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ЛАБОРАТОРИЯ ТЕОРЕТИЧЕСКОЙ ФИЗИКИ

ON THE INVESTIGATION OF RELAXATION EFFECTS IN HEMATITE IN STRONG PULSED MAGNETIC FIELDS USING NEUTRON DIFFRACTION

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ON THE INVESTIGATION OF RELAXATION EFFECTS IN HEMATITE IN STRONG PULSED MAGNETIC FIELDS USING NEUTRON DIFFRACTION

I. In order to determine the properties of magnetic materials in very high magnetic fields, often pulsed fields are used. The behaviour of the spin system is handled quasistatically. However, in a more detailed description, we have to take into account dynamical processes which are originated in the sudden variation of the pulsed magnetic field.

In this paper we will calculate the relaxation time of the spin system of a Dzyaloshinsky antiferromagnet using the nonequilibrium statistical operator of Zubarev /1/. As an example we consider hematite for temperatures below the Morin temperature. In this case recently the relaxation of the staggered magnetization vector was observed at the JINR Dubna by neutron diffraction technique.

Many properties of hematite $(\alpha - Fe_2O_3)$ are well investigated /2/. The iron ions are situated along the trigonal axis (z-axis) of the rhomboedrical elementary cell containing two molecules Fe_2O_3. Below the Néel temperature $T_N \approx$ 960 K hematite is essentially antiferromagnetic. Let us denote the sublattice magnetization by $\vec{M}_1, \vec{M}_2$, the total magnetization by $\vec{M} = \vec{M}_1 + \vec{M}_2$ and the staggered magnetization by $\vec{N} = \vec{M}_1 - \vec{M}_2$. Because of the crystal symmetry a Dzyaloshinsky-Moriya antisymmetric exchange term is possible, which produces a weak ferromagnetism above the Morin temperature $T_M \approx$ 260K.

Below T_M we have a pure antiferromagnet, the spins are directed along the z-axis. In this region, if we apply a magnetic field higher than a critical field $H_c \approx$ 68 kG in z-direction, the vector $\vec{N}$ flips from the z-direction into the plane perpendicular to z. After switching off this field, $\vec{N}$ will relax to its equilibrium position.

If we consider these switching processes in a time-dependent magnetic field, the spin system can not follow immediately the variation of the magnetic field. We will obtain a nonequilibrium state, which goes over into the equilibrium state after the relaxation time.

To investigate the dynamics of this process we start with the Hamiltonian

$$H = \sum_{\langle i,j \rangle} J \vec{S}_i \vec{S}_j - \sum_{\langle i,j \rangle} \vec{D} \vec{S}_i \times \vec{S}_j - \sum_{i,j} K (S_i^{z\,2} + S_j^{z\,2}) - g \mu_B H_E \sum_{i,j} (S_i^z + S_j^z) . \quad (1)$$

The external magnetic field H_E is directed parallel to the z-axis. The N/2 spins of the sublattices 1,2 are denoted by i and j, and the interaction is taken different from zero only between next neighbours. The effective exchange, the Dzyaloshinsky, and the single ion anisotropy field are $H_J \approx 1.2 \cdot 10^7$ G, $H_D \approx 2.4 \cdot 10^4$ G, and $H_K \approx 2.2 \cdot 10^2$ G, respectively /4/ .

The description of the dynamical behaviour of the spin system in the nonequilibrium state will be performed in two steps. Firstly a phenomenological equation of motion is derived from the microscopical description. Then, this equation of motion has to be solved for the special case of given initial conditions.

II. Equations of motion of the magnetization vectors has been derived from the microscopical description for ferromagnetic /3/ and antiferromagnetic systems /4/ . The method of the nonequilibrium statistical operator of Zubarev /1/ , as described in /4/ , can also be applied to the Dzyaloshinsky antiferromagnet. In this paper, we only will give the results for a Dzyaloshinsky antiferromagnet, which were obtained by following the calculations of /4/ .

