Magnetic properties of Dy$_x$U$_{1-x}$Co$_2$ system

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The magnetic properties of Dy$_x$U$_{1-x}$Co$_2$ system, investigated in the 4-800 K temperature range and fields up to 80 kOe indicate ferrimagnetic order for $x$<0.2. The Curie temperatures decrease with decreasing Dy content. The internal magnetic field dependence of the Co moment indicates that cobalt moment becomes saturated above 1500 kOe. The results confirm the induced character of cobalt moment.

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1. Introduction

The magnetic properties of the cubic MgCu$_2$ Laves phase compounds between the rare earth and transition metals indicated that ferrimagnetic and ferrimagnetic orders occur for light and heavy rare earth compounds, respectively. DyCo$_2$ presents the ferromagnetic behavior, the cobalt moment is opposite to that of Dy [1]. In this paper the influence of localized 4f magnetic moment of Dy on Co moment is discussed studying the magnetic behavior of Dy$_x$U$_{1-x}$Co$_2$ system. UC$_2$ has a temperature independent susceptibility and crystallizes in the cubic of MgCu$_2$ type symmetry [2,3]. We followed to determine the magnetic moment on Co atoms and to obtain indications on the strength of the exchange interactions.

2. Experimental

The polycrystalline samples were synthesized by simultaneously melting the constituents in an argon arc furnace. Several times melting assured a good homogeneity. The samples were thermally treated at 1100 K for five days. The X ray diffraction pattern showed the presence of MgCu$_2$ type structure [4].

The magnetic measurements were carried out in the 4-800 K range and fields up to 80 kOe. The spontaneous magnetization $M$, was determined from the magnetization isotherms according to the approaching to saturation law: $M = M_s (1 - a / H)$, where a is the coefficient of the magnetic hardness. Above the magnetic transition temperatures, the susceptibility $\chi$ values were determined from their field dependence, according to the relation $\chi = \chi + cM$, H$^{-1}$ by extrapolating the measured values $\chi$ to H$^{-1}$ →0. We designated by c the presumed impurity content and $M_s$ is its saturation magnetization. By this method any possible alteration of the magnetic susceptibility due to the presence of the magnetic ordered impurity content was avoided.

3. Results

DyCo$_2$ is reported as a ferrimagnet with a saturation magnetization of 6.6 $\mu_B / \mu$ indicating a superposition of 8.8 $\mu_B$ the Dy magnetic moment and opposite moments of 1 $\mu_B$ per Co atom. The Curie temperature is reported as 135 K, 159 K [1.5].

The temperature variations of the magnetizations are plotted in Fig. 1. Replacing Dy by uranium the magnetizations and the Curie temperatures decrease. The transition temperatures are nonlinearly composition dependent (Fig. 2). Assuming that the Dy moment is not altered by uranium substitution, the Co magnetic moments were obtained. The composition dependence of these values plotted in Fig. 3 indicates the influence of the Dy on Co magnetic moment value. The ordered phase parameters are included in Table 1. The paramagnetic measurements [4] show that the reciprocal susceptibility has a trend characteristic to the ferrimagnetic ordering. The effective magnetic moments calculated from the linear high temperatures dependence of the reciprocal susceptibility are presented in Table 1.

<table>
<thead>
<tr>
<th>$X$</th>
<th>1</th>
<th>0.8</th>
<th>0.6</th>
<th>0.4</th>
<th>0.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_c$(K)</td>
<td>150</td>
<td>130</td>
<td>85</td>
<td>50</td>
<td>20</td>
</tr>
<tr>
<td>$\mu / (\mu_B / \mu)$</td>
<td>6.8</td>
<td>5.04</td>
<td>3.38</td>
<td>1.92</td>
<td>0.9</td>
</tr>
<tr>
<td>$\mu_{Co} / (\mu_B / \mu)$</td>
<td>1</td>
<td>1</td>
<td>0.9</td>
<td>0.8</td>
<td>0.4</td>
</tr>
<tr>
<td>$\mu_{exp} / Co$</td>
<td>2.5</td>
<td>2.45</td>
<td>2.44</td>
<td>2.6</td>
<td>2.7</td>
</tr>
<tr>
<td>$H_{int}$(kOe)</td>
<td>1880</td>
<td>1500</td>
<td>1128</td>
<td>752</td>
<td>376</td>
</tr>
</tbody>
</table>

