Parametrical NMR instability in simple metals at nanokelvin temperatures

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The paramagnetic state of a highly polarized copper nuclear-spin system under transverse pumping is theoretically studied at ultralow temperatures. The parametrical instability is investigated and the threshold amplitudes are calculated versus the detuning of pumping frequency from the Larmor frequency. It is shown that sample shape greatly affects the values of the instability threshold.

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I. INTRODUCTION

In the last few years several world records have been broken at both positive and negative sides of absolute zero (see, e.g., the review article Ref. 1) in simple metals. These achievements together with advanced instrumentation for measuring and analyzing various NMR data allows one to calculate the constants of indirect nuclear-spin-spin interactions mediated by conduction electrons such as isotropic Ruderman-Kittel[2] as well as anisotropic indirect exchange interactions.[3] As theoretically expected several ordered phases of nuclear spins are experimentally discovered in copper[4–6] and silver.[7,8] The exception is rhodium where no sign of nuclear ordering has been observed[9] though the system was cooled to 250 pK in zero magnetic field.

It should be mentioned that in order to verify the theory describing the spin-spin interaction processes in simple metals one could investigate not only the spin ordering in zero magnetic field but also study the spin systems nonlinear behavior in strong static magnetic fields (providing a highly polarized paramagnetic state of nuclear spins) under the weak transverse pumping. So at this point, it seems most appropriate to study in detail different nonlinear processes induced by the spin-spin interaction forces, especially parametrical instability in the media.

As is well known, the Suhl's parametrical instability theory[10] is based upon the following statement: when the value of a spatially uniform pumping field exceeds a certain threshold the nonlinear coupling between uniform and nonuniform (with definite wave number) modes induces exponential growth of the nonuniform mode with fastest growing rate. If three magnon processes dominate one deals with the first kind of Suhl instability (parametrical magnons carry half the frequency of the transverse pumping field), while the second kind of Suhl instability describes the processes caused by four magnon interactions (parametrical magnons carry the same frequency as a transverse pumping field). In the presence of a strong magnetic field there is a huge gap in the spin excitation spectrum and three magnon decay processes are suppressed. However, four magnon processes are allowed and they will cause the parametrical instability. Thus in the present paper the second kind of Suhl instability under the transverse pumping is theoretically studied in the highly polarized paramagnetic state of a nuclear-spin system in simple metals. The effect could be easily detected experimentally: after the amplitude of the pumping field exceeds the threshold the number of parametrical spin excitations grows rapidly and this will affect a sharp increase of the imaginary part of magnetic susceptibility.

Similar problems have been investigated by a number of authors (see, e.g., Ref. 11) for electronic spin systems in magnets as well as more recently for nuclear-spin systems in electronic diamagnets.[12,13] Like electronic spin systems, in simple metals there exist isotropic exchange and anisotropic dipolar interactions between nuclear spins. But the physical picture is rather different, because in the case of electronic spins the exchange interaction is much larger than the dipolar one, while in the case of nuclear spins they could be characterized by the same scale. As we will show below this circumstance causes a strong shape dependence of instability thresholds in simple metals. Thus we investigate how the different peculiarities characterizing Ruderman-Kittel and dipolar interactions could be manifested via the phenomenon of parametrical instability. The analitical results presented here are valid for copper and silver and we perform numerical estimations and plot the graphs for copper, although we note that the consideration could be simply extended for the nuclear spin-system in rhodium taking into account anisotropic exchange forces.

II. BASIC CONSIDERATION

Let us consider a spin system of a metallic sample placed in a strong static magnetic field H_0 (for which nondiagonal processes are suppressed) which is undergoing transverse pumping. H_0 is applied along the [0 0 1] axis (z direction) of the fcc lattice. The secular part of the nuclear-spin system Hamiltonian has the form[1]

$$\mathcal{H}=-\gamma\hbar H_0\sum_f^N I_f^z-\frac{1}{2}\sum_{fg}^N J_{fg}(\vec{I}_f\vec{I}_g)-\frac{1}{2}\sum_{fg}^N D_{fg}(2I_f^zI_g^z-I_f^+I_g^-)-\frac{\hbar\omega_1}{2}\sum_f^N(I_f^+e^{i\omega t}+I_f^-e^{-i\omega t}),\qquad(1)$$

