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Ordered phase of nuclear spins in uniaxial ferromagnets at ultralow temperatures

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Abstract. Paper investigates the onset of nuclear magnetic ordering caused by the indirect Suhl-Nakamura interaction in ferromagnets. The necessary condition for nuclear spin ordering with definite ordering vector is obtained. Particularly, it is shown that ferromagnetically ordered phase of nuclear spins could be observed only in case of disk shaped samples. The spectrum of the nuclear spin excitations is also found.

PACS. 75.10.-b General theory and models of magnetic ordering – 75.30.Ds Spin waves

1 Introduction

In references [1,2] the ferromagnetically ordered phase of nuclear spins in ferromagnets at ultralow spin temperatures was predicted. Such ordering is caused by the nuclei's indirect Suhl-Nakamura interaction [3,4] and takes place in case of compensation of hyperfine field on nuclei by the external static magnetic field. The nuclear ordering occurred has the direction nonparallel to the magnetization of electron spins. The nuclear spin-wave spectrum in the ordered phase was calculated in references [2,5].

However, to the best of our knowledge, the above theoretical predictions have not been confirmed experimentally. In our opinion, this could be caused by the following: in references [1,2,5] the periodical boundary conditions were used neglecting demagnetization effects, consequently the sample shape was not taken into account.

According to the result of the present article the demagnetization field plays a significant role in the formation of nuclear ordering. As it will be shown the nuclear ferromagnetically ordered phase could be observed only in disk shaped samples with applied static magnetic field perpendicular to the disk plane. The modified dispersion relation for nuclear spin excitations will be also obtained.

It should be also mentioned that using the dynamical polarization and adiabatical demagnetization methods [6] the required ultralow nuclear spin temperatures can be reached easily.

2 Origin expressions

Let us consider the electron-nuclear spin system in uniaxial ferromagnet. We choose x direction parallel to the axis of magnetic anisotropy and examine the case when a static magnetic field H_0 is applied along z axis. In this case the Hamiltonian has the following form:

$$\mathcal{H} = gH_0 \sum_f^N S_f^z - \gamma H_0 \sum_f^N I_f^z - K \sum_f^N \left(S_f^x\right)^2 + \frac{1}{2} \sum_{fg}^N J_{fg} \left(\mathbf{S}_f \mathbf{S}_g\right) + A \sum_f^N \left(\mathbf{S}_f \mathbf{I}_f\right) + \mathcal{H}_{dd}, \quad (1)$$

where $(-g)$ and γ are gyromagnetic ratios for electrons and nuclei, respectively; N is a number of sites in a sample; $\mathbf{S}_f$ and $\mathbf{I}_f$ are electron and nuclear spin operators disposed in the lattice site f, respectively; J_{fg} is the exchange interaction constant (for ferromagnet $J_{fg} < 0$); K is a constant of magnetic anisotropy ($K > 0$); A is a constant of hyperfine interaction ($A < 0$) and we work in the unit system $\hbar = k_{\mathrm{B}} = 1$. The last term in (1) describes the dipole-dipole interaction between electron spins (*i.e.* demagnetization forces) which was neglected in references [1,2,5]. This term has a well known form [7]:

$$\mathcal{H}_{dd} = \sum_{f,g}^N \frac{g^2}{2r_{fg}^3} \left\{ \left(\mathbf{S}_f \mathbf{S}_g\right) - 3 \frac{\left(\mathbf{S}_f \mathbf{r}_{fg}\right)\left(\mathbf{S}_g \mathbf{r}_{fg}\right)}{r_{fg}^2} \right\},$$

where $r_{fg} \equiv |\mathbf{r}_{fg}|$; $\mathbf{r}_{fg} = \mathbf{r}_f - \mathbf{r}_g$; $\mathbf{r}_f$ and $\mathbf{r}_g$ are the radius vectors of sites f and g, respectively.

As ultralow spin temperatures are considered the spin-wave approximation is used for electron spin excitations:

$$S_f^{\pm} = \sqrt{2S} a_f^{\pm}, \quad S_f^z = -S + a_f^+ a_f^-,$$

where $S_f^{\pm} = S_f^x \pm \mathrm{i} S_f^y$; S and I are electron and nuclear spins, respectively. Further, using the momentum

