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Paramagnetic anisotropy of dysprosium- and holmium-yttrium alloys

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The paramagnetic susceptibility of Y–2-at.%-Dy, Y–25-at.%-Dy, Y–50-at.%-Dy, and Y–2-at.%-Ho solid-solution single crystals have been measured between 100 and 360 K. In the alloys with Dy, the anisotropy of the reciprocal susceptibility increases only very slowly with the concentration. This may mean that bilinear exchange anisotropy is small. The thermal variation of the second-order one-ion anisotropy parameter u_{2z}^0 is in agreement with the screened point-charge model. When evaluated at sufficiently high temperature, the variation of u_{2z}^0 from one rare earth to another is found to be close to that corresponding to this model.

I. INTRODUCTION

The study of the magnetic properties of the alloys of rare earths with yttrium and lutetium is of great interest in the analysis of the magnetic anisotropy of these metals. The studies on these alloys permit one to distinguish the exchange anisotropy from the one-ion anisotropy. The latter can be better obtained from dilute alloys, where the magnetostriction is less important than in the pure rare earths.

Until now, only Høg and Touborg[1–4] have measured the susceptibilities of dilute alloys of rare earths in Y, Lu, and Sc below 100 K. In these alloys the exchange anisotropy is certainly negligible. Høg and Touborg have computed the four crystalline-potential parameters by the Van Vleck method.[5]

We have decided to complete these data by measuring the paramagnetic susceptibilities of alloys of increasing concentration of rare earths in Y, between 100 and 360 K. Several things are of interest here. At these temperatures only the two first terms of the $1/T$ expansion of the reciprocal susceptibilities are significant.[6,7] Two selection rules require that the constant term of the anisotropy of the reciprocal susceptibility $1/\chi_z - 1/\chi_x$ between the direction $\vec{z}$ of the sixfold c axis and an $\vec{x}$ direction in the basal plane depend only on the two parameters u_{2z}^0 and $n_{zz} - n_{xx}$ of the atomic Hamiltonian.[6,8] These parameters represent the second-order one-ion and the bilinear exchange anisotropies, respectively. The Y ions are trivalent and their electronic structure is close to that of the rare earths. As a first approximation we can consider that the crystalline potential on one rare-earth ion and the exchange interaction between two ions are independent of the concentration for a given value of the lattice parameters. In this case the one-ion anisotropy energy is proportional to the concentration and the exchange anisotropy energy to the square of the concentration. Within the above approximation we can evaluate the relative contributions of the second-order one-ion and bilinear exchange anisotropies from measurements on alloys of increasing rare-earth concentrations at high temperature.

Only the crystalline potential u_{2z}^0 can be reached from high-temperature measurements; therefore it can be obtained with a far greater accuracy than at low temperature, where the contributions of the four parameters are mixed. The Van Vleck-method calculation must be done on a wide interval of temperatures where the lattice parameters vary; contrarily to our method, it can be used only for the very-low-concentration range where the exchange interactions are very small, since they are treated as perturbations, and can be evaluated with only poor accuracy. Besides, at these temperatures where the magnetization becomes important on each ion, general or local magnetostriction effects can occur, and the difference of polarization of the conduction band induced by the ionic magnetic moments may change the value of u_{2z}^0.

We have determined on our alloys the variation of $1/\chi_z - 1/\chi_x$ with rare-earth concentration at 0 °C to obtain the contributions of the two anisotropies. The variation of $1/\chi_z - 1/\chi_x$ with the temperature and its change from Dy to Ho was compared with the prediction of the point-charge model.

We then compared our results with those of Touborg and Høg on their alloys with Y and Lu and with the anisotropy of alloys of rare earths with gadolinium.

II. EXPERIMENTAL PROCEDURE

The four crystals were purchased from Metals Research. They are 4-mm-diam spheres of Y–2-at.% Dy, Y–25-at. % Dy, Y–50-at. % Dy, and Y–2-at. % Ho, made from sublimed material of 99.9% purity.

