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Magnetic state of $R_{1-x}Y_xCo_2$ compounds near the critical concentration

N.V. Baranov^a, A.N. Pirogov^b

*Institute of Physics and Applied Mathematics, Ural State University, 620083 Ekaterinburg, Russia bInstitute of Metal Physics, Russian Academy of Sciences, Ural Branch, 620219 Ekaterinburg GSP-170, Russia

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Abstract

The substitution of R ions in RCo₂ compounds for non-magnetic yttrium leads to the disappearance of the magnetic moment in the itinerant d-electron subsystem at a critical Y concentration. The magnetic state of the Ho_{1-x}Y_xCo₂ and Er_{1-x}Y_xCo₂ compounds near the corresponding critical concentrations $x_c = 0.58$ and $x_c = 0.47$ has been studied by means of neutron diffraction, electrical resistivity and a.c. susceptibility measurements. It is found that a long-range magnetic order coexists with a shortrange order in a narrow concentration range near x_c (1 at.% for (Ho,Y)Co₂). The change in magnetic order with Y concentration in both systems occurs through a magnetic phase transition of first order. The properties of the (Ho,Y)Co₂ and (Er,Y)Co₂ compounds near x_c are strongly affected by the applied magnetic field.

Keywords: Itinerant magnetism; First order magnetic phase transition; Spin fluctuations

1. Introduction

Cubic Laves phases RCo_2 , where R is a rare earth metal or yttrium, have been the subject of a large number of experimental and theoretical studies owing to the coexistence of two interacting magnetic subsystems, namely the localized magnetic moments of rare earth ions and the itinerant 3d electrons of cobalt [1–3].

When the R ions have magnetic moments, the RCo₂ compounds are ferro- or ferrimagnets where the magnetic moment of Co, μ_{Co} , may reach a value of 1 μ_{B} under the influence of an effective field H_{eff} acting from the R subsystem. The $\mu_{Co}(H_{eff})$ dependence shows a metamagnetic behaviour, because the Fermi level $(E_{\rm F})$ lies on a sharply falling part of the density of states N(E) curve with a positive curvature. With H_{eff} increasing up to a critical value $H_{eff}^{\rm c}$, μ_{Co} increases rapidly to 1 $\mu_{\rm B}$. The RCo₂ compounds with non-magnetic R ions (Lu and Y) are Pauli exchange-enhanced paramagnets [4]. Measurements of the magnetization in ultrahigh magnetic fields [5] have shown that the value of μ_{Co} jumps to 0.5 $\mu_{\rm B}$ in YCo₂ and to 0.7 $\mu_{\rm B}$ in LuCo₂ when the field reaches a critical value of about 70 T.

The substitution of R ions in RCo_2 for non-magnetic Y ions allows one to change the H_{eff} value and makes it possible to study the problem of the onset of a

magnetic moment in the subsystem of itinerant delectrons. The critical Y concentration x_c at which the sharp fall in μ_{Co} is observed in $R_{1-x}Y_xCo_2$ compounds depends on the type of R ion [6]. It has also been found that the temperature dependences of the electrical resistivity [7,8], specific heat [6], and thermal expansion [8] vary markedly near x_c and that the magnetic state of compounds with $x > x_c$ can be characterized as that of a spin glass [9].

The aim of the present paper is to determine how the induced character of the moment of Co atoms affects the behaviour of $R_{1-x}Y_xCo_2$ in the vicinity of the critical concentration. We have measured the electrical resistivity, a.c.-susceptibility and neutron diffraction of $Er_{1-x}Y_xCo_2$ and $Ho_{1-x}Y_xCo_2$.

2. Experimental details

The $\operatorname{Er}_{1-x} Y_x \operatorname{Co}_2$ and $\operatorname{Ho}_{1-x} Y_x \operatorname{Co}_2$ compounds were prepared by arc melting in a helium atmosphere and homogenized at 1220 K for 50 h.

The electrical resistivity was measured on samples of dimensions $1 \times 1 \times 6$ mm³ by the four-probe method in the temperature range from 1.8 to 300 K and magnetic fields up to 6 T. The a.c. susceptibility was measured at a frequency of 1 kHz and a field amplitude of 1×10^{-4} T.

Neutron diffraction studies were performed on a device mounted on one of the horizontal channels of the IVV-2M reactor at a wavelength of 2.42 Å in the angle interval $2\theta = 10^{\circ}-125^{\circ}$ and at temperatures from 4.2 to 300 K.

