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THEORY OF ANTIFERROMAGNETIC RESONANCE IN HYPERFINE-ENHANCED NUCLEAR MAGNETS

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Abstract. - The antiferromagnetic resonance in hyperfine-enhanced nuclear magnets is investigated by setting up the coupled equations of motion of the nuclear and the induced electronic magnetic moments. There are electron-like and nuclear-like resonances and the latter interprets the observed feature of the antiferromagnetic resonance in Cs2HoNaC16.

1. Introduction

The hyperfine-enhanced nuclear magnets have unusual magnetic properties and attract wide attention [1]. The electronic system in such magnets have the nonmagnetic ground state (singlet $\Gamma_1$ or nonmagnetic doublet $\Gamma_3$) and it is expected to have large hyperfine (hf) coupling constant. Crystals containing the rare earth ions with even numbers of 4f-electrons, like Pr$^{3+}$, Ho$^{3+}$, or Tm$^{3+}$, are candidates.

The Ho ion in Cs$_2$HoNaC16 has the nonmagnetic doublet $\Gamma_3$ as the ground state. This becomes the antiferromagnet below $T_N = 4.8$ mK. The antiferromagnetic resonance has been observed in this compound [2] and the resonance line centers at about 250 MHz in zero field, and this may manifest the hf enhanced nuclear magnet.

Bleaney [3] made a theory of resonance for such systems by considering that the nuclear and the induced electronic moments in a rare earth ion couple rigidly to show an effective magnetic moment. In the dynamical motion of the nuclear and the electronic magnetic moments, the picture of this rigid coupling may not be legitimate because the hf coupling is not extremely strong. Here, let us solve the coupled equations of motion of the nuclear and the induced electronic magnetic moments. Apparently, the number of solutions are doubled and we obtain the nuclear-like and the electron-like solutions and the solutions will be compared with the experiment of the antiferromagnetic resonance.

2. Formulations

The system of interest Cs$_2$HoNaC16 has the electronic interaction between rare earth ions which is mostly dipolar, and it is conjectured that there exists a small lattice distortion from cubic symmetry at low temperature [2, 4]. The Hamiltonian of the system is then given as

$$H = H^{(e)}_{\text{anis}} + \frac{1}{2} \sum_{m,n} J_{mn} \cdot K_{mn} \cdot J_n - \sum_n A_l n \cdot J_n$$

$$+ \frac{1}{2} \sum_{m,n} I_m \cdot K_{mn}^{(e)} \cdot J_n + \frac{1}{2} \sum_{m,n} I_m \cdot K_{mn}^{(h)} \cdot I_n$$

$$- \sum_n F I_n^2 - \sum_n H \cdot (\mu J_n + \mu_N I_n),$$

where $H^{(e)}_{\text{anis}}$ is the crystal-field for the 4f-electrons, the second, the fourth, and the fifth terms are dipolar interactions (K's are tensors), the third is the hf interaction, the sixth the quadrupole interaction of the nuclear spins, the last term the Zeeman energy, and $H$ is the applied static magnetic field. The dipolar interaction between nuclear spins ($K^{(N)}$ term) is much smaller than other dipolar interactions and will be neglected. At low temperature, the $J_n$ contributes as the electronic moment induced by the hyperfine interaction for the singlet ground state and the prescription to obtain this has been given previously [1]. From (1) the phenomenalistic equations of motion for the electronic and the nuclear magnetic moments are

$$\frac{1}{\gamma} \frac{d}{dt} M^\pm = M^\pm \times \left( H + H^{\pm}_e + H^{\pm}_h + H^{\pm}_N \right),$$

$$\frac{1}{\gamma_N} \frac{d}{dt} m^\pm = m^\pm \times \left( H + h^{\pm}_e + h^{\pm}_a + h^{\pm}_N \right),$$

where $M^\pm = g\mu_B \langle J^\pm \rangle$ and $m^\pm = g_N\mu_N \langle I^\pm \rangle$ are induced electronic and the nuclear magnetic moments of the two sublattices, $\gamma$ and $\gamma_N$ are the respective gyromagnetic ratios, and the four quantities in the brackets are the external, the exchange-type (dipolar), the anisotropy, and the hf fields, respectively. These fields may be expressed as follows.

