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Nuclear spin relaxation at ultralow temperatures

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Nuclear spin relaxation induced by hyperfine coupling is studied theoretically at positive and negative submicrokelvin temperatures. By avoiding the assumption of the high-temperature limit, adopted in conventional theories, we derive a formula in which the relaxation rate is expressed in terms of thermal averages of nuclear spin energies. The exchange interaction induces an asymmetry in the energy spectrum, which leads to relaxation rates that depend on whether the nuclear spin temperature is positive or negative. High-temperature expansion methods and Monte Carlo simulations are applied to explain the anomalous results by Hakonen *et al.* in rhodium qualitatively. [S0163-1829(99)12313-0]

I. INTRODUCTION

The success in finding nuclear spin ordering in noble metals has opened a new field in ultralow-temperature physics.¹ After the discovery of antiferromagnetic order in copper below 58 nK,² silver was found to undergo phase transitions at 560 pK to antiferromagnetic (AF) order and at -1.9 nK to ferromagnetic (F) order.^{3,4} In these experiments negative temperatures were produced by rapid inversion of the external field. At $T < 0$ the system is stabilized by maximizing the free energy⁵ so that high-energy excitations become important, in contrast to $T > 0$ where the equilibrium is established by the free-energy minimum and low-energy excitations are important.

The experimental studies have been extended to the search for nuclear ordering in rhodium. Although the nuclear order has not been achieved in experiments down to 280 pK and up to -750 pK, it was found that the paramagnetic susceptibility displays AF Curie-Weiss behavior at $T > 0$ and a crossover from F to AF tendency at $T < 0$.⁶ Furthermore, Hakonen *et al.* found that, at the extreme temperatures, the nuclear spin relaxation is about two times slower at $T < 0$ than at $T > 0$.⁷ When the temperature of spins decreases and becomes comparable with the internal field seen by the nuclei, the assumption of high temperature adopted in the conventional theories cannot be applied anymore. At these temperatures, a deviation from the Korringa law is expected to occur.⁸ However, as far as the relaxation with infinitesimal difference of temperatures between the nuclear spins and the conduction electrons is considered, as in the conventional theories,⁹ one cannot make a distinction between positive and negative temperatures. On the other hand, the two samples used in the experiments⁷ contained 6 and 14 ppm of iron impurities. Although it is known that magnetic impurities increase the relaxation rate in metals,^{10,11} it seems not very successful to pinpoint them as the origin of the anomaly at $T < 0$. Hence the anomaly has remained unexplained and motivates the present study.

II. FORMULA FOR NUCLEAR-SPIN RELAXATION

We consider the rate of heat flow from nuclear spins (system) to conduction electrons (reservoir) following Leggett and Vuorio.¹² We assume that the nuclear spin system is in internal thermal equilibrium at temperature T , different from the temperature of the reservoir T_e . In the experiments,⁷ performed in magnetic fields less than $400 \mu\text{T}$, T_e was about $100 \mu\text{K}$, whereas T was on the order of ± 1 nK. We denote the inverse of temperature ($1/k_B T$) as β and β_e for the system and reservoir, respectively.

We assume that the heat flow is mediated by the hyperfine coupling

$$H = A \sum_i \mathbf{I}_i \cdot \mathbf{s}_i, \quad (1)$$

where \mathbf{I}_i and \mathbf{s}_i are the nuclear spin operator and the conduction-electron-spin operator at site i . Leggett and Vuorio¹² wrote down an expression for the heat-flow rate from the system to the reservoir on the basis of the golden rule,

$$\begin{aligned} \frac{dQ}{dt} = & \frac{2\pi}{\hbar} A^2 \sum_i \sum_{\alpha=x,y,z} \sum_{n,n'} \sum_{m,m'} P_n^{(n)} P_m^{(e)} (E_n - E_{n'}) \\ & \times |\langle n | I_{i\alpha} | n' \rangle|^2 |\langle m | s_{i\alpha} | m' \rangle|^2 \delta(E_n + E_m - E_{n'} - E_{m'}), \end{aligned} \quad (2)$$

where n and n' refer to the states of the system, m and m' to the reservoir. $P_n^{(n)}$ and $P_m^{(e)}$ denote the canonical distribution for the system and the reservoir, respectively. Leggett and Vuorio¹² expanded the r.h.s. of Eq. (2) in $\Delta\beta = \beta - \beta_e$ and retained only the first-order terms in order to apply the theory to the case where the temperature of the system is close to that of the reservoir. However, in the present case, $\Delta\beta \approx \beta$ and $|\Delta\beta| \gg \beta_e$, so that we proceed with the calculation of Eq. (2) without expanding in $\Delta\beta$. In a similar manner

as done by Leggett and Vuorio,¹² we introduce correlation functions for the nuclear spins and the conduction-electron spins:

$$\Phi_{i\alpha}^{(n)}(\omega) = \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} \langle I_{i\alpha}(t) I_{i\alpha} \rangle e^{i\omega t} dt, \quad (3)$$

$$\Phi_{i\alpha}^{(e)}(\omega) = \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} \langle s_{i\alpha}(t) s_{i\alpha} \rangle e^{i\omega t} dt, \quad (4)$$

where $\Phi_{i\alpha}^{(n)}(\omega)$ and $\Phi_{i\alpha}^{(e)}(\omega)$ are defined by thermal averaging of the nuclear spin and the conduction-electron Hamiltonians at β and β_e , respectively.