The susceptibilities were measured between 100 and 360 K: by Aléonard with a translation balance,[9] in the $\vec{z}$ direction of the sixfold c axis and in a direction $\vec{x}$ in the basal plane. The applied magnetic field was between 3000 and 5000 Oe. In this range the measured susceptibility is equal to the initial susceptibility. The accuracy of the orientation of the crystals was of the order of 1°, and the susceptibilities were measured accurately within 0.2%. Between the c axis and the basal plane, the susceptibility varies sinusoidally with a period of π, for all the alloys in the paramagnetic region, in agreement with the theory.

The contribution of Y to the susceptibility[3] was substracted, assuming that the susceptibilities of the Y ions are the same as in pure Y. The curves $1/\chi_z$ vs T and $1/\chi_x$ vs T, where T is the temperature in Kelvin and $1/\chi_z$ and $1/\chi_x$ the contributions of the rare earths to the reciprocal susceptibilities are given in Figs. 1–4. The values of χ_z and χ_x are in cgs units (erg/G^{-2}) and are for 1-g atom of rare earth. The reciprocal susceptibilities vary linearly with temperature over the entire interval of measurements for Y–2-at. % Ho, above 250 K for Y–2-at. % Dy, and above 200 K for the two other alloys. They can be represented by $1/\chi_z = (1/C_z)(T - \Theta_{pz})$ and $1/\chi_x = (1/C_x)(T - \Theta_{px})$, where C_z and C_x are the experimental Curie constants and Θ_{pz} and Θ_{px} the experimental Curie temperatures. C_z and C_x are only slightly larger than their theoretical values. The values of these constants are given in the Table I. At lower temperatures, the higher-order terms of the thermal expansions of the susceptibilities become important, and the ordering of Y–50-at. % Dy begins below 115 K.

III. INTERPRETATION OF SUSCEPTIBILITIES OF OUR ALLOYS

To zeroth order in $1/T$, the expression of the susceptibility obtained by an exact calculation[6] is the same as that obtained in the molecular field approximation.[7] The reciprocal susceptibilities of a single crystal of solid solution of rare earth with Y along the $\vec{z}$ axis and an $\vec{x}$ axis in the basal plane are given for 1-g atom of rare earth by the simple expressions

$$\frac{1}{\chi_z} = \frac{1}{C}\left(T + \mathcal{C}\,\frac{2J(J+1)}{3Nk}\,n_{zz} + \frac{(2J-1)(2J+3)}{5k}\,u^0_{2z}\right) + O\left(\frac{1}{T}\right),$$
$$\frac{1}{\chi_x} = \frac{1}{C}\left(T + \mathcal{C}\,\frac{2J(J+1)}{3Nk}\,n_{xx} - \frac{(2J-1)(2J+3)}{10k}\,u^0_{2z}\right) + O\left(\frac{1}{T}\right) \qquad (1)$$

for an atomic concentration $\mathcal{C}$ of rare earth. C is the spectroscopic Curie constant and k is the Boltzman constant. u^0_{2z} is the coefficient of $O^0_{2z}(J) = 3J_z^2 - J(J+1)$ in the expansion of the crystalline potential when $\vec{z}$ is the quantization axis and n_{zz} and n_{xx} are the sum on one ion gram of rare earth of all the bilinear parts of the exchange interac-

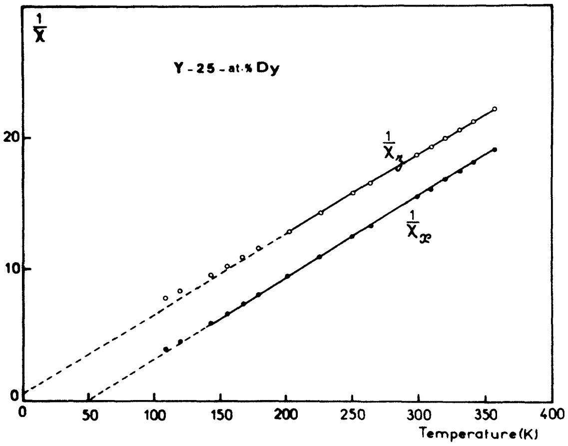

FIG. 2. Contributions of Dy to reciprocal susceptibilities of Y–25-at. % Dy.