where γ is the gyromagnetic ratio for nuclei, $\vec{I}_f$ is the operator of the spin located in the lattice site f, $I_f^{\pm}=I_f^x\pm iI_f^y$, N is the number of spins in a lattice, J_{fg} are the constants of Ruderman-Kittel interaction,

$$D_{fg}=\frac{\gamma^2\hbar^2}{2|\vec{r}_f-\vec{r}_g|^3}(3\cos^2\vartheta_{fg}-1)$$

are dipolar constants, $\vec{r}_f$ is the radius vector of the spin situated in the site f, ϑ_{fg} is the angle between $\vec{r}_f-\vec{r}_g$, and z, ω, and ω_1 are a frequency and amplitude (in frequency units) of the transverse pumping field.

Let us mention that the probability of three magnon processes is minute and the first kind of Suhl instability does not take place. Therefore, the nonsecular terms in Hamiltonian (1) are neglected.

Let us use the momentum presentation

$$\vec{I}_k=\frac{1}{N}\sum_f^N \vec{I}_f\, e^{i\vec{k}\vec{r}_f}$$

and rewrite the Hamiltonian (1) in the form [it coincides with the secular part of expression (103) from Ref. 1]

$$\mathcal{H}=-\hbar\omega_0 N I_0^z-\frac{1}{2}\sum_q \{(J_q+2D_q)I_q^z I_q^z+(J_q-D_q)I_q^+I_q^-\}$$
$$-\frac{\hbar\omega_1}{2}N(I_0^+e^{i\omega t}+I_0^-e^{-i\omega t}), \tag{2}$$

where $\omega_0=\gamma H_0$ is a Larmor frequency and

$$J_q=\sum_g^N J_{fg}e^{i\vec{q}(\vec{r}_g-\vec{r}_f)},\qquad D_q=\sum_g^N D_{fg}e^{i\vec{q}(\vec{r}_g-\vec{r}_f)}. \tag{3}$$

In expression for J_q only the summation over 12 neighboring sites will be considered. In principle the next neighborhood also could be taken into account but this complicates the calculations and does not lead to significant changes in the final results. Strictly speaking, the far acting effects should be taken into account for the dipolar forces. However, as we will see below, only the modes close to the boundary of the Brillouin-zone are parametrically excited and so we have two kinds of modes: the uniform mode with $q=0$ and strongly nonuniform modes of parametrical excitations with $2aq\sim 1$ ($2a$ is a lattice parameter for the fcc structure). For the latter ones the exponent [see formula (3) for D_q] oscillates strongly and the sum over the sites out of the Lorentz sphere is equal to zero. While in the case of uniform mode ($q=0$) the sum over all sites of the lattice should be taken into account. Thus summarizing the expressions for J_q and D_q could be written as follows:

$$J_q=4J_1(c_xc_y+c_xc_z+c_yc_z),$$
$$D_{q\neq 0}=-D_1(2c_xc_y-c_xc_z-c_yc_z),$$
$$D_0=-\frac{2\pi}{3}D_1\xi, \tag{4}$$

where $c_\alpha\equiv\cos q_\alpha a$ ($\alpha=x,y,z$), $J_1=-12,7$ nK is the Ruderman-Kittel constant for nearest-neighboring nuclear-spin pairs, $D_1=\gamma^2\hbar^2/a^3=25,4$ nK is a dipolar constant and $-1\leqslant\xi\leqslant 2$ is a static demagnetizing factor depending on sample shape. For instance, $\xi=-1$ for wire samples with applied static magnetic field along wire and $\xi=2$ for thin disks with applied field along the direction perpendicular to the disk plane (see, e.g., Ref. 14).

Using the commutation relations

$$[I_{k_1}^z,I_{k_2}^+]=\frac{1}{N}I_{k_1+k_2}^+,\qquad [I_{k_1}^+,I_{k_2}^-]=\frac{2}{N}I_{k_1-k_2}^z$$

one can simply obtain from Eq. (2) the motion equation for I_k^+ in the rotating (with frequency ω) frame of references:

$$\frac{dI_k^+}{dt}=-i\Delta I_k^++\frac{i}{\hbar}\sum_q (J_q-D_q)I_q^+I_{k-q}^z$$
$$-\frac{i}{\hbar}\sum_q (J_q+2D_q)I_{k-q}^+I_q^z+i\omega_1 I_k^z, \tag{5}$$

where $\Delta=\omega_0-\omega$ is a detuning of the frequency of the pumping field.

