Crystal field and exchange in the cubic HoAl₂ compound

B. Barbara, J. X Boucherle, M.F. Rossignol, J. Schweizer

► To cite this version:

B. Barbara, J. X Boucherle, M.F. Rossignol, J. Schweizer. Crystal field and exchange in the cubic HoAl₂ compound. *Physica B+C*, 1977, 86-88, pp.83 - 84. 10.1016/0378-4363(77)90236-4 . hal-01660389

HAL Id: hal-01660389

<https://hal.science/hal-01660389>

Submitted on 10 Dec 2017

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

CRYSTAL FIELD AND EXCHANGE IN THE CUBIC HoAl_2 COMPOUND

B. BARBARA*, J.X. BOUCHERLE†, M.F. ROSSIGNOL* and J. SCHWEIZER‡

Magnetization and polarized neutron diffraction measurements performed on a single crystal of HoAl_2 reveal the existence of a first order reorientation of the magnetic moments at 20 K. These results are interpreted in terms of a cubic crystal field and exchange interactions (molecular field). It is concluded that biquadratic exchange is non-negligible.

The compound HoAl_2 crystallizes in the cubic Laves phases and orders ferromagnetically at 35 K. In relation to the other RAI_2 compounds its magnetic behaviour is unusual: the magnetization axis is $[110]$ at low temperature and becomes $[100]$ at 20 K, up to the Curie temperature. This change of easy axis of magnetization, which gives rise to a specific heat anomaly at 20 K [1], was first shown by magnetization [2] and polarized neutron diffraction [3] experiments on a single crystal.

The present communication concerns the interpretation of our magnetization measurements performed on a single crystal of HoAl_2 at the Service National des Champs Intenses of Grenoble. Careful X-ray measurements having shown that any possible distortion is less than 10^{-4} [4], we consider that HoAl_2 remains cubic in the ordered state and then the crystal field acting on the Ho^{3+} ions is taken with a cubic symmetry. The exchange interactions, considered in the limits of the molecular field approximation, are taken as isotropic in a first approach. The effective hamiltonian for a fourfold quantization axis is

$$\mathcal{H}_1 = B_4^0(O_4^0 + 5O_4^4) + B_6^0(O_6^0 - 21O_6^4) - g_J\mu_B\mathbf{J}(\mathbf{H} + \mathbf{H}_m), \quad (1)$$

where B_4^0 and B_6^0 are the crystal field parameters, O_l^m the Stevens' operators and $\mathbf{J}$ the angular momentum operator. $\mathbf{H}$ and $\mathbf{H}_m$ are the applied and molecular fields, respectively. The latter satisfies the consistency condition $\mathbf{H}_m = \lambda\langle\mathbf{J}\rangle$, where $\lambda = 3k\theta_p/g_J\mu_B J(J+1)$ and θ_p is the paramagnetic Curie temperature. Using the two-dimensional mean field method [5], the diagonalization of this hamiltonian for given values of B_4^0

and B_6^0 permits in particular the determination of the holmium moment $\mu(\mathbf{H}, T)$ for any magnetic field $\mathbf{H}$ and temperature T , and that of the free energy $F_{(T)}^{[hkl]}$ at each temperature and with μ parallel to one of the principal crystallographic directions $[hkl]$ of the cube. The crystal field parameters B_4^0 and B_6^0 were determined by using two criteria: (i) the comparison between the measured and calculated magnetization curves, as for the other RAI_2 compounds, and (ii) the equality of the calculated free energy $F_{(T)}^{[110]}$ and $F_{(T)}^{[100]}$ at 20 K, a specific condition for HoAl_2 , imposed by the existence of the change of easy axis of magnetization. The values obtained $B_4^0 = -0.68 \times 10^{-4}$ meV, $B_6^0 = 0.63 \times 10^{-6}$ meV ($W = 0.15$ K, $x = -0.32$ in the notations of Lea et al. [6]) and $\theta_p = 32$ K give an excellent fit of the magnetization curves (fig. 1) and verify the condition $\Delta F(T) = F_{(T)}^{[100]} - F_{(T)}^{[110]}$ zero at 20 K (fig.

Fig. 1. Measured ($\square\Delta\circ$) and calculated (—) magnetization curves in HoAl_2 .

