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Antiferromagnetic Resonance and Anisotropic Exchange Interactions in $CoCl_2 6H_2O$

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Antiferromagnetic resonance experiment was performed on a single crystal of $CoCl_2 6H_2O$ in the frequency range of 9.5 to 47 kMc/sec at liquid helium temperature. The result shows certain systematic deviations from the Nagamiya-Yosida theory and these deviations can be removed by introducing a large anisotropic exchange interaction of the symmetric tensor form in place of the ordinary small anisotropy energy. The effective fields acting on sublattice magnetizations, M^+ and M^-, are assumed to be of the form: $H_E^{\pm} = -AM^{\mp} - \Gamma M^{\pm} - A'M^{\mp} - \Gamma' M^{\pm}$, where A' and Γ' are newly introduced anisotropic molecular field tensors. It was found that the symmetry axes of A' and Γ' coincide with those of the g-tensor and their components are given by $A_x' = 0.25$A, $A_y' = 0.20$A, $A_z' = -0.45$A, $\Gamma_x' = 0.19$A, $\Gamma_y' = 0.18$A and $\Gamma_z' = -0.37$A, where x, y and z correspond to the c-, b- and a'-axes, respectively.

Antiferromagnetic resonance experiment was performed on a single crystal of $CoCl_2 6H_2O$ in the frequency range of 9.5 to 47 kMc/sec at liquid helium temperatures[1]. The antiferromagnetic behavior of this salt was first discovered by Haseda and Kanda[2] from the measurement of the powder susceptibility. The x-ray analysis of the crystal structure[3], the specific heat measurement[4], the single crystal susceptibility data[5] and the proton resonance experiments[6] were reported recently. From these experimental results, it has become clear that this salt is antiferromagnetic below 2.29°K, and the spin orientation at low temperatures was determined to be parallel to the c-axis. Unfortunately, however, no neutron diffraction experiment has been reported so that we do

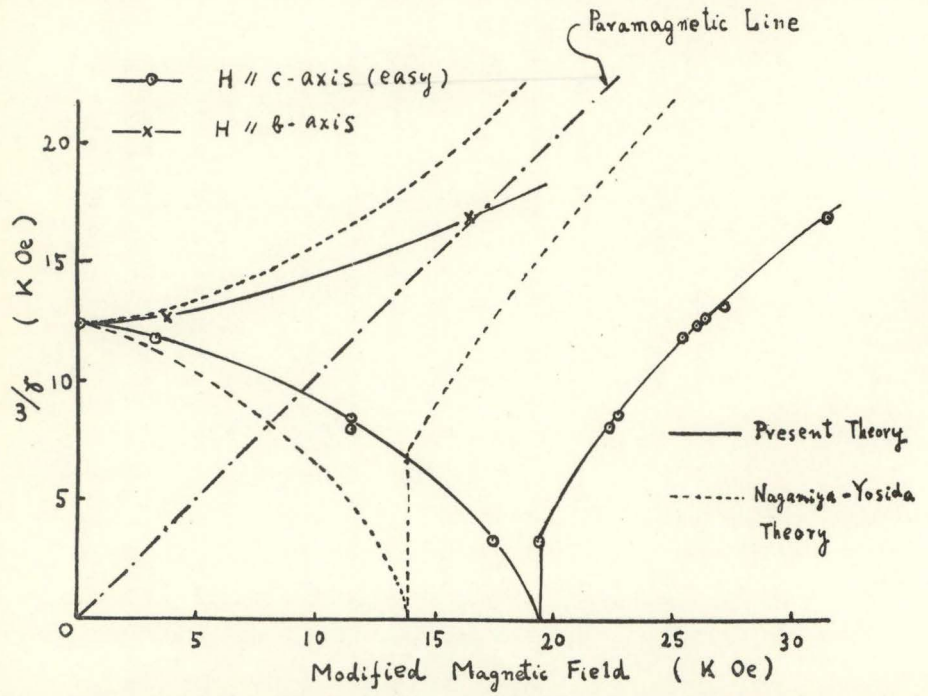

Fig. 1. Frequency-field diagram of the resonance points at 1.5°K. Dotted lines are drawn after Nagamiya-Yosida theory and full lines are drawn after the present theory.