Averaging the expression (5) quantum statistically one can decouple the nonlinear terms (see, e.g., Ref. 11). Let us make the following definitions: $\vec{m}_k\equiv\langle\vec{I}_k\rangle/I_0$, $\langle\cdots\rangle$ denotes quantum-statistical averaging,

$$R_q=\frac{I_0}{\hbar}(J_q-D_q),\qquad T_q=\frac{I_0}{\hbar}(J_q+2D_q). \tag{6}$$

As it could be easily shown (see, e.g., Ref. 15) the decoupling procedure is valid if $|m_k^+|^2\gg 1/N$ and thus it is justified and macroscopical quantities are considered.

In order to solve the problem analytically let us examine small deviations from the static state. Therefore we work in the limit $\Sigma_k|m_k^+|^2\ll 1$, thus only small tipping angles are examined and m_k^z could be presented as follows:

$$m_k^z=1-\frac{1}{2}\sum_q m_{k+q}^+m_q^-\,.$$

At the initial moment the number of magnons with wave number $q\neq 0$ is extremely small according to the thermal distribution. Thus only the homogeneous mode exists which behaves following the motion equation easily derived from Eq. (5):

$$\frac{dm_0^+}{dt}=-i(\Delta+T_0-R_0-i\eta)m_0^++i\omega_1. \tag{7}$$

Here we introduce damping parameter $\eta\equiv T_2^{-1}$ (T_2 is a linewidth) and we assume that the damping for all of the modes is the same.[10] Parameter η substitutes the nonlinear terms in Eq. (5) taking into account the interaction with thermal magnons. In case of high spin polarizations the damping parameter could be easily calculated in spin-wave approximation as the inverse lifetime of the mode (see, e.g., Ref. 16) and is approximately equal to

$$\eta\approx(1-p_0)D_1, \tag{8}$$

where p_0 is the nuclear-spin system polarization.

Equation (7) has a stationary solution in the form

$$m_0^+ = \frac{\omega_1}{\Delta + T_0 - R_0 - i\eta}. \tag{9}$$

Now we are ready to write down the motion equations for parametrically excited modes keeping only nonlinear terms which include the homogeneous mode:

$$\left(\frac{d}{dt} + \tilde{\eta} + i\tilde{\omega}_k\right) m_k^+ + iS_k (m_0^+)^2 m_{-k}^- = 0,$$

$$\left(\frac{d}{dt} + \tilde{\eta} - i\tilde{\omega}_k\right) m_{-k}^- - iS_k^* (m_0^-)^2 m_k^+ = 0, \tag{10}$$

where

$$\tilde{\eta} = \eta\left(1 + \frac{|m_0^+|^2}{2}\right),$$

$$\tilde{\omega}_k = \Delta + T_0 - R_k + \frac{|m_0^+|^2}{2}[R_k - T_k + \Delta],$$

$$S_k = -\frac{1}{2}[T_k - T_0 - \Delta + i\eta], \tag{11}$$

and S_k^* denotes complex conjugated quantity of S_k. Substituting m_k^+, $m_k^- \sim e^{\nu_k t}$ in Eq. (10) we get the following solution for the growing rate ν_k:

$$\nu_k = \sqrt{|S_k|^2 |m_0^+|^4 - (\tilde{\omega}_k)^2} - \tilde{\eta}. \tag{12}$$

III. RESULTS

If the growing rate ν_k is real and positive the amplitude of parametrically excited mode exponentially grows, i.e., parametrical instability occurs. The maximum increment is reached if the resonance condition for parametrical excitations holds [see expression (12)]:

$$\tilde{\omega}_k = 0. \tag{13}$$

Thus from Eqs. (12) and (13) we get the threshold value for the tipping angle ϑ of the homogeneous mode:

$$\sin^2 \vartheta_{cr} = |m_0^+|^2_{cr} = \left.\frac{\tilde{\eta}}{|S_k|}\right|_{min}, \tag{14}$$