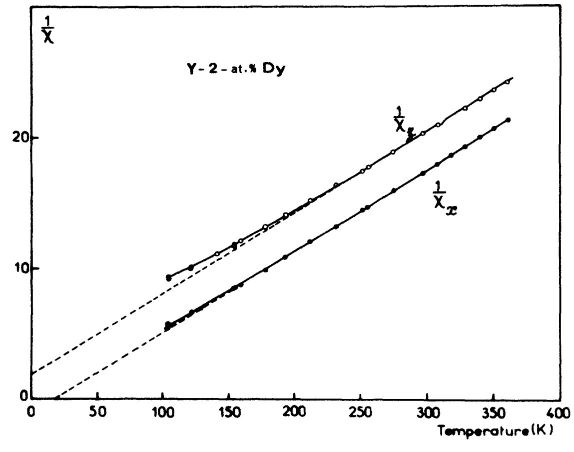

FIG. 1. Contribution of Dy to reciprocal susceptibilities of Y–2-at. % Dy.

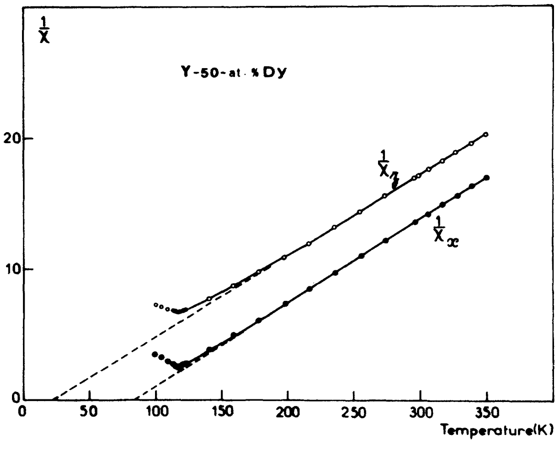

FIG. 3. Contributions of Dy to reciprocal susceptibilities of Y–50-at. % Dy.

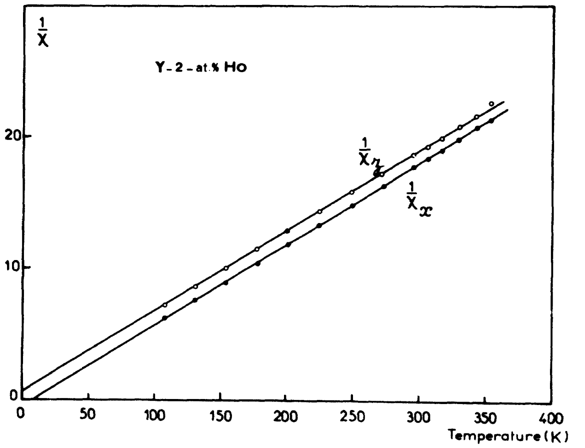

FIG. 4. Contributions of Ho to reciprocal susceptibilities of Y–2-at. % Ho.

tions when the magnetic moments are, respectively, along the $\vec{z}$ and $\vec{x}$ axes

$$n_{zz}=\sum_{(ij)i\neq j} n_{ijzz} \text{ and } n_{xx}=\sum_{(ij)i\neq j} n_{ijxx}.$$

The bilinear exchange Hamiltonian between two magnetic ions i and j is $\mathcal{H}=\vec{J}_i\cdot\vec{\vec{n}}_{ij}\cdot\vec{J}_j$.

A. Anisotropy of the susceptibility at 0°C

The quantities u^0_{2z}, n_{zz}, and n_{xx} vary with the temperature, since they depend on the lattice parameters c along the c axis and on a, the distance between two atoms in the basal plane. In the screened point-charge model, when expressed in erg/atom, u^0_{2z} is given by[8,10]

$$u^0_{2z}=4.12\,\frac{e^2}{4}\,\alpha_J\, Z\langle r^2\rangle\,\frac{c/a-\sqrt{\frac{8}{3}}}{a^3}\,, \tag{2}$$

where e is the electronic charge in cgs electrostatic units, α_J is a coefficient tabulated by Hutchings,[11] whose value depends on the rare earth, Z is a coefficient that is equal to 3 in the absence of screening, and $\langle r^2\rangle$ is the average value in cm^2 of the distance squared between a $4f$ electron and the center of the ion.