3. Results and discussion

According to previous studies [10,11], the critical concentrations for the $Er_{1-x}Y_xCo_2$ and $Ho_{1-x}Y_xCo_2$ systems are $x_c = 0.47$ and $x_c = 0.58$ respectively. Figs. 1 and 2 show the temperature dependences of the electrical resistivity of the compounds near these concentrations within the temperature range 2–60 K. At higher temperatures where the compounds are paramagnetic the shape of the $\rho(T)$ curves does not depend on the R content or the type of R ion. This behaviour has been observed in all $R_{1-x}Y_xCo_2$ systems and is explained by a predominant contribution to the electrical resistivity from s-electron scattering on spin fluctuations of the itinerant d-electrons via an s-d exchange interaction [12]. As follows from Figs. 1 and 2, both systems exhibit the minimum ρ above the magnetic ordering temperature



Fig. 1. Temperature dependences of electrical resistivity of $\text{Er}_{1-x}Y_x\text{Co}_2$: curves 1, B=0; for x=0.45, curve 2, B=0.4 T; curve 3, B=3.0 T; for x=0.5, curve 2, B=6.0 T; dashed curve corresponds to measurements at B=0 after application of field B=5 T.



Fig. 2. Temperature dependences of electrical resistivity of $Ho_{1-x}Y_xCo_2$: curves 1, B=0; curve 2, B=2 T; curve 3, B=5 T; dashed curve corresponds to measurements at B=0 after application

of field B = 5 T.

 $T_{\rm c}$. In the Dy_{1-x}Y_xCo₂ system an analogous behaviour has been observed [8]. On our opinion this minimum on the $\rho(T)$ curves results from the induced character of the magnetic moments of Co atoms. This suggestion is supported by the fact that the $\rho(T)$ dependences for isostructural $Er_{1-x}Y_xNi_2$ compounds do not exhibit such minima. Unlike the Co atoms in RCo₂, in RNi₂ compounds the Ni atoms are non-magnetic even in compounds with magnetic R ions. According to band structure calculations, this is caused by the fact that in RNi₂ the Fermi level is located on the flat part of the N(E)curve with low values of density of states [13]. As can be seen from Fig. 3, the $\rho(T)$ curve for ErNi₂ is typical of metallic ferromagnets and shows an anomaly at $T = T_c = 8.2$ K. In $\text{Er}_{1-x} Y_x \text{Ni}_2$ the magnetic contribution to the electrical resistivity is caused by s-electron scattering on localized Er magnetic moments. Taking into account the peculiarities of the electronic structure of RCo_2 and the metamagnetic character of the $\mu_{Co}(H_{eff})$ curve, one can explain the minima on the $\rho(T)$ curves near x_c in $R_{1-x}Y_xCo_2$ via the increasing contribution of scattering on localized spin density fluctuations in the Co subsystem upon approaching T_c . These spin density fluctuations in the Co subsystem are caused by fluctuations of H_{eff} and by the existence of a region (clusters) with a short-range order in the R subsystem



Fig. 3. Temperature dependences of electrical resistivity of $\text{Er}_{1-x}Y_x\text{Ni}_2$. The inset shows the concentration dependence of the magnetic ordering temperature.

due to the inhomogeneous distribution of Y over the lattice. With decreasing temperature H_{eff} may reach its critical value, which induces a magnetic moment on Co atoms located within the clusters, i.e. localized spin density fluctuations in the Co subsystem. The increase in the electrical resistivity with decreasing temperature to $T_{\rm e}$ can be connected with the increasing number and volume of these clusters. This situation can take place when the size of clusters is essentially less than the electron free path [14].

The appearance of a long-range magnetic order at $T = T_{c}$ and the formation of an infinite cluster are accompanied by a decrease in ρ . However, in the concentration range $x \approx x_c$ even at low temperatures (T < 4.2 K) the long-range magnetic order does not occur within the whole volume of the sample and there are still some parts of the sample with a short-range order. This is supported by the fact that the residual electrical resistivity of these compounds is very high and is greatly affected by an applied magnetic field. This is seen from Figs. 1 and 2, where the $\rho(T)$ curves at various magnetic fields are shown for Er_{0.55}Y_{0.45}Co₂ and Ho_{0.423}Y_{0.577}Co₂. The magnetic field results in a significant decrease in the electrical resistivity both in the low temperature range and at $T \approx T_c$. In the latter case we observe the disappearance of the minimum on the $\rho(T)$ curves owing to the suppression of spin density fluctuations and with increasing cluster size in an applied magnetic field.

