



磁氣的に飽和した常磁性体におけるコヒーレントな スピン運動

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Coherent Spin Motion in a Magnetically Saturated Paramagnet

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高柳 滋・伊達 宗行* : 磁氣的に飽和した常磁性体における
コヒーレントなスピン運動

In this short note we report clear evidence of coherent spin motions in a saturated paramagnetic system under a strong magnetic field. This was revealed because in electron spin resonance the spin system sees a "ferromagnetic" anisotropy instead of paramagnetic one. A single crystal of clathrate compound $\text{Mn}(\text{NH}_3)_2\text{Ni}(\text{CN})_4 \cdot 2\text{C}_6\text{H}_6$ that had been grown at the Watanabe Laboratory of Hokkaido University was used in our experiment. This crystal is isomorphous with $\text{Ni}(\text{NH}_3)_2\text{Ni}(\text{CN})_4 \cdot 2\text{C}_6\text{H}_6$ ¹⁾, having a base centered spin arrangement in the c-plane with a tetragonal axis along the c-direction and shows Mn spin paramagnetism with an antiferromagnetic Curie-Weiss constant Θ of -0.8°K .²⁾ Ni ions in this compound are believed to be diamagnetic. An ESR study was carried out using microwaves of 35 GHz region at liquid helium temperatures. Since the induced paramagnetic moment M due to an external magnetic field H_0 was fairly large (about 80 % magnetization at 1.5°K), the demagnetization effect³⁾ could not be neglected. A thin disk-shaped specimen with a wide c-plane was used with H_0 in the ac-plane. Examples of the experimental results are shown in Fig. 1. If the angular dependences were due to the dipolar effect, i. e. the demagnetization effect only, the resonance should follow the dotted lines which are normalized at the a-axis, and the ratio $|\Delta H_c/\Delta H_a|$ should be 2 where ΔH_c and ΔH_a represent shifts from free spin resonance point at $H//c$ and $H//a$, respectively. As can be seen in Fig. 1, however, the experimental results do not coincide with the dotted lines.

To explain such deviations, a crystalline anisotropy DS_z^2 must be taken into account. Usually, fine and hyperfine structures are masked by exchange narrowing and no shift is expected at high temperatures. Under paramagnetic saturation, however, a shift due to DS_z^2 occurs because almost all spins are in a $|-5/2\rangle$ state and a transition $|-5/2\rangle \rightarrow |-3/2\rangle$ dominates. It has to be emphasized that the angular dependence due to DS_z^2 is proportional to $(3\cos^2\theta - 1)$ ⁴⁾ so that the ratio $|\Delta H_c/\Delta H_a|$ remains at 2 even if the DS_z^2 term were to be reflected.

Experimentally, the ratios at 4.2 and 1.5°K were 1.6 and 1.2, respectively.

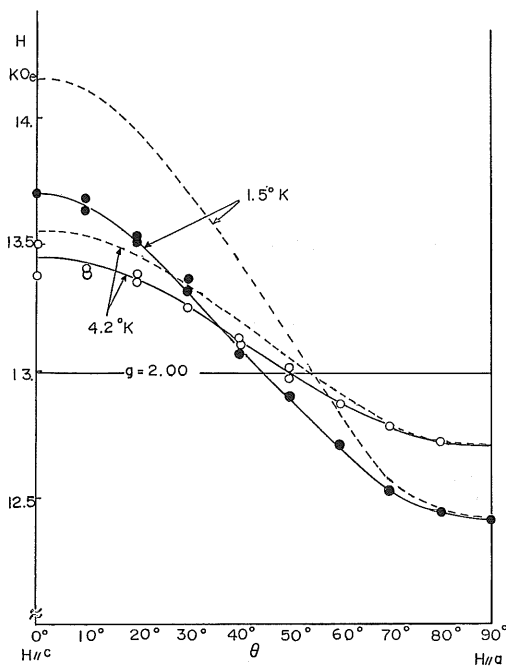


Fig. 1. Angular and temperature dependences of the resonance point at 36.4 GHz. Dotted lines show theoretical curves due to dipolar interaction only.

Next, let us consider the DS_z^2 term in a ferromagnetic sense. Introducing an effective anisotropy energy of $K\gamma^2$, $K < 0$, where γ is a direction cosine between H_0 and the c -axis, the anisotropy field is calculated as $-2K/M$ and 0 for $H_0//c$ and $H_0//a$, respectively. As can easily be seen, the ratio $\Delta H_c/\Delta H_a$ can be modified by introducing K . An uniaxial anisotropy constant K extrapolated to 0°K was found to be 3.2×10^4 erg/cc; this corresponds to D of -1.4×10^{-2} cm $^{-1}$, using a formula $K = DNS(S - 1/2)$, N being the spin number/cc. The D value obtained is very natural compared with those of the usual Mn compounds.

The anisotropy of $\text{Mn}(\text{NH}_3)_2 \cdot \text{Ni}(\text{CN})_4 \cdot 2\text{C}_6\text{H}_6$ under paramagnetic saturation can therefore be interpreted by a "ferromagnetic" or a collective model rather than a paramagnetic one. This means that many spins coherently precess near their equilibrium positions although the life time and range of the coherent motions are not yet clear.

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