and the minimal value over $\vec{k}$ should be taken keeping in mind the condition (13). Taking into consideration that only small tipping angles $\sin^2\vartheta = |m_0^+|^2 \ll 1$ are examined the approximate analytical solution could be derived from Eqs. (14), (6), and (4):

$$|m_0^+|^2_{cr} = \frac{2\hbar\eta}{3I_0 D_1}\left[2 + \frac{\hbar\tilde{\Delta}}{I_0(D_1 - 4J_1)}\right]^{-1}, \qquad \tilde{\Delta} = \Delta + T_0, \tag{15}$$

where $\tilde{\Delta}$ varies within the frequency range

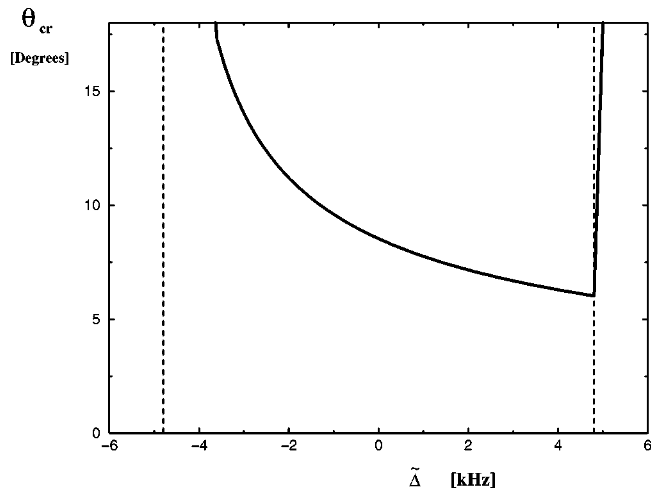

FIG. 1. Critical values of tipping angles for which parametrical instability occurs versus $\tilde{\Delta}$. Vertical dashed lines indicate the spin-wave spectrum zone's boundaries. Note that we consider only small tipping angles $\sin^2\vartheta_{cr} = |m_0^+|^2 \ll 1$.

$$-2\frac{I_0}{\hbar}(D_1 - 4J_1) \leqslant \tilde{\Delta} \leqslant 2\frac{I_0}{\hbar}(D_1 - 4J_1). \tag{16}$$

Outside of this range the condition (13) could not be fulfilled and the threshold amplitude grows rapidly (see Fig. 1). Within the frequency range (16) the wave-number vectors of parametrically unstable modes could be also determined:

$$c_y = c_z = \pm 1, \quad c_x = \mp 1 \mp \frac{\hbar\tilde{\Delta}}{I_0(D_1 - 4J_1)} \quad \text{for}$$

$$-2\frac{I_0}{\hbar}(D_1 - 4J_1) \leqslant \tilde{\Delta} \leqslant 0,$$

$$c_x = c_y = \pm 1, \quad c_z = \mp \frac{\hbar\tilde{\Delta}}{2I_0(D_1 - 4J_1)} \quad \text{for}$$

$$0 < \tilde{\Delta} \leqslant 2\frac{I_0}{\hbar}(D_1 - 4J_1). \tag{17}$$

Afterwards, using the expressions (9) and (15) the formula for the threshold amplitude of the pumping field could be easily obtained:

$$\omega_1^{cr} = \sqrt{\frac{2\hbar\eta}{3I_0 D_1}} \sqrt{\frac{(\Delta + T_0 - R_0)^2 + \eta^2}{2 + \dfrac{\hbar(\Delta + T_0)}{I_0(D_1 - 4J_1)}}}, \tag{18}$$

where as earlier $\tilde{\Delta} = \Delta + T_0$ varies between the boundaries (16).

In the present paper we make the numerical estimate in the case of copper. In Fig. 1 the dependence of $|m_0^+|^2_{cr}$ upon the $\tilde{\Delta}$ is presented. There and further we choose the damping constant as follows $\hbar\eta/D_1 = 0{,}1$, i.e., according to Eq. (8) the high polarized paramagnetic phase of copper nuclei $p_0 \simeq 90\%$ is considered.