not know the super-structure of the spins. Paramagnetic resonance was performed by the present author[1] and the principal g-values determined by the experiment are $g_a'=2.9$, $g_b=5.0$ and $g_c=5.0$ which are consistent with the susceptibility data. Thus, in the antiferromagnetic resonance in $CoCl_2$-$6H_2O$, the contribution of the orbital angular momentum should play a role as is anticipated from the large deviation of the g-values from the free spin value.

The result of the antiferromagnetic resonance can be understood by the theory developed by Nagamiya and Yosida[7] and also by Ubbink *et al.*[7], except for certain systematic deviations from the theory. These deviations can be removed by introducing a large anisotropic exchange interaction of the symmetric tensor form in place of the ordinary small anisotropy energy. The equation of motion of the two sublattice magnetizations are

$$\frac{1}{\gamma}\frac{d}{dt}M^{\pm}=M^{\pm}\times(H+H_E^{\pm})\,, \qquad (1)$$

where γ is the gyromagnetic ratio, M is the magnetization of the $+$ and $-$ sublattices, H and H_E represent the external magnetic field and the exchange field, respectively. The exchange field acting on the sublattice magnetizations, M^+ and M^-, are assumed to be of the form

$$H_E^{\pm}=-AM^{\mp} -\Gamma M^{\pm}-A'M^{\mp}-\Gamma' M^{\pm}\,, \qquad (2)$$

where A and Γ are the isotropic molecular field constants and A' and Γ' are newly introduced anisotropic molecular field tensors given by

$$\left.\begin{aligned} A'&=\begin{pmatrix} A_x' & 0 & 0 \\ 0 & A_y' & 0 \\ 0 & 0 & A_z' \end{pmatrix}, \\ \Gamma'&=\begin{pmatrix} \Gamma_x' & 0 & 0 \\ 0 & \Gamma_y' & 0 \\ 0 & 0 & \Gamma_z' \end{pmatrix}, \end{aligned}\right\} \qquad (3)$$

where x, y and z correspond to the crystallographic c-, b- and a'-axes, respectively and the diagonal sum of each tensor is assumed to be zero. It may be noted that the anisotropy coefficients K_1 and K_2 used in the Nagamiya-Yosida theory correspond to the 4-th term in Eq. (2), but that the third term expressed by $A'M$ is newly introduced in the present treatment. The experimental result shows that the symmetry axes of A' and Γ' coincide with those of the g-tensor and their components are given by

$$\left.\begin{aligned} &A_x'=0.25A, \quad A_y'=0.20A, \quad A_z'=-0.45A, \\ &\Gamma_x'=0.19A, \quad \Gamma_y'=0.18A, \quad \Gamma_z'=-0.37A, \end{aligned}\right\} \qquad (4)$$

The results calculated with these values are represented by full lines in Fig. 1, which are in satisfactory agreement with the experimental points.

Now let us consider the origin of the large anisotropic exchange constants A' and Γ'. As is seen in Eq. (4), these tensors have the same symmetry with that of the g-tensor. Accordingly such anisotropic exchange interaction may come mainly from the isotropic exchange interactions, which become anisotropic in the effective spin hamiltonian which is obtained when the true spin is replaced by the effective spin of 1/2. In $CuCl_2 2H_2O$, in which the anisotropy of the g-tensor is not so large as in $CoCl_2 6H_2O$, these anisotropic parts of the exchange interactions are small compared with the isotropic parts, so that the old theory was satisfactorily applicable.

The experiment was also performed in $CoBr_2 6H_2O$ and the result was similar with that of $CoCl_2 6H_2O$.

References

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DISCUSSION

C. J. Gorter: If I remember well we have in Leiden also used similar formulae for anisotropic molecular fields. It might be useful to compare the different sets of formulae.