Thus it is more accurate to interpret the anisotropy of the paramagnetic susceptibility at a given temperature. We choose 0 °C, which is well within the domain of linearity of $1/\chi_z - 1/\chi_x$ vs T. The lattice parameters of Y are very close to those of Dy, especially at 0 °C, where the values of a are 3.5923 and 3.6449 Å and those of c are 5.6545 and 5.7305 Å for Dy and Y, respectively[12]; the value of $(c/a-\sqrt{\frac{8}{3}})/a^3$ is -1.270×10^{-3} for Dy and -1.255×10^{-3} for Y in Å^{-3}. Thus it is not necessary to make any correction for the variation of a and c with the alloys.

We see in Table II that $(1/\chi_z - 1/\chi_x)$ (0 °C) increases regularly with the concentration, this increase being almost linear for the three Dy-Y alloys. The variation of about 20% from Y–2-at. % Dy to pure Dy may be due to the appearance of exchange anisotropy with increasing concentration. The one-ion anisotropy, which is present alone at low concentration, always remains preponderent. The variation of $1/\chi_z - 1/\chi_x$ from Y–2-at. % Ho to pure Ho is smaller than in the case of Dy. It is smaller yet if we compare u^0_{2z} of the alloy with u^0_{2z} in pure Ho for the same a and c (Table III). This variation is not known with enough accuracy to be interpreted.

In Table II we give also the values of u^0_{2z}/k (0 °C) corresponding to $(1/\chi_z - 1/\chi_x)$ (0 °C) when the exchange anisotropy is neglected, and of $u^0_{2z}/k\alpha_J$ (0 °C). This last quantity is independent of the rare earth in the point-charge model. It actually varies by about 17% from Dy to Ho, which is relatively small.

B. Thermal variation of u^0_{2z}

In the point-charge model, u^0_{2z} varies with the temperature as $(c/a-\sqrt{\frac{8}{3}})/a^3$. In a previous pa-

TABLE I. Experimental data.

Alloys	C_z (cgs, g atom)	C_x (cgs, g atom)	C_{spect} (cgs, g atom)	Θ_{pz} (K)	Θ_{px} (K)	$\Theta_{px}-\Theta_{pz}$	$\frac{1}{3}(\Theta_{pz}+2\Theta_{px})$
Y–2-at. % Dy	16.00	15.97	14.17	−29	19	48	3
Y–25-at. % Dy	16.56	16.22	14.17	−9	49	58	29.7
Y–50-at. % Dy	16.17	15.67	14.17	21	83	62	62.3
Dy	14.85[a] 14.55[b]	14.4[a] 14.21[b]	14.17	109[a] 118[b]	167[a] 171[b]	58[a] 53[b]	147.7[a] 153.3[b]
Y–2-at. % Ho	16.28	16.17	14.07	−9	9	18	3
Ho	15.2[a] 14.50[b]	15.1[a] 14.42[b]	14.07	75[a] 89[b]	91.5[a] 104[b]	16.5[a] 15[b]	86[a] 99[b]

[a] From R. Aléonard *et al.* (Ref. 15).

[b] From R. Aléonard (Ref. 16).

TABLE II. Anisotropy of various alloys (present measurements).

Alloys	$\frac{1}{\chi_z}-\frac{1}{\chi_x}$ (0 °C) (cgs, 1 g atom)	b	$\frac{u^0_{2z}}{k}$ (0 °C) (K)	$\frac{u^0_{2z}}{k\alpha_J}$ (0 °C) (K)
Y–2-at.% Dy	2.97	34	0.557	−87.7
Y–25-at. % Dy	3.22	56	0.60	−95
Y–50-at. % Dy	3.46	56	0.65	−102.2
Dy	3.71[a] 3.47[b]	47[c]	0.695[a] 0.652[b]	−109.5[a] −102.7[b]
Y–2-at. % Ho	1.00	59	0.164	−73.7
Ho	1.10[a] 0.97[b]	36[c]	0.181[a] 0.16[b]	−81.5[a] −72[b]

[a] From R. Aléonard *et al.* (Ref. 15).
[b] From R. Aléonard (Ref. 16).
[c] From P. Boutron (Ref. 8).

per[8] we gave a theoretical expression for n_{zz} and n_{xx} from Specht's Hamiltonian.[13] We ignore their dependence on a and c, but since the exchange anisotropy seems small, we thought it would be interesting to compare the thermal variation of $1/\chi_z - 1/\chi_x$ with that of $(c/a-\sqrt{\frac{8}{3}})/a^3$.