The time derivation of the mean values of the operators $\vec{M}$ and $\vec{N}$ can be divided into two parts, a nondissipative (pr.) and a dissipative (dmp.) one. If we select only linear terms of these operators and the lowest order in K/Jz , D_z^2/JK (z-number of next neighbours), we obtain for $\epsilon_0 / k_B T \gg 1$

$$\dot{M}_x^{pr.} = \frac{g\mu_B}{\hbar} H_E M_y + \frac{2S}{\hbar} K N_y + \frac{SDz}{\hbar} M_x ,$$

$$\dot{N}_x^{pr.} = \frac{g\mu_B}{\hbar} H_E N_y + \frac{2S}{\hbar} (K + Jz) M_y - \frac{SDz}{\hbar} N_x ,$$

$x \leftrightarrow y$ with $\frac{d}{dt} \leftrightarrow -\frac{d}{dt}$ and $D \leftrightarrow -D$;

$$\dot{M}_x^{dmp.} = -\lambda' \left(M_x + \frac{1}{4} \frac{D^2 z}{JK} M_x + \frac{1}{4} \frac{D}{J} \frac{D^2 z}{JK} N_y \right) , \tag{2}$$

$$\dot{N}_x^{dmp.} = -\lambda' \left(N_x - \frac{1}{4} \frac{D^2 z}{JK} N_x - \frac{1}{4} \frac{D}{J} \frac{D^2 z}{JK} M_y \right) ,$$

$x \leftrightarrow y$ with $D \leftrightarrow -D$;

$$\lambda' = \frac{z^{1/2} K^{3/2} e^{-\epsilon_0/k_B T}}{2\hbar \pi^3 S^3 J^{5/2} (k_B T)^2} \left(1 - \frac{1}{4}\frac{D^2 z}{JK} \pm \dots\right)$$

$$\epsilon_0 = 2S(KJz + K^2)^{1/2} \left(1 - \frac{1}{8}\frac{D^2 z}{JK} \pm \dots\right).$$

These equations can be transformed into a Landau-Lifshitz equation for the sublattice magnetization $\vec{M}_1$, $\vec{M}_2$:

$$\dot{\vec{M}}_{1,2} = \gamma[\vec{M}_{1,2} \times \vec{H}'_{1,2}] - \frac{\lambda}{|\vec{M}|}[\vec{M}_{1,2} \times [\vec{M}_{1,2} \times \vec{H}''_{1,2}]]$$

with

$$\vec{H}'_1 = H_E \vec{e}_z - \frac{2Jz}{g\mu_B N}\vec{M}_2 + \frac{4K}{g\mu_B N} M_1^z \vec{e}_z - \frac{2Dz}{g\mu_B N}\vec{e}_z \times \vec{M}_2 ,$$

$$H''_1 = \frac{1}{g\mu_B N}\left(4KM_1^z \vec{e}_z - \frac{D^2 z}{J}\vec{M}_2 - \frac{D}{J}\frac{D^2 z}{J}\vec{e}_z \times \vec{M}_2\right), \qquad (3)$$

$1 \leftrightarrow 2$ with $D \leftrightarrow -D$; $\gamma = \frac{g\mu_B}{h}$, $\lambda = \frac{g\mu_B}{2SK}\lambda'$,

We have found two different effective fields $\vec{H}'$ and $\vec{H}''$. If we consider the Hamiltonian (1), it is clear that H_E will not contribute to dissipative effects. The internal exchange field originated in J is not contained in zeroth order of D in $\vec{H}''$, because $\vec{M}$ commutes with the exchange interaction term, and the isotropic exchange does not single out any direction to which $\vec{N}$ can relax.

The equation (3) of the Landau-Lifshitz type was obtained from the microscopical description in the case of small declination of the magnetization vectors from the ground state. However, the parameters λ, $\vec{H}'$, $\vec{H}''$ can be considered as constants, not depending on the direction of $\vec{M}_1$, $\vec{M}_2$ and the time-variation rate of these quantities, as usually done in the phenomenological description.

