concentration of T-elements than in R-Co films, due to the smaller splitting of the $3d$-band. The dependence $\langle\mu_{\mathrm{Ni}}\rangle(x)$ was used to analyze Gd magnetism in the composition of Gd-Ni films. It is found that a deviation from collinearity takes place in the magnetic structure of the Gd subsystem, which is probably caused by a weak exchange between the sublattices.

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Conflict of interest

The authors declare no conflict of interest.

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