The thermal variation of a and c of Dy and Y has been measured above 20 °C by Spedding *et al.*[12] The equation of the tangent to the curve $(c/a-\sqrt{\frac{8}{3}})/a^3$ vs T at a given temperature t_0 in °C can be written

$$(c/a-\sqrt{\tfrac{8}{3}})/a^3 = A_0(t_0)\,[1-b_0(t_0)\times 10^{-5}\,t], \qquad (3)$$

where t is the temperature in °C.

For yttrium, $(c/a-\sqrt{\frac{8}{3}})/a^3$ varies almost linearly with t from 0–400 °C, and $b_0(0\,°\mathrm{C}) = 39$. Above 400 °C, the slope decreases slightly. For dysprosium, the slope decreases from 0 to 400 °C, and then remains constant; $b_0(0\,°\mathrm{C}) = 29$ and $b_0(500\,°\mathrm{C}) = 45$. According to Darnell and Moore,[14] $b_0(0\,°\mathrm{C}) = 36$.

The thermal variation of a and c for our alloys is unknown. In a first approximation we can assume that the slope varies linearly with the concentration. But we can think that for Dy the slope under 400 °C is modified by the appearance of magnetism. The ordering point is much lower for our alloys. We can consider that for the alloys with Dy as well as the diluted alloy with Ho, $b_0(0\,°\mathrm{C}) \simeq 40$, which is intermediate between the values for Y at 0 °C and Dy at 500 °C.

The thermal variation of $1/\chi_z - 1/\chi_x$ becomes linear above the same temperature as $1/\chi_z$ and $1/\chi_x$ for all the alloys. The slope of the curve can be determined with a relatively good accuracy for the alloys with Dy; it is less good for the alloy with Ho. We can write

$$1/\chi_z - 1/\chi_x = A(1-b\times 10^{-5}t)\ . \qquad (4)$$

The values of b are given in Table II. They are relatively close to the value of $b_0(0\,°\mathrm{C})$. The thermal variation of a and c is not known accurately.

The difference $1/\chi_z - 1/\chi_x$ and therefore u^0_{2z} vary with the temperature as $(c/a-\sqrt{\frac{8}{3}})/a^3$ within the experimental error, as they do for the pure rare earths.[8,15,16]

C. Variation of exchange interactions with concentration

If we neglect the exchange anisotropy, we can define the quantity $n = n_{xx} = n_{zz}$; n is the sum of the exchange interactions in the alloy. It can be calculated at a given temperature T from $(1/\chi_z + 2/\chi_x)$ vs T. But since the constant term is small compared to T in the paramagnetic region, a small error on C will give a large error on $\mathcal{C}n$. Then, unlike u^0_{2z}, n is better defined by the Curie temperatures,

TABLE III. Anisotropy of various alloys (Touborg and Hǿg).

Alloys	$\frac{u^0_{2z}}{k\alpha_J}$ (LT) (K)	$\frac{u^0_{2zm}}{k\alpha_J}$ (K)	T_m (K)
Y–0.166-at. % Tb	−94[b]	−130	90
Y–1.23-at. % Tb		−130	90
Tb[a]		−122[e] −120[f]	273
Y–0.14-at. % Dy	−50.5[b]	−115	100
Y–0.898-at. % Dy	−53[b]	−129	100
Y–3.60-at. % Dy		−122	100
Dy[a]		−109[e] −103[f]	273
Y–0.14-at. % Er	−122[b]	−110	100
Y–0.309-at. % Er	−117[b]	−109.5	90
Y–0.993-at. % Er		−104	90
Y–2-at. % Er	−134[c]	(neutrons)	
Er[a]		−129[e] −110[f]	273
Lu[a]–0.634-at. % Tb	−77[d]	−110	90
Lu[a]–0.481-at. % Dy	−27[d]	−103	90
Lu[a]–0.978-at. % Ho	−44[d]	−62.5	90
Ho[a]		−76.8[e] −68.1[f]	273
Lu[a]–0.538-at. % Er	−77[d]	−83	90

