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The measurement of hyperfine field of ^{60}Co in $MnCl_2 \cdot 4H_2O$ crystal by the method of nuclear orientation

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Abstract

The hyperfine field of Co in metallic magnets such as Fe and Ni has been extensively studied; there have been far fewer experiments about its hyperfine field in insulating magnets. In an attempt to understand this problem, using low-temperature nuclear orientation method, the hyperfine field in antiferromagnetic insulating $MnCl_2 \cdot 4H_2O$ crystals, was investigated and determined. The hyperfine field from the anisotropy of the 1170 and 1330 keV lines of ^{60}Co was found to be $B_{hf} = 28.6 \pm 1^{+0.8}_{-0.3}$ T.

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1. Introduction

The method of low-temperature nuclear orientation (LTNO) relies on the interactions of the nuclear magnetic dipole and electric quadrupole moments with magnetic and electric field gradients, respectively. Unpaired electrons within unfilled atomic shells often give rise to strong magnetic and electric fields, commonly referred to as hyperfine fields. As temperature of the nuclear spin system reaches values in the (milli Kelvin) range, a significant polarization of the nuclear spins along the local hyperfine field is attained. The process of nuclear orientation (NO) can be utilized in practice as an experimental technique in ferro- or antiferromagnetically ordered single crystals. In antiferromagnets below the critical temperature, the electron spins in the crystal order alternately parallel and antiparallel along a particular direction. On cooling the crystal below its Neél temperature at about 100 mK, the nuclei will become aligned parallel to the direction of the spontaneous magnetization of the sublattices. Daniels and LeBlank [1] reported the first successful alignment of nuclei in an antiferromagnetic crystal.

In order to orient the nuclear magnetic moment of ^{60}Co and ^{54}Mn, we have used $MnCl_2 \cdot 4H_2O$ crystal which is a collinear antiferromagnet with $T_N = 1.6$ K. The crystal structure has been studied by X-ray diffraction [2]. At room temperature the structure is monoclinic ($\beta = 99.74^\circ$) with four magnetic ions per unit cell. The magnetic structure of the antiferromagnetic phase has been also studied by neutron diffraction [3].

The main goal of this paper is to determine the hyperfine field of ^{60}Co in $MnCl_2 \cdot 4H_2O$ crystal. In this experiment the isotopes of ^{60}Co and ^{54}Mn have been utilized, in which their decay schemes are well understood [4]. The decay of ^{54}Mn ($I = 3$) is specified by electron capture to the 835 keV excited state of ^{54}Cr and then by γ-decay (E2 transition) to the ground state. The magnetic moment of ^{54}Mn is 3.281 ± 0.013 nm [5] and its hyperfine field in $MnCl_2 \cdot 4H_2O$ crystal has been measured to be 60.05 T [6]. The ^{60}Co ($I = 5$ and $\mu = 3.799 \pm 0.008$ nm) decays by emission of beta particles to an excited state of ^{60}Ni. Two gamma rays (1170

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and 1332 keV) are promptly emitted as the ^{60}Ni nucleus decays to its ground state.

2. Theoretical considerations

Consider a system of magnetic ions with electron spins **S** that are ordered collinearly by exchange interaction. Assume that the nuclear spins are oriented by an isotropic magnetic hyperfine interaction, which can be described by the Hamiltonian

$$H = A\,\mathbf{I}\cdot\mathbf{S}, \tag{1}$$

where A is the strength of the hyperfine interaction, **I** is the spin of the nuclei and **S** is the electron spins that are polarized by exchange interaction. This interaction can be represented by an effective magnetic field [7]:

$$H = -\boldsymbol{\mu}\cdot\mathbf{B}_{\mathrm{hf}}, \tag{2}$$

where $\mathbf{B}_{\mathrm{hf}}$ is the hyperfine field and μ is the magnetic moment related to the nuclear spin. If some of the magnetic ions replaced with radioactive isotopes (in this case ^{60}Co and ^{54}Mn) these isotopes will be also oriented. The normalized intensity of gamma rays emitted from oriented radioactive nuclei at angle θ to the quantization axis is anisotropic and can be written as [8]