As follows from Figs. 1 and 2, at low temperature (T < 4.2 K) the electrical resistivity decreases in an applied magnetic field by almost 50%, but after removal of the field the sample is not restored to its initial resistivity. As previously shown [10,11], this behaviour is caused by splitting of the Co d-band and an increase in the Co magnetic moment from 0.3 to 1 $\mu_{\rm B}$, accompanied by the suppression of spin fluctuations. Moreover, the field-induced metamagnetic transition in the d-electron subsystem is accompanied by an increase in the unit cell volume [10] of compounds with $x \approx x_c$ as well as by an irreversible decrease in the coefficient of the linear term of the low temperature specific heat γ ($\Delta\gamma/\gamma = -44\%$ [15]).

The irreversibility in the behaviour of Er_{0.55}Y_{0.45}Co₂ and Ho_{0.423}Y_{0.577}Co₂ in an applied field may be connected with the hysteresis in the dependence $\mu(H_{\text{eff}})$ near $H_{\rm eff}^{\rm c}$. According to calculations, such a hysteresis may reach a value of 25 T [16]. The magnetic state of both systems with an yttrium content close to x_c may be characterized by the existence of clusters with $\mu_{Co} \neq 0$ in a paramagnetic matrix at $T > T_c$ and by the existence of clusters with $\mu_{\rm Co} = 0$ in a ferrimagnetic matrix at $T < T_{c}$. This assumption about the magnetic state of $R_{1-x}Y_xCo_2$ near the critical concentration is supported by neutron diffraction measurements. Figs 4 and 5 show the neutron diffraction patterns for $Ho_{1-x}Y_xCo_2$ and $Er_{1-r}Y_rCo_2$ measured at 4.2 K near the (111) and (220) reflections. The neutron diffraction patterns for $Ho_{0.43}Y_{0.57}Co_2$ and $Er_{0.6}Y_{0.4}Co_2$ (x < x_c) exhibit only Bragg reflections with the instrumental angle width. These reflections result from a long-range magnetic order and nuclear scattering. The wave vector of the magnetic structure is $k = (2\pi/a)$ (0,0,0). The neutron diffraction patterns for Ho_{0.15}Y_{0.85}Co₂ and Er_{0.4}Y_{0.6}Co₂ $(x>x_c)$ exhibit only nuclear scattering. The neutron diffraction patterns for $Ho_{1-x}Y_xCo_2$ with x=0.58, 0.6and 0.7 and for $\operatorname{Er}_{1-x} Y_x \operatorname{Co}_2$ with x = 0.45 and 0.5 ($x \approx x_c$) are characterized by magnetic diffuse scattering due to inhomogeneities of magnetization. The maxima of diffuse scattering have an angle width larger than the instrumental one. The mean size of magnetic inhomogeneities calculated from the angle width is about 2 nm.

The flat decrease in the intensity of diffuse scattering, I_d , with temperature for the compositions with $x \ge x_c$ (insets in Figs. 4 and 5) may be connected with the disappearance of magnetic order in clusters of different sizes. The smaller the cluster size, the higher is the disordering temperature.

It should be noted that the $I_d(T)$ dependence correlates with the temperature dependence of the electrical resistivity. As can be seen from Fig. 1 (x=0.5) and from the inset in Fig. 4 (x=0.5), an increase in the electrical resistivity with decreasing temperature takes place at the same temperature at which neutron



Fig. 4. Neutron diffraction patterns near (111) and (220) reflections at 4.2 K for $\text{Er}_{1-x}Y_x\text{Co}_2$: curve 1, x=0.4; curve 2, x=0.45; curve 3, x=0.5; curve 4, x=0.6. The inset shows the temperature dependence of the integral intensity of diffuse scattering near the (111) reflection for the sample with x=0.5.

diffuse scattering is visible. This correlation between $\rho(T)$ and $I_d(T)$ confirms our suggestion about the origin of the resistance minima in $R_{1-x}Y_xCo_2$ near the critical Y content.

The neutron diffraction patterns for $Ho_{0.423}Y_{0.577}Co_2$ and $Er_{0.55}Y_{0.45}Co_2$ exhibit both Bragg magnetic reflections (with k=0) and diffuse maxima, thus pointing to a coexistence of long- and short-range magnetic orders.

The distribution of the intensities of scattered neutrons around reciprocal lattice sites is given by [17]

$$I(\boldsymbol{q}) \sim A^2(\tau) f^2(\boldsymbol{q}) \exp\left(\frac{-(\boldsymbol{q}-\tau)^2}{\Delta^2}\right) + \frac{a^2(\tau) f^2(\boldsymbol{q}) \kappa^2}{(\boldsymbol{q}-\tau)^2 + \kappa^2} \qquad (1)$$



 2θ , degr.