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4. Discussion

UCO₂ compound is an enhanced paramagnet with a susceptibility \( \chi = 10.62 \times 10^{-3} \text{ emu/mol} \) [2,6]. An evaluation of the uranium magnetic moment is possible using the Straub and Harrison method [7] taking into account the interactions between s,p,d and f atomic orbitals. The criterion for determination the magnetic state comparing the covalent energy to the critical one (1.38 eV) seems to be good for uranium compounds [8]. The distances between uranium atom and the nearest neighbours of U and Co, involved in the calculation of \( V_\alpha \) and \( V_\beta \), were obtained using the atom positions for UCO₂ reported in [3]. The values of 0.772 eV and 1.02 eV lead to a covalent energy of 1.25 eV determined by strong delocalization and intense hybridization and close to the magnetic instability limit.

\[
\mu_B = M_D(0) H_D(1/kT) \quad \text{and} \quad \mu_C = \mu_C(0) H_C(1/kT).
\]

\( B(x) \) represents Brillouin function. \( \mu_D(0) = 8.8 \mu_B \) and \( \mu_C(0) = \frac{\mu_C}{2} \mu_B \). Near transition temperature the approximations made in \( B(x) \) leads to the expression involving \( T_C \):

\[
\beta - \frac{kT_C}{2\mu_C(0)} \left( \alpha - \frac{45kT_C}{17\mu_C(0)} \right) - \gamma^2 = 0. \]

Fitting the experimental data \( \mu = \mu_D - 2\mu_C \) for DyCo₂ the parameters obtained are: \( \alpha = 15.44 \mu_B^2 \) and \( \beta = 243.66 \mu_B^2 \) and \( \gamma = -41.7 \). For DyCo₂ the theoretical curve is presented in Fig. 1. For Dy₃U₁Co₂ using the experimental values for \( T_C \), the \( \mu_C(0) \) for different Dy content were evaluated. These values, plotted in Fig. 3, may be compared with the experimental ones.

The magnetic properties of RC₀₂ systems were discussed in the two models [10]. The exchange enhanced paramagnetism model describes the magnetic behavior around the transition temperatures. At higher temperatures from the linear Curie-Weiss type behavior the effective magnetic moments of Co atoms presence were evidenced. For this high temperature region the spin fluctuations model is proper to describe the paramagnetic properties of ferrimagnetic RC₀₂ (R-rare earth) compounds [10].

According to exchange enhanced paramagnetism model [11,12] Dy possesses a well-localized magnetic moment having its free ion value and Co generates electronic energy bands showing an exchange-enhanced paramagnetism susceptibility. The Curie temperatures is expressed as:

\[
T_C = x \frac{N \mu_e^2}{3k} \left( g_j - 1 \right) J(J+1)(2J_{Dy}Dy JCoCo + J_{Dy}Co \chi_d(T_C)). \]

\( N \) is Avogadro’s number, \( J \) the quantum number of the total angular momentum of Dy 4f electrons, \( g_j \) is Landé’s factor and \( \chi_d \) is the susceptibility of d electrons at \( T_C \). \( D_yDy \) may be correlated to molecular field coefficient \( \alpha \) [9] having the value 6.25 mol. ft. emu⁻¹ and leading to spin-spin exchange interaction constants \( J_{DyDy} = g_j^2 \chi_d(T_C) \) (\( g_j - 1 \)² 50 mol. ft. emu⁻¹). For \( J_{DyCo} = g_j n_{DyCo} (g_j - 1) \) using \( \gamma \), the value is 135 mol. ft. emu⁻¹. These values are very close of those reported in [1] and of those considered discussing the properties of (RY)Co₂ and R(3d⁺) compounds [12]. The Curie temperatures according to the expression, is not linearly composition dependent because of temperature and composition variation of \( \chi_d \) (Fig. 2).

The molecular field acting on Co moment due to exchange interaction with the localized 4f spins, neglecting other exchange interactions is dependent on the Dy content: \( H_C = n_{DyCo} M_{Dy} = xN_{DyCo} (g_j - 1) \mu_B \). The internal field for DyCo₂ is 1884 kOe. The exchange field variation of Co moment for Dy₃U₁Co₂ is represented in Fig. 4. In order to compare the behavior with other system where Dy is replaced by a nonmagnetic element there are plotted the
experimental Co moments for Dy$_x$Y$_{1-x}$Co$_2$ [13] and Dy$_x$Zr$_{1-x}$Co$_2$ [14]. The same trend is observed in all these systems and proves that the Co moment saturates over 1500 kOe.

5. Conclusions

The low temperature measurements show that Co moment in Dy$_x$U$_{1-x}$Co$_2$ system is opposite oriented to Dy one determining a ferrimagnetic type behavior. The Co moments are decreasing as the uranium is substituting for dysprosium. These moments are induced by the exchange field exerted by the localized 4f moments and saturates over 1500 kOe.

References


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