* Laboratoire de Magnétisme, C.N.R.S., 166X, 38042-Grenoble-Cedex, France

† D.R.F.-D.N., Centre d'Etudes Nucléaires 85X, 38041-Grenoble-Cedex, France

‡ Institut Laue-Langevin, 156X, 38042-Grenoble-Cedex, France

2). However, this model does not give a satisfactory representation of the measured magnetic anisotropy between the directions [100] and [110]. This quantity can be determined by evaluating the area enclosed by the magnetization curve obtained along the hard magnetization axis and the line of saturation of the same magnetization curve extrapolated to zero internal field [7]. At each temperature this anisotropy can be compared with the calculated free energy difference $\Delta F(T)$; there is no agreement at low temperature (fig. 2). In fact the variation of the calculated ratio $\Delta F(T)/\Delta F(0)$ depends very few of the values of W and x , showing that the disagreement is not due to any inaccuracy in the values of the crystal field parameters. The hamiltonian, eq. (1), sufficient for the interpretation of the magnetization is not satisfactory as an explanation of the thermal variation of the anisotropy of the energy.

As the RAl_2 compounds have small anisotropic interactions [8] and the rare earth intermetallic compounds usually present a biquadratic exchange in addition to the isotropic bilinear exchange [9], we have in a second approach included in eq. (1) a biquadratic term in the molecular field approximation:

$$\mathcal{H}_2 = \mathcal{H}_1 + \mathcal{H}_{(2)}^{\text{ex}},$$

with $\mathcal{H}_{(2)}^{\text{ex}} = -\mathcal{K}\langle O_2^0 \rangle O_2^0$; O_2^0 is the second order Stevens' operator and $\mathcal{K}$ a parameter having the dimension of an energy analogous to the $\mathcal{T} = \lambda g_J \mu_B$ of the bilinear exchange ($\sum \mathcal{T}_{ij} \mathbf{J}_i \mathbf{J}_j$). Diagonalizing $\mathcal{H}_2$ within the $J = 8$ manifold of Ho^{3+} , as was done for $\mathcal{H}_1$, one gets by a self-consistent method the magnetization μ and the free energy F . We observe that neither the

Fig. 2. Thermal variation of the measured anisotropy of the energy. Calculated curves of the free energy difference $\Delta F(T)$ with or without ($\mu = 0$) biquadratic exchange.

magnetization nor the temperature for which $\Delta F(T) = 0$ are very sensitive to small biquadratic exchange. On the contrary, the values of $\Delta F(T)$ for $T \neq 20$ K are very sensitive to small values of the parameter. The best fit for the thermal variations of the anisotropy of the energy is obtained for the exchange parameters $\mathcal{T} = 1.5$ K (giving $\theta_p = 36$ K, in perfect agreement with the measured value) and $\mathcal{K} = -1.01$ mK. This last value can be compared with the value of 0.6 mK obtained by Lüthi [10] for HoSb .

In conclusion:

(1) In contrast to the magnetization, the anisotropy of the energy is very sensitive to small biquadratic exchange.

(2) The thermal variation of the anisotropy of the energy can be described by $\mathcal{H}_2$ but not by $\mathcal{H}_1$. Nevertheless both hamiltonians give a change of easy axis of magnetization from [110] to [100] when the temperature increases.

(3) The bilinear isotropic exchange parameter $\mathcal{K}$, which is related to the paramagnetic Curie temperature, corresponds to 32 K in the first approach and to 36 K in the second one; the experimental value is 36 K.

(4) Assuming that the rotation of the easy axis is a first order transition, the thermal variation of the calculated entropy exhibits at 20 K a discontinuity which corresponds to a latent heat of 6.8 J/mol from $\mathcal{H}_1$ and of 2.8 J/mol from $\mathcal{H}_2$. These values must be compared with the latent heat evaluated from the measurements of Hill et al. [1]: 2.3 J/mol.

References

- [1] T.W. Hill, W.E. Wallace, R.S. Craig and T. Inoue, J. Solid State Chem. 8 (1973) 364.
- [2] B. Barbara, J.X. Boucherle and M.F. Rossignol, Phys. Lett. 55A (1975) 321.
- [3] B. Barbara, J.X. Boucherle, M.F. Rossignol and J. Schweizer, Neutron Scattering Conf., Gattingburg, Te. (June 1976) to appear.
- [4] B. Barbara, D.K. Ray, M.F. Rossignol and F. Sayetat, to be published.
- [5] P. Bak, Risø, Report no. 312 (1974). H.G. Purwins, E. Walker, B. Barbara, M.F. Rossignol and P. Bak, J. Phys. C: Solid State phys. 7 (1974) 3573.
- [6] K.R. Lea, M.M.J. Leask and W.P. Wolf, J. Phys. Chem. Solids 23 (1962) 1381.
- [7] B. Barbara, M.F. Rossignol, H.G. Purwins and E. Walker, Phys. Stat. Sol. 22 (1974) 553.
- [8] N. Kaplan, E. Dorman, K.H.J. Buschow and D. Ledenbaum, Phys. Rev. B7 (1973) 40.
- [9] H.H. Teitelbaum and P.M. Levy, to be published.
- [10] B. Lüthi, to be published.