Fig. 5. Neutron diffraction patterns at 4.2 K for $Ho_{1-x}Y_xCo_2$: curve 1, x=0.57; curve 2, x=0.577; curve 3, x=0.58; curve 4, x=0.85. The inset shows the temperature dependence of the integral intensity of diffuse scattering near the (111) reflection for the sample with x=0.58.

where the first term corresponds to Bragg neutron scattering, the second term corresponds to diffuse neutron scattering, q is the transferred momentum, τ is the vector of the reciprocal lattice, Δ is the distribution operator and κ is the inverse correlation radius for inhomogeneities.

In general, to analyse the total scattering near the reciprocal lattice site, one should expand it into a gaussian function with the halfwidth determined by the apparatus resolution and a Lorenz function with the physical halfwidth κ . In our case we can make some qualitative conclusions about the behaviour of the diffuse scattering intensity with x variation by utilizing the results of Ref. [18]. On the basis of the nuclear magnetic

resonance (NMR) data from Ref. [18], one can assume that I_d is proportional to the sample fraction occupied by finite magnetic clusters. To find this fraction, let us turn to the methods of percolation theory, applied, for example, to analyse the magnetic properties of alloys of critical concentration [19].

As mentioned above, owing to fluctuations in the distribution of Y ions, the $R_{1-x}Y_xCo_2$ samples are likely to contain regions in which the field of a metamagnetic transition in the Co subsystem is less than the critical one. These regions will be non-magnetic. The regions for which the condition $H_{\text{eff}} > H_{\text{eff}}^c$ is valid are magnetic. They occupy a volume V(x) which consists of an infinite cluster P(V) and finite clusters V-P(V). The value of P(V) determines the fraction of the sample volume with a long-range magnetic order and consequently the intensity of the Bragg reflections.

Let the concentration distribution z(r) be described by a gaussian function of the density of local probability values [19]. In this case the volume occupied by magnetic regions at 0 K is

$$V(x) = \int_{-\infty}^{x_0} (2\pi \langle \delta x^2 \rangle)^{-1/2} \exp\left(-\frac{(z-x)^2}{2\langle \delta x^2 \rangle}\right) dz$$
(2)

where x_0 denotes the critical concentration without composition fluctuations. An infinite cluster and consequently a long-range magnetic order occur when V(x)exceeds some critical value $V_c(x)$. Numerical calculations performed for various gaussian distributions show [20] that $V_c(x)$ is universal:

$$V_{\rm c}(x) = 0.17 \pm 0.01 \tag{3}$$

In this case

$$V(\mathbf{x}_{\rm c}) = V_{\rm c} \tag{4}$$

Assuming that the Co magnetic moment equals $\mu_{Co}(0)$ for $z < x_0$ and $\mu_{Co} = 0$ for z > x, the magnetization of the Co subsystem for an infinite cluster vs. concentration is

$$\mu_{\rm Co}({\bf x}) = \mu_{\rm Co}(0)P(V) \tag{5}$$

According to calculations [19] one may assume that

$$P(V) = \begin{cases} V, & 1 > V > 0.3 \\ [0.613 - (0.3 - V)](V - V_c)^{0.35}, & 0.3 > V > V_c \end{cases}$$
(6)

Fig. 6 shows the experimental and calculated $\langle \delta x^2 \rangle^{1/2}$ (by Eqs. (2), (5) and (6)) values of $\mu_{Co}(x)/\mu_{Co}(0)$ for $Ho_{1-x}Y_xCo_2$ and $Er_{1-x}Y_xCo_2$. It is seen that for $x < x_c$ they agree satisfactorily. In the calculation we assumed that $\langle \delta x^2 \rangle^{1/2} = [x(1-x)/N]^{1/2} = 0.04$, $x_0 = 0.542$ and $\langle \delta x^2 \rangle^{1/2} = 0.11$, $x_0 = 0.365$ for $Ho_{1-x}Y_xCo_2$ and $Er_{1-x}Y_xCo_2$ respectively (N is the number of atoms in a finite cluster). Using the obtained values of V(x) and



Fig. 6. Concentration dependences of fraction of infinite cluster, P(V), and magnetic moment of Co subsystem, $\mu_{Co}(x)/\mu_{Co}(0)$, for $\operatorname{Er}_{1-x}Y_x\operatorname{Co}_2$ (circles) and $\operatorname{Ho}_{1-x}Y_x\operatorname{Co}_2$ (triangles): \bigcirc , calculated from data on magnetovolume anomaly [21]; $\textcircled{\bullet}$, \blacktriangledown , obtained from neutron diffraction [10,11]; \triangle , obtained from magnetization measurements [7].