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presentation

$$a^{\pm}_{\mathbf{p}} = \sum_{f}^{N} \frac{1}{N} a^{\pm}_{f} \mathrm{e}^{\pm \mathrm{i}\mathbf{p}\mathbf{r}_f}, \qquad J_{\mathbf{p}} = \sum_{g}^{N} J_{fg} \mathrm{e}^{\mathrm{i}\mathbf{p}\mathbf{r}_{fg}}$$

and diagonalizing the electronic part of Hamiltonian (1) the following can be obtained:

$$\begin{aligned}\mathcal{H} = & \sum_{\mathbf{p}} \Omega_{\mathbf{p}} b^{+}_{\mathbf{p}} b_{\mathbf{p}} - \gamma \delta H \sum_{f}^{N} I^{z}_{f} \\ & + A \sqrt{\frac{S}{2N}} \sum_{f}^{N} \sum_{\mathbf{p}} \left\{ \mathrm{e}^{\mathrm{i}\mathbf{p}\mathbf{r}_f} \left(I^{+}_{f} \mu_{\mathbf{p}} - I^{-}_{f} \nu^{*}_{\mathbf{p}} \right) b_{\mathbf{p}} \right. \\ & \left. + \mathrm{e}^{-\mathrm{i}\mathbf{p}\mathbf{r}_f} \left(I^{-}_{f} \mu_{\mathbf{p}} - I^{+}_{f} \nu_{\mathbf{p}} \right) b^{+}_{\mathbf{p}} \right\}, \end{aligned} \tag{2}$$

where $b^{+}_{\mathbf{p}}$ and $b_{\mathbf{p}}$ are the diagonalized operators of magnon creation and annihilation, respectively; $I^{\pm}_{f} = I^{x}_{f} \pm \mathrm{i} I^{y}_{f}$; $\delta H = H_0 + (AS/\gamma)$ is a total static magnetic field on nuclei; $\mu_{\mathbf{p}}$ and $\nu_{\mathbf{p}}$ are diagonalizing constants [8]:

$$\mu_{\mathbf{p}} = \sqrt{\frac{E_{\mathbf{p}} + \Omega_{\mathbf{p}} - KS}{2\Omega_{\mathbf{p}}}},$$
$$\nu_{\mathbf{p}} = \frac{C_{\mathbf{p}} - KS}{\sqrt{2\Omega_{\mathbf{p}}(E_{\mathbf{p}} + \Omega_{\mathbf{p}} - KS)}}.$$

Here $\Omega_{\mathbf{p}}$ expresses the dispersion relation for electron spin excitations [5,8] (it should be noted that we restrict ourselves to the consideration of long wavelength limit $|\mathbf{p}|a \ll 1$, a is a lattice parameter):

$$\Omega_{\mathbf{p}} = \sqrt{E^2_{\mathbf{p}} - |C_{\mathbf{p}}|^2 - \omega_{\mathrm{A}}(E_{\mathbf{p}} - \mathrm{Re}C_{\mathbf{p}})};$$
$$E_{\mathbf{p}} = \omega_{\mathrm{H}} - \omega_{\mathrm{ex}} a^2 |\mathbf{p}|^2 - (D_0 + D_{\mathbf{p}}/2)$$

and [9,10]

$$C_{\mathbf{p}\neq 0} = \frac{\omega_{\mathrm{M}}}{2} \sin^2 \vartheta_{\mathbf{p}} \mathrm{e}^{-2\mathrm{i}\varphi_{\mathbf{p}}}, \qquad C_0 = 0,$$
$$D_{\mathbf{p}\neq 0} = \omega_{\mathrm{M}} \left(\cos^2 \vartheta_{\mathbf{p}} - 1/3 \right), \quad D_0 = \omega_{\mathrm{M}} \xi / 3.$$

In above formulae the following definitions are made: $\omega_{\mathrm{H}} = gH_0$; $\omega_{\mathrm{M}} = 4\pi g^2 S/a^3$; $\omega_{\mathrm{ex}} = SJ_0/6$ is the characteristic frequency of exchange interaction in case of simple cubic lattice; $\omega_{\mathrm{A}} = 2KS$ is the anisotropic frequency; $\vartheta_{\mathbf{p}}$ and $\varphi_{\mathbf{p}}$ are orbital and polar angles of $\mathbf{p}$, respectively; parameter ξ depends upon the sample shape and varies between the boundaries: $-1 \leq \xi \leq 2$ (*e.g.* for spherical samples $\xi = 0$, for disk samples with applied magnetic field perpendicular to the disk plane $\xi = 2$).

