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MAGNETOELECTRIC EFFECT OF Cr_2O_3

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A formula for the temperature dependent magnetoelectric susceptibility of Cr_2O_3 is obtained using the method of renormalized spin waves of Nagai and compared with the formula in random phase approximation. The three coefficients a^g, a^{LS}, and a^J which respectively represent the change in the g-factor, in the crystal field splitting and in the exchange integral due to the electric field parallel to the easy axis, are determined by comparing the theoretical with experimental susceptibility curves, as well as the antiferromagnetic resonance shift.

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1. Introduction

It is well known that the values of coefficients a^g, a^{LS}, a^J can be determined from the temperature dependence of the magnetoelectric susceptibility and from the electric shift of the antiferromagnetic resonance (AFRM). The necessary theoretical formulae depend on the exchange parameters which in case of Cr_2O_3 are well known from the inelastic neutron scattering experiments [1, 2]. Till now these parameters were not used in the calculations of the magnetoelectric effects (earlier papers considered them usually as fitting parameters). The main aim of the present paper is to determine a^g, $2a^{LS}$, a^J on the basis of the well known exchange parameters using the improved theoretical formulae.

2. Theory

The energy of spin waves and the magnetoelectric susceptibility of a two-sublattice antiferromagnet (Cr_2O_3) in the magnetic H^z and the electric field E^z parallel to the direction of spin orientation will be calculated on the basis of the following Hamiltonian [3, 4]:

$$H = \frac{1}{2}\sum_{i,j} K_{i,j}\boldsymbol{S}_i\boldsymbol{S}_j - g\mu_{\mathrm{B}}\sum_j H^z S_j^z + \frac{g^2\mu_{\mathrm{B}}}{J_d}a^g H^z E^z\sum_j \varepsilon(j)S_j^z - \frac{1}{2}I_{\mathrm{a}}\sum_j (S_j^z)^2$$
$$+g\mu_{\mathrm{B}}a^{LS}E^z\sum_j\varepsilon(j)(S_j^z)^2 + \frac{1}{2}g\mu_{\mathrm{B}}E^z\sum_{ij}[\varepsilon(i)+\varepsilon(j)]a^j(i,j)\boldsymbol{S}_i\boldsymbol{S}_j. \qquad (1)$$

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$K_{i,j}$ are the exchange parameters for Cr_2O_3 (we restrict to five interaction constants: K_1, K_2, K_3, K_4, K_5), $J_d = K_1+3K_2+3K_3+K_5 = 556.7\ \mathrm{cm}^{-1}$ [5] (Foner gives $J_d = 620\ \mathrm{cm}^{-1}$ [6]). I_a is the anisotropy energy coefficient, $\varepsilon(j) = \pm 1$ for the upper and lower sublattices, respectively. The Hamiltonian (1) can be diagonalized easily using the standard approximations and methods. The method of Nagai [7] was applied by Tripathi et al. [4] to obtain the energy of spin waves in the presence of the magnetic and electric fields. We modified their formulae taking into account the differences between the average $\langle a_k^+ a_k \rangle$ and $\langle b_k^+ b_k \rangle$ caused by the fields. The notation $\langle \rangle$ stands for the statistical average over grand canonical ensemble. We get

$$\varepsilon_{\boldsymbol{k}}^{\pm} = \pm \frac{\Omega_{\boldsymbol{k}}^{+}(T) - \Omega_{\boldsymbol{k}}^{-}(T)}{2} + \sqrt{\left(\frac{\Omega_{\boldsymbol{k}}^{+}(T) + \Omega_{\boldsymbol{k}}^{-}(T)}{2}\right)^2 - |B_{\boldsymbol{k}}(T)|^2}. \qquad (2)$$

The quantities $\Omega_{\boldsymbol{k}}^{+}(T)$, $\Omega_{\boldsymbol{k}}^{-}(T)$, $B(T)$ depend on the exchange parameters and on external fields. The electric shift in AFMR is obtained by setting $\boldsymbol{k} = 0$ in (2).

In order to obtain the formula for the magnetoelectric susceptibility two approximate methods were applied:

1. The spin wave theory renormalized according to Nagai gave

$$\chi_{\parallel}^{\mathrm{ME}}(T) = -N_0 \frac{g^2 \mu_{\mathrm{B}}^2}{J_d} a_{\parallel}^{g} \langle S_l^z \rangle_0 - \chi_{\parallel}^{\mathrm{M}}(T) \Bigg\{ 2a_{\parallel}^{LS} \langle S_l^z \rangle_0$$

$$+ S a_{\parallel}^{J} \left[1 + \frac{1}{2}(v_1 + v_2 - u_1 - u_2)\right] \frac{\frac{1}{N_0} \sum_{\boldsymbol{k}} [1 - \gamma_{\mathrm{s}}(\boldsymbol{k})] \langle \partial n_{\boldsymbol{k}} \rangle_0 / \partial \varepsilon_{\boldsymbol{k}}^{\mathrm{N}}(T, 0)}{\frac{1}{N_0} \sum_{\boldsymbol{k}} \partial \langle n_{\boldsymbol{k}} \rangle_0 / \partial \varepsilon_{\boldsymbol{k}}^{\mathrm{N}}(T, 0)} \Bigg\}. \qquad (3)$$

where $\langle S_l^z \rangle_0$ is the average value of spin operator S_l (spin on l sublattice) at zero fields, $S = 3/2$, the quantities u_1, u_2, v_1, v_2 characteristic of the Nagai theory, depend on the temperature (they are calculated at zero fields). $\varepsilon_{\boldsymbol{k}}^{\mathrm{N}}(T, 0)$ is the temperature dependent magnon energy at zero external fields (the formula (2) for $E^z = H^z = 0$), $\langle n_k \rangle_0 = 1/\left\{\exp\left[\varepsilon_{\boldsymbol{k}}^{\mathrm{N}}(T,0)/k_{\mathrm{B}}T\right] - 1\right\}$, the magnetic susceptibility $\chi_{\parallel}^{\mathrm{M}}(T)$ obtained by this method is given in [8], N_0 is the number of spins in the sublattice, $\gamma_{\mathrm{s}}(\boldsymbol{k}) = \cos(\boldsymbol{k}\boldsymbol{a}_1) + \cos(\boldsymbol{k}\boldsymbol{a}_2) + \cos(\boldsymbol{k}\boldsymbol{a}_3)/3$ ($\boldsymbol{a}_1$, $\boldsymbol{a}_2$, $\boldsymbol{a}_3$ are the primitive lattice vectors).

2. Random-phase approximation (RPA) calculation gives a similar formula as in (3), where now $\varepsilon_{\boldsymbol{k}}^{\mathrm{RPA}}(T,0)$, $\langle S_l^z \rangle_0^{\mathrm{RPA}}$, 0, 0, 0, 0 are inserted instead of $\varepsilon_{\boldsymbol{k}}^{\mathrm{N}}(T,0)$, S, u_1, u_2, v_1, v_2, respectively.

3. Results

The three dimensionless coefficients a^g, a^{LS}, a^J are evaluated by fitting the theoretical formula for the magnetoelectric susceptibility (3) to the experimental points [9] (see Fig. 1). The formula for $\chi_{\parallel}^{\mathrm{ME}}(T)$ based on the spin wave of Nagai cannot be used up to T_N ($T_\mathrm{N} = 308$ K for Cr_2O_3). The fitting performed up to 200 K gave

$$a^g = 0.0344, \quad a^{LS} = -0.240, \quad a^J = -2.07.$$

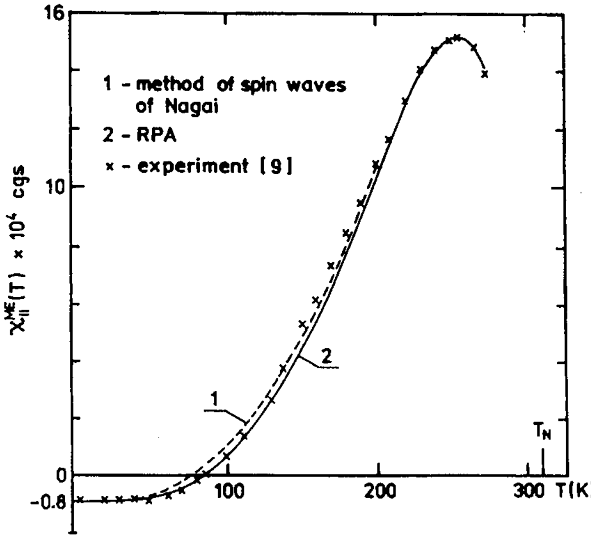

Fig. 1. Temperature dependence of the parallel magnetoelectric susceptibility for Cr_2O_3.

In the RPA method, due to the replacement of the constant $S = 3/2$ in (3) by the temperature dependent term $\langle S_l^z \rangle_0^{\mathrm{RPA}}$, one can fit $\chi_{\parallel}^{\mathrm{ME}}(T)$ to the experimental curve in the whole temperature range and the values of the coefficients are

$$a^g = 0.0344, \quad a^{LS} = -0.172, \quad a^J = -2.07.$$

From the AFMR shift [9] and the value of $\chi_{\parallel}^{\mathrm{ME}}(T)$ at 4.2 K one obtains two coefficients, namely

$$a^g = 0.0344, \qquad a^{LS} = 0.00358.$$

This great discrepancy in a^{LS} may be caused by the temperature dependence of the coefficients which were assumed here to be constant.

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