[a] These data correspond to a and c of Y (or Dy) at 0 °C.
[b] From J. Hǿg *et al.* (Ref. 1).
[c] From O. Rathmann *et al.* (Ref. 17).
[d] From P. Touborg *et al.* (Ref. 4).
[e] From R. Aléonard *et al.* (Ref. 15).
[f] From R. Aléonard (Ref. 16).

$$\mathcal{C}n \propto \tfrac{1}{3}(\Theta_{pz}+2\Theta_{px}) . \qquad (5)$$

The average paramagnetic Curie temperature is given in Table I. It is proportional to the rare-earth concentration within a good approximation, in agreement with the theory. It is also in agreement with the variation of the exchange anisotropy, if the increase of $1/\chi_z - 1/\chi_x$ with $\mathcal{C}$ is due to that anisotropy.

IV. COMPARISON WITH ANISOTROPY OF OTHER ALLOYS OF RARE EARTHS WITH Y, Lu, AND Sc

Touborg and Høg[1-4] have measured the initial susceptibilities of diluted alloys of Tb, Dy, Ho, and Er with yttrium, lutetium and scandium between a few degrees Kelvin and 100 K. They have calculated the four crystalline-potential parameters of these rare earths by the Van Vleck method. The values of $u_{2z}^0/k\alpha_J$ [low temperature (LT)] they obtained are listed in Table III (their quantity B_2^0 is equal to u_{2z}^0/k). We have also calculated at a temperature T_m close to their maximum temperature of measurement a quantity u_{2zm}^0 also listed in Table III; u_{2zm}^0 is defined from $(1/\chi_z)_{T_m} - (1/\chi_x)_{T_m}$ by Eq. (1), as u_{2z}^0 should be, when the exchange anisotropy and the complementary term $O(1/T)$ are neglected. Since at that temperature this last term becomes important, u_{2zm}^0 is only a very rough approximation of u_{2z}^0.

Theoretically, in the point-charge model $u_{2z}^0/k\alpha_J$ should be the same for all of the rare earths, and should not vary when they are diluted in Y or Lu, if u_{2z}^0 is expressed for given values of a and c as is done in Table III [the experimental value is replaced by another one proportional to $(c/a - \sqrt{\frac{8}{3}})/a^3$].

In practice we see from Table III that the values of $u_{2z}^0/k\alpha_J$ obtained by Touborg and Høg vary greatly from one rare earth to another. Instead, the relative variations of $u_{2zm}^0/k\alpha_J$ are relatively small, except for Ho in Lu. We may then expect that if u_{2zm}^0 could have been evaluated at higher temperatures, the variation should have been even less.

For Dy, the value of $(1/\chi_z - 1/\chi_x)$ (100 K) measured by Touborg and Høg is almost the same as that measured by Aléonard on our Dy-Y alloy. It can be expected that for the other rare earths, the true value of u_{2z}^0, obtained at high temperature, is smaller than u_{2zm}^0, as for Dy, and then closer to the value obtained by Touborg and Høg except for Er in Y.

Then it seems that the variation of u_{2z}^0 could be closely approximated by the point-charge model. The values of u_{2z}^0 obtained at low temperature seem correct, since for Er the neutron diffraction gives almost the same value of u_{2z}^0.[17] But u_{2z}^0 does not have the same value at low as at high temperature, as is the case for pure rare earths, probably because the different polarization of the conduction band changes its value and because its experimental value includes magnetostriction contributions.