3 Ordered phase of nuclear spins

In order to obtain the Hamiltonian describing indirect interaction between nuclear spins let us average (2) over the fast oscillations of electron spins (for the details concerning this procedure see Ref. [11]). Thereby we get the Hamiltonian with modified (*via* including demagnetization effects) Suhl-Nakamura interaction:

$$\mathcal{H}^{\mathrm{eff}}_{n} = -\gamma \delta H I^{z}_{0} - \frac{1}{2N} \sum_{\mathbf{p}} \mathbf{I}_{-\mathbf{p}} \hat{V}_{\mathbf{p}} \mathbf{I}_{\mathbf{p}}, \tag{3}$$

where the following momentum presentation is used for nuclear spins:

$$\mathbf{I}_{\mathbf{p}} = \sum_{f}^{N} \mathbf{I}_{f} \mathrm{e}^{\mathrm{i}\mathbf{p}\mathbf{r}_f}$$

and

$$\hat{V}_{\mathbf{p}} = \begin{pmatrix} V^{xx}_{\mathbf{p}} & V^{xy}_{\mathbf{p}} & 0 \\ V^{yx}_{\mathbf{p}} & V^{yy}_{\mathbf{p}} & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

Here the tensor's components are presented by the formulae:

$$V^{xx}_{\mathbf{p}} = A^2 S \frac{E_{\mathbf{p}} - \mathrm{Re}C_{\mathbf{p}}}{\Omega^2_{\mathbf{p}}},$$
$$V^{yy}_{\mathbf{p}} = A^2 S \frac{E_{\mathbf{p}} + \mathrm{Re}C_{\mathbf{p}} - \omega_{\mathrm{A}}}{\Omega^2_{\mathbf{p}}},$$
$$V^{xy}_{\mathbf{p}} = V^{yx}_{\mathbf{p}} = A^2 S \frac{\mathrm{Im}C_{\mathbf{p}}}{\Omega^2_{\mathbf{p}}}.$$

The theory of the onset of magnetic ordering is discussed in detail in recent review article reference [6]. Particularly, it is well known that magnetic order takes place over ordering vector $\mathbf{k}$ for which the eigenvalue of $\hat{V}_{\mathbf{k}}$ reaches maximum value. In our case the maximum eigenvalue for $\mathbf{k} \neq 0$ can be presented in the form:

$$\lambda_{\max}(\mathbf{k} \neq 0) = \frac{A^2 S}{\omega_{\mathrm{H}} - \omega_{\mathrm{A}} - \omega_{\mathrm{M}}(\xi + 1)/3}, \tag{4}$$

while the maximum eigenvalue of $\hat{V}_{\mathbf{k}}$ for $\mathbf{k} = 0$ has a form

$$\lambda_{\max}(\mathbf{k} = 0) = \frac{A^2 S}{\omega_{\mathrm{H}} - \omega_{\mathrm{A}} - \omega_{\mathrm{M}} \xi / 2}. \tag{5}$$

Besides, the value (4) is reached in case of $|\mathbf{k}| \to 0$ and $\sin \vartheta_{\mathbf{k}} = 0$.

The ferromagnetic ordering of nuclear spins (caused by indirect Suhl-Nakamura interaction) takes place only if

$$\lambda_{\max}(\mathbf{k} = 0) \geq \lambda_{\max}(\mathbf{k} \neq 0). \tag{6}$$

Otherwise, no permanent nuclear spin structure (caused by Suhl-Nakamura interaction) with definite ordering vector exists. Indeed, in the last case, as it is mentioned above, the ordered state is characterized by the ordering vector $|\mathbf{k}| \to 0$. But in this case our whole consideration is not valid, because in a such wavelength range only the inhomogeneous Walker modes [12] are excited. These modes can not be specified by the definite wave vector. Consequently if inequality (6) does not hold the complex ordered

phases of nuclear spins depending upon the sample shape are realized, but no ordered nuclear spin structure characterized by a definite ordering vector exists. On the other hand, if inequality (6) holds the ordering vector is equal to zero and thus the ferromagnetic ordering of nuclear spins could be taken place.

Inequality (6) is satisfied for $\xi \geq 2$, *i.e.* only for disk shaped samples, when the external magnetic field is applied normally to the disk plane (let us remind that ξ varies between the boundaries $-1 \leq \xi \leq 2$).

Using the general results of [6] one can conclude that the expression for the critical value of the effective magnetic field at zero temperature has the form:

$$\gamma\delta H_{\rm c} = \frac{A^2 SI}{\omega_{\rm H} - \omega_{\rm M} - \omega_{\rm A}}, \tag{7}$$

while the transition temperature at zero effective magnetic field ($\delta H = 0$) is expressed by the equality:

$$T_{\rm c} = \frac{A^2 SI(I+1)/3}{\omega_{\rm H} - \omega_{\rm M} - \omega_{\rm A}}\,. \tag{8}$$

Expressions (7, 8) are slightly modified in comparison with the respective ones obtained in references [1,2] and thus, their numerical values are almost the same.