Now, we have to solve the equation of motion (3) for given initial conditions. In a similar case, however, starting from a phenomenological Landau-Lifshitz equation in which the effective fields $\vec{H}'$ and $\vec{H}''$ are assumed to be equal, Nishikubo et al. /5/ have solved the equation of motion analytically and by numerical integration.

Omitting terms containing D, we easily can solve equation (2). For the magnetization vectors we obtain an equation of motion of the same form as a classical pendulum with viscous damping. For small oscillations the damping parameter is equal to the inverse of the relaxation time τ passing when the amplitude is decreased by e^{-1}: $\tau = 1/\lambda'$.

III. The damping parameter λ' (2) depends on S,J,D,K,T in such a manner, that for low temperatures, small K, and large values of J and S the damping parameter becomes small. Therefore the relaxation time in hematite will be relatively large. Using the values for hematite given above /2/, the relaxation time at T = 77K results from equation (2) as about $\tau \approx 10^{-4}$ s.

Of course, there are also other relaxation effects not described by the Hamiltonian (1), which diminish the relaxation time. For instance, from experiments in yttrium-iron garnets it is well known that random volume and surface distributions of magnetic inhomogeneities have a pronounced effect on the observed spin-wave damping. In antiferromagnets it is possible, therefore the damping parameter for the magnetization vector $\vec{M}$ differs essentially from the damping parameter for the staggered magnetization vector $\vec{N}$.

A relative large value of the relaxation time of the staggered magnetization vector $\vec{N}$ is not in contradiction to experiments performed up to now in hematite.

As shown by Eastman /6/ the line-width in antiferromagnetic resonance experiments is not directly related to the relaxation of the spin system. Heeger /7/ has measured the spin-wave instability in antiferromagnetic resonance in $KMnF_3$. He obtained a relaxation time approximately three orders of magnitude larger than derived from line-width of AFMR. A similar result was obtained for $CuCl_2 \cdot 2H_2O$ /8/.

In experiments with hematite in pulsed magnetic fields, in which the total magnetization vector $\vec{M}$ is observed, an hysteresis appears in the dependence of the magnetization on the magnetic field /9, 10/. Besser et al. /9/ have mentioned this effect, but have not investigated the origin of it.

More important are experiments in which the antiferromagnetic vector $\vec{N}$ is directly observed during the spin-flop process. Shchurov /11/ has measured the magnetostriction of a single crystal of hematite in high pulsed fields, which is connected with the direction of the vector $\vec{N}$. A relaxation effect is clearly shown in some of his figures (figs.9,10,38,39,40 of /11/), but not discussed in the text.

Recently, in the Laboratory of Neutron Physics at the JINR (Dubna) experiments were performed, in which the variation of the direction of $\vec{N}$ in pulsed magnetic fields was observed using neutron diffraction method. In these experiments, Nisiol et al. have used pulsed fields up to 65 kG and about 2 ms duration, applied into the direction of the second order axis of a hematite single crystal at the temperature T = 77 K. They observed, that the intensity of neutron diffraction, which corresponds to the magnitude of the angle between the staggered magnetization vector $\vec{N}$ and the trigonal axis, essentially differs from zero at the time interval of $2\text{-}3.10^{-4}\,s$ after the pulsed magnetic field vanishes.

It seems to be possible to study the behaviour of the spin system during the spin-flipping process in more detail. It would be interesting to observe the antiferromagnetic vector $\vec{N}$ at low temperatures in pulsed magnetic fields parallel to the trigonal axis and maximum values higher than H_c . In the quasistatic description the time dependence of the vector $\vec{N}$ follows a step function. If it were possible to observe relaxation effects in this spinflop process, we would get some insight into the dynamical behaviour of the spin system.

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