The parameter u_{2z}^0 also includes a contribution of the exchange anisotropy which is of second order of one ion and of zeroth order of the other one. This contribution is a part of the rare-earth–conduction-electron exchange. It does not vary with the concentration, and it varies from one rare earth to another as the one-ion anisotropy. Thus it cannot be distinguished from this by paramagnetic measurements. It can be assimilated to a part of the screening potential as well as the other contributions.[6]

V. COMPARISON WITH ANISOTROPY OF RARE-EARTH–GADOLINIUM ALLOYS

Levitin *et al.*[18] have measured the paramagnetic susceptibilities of Gd-Dy–alloy single crystals; Kadomatsu *et al.*[19] those of Gd-Er alloys, Fujii[20] those of Gd-Ho and Gd-Dy alloys, and Tajima[21] that of one Gd-Tb alloy. These alloys contain two different magnetic ions; thus their paramagnetic susceptibilities must be expressed theoretically as those of a ferrimagnetic crystal. In a ferrimagnet the asymptotic Curie point Θ_a and the paramagnetic or ordering Curie point Θ_p have very different theoretical expressions, even in the molecular-field approximation.[22]

Levitin *et al.*[18] give the theoretical expression of the anisotropy of Θ_p in the presence of one-ion and exchange anisotropies. They establish that $\Theta_{pm}(\Theta_{pz} - \Theta_{px})$, where $\Theta_{pm} = \frac{1}{3}(\Theta_{pz} + 2\Theta_{px})$, varies linearly with the concentration if there is only one-ion anisotropy. The experimental variation of $\Theta_{pm}(\Theta_{pz} - \Theta_{px})$ is almost linear, and the one-ion anisotropy is dominant, in agreement with the results on Y and Lu alloys.

Unfortunately, the deviation from linearity is more important for Gd-Ho alloys, and in Gd-Er alloys there is even a maximum of $\Theta_{pm}(\Theta_{pz} - \Theta_{px})$ near 75-at. % Er.

These discrepancies probably stem from the fact that the theoretical expression of the anisotropy of the susceptibility near the ordering point in the molecular-field approximation is very approximate. It would be much better to determine $\Theta_{az} - \Theta_{ax}$ from paramagnetic measurements at much higher temperatures.

Tajima[23] measured the anisotropy of heavy rare-earth alloys with Gd at very low temperature. If the exchange anisotropy is neglected, u_{2z}^0 is about 1.5 times larger in these alloys at low temperature than in pure rare earths in the paramagnetic region. One might think that this difference is due to the exchange anisotropy[8] since in dilute alloys one could estimate that the magnetostriction effects are very small. Now experimental data are also available on these alloys at high temperatures and on the alloys in Y and Lu. From the new data it appears that this factor 1.5 is more probably due to a variation of u_{2z}^0 in these alloys as in the pure rare earths from low temperatures to the paramagnetic region, for a reason yet unknown.

VI. CONCLUSION

We have measured the paramagnetic susceptibilities of four alloys of rare earths with Y up to 360 K. The quantity $1/\chi_z - 1/\chi_x$ increases slowly with the concentration of Y in Dy at a given temperature; the bilinear exchange anisotropy seems small compared with the second-order one-ion anisotropy. The parameter u_{2z}^0 varies experimentally as $(c/a-\sqrt{\frac{8}{3}})/a^3$, in agreement with the theory, and varies from Dy to Ho as in the point-charge model, within a good approximation. A rough estimate of u_{2z}^0 in the paramagnetic region at about 100 K, the highest temperature where measurements have been done on other alloys, confirms that the bilinear exchange anisotropy is small and that the variation of u_{2z}^0 is close to that in the point-charge model, which is not so bad at room temperature. The situation is different at very low temperatures, where u_{2z}^0 does not have the same value in dilute alloys as in pure rare earths, than at high temperatures.

It would be interesting to confirm the validity of the screened point-charge model in the range of temperatures where the magnetic interactions are small. For this, it should be necessary to know the paramagnetic susceptibilities of many alloys of all of the heavy rare earths at various concentrations with Y, Lu, and Gd, at room temperature and above, where the other terms of the thermal expansions of $1/\chi_z$ and $1/\chi_x$ are small compared to the first two. If very accurate measurements were available, we could perhaps at last determine the value of the small contribution of the bilinear exchange anisotropy.

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