$$H^{\pm}_e = -\Gamma \cdot M^\mp - \Delta \cdot M^\pm,$$

$$H^{\pm}_a = \varepsilon_s \frac{B}{M_0} M^\pm, \quad H^{\pm}_N = c m^\pm,$$

$$h^{\pm}_e = -\Gamma_N \cdot M^\pm - \Delta_N \cdot M^\pm,$$

$$h^{\pm}_a = \varepsilon_s \frac{b}{m_0} m^\pm, \quad h^{\pm}_N = c M^\pm,$$
where $\Gamma$ and $\Delta$ are tensors, $M_0$ and $m_0$ the respective magnetic moments at $T = 0$ K, and $\varepsilon_2$ the unit vector. The anisotropy field $H_a$ expresses the effects of the lattice distortion and of the spontaneous spin polarization with which the $I_3$ doublet splits, and $h_a$ comes from the nuclear quadrupole interaction. The anisotropy field coming from the dipolar interaction is buried in $H_a$ and $h_a$ terms.

The usual technique to solve (2) is to introduce the variables $M = M^+ + M^-$, $M' = M^+ - M^-$, $m = m^+ + m^-$, and $m' = m^+ - m^-$ and neglect the small terms of the order of $M^2$ and $m^2$. Thus, for the component of $x$ and $y$, the equations (2) written with the new variables constitute the eight simultaneous equations. If we approximate these by the first four and the last four equations neglecting the off-diagonal terms that connect between these two groups, we obtain the familiar solutions; all solutions split linearly with $H$ and the first type of solutions is similar to the expression of the usual antiferromagnetic resonance and the second one the solution that is essentially the nuclear spin resonance involving the large internal field of the hf interaction and the quadrupole interaction.

3. Numerical solutions and comparison with experiments

The eight simultaneous equations are solved numerically for reasonable sets of parameters and a typical set of solutions is shown in figure 1. Here all magnetic fields are reduced by $\gamma m_0 \approx 460 (I) \approx 1600$ gauss for $\langle I \rangle \approx 7/2$, and the parameters are set as $cM_0 = 500$, $M_0 (\Gamma - \Delta)_{zz} = 0.01$, $M_0 (\Gamma + \Delta)_{zz} = 0.05$, $B/M_0 = 0.4$, and $b/m_0 = 0.2$ in unit of $cm_0$ and $\gamma/\gamma_N = 1.9 \times 10^3$. The eigenvalues stand for $\omega/\gamma m_0$ and we set as $(\Gamma - \Delta)_{zz} \approx (\Gamma - \Delta)^{-1/2}$, because of symmetry. The electronic dipolar field has been calculated by Suzuki et al. by assuming $g_B (J) \approx 1M_B$ and is estimated as $H (dip) \approx 66$ gauss, which is close to the above value.

The observed antiferromagnetic resonance line is at $\nu_0 \approx 250$ MHz in zero field and split with $H$ to show $\nu = 280$ and 245 MHz ($\Delta \nu = 35$ MHz) under $H \approx 100$ gauss ($H/cm_0 \approx 0.0625$). These values can be expressed as $\omega_0/\gamma m_0 = 0.0894$ and $\Delta \nu/\nu_0 = 0.154$. If we take the lower nuclear-like solution in the present calculation, the corresponding values are 0.0695 and 0.215 for the case of the corresponding $H$ in fair agreement with experiment.

Thus, the present picture can give the feature of the observed antiferromagnetic resonance. The essential difference from the picture of the rigid coupling (cf. Sect. 1) arises in the existence of the high frequency resonance of the electron spin nature and the $H$-dependence of the antiferromagnetic resonance at high fields, although the theory must be refined to discuss this latter problem in detail.

Fig. 1. - A typical result of the resonance frequencies against the external magnetic field. The values of parameters are given in the text and all quantities are reduced by $cm_0 \approx 1600$ gauss. The solid circle marked on the crossing point of two curves indicates the complex solution of $\omega$.

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