In expression (15) the sample shape dependence is included into the shifted value of frequency detuning $\tilde{\Delta}$ in order to define the conditions of validity of the small tipping angle approximation. Thus the graph $|m_0^+|^2_{cr}$ versus $\tilde{\Delta}$ does not ''feel'' the sample shape because the shape dependence exists only in $\tilde{\Delta}$. Now using the formula (18) we examine the dependence of ω_1^{cr} upon the unshifted detuning Δ and sample shape. The corresponding graphs are presented in Fig. 2. Note that in the case of thin disk samples with applied static magnetic field perpendicular to the disk plane ($\xi=2$) the minimum threshold is very small [see Fig. 2(a)]:

$$(\omega_1^{cr})_{min} \simeq \frac{\eta}{3}\sqrt{\frac{2\hbar\eta}{D_1}},$$

and if we choose again $\hbar\eta/D_1=0{,}1$ then $(\omega_1^{cr})_{min}\simeq 5\ \mu\mathrm{T}$ for the detuning $\Delta=R_0-T_0\simeq 11$ kHz. The appearance of such a minute threshold is caused by the fact that both resonance conditions for the homogeneous mode $\Delta+T_0-R_0=0$ [see expression (9)] and for the parametrically unstable mode $\tilde{\omega}_k=0$ could be reached simultaneously. As calculations shows, such a situation is realized if $\xi>0$. Otherwise, e.g., for wire samples with a static demagnetizing factor $\xi=-1$ [see Fig. 2(b)] the threshold amplitude is rather higher, $\omega_1^{cr}\simeq 600\ \mu\mathrm{T}$, and almost constant.

IV. CONCLUSIONS

In the present paper the unstable behavior of the weak pumping process of the highly polarized paramagnetic state of a nuclear-spin system in simple metals is theoretically studied. Developing the formalism presented here it will be possible to study various ordered states of a copper nuclear-spin system using the ''instability tool.'' Moreover, the parametrical instability phenomenon is a real indicator of existence of different chaotic regimes[17,18] in the system for larger amplitudes (than the threshold one) of the pumping magnetic field.

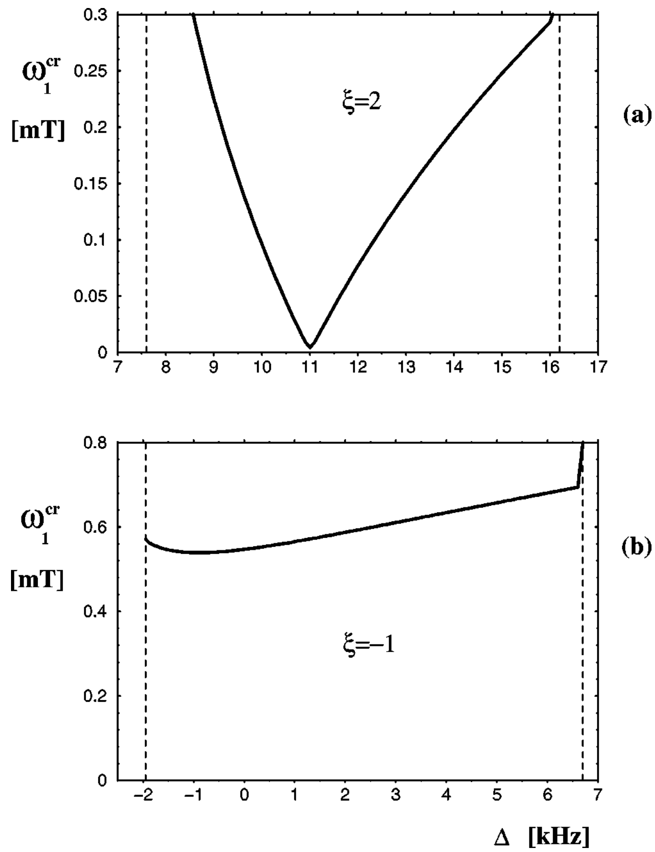

FIG. 2. Thresholds for the amplitude of the pumping magnetic field versus detuning of its frequency Δ from the Larmor frequency. (a) The case of the thin disk sample with applied static magnetic field perpendicular to the disk plane (static demagnetizing factor $\xi=2$) and (b) the case of wire with applied static magnetic field along the sample ($\xi=-1$). The dashed vertical lines indicate the boundaries of the range of Δ, where the condition $|m_0^+|^2 \ll 1$ holds.

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