P(V), we plot the concentration dependence of the volume occupied by finite clusters in Fig. 6. It is seen that at $x=x_c$ this volume is maximum for both the compounds. For $x < x_c$ the infinite cluster coexists with the finite magnetic clusters.

Upon magnetization in an applied field the finite clusters should behave as super paramagnetic particles and this should be manifested in the magnetization vs. field curves. As shown in Ref. [11], this is the case for $x < x_c$. There is an essential difference between the calculated $\mu_{\rm Co}(x)/\mu_{\rm Co}(0)$ curve and the experimental magnetic measurement data [7] for $x > x_c$. This is seemingly due to the fact that in determining $\mu_{\rm Co}$, the decrease in the R subsystem magnetism with increasing x at $x > x_c$ as well as the probable increase in $\mu_{\rm Co}$ near x_c in an applied magnetic field was neglected.

Using the data of the concentration dependence of V-P(V) from Fig. 6 (inset) and assuming $I_d \approx V-P(V)$, by Eq. (1) we may get the calculated $I_d(x)$ curves e.g. for Ho_{1-c}Y_cCo₂. These are shown in Fig. 7 by solid line. It is seen that the calculated curves describe the experimental situation well qualitatively. The results obtained allow us to suggest that the H_{eff} fluctuations and metamagnetic behaviour of the μ_{Co} (H_{eff}) curve are responsible for the appearance of the regions with a short-range magnetic order in $R_{1-x}Y_x$ Co₂.

One more peculiarity in the behaviour of the systems is worth mentioning. Fig. 8 shows the temperature dependences of the a.c. susceptibility for $\text{Er}_{0.55}Y_{0.45}\text{Co}_2$ and $\text{Ho}_{0.423}Y_{0.577}\text{Co}_2$. As shown in Ref. [7], for



Fig. 7. Experimental (\bigcirc) and calculated (\longrightarrow) maxima of magnetic diffuse scattering around reciprocal lattice site (111) for Ho_{1-x}Y_xCo₂.



Fig. 8. Temperature dependences of a.c. susceptibility of (1) $E_{T_{0.55}}Y_{0.45}Co_2$ and (2) $Ho_{0.423}Y_{0.577}Co_2$: solid line indicates magnetic ordering temperature T_c obtained from $\rho(T)$ dependences; dashed line corresponds to T_c obtained from neutron diffraction.

Ho_{1-x}Y_xCo₂ with small x the temperature of the susceptibility maximum and the temperature of the sharp drop in electrical resistivity coincide. However, this is not the case for higher x. In Fig. 8 we have marked by solid lines the temperatures of the sharp drop in ρ with decreasing T. Also indicated is the temperature at which magnetic neutron scattering disappears for Er_{0.55}Y_{0.45}Co₂ (dashed line). The above temperatures are seen to be different. Different values of T_c obtained

by different methods are caused by the inhomogeneous and complicated magnetic structure of compounds near the critical concentration.

4. Conclusions

Taking into account previous results [10,11,15] and the data obtained in the present study, we can draw the following conclusions.

(1) A coexistence of short- and long-range magnetic orders is observed near the critical concentration in $R_{1-x}Y_xCo_2$ ($R \equiv Er$, Ho). The magnetic state of these compounds is changed upon Y substitution within a narrow concentration range (1 at.% for (Ho,Y)Co₂) through a first-order magnetic phase transition. This behaviour is a result of the itinerant metamagnetism of the Co subsystem.

(2) The critical concentrations at which the Co magnetic moments disappear are $x_c = 0.47$ and $x_c = 0.58$ for the Er and Ho systems respectively. The value of the critical field H_{eff}^c obtained from these data is close to 70 T, in agreement with the data obtained from measurements of the magnetization of YCo₂ in ultrahigh magnetic fields [5].

(3) The magnetic state of the $R_{1-x}Y_xCo_2$ compounds near x_c can be described by means of percolation theory.

(4) Near the critical concentration an irreversible splitting of the Co d-band by a small magnetic field is observed. This splitting is accompanied by an increase in the unit cell volume and the suppression of spin density fluctuations. It causes an abrupt decrease in the electrical resistivity $(\Delta \rho / \rho \approx -50\%)$ and a decrease in the coefficient γ in the linear term of the expression for the low temperature specific heat $(\Delta \gamma / \gamma = -44\%$ for $\text{Er}_{0.5}\text{Y}_{0.45}\text{Co}_2)$.

(5) The above features of the magnetic state found for the Ho_{1-x}Y_xCo₂ and Er_{1-x}Y_xCo₂ compounds are likely to be typical also of other R_{1-x}Y_xCo₂ (R = Tm, Dy) compounds characterized by a first-order phase transition at $T = T_c$.

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