$$W(\theta) = \sum_{K=even} B_k A_k P_k(\cos\theta), \tag{3}$$

where A_k are parameters associated only with the radioactive decay, P_k are the ordinary Legendre polynomials and B_k are "orientation parameters" defined for axial symmetry as

$$B_k(B_{\mathrm{hf}},T) = \sqrt{(2I+1)(2k+1)} \times \sum_{m=-I}^{I}(-1)^{I+m}\begin{pmatrix} I & I & k \\ -m & m & 0\end{pmatrix} P_m. \tag{4}$$

In this equation the matrices are Wigner 3-j symbols and P_m is the normalized population of the substate with magnetic quantum number $m(-I \leqslant m \leqslant I)$.

In thermal equilibrium, at temperature T, the populations of the nuclear Zeeman levels are given by Boltzman distribution

$$p_m = \frac{\exp(-\Delta E m/K_{\mathrm{B}}T)}{\sum_m \exp(-\Delta E m/K_{\mathrm{B}}T)}, \tag{5}$$

where $\Delta E = \mu B_{\mathrm{hf}}/I$ is the separation in energy between the Zeeman magnetic sublevels. Eqs. (4) and (5) indicate that B_k depend on the temperature and the hyperfine field. Therefore, by measuring the temperature and the normalized intensity of the gamma rays in a particular direction and comparing it with Eq. (3), one can obtain the hyperfine field.

3. Experimental procedure

The $MnCl_2\cdot 4H_2O$ crystal was grown from saturated and mixed solutions of the salt that containing the radioactive isotopes of ^{60}Co and ^{54}Mn (a few (micro Curie)). The crystal was plate-like and the flat area containing the c and b crystalline directions. The dimensions along the c, b and a^* (normal to the b–c plane) directions were approximately 12, 10 and 3 mm, respectively. The crystal was fixed to a cold finger of the dilution refrigerator and cooled to temperatures below 100 mK. The easily identifiable c-axis was mounted vertically and a Ge(Li) detector was mounted in the downward vertical direction to count γ-rays emitted from the oriented radioactive nuclear spins. Since the easy axis of magnetization of the $MnCl_2\cdot 4H_2O$ crystal is not exactly along the c-axis and it has an angle of about 7° with the c-axis towards the a-axis [3], the W (7°) were measured instead of W (0°).

4. Results and conclusions

The data were collected by a Ge(Li) detector and a multichannel analyzer (MCA) over 1000 s lifetime intervals and then analyzed by a personal computer. For the 835 keV γ-rays emitted in the decay of ^{54}Mn the series of Eq. (3) terminates at $k = 4$ with $A_0 = 1$, $A_2 = -0.495$ and $A_4 = -0.447$. From these parameters and measured anisotropy $W(7^\circ) = 0.840 \pm 0.002$ and comparing it with Eq. (3), the base temperature of the crystal was found to be $T = 49.4 \pm 0.4^{+0.7}_{-1.6}$ mk. For both γ-rays ^{60}Co only the decay parameters of $A_o = 1$, $A_2 = -0.4206$ and $A_4 = -0.2428$ are nonzero [9]. Substituting these parameters and the base temperature in Eq. (3) and comparing the calculated anisotropy with measured one, $W(7^\circ) = 0.956 \pm 0.003$, which is the average anisotropy of the 1170 and 1330 keV lines, the hyperfine of ^{60}Co was found to be $B_{\mathrm{hf}} = 28.6 \pm 1^{+0.8}_{-0.3}$ T. The first error in T and B_{hf} is statistical and the second is systematic error. A possible misalignment of the crystal about $\pm 7^\circ$ was used to calculate the systematic error. The calculated value of hyperfine field with ab initio methods is 24 T, which the contributions of the core and valance electrons are 19.6 and 4.4 T, respectively [10]. The difference between the measured and calculated hyperfine field for ^{60}Co revealing the presence of large internal effective magnetic field acting on the nuclei due to the host crystal.

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