In case of $|\delta H| < \delta H_{\rm c}$ and below the ordering temperature the ferromagnetic ordering of nuclear spins takes place along the vector $\mathbf{n}$. It lies in xz plane and

$$\cos\Psi = \frac{\gamma\delta H}{\langle I\rangle V_0^{xx}},$$

where Ψ is an angle between $\mathbf{n}$ and z axis; $\langle I\rangle$ is a static averaged value of nuclear polarization.

Let us choose the new frame of references where the direction of averaged nuclear spin coincide with the z' axis:

$$\begin{aligned}
(I_{\mathbf{p}}^x)' &= I_{\mathbf{p}}^x \cos\Psi - I_{\mathbf{p}}^z \sin\Psi,\\
(I_{\mathbf{p}}^y)' &= I_{\mathbf{p}}^y,\\
(I_{\mathbf{p}}^z)' &= I_{\mathbf{p}}^z \cos\Psi + I_{\mathbf{p}}^x \sin\Psi.
\end{aligned}$$

Then diagonalizing (3) in analogy with reference [5] the following dispersion relation can be obtained for linear spin excitations in the nuclear ordered phase:

$$\begin{aligned}
\Omega_{\mathbf{p}}^n = I\Big\{&(V_0^{xx} - V_{\mathbf{p}}^{yy})(V_0^{xx} - V_{\mathbf{p}}^{xx}\cos^2\Psi)\\
&- (V_{\mathbf{p}}^{xy})^2 \cos^2\Psi\Big\}^{1/2}.
\end{aligned} \tag{9}$$

This spectrum is characterized by the following gap and upper limit:

$$\begin{aligned}
\Omega_0^n &= I|\sin\Psi|\sqrt{V_0^{xx}(V_0^{xx} - V_0^{yy})},\\
\Omega_\infty^n &= IV_0^{xx}.
\end{aligned}$$

Let us consider a weakly anisotropic ferromagnet ($\omega_{\rm A} \ll \omega_{\rm M} \ll \omega_{\rm H}$) when the full compensation of hyperfine field on nuclei occurs, *i.e.* $\gamma H_0 = |A|S$, thus $\cos\Psi = 0$. Examining the long wavelength excitations $\omega_{\rm ex} a^2|\mathbf{p}|^2 \ll \omega_{\rm H}$ the following expression can be obtained from (9):

$$\Omega_{\mathbf{p}}^n = \frac{I\gamma^2}{Sg^2}\sqrt{\omega_{\rm H}\big(\omega_{\rm A} + \omega_{\rm ex} a^2|\mathbf{p}|^2 + \omega_{\rm M}\sin^2\vartheta\sin^2\varphi\big)}. \tag{10}$$

In order to compare our results with reference [1] let us consider ferromagnet $^{(151)}$EuO which is characterized by the following constants: $S = 7/2$, $I = 5/2$, $\omega_{\rm H} = g|A|S/\gamma = 2.4 \times 10^{12}$ s^{-1}, $\omega_{\rm A} = 0$, $\omega_{\rm ex} = 7.3 \times 10^{12}$ s^{-1}, $\omega_{\rm M} = 1.1 \times 10^{11}$ s^{-1}, $a = 5.2 \times 10^{-8}$ sm, $g = 1.8 \times 10^7$ (s/Oe)$^{-1}$, $\gamma = 6.6 \times 10^3$ (s/Oe)$^{-1}$. Then one can get from (8) the value for ordering temperature $T_{\rm c} = 2.2 \times 10^{-6}$ K, which is almost the same as in reference [1]. We also can obtain from (10) the dispersion relation:

$$\Omega_{\mathbf{p}}^n \simeq T_1\sqrt{a^2|\mathbf{p}|^2 + T_2\sin^2\vartheta\sin^2\varphi},$$

where $T_1 = 4.3 \times 10^5$ s^{-1} and $T_2 = 1.5 \times 10^{-2}$. Finally we conclude that demagnetization effects (which are manifested in appearance of the angular dependence) are significant in whole wavelength range $a|\mathbf{p}| \ll 1$ where the continual approach is valid.

4 Conclusions

The main result of the present article is the following: the nuclear ferromagnetically ordered phase caused by the Suhl-Nakamura interaction could be observed only in disk shaped samples where applied external magnetic field is directed perpendicularly to the disk plane. For the purpose of the present article the anisotropic axis is considered to be perpendicular to the direction of external field. However, as it can be easily shown, the main result remains the same for any orientation of the axis of magnetic anisotropy.

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