In terms of the correlation functions, Eq. (2) is expressed as

$$\frac{dQ}{dt} = -2\pi A^2 \sum_i \sum_{\alpha=xyz} \int_{-\infty}^{\infty} \hbar \omega \Phi_{i\alpha}^{(n)}(\omega) \Phi_{i\alpha}^{(e)}(-\omega) d\omega. \quad (5)$$

By the fluctuation-dissipation theorem, the correlation functions defined in Eqs. (3) and (4) relate with the local susceptibilities, given in the units of $g\mu_B$ and $g_N\mu_N$:

$$\Phi_{i\alpha}^{(n,e)}(\omega) = \frac{1}{\pi} \frac{\text{Im} \chi_{i\alpha i\alpha}^{(n,e)}(\omega)}{1 - e^{-\beta\hbar\omega}}. \quad (6)$$

Though $\text{Im} \chi_{i\alpha i\alpha}^{(n)}(\omega)$ changes its sign depending on $\beta < 0$ or $\beta > 0$, $\Phi_{i\alpha}^{(n)}(\omega)$ remains positive. If we substitute Eq. (6) into Eq. (5), we can confirm that dQ/dt vanishes when $\beta = \beta_e$, since $\text{Im} \chi(\omega)$ is odd in ω . Therefore we may rewrite Eq. (5) to the form

$$\begin{aligned} \frac{dQ}{dt} = & -2\pi A^2 \sum_i \sum_{\alpha=xyz} \int_{-\infty}^{\infty} \hbar \omega [\Phi_{i\alpha}^{(n)}(\omega) - \overline{\Phi_{i\alpha}^{(n)}(\omega)}] \\ & \times \Phi_{i\alpha}^{(e)}(-\omega) d\omega, \end{aligned} \quad (7)$$

where $\overline{\Phi_{i\alpha}(\omega)}$ denotes the thermal average at $\beta = \beta_e$. Here we notice that the characteristic frequency of the nuclear spins is much lower than that of the conduction electrons. Therefore it is legitimate to replace $\Phi_{i\alpha}^{(e)}(-\omega)$ with $\Phi_{i\alpha}^{(e)}(0)$ and to put it outside the integral in Eq. (7). Then, with aid of Eq. (6), it is allowed to write

$$\begin{aligned} \frac{dQ}{dt} = & -2A^2 \sum_i \sum_{\alpha=xyz} \lim_{\omega \rightarrow 0} \frac{\text{Im} \chi_{i\alpha i\alpha}^{(e)}(\omega)}{\beta_e \hbar \omega} \\ & \times \int_{-\infty}^{\infty} \hbar \omega [\Phi_{i\alpha}^{(n)}(\omega) - \overline{\Phi_{i\alpha}^{(n)}(\omega)}] d\omega, \end{aligned} \quad (8)$$

where $\Phi_{i\alpha}^{(n)}(\omega)$ does not depend on i as far as the system is in the paramagnetic phase and, as the conduction-electron system is paramagnetic and in a weak external field, $\chi_{i\alpha i\alpha}^{(e)}(\omega)$ does not depend on α . As a result we can write Eq. (8) as

$$\begin{aligned} \frac{dQ}{dt} = & -2A^2 \left(\frac{1}{N} \sum_i \lim_{\omega \rightarrow 0} \frac{\text{Im} \chi_{ii}^{(e)}(\omega)}{\beta_e \hbar \omega} \right) \\ & \times \sum_i \sum_{\alpha=xyz} \int_{-\infty}^{\infty} \hbar \omega [\Phi_{i\alpha}(\omega) - \overline{\Phi_{i\alpha}(\omega)}] d\omega, \end{aligned} \quad (9)$$

where N denotes the total number of spins. For a noninteracting electron gas,

$$\lim_{\omega \rightarrow 0} \frac{\text{Im} \chi_{ii}^{(e)}(\omega)}{\hbar \omega} = \frac{\pi}{2} \rho_F^2, \quad (10)$$

where ρ_F is the density of states at the Fermi energy per spin. In the case when magnetic impurities are present, they interact with the conduction electrons and remain in thermal equilibrium at β_e . It is well known that magnetic impurities enhance the relaxation rate of neighboring nuclear spins in metals.^{10,11} Therefore to define a unique spin temperature in the presence of magnetic impurities, rapid spin diffusion is necessary among nuclear spins.¹⁰ Hereafter we confine ourselves to this case. Then, the effect of magnetic impurities appears via $\text{Im} \chi_{i\alpha i\alpha}^{(e)}(\omega)$ which acts equally at $T > 0$ and $T < 0$ as can be seen from Eq. (9). The difference in relaxation rate between $T > 0$ and $T < 0$ must then come from the second factor in Eq. (9) which consists of the correlation function of nuclear spins. We consider this in the following.

As it can be shown rigorously by returning to the Lehmann representation that

$$\sum_{\alpha=xyz} \int_{-\infty}^{\infty} \hbar \omega \Phi_{i\alpha}(\omega) d\omega = -\frac{1}{\hbar} \langle [H, \mathbf{I}_i] \mathbf{I}_i \rangle_{\beta}, \quad (11)$$

we obtain from Eq. (9)

$$\frac{dQ}{dt} = \frac{1}{\tau_0} \sum_i [\langle [H, \mathbf{I}_i] \mathbf{I}_i \rangle_{\beta} - \langle [H, \mathbf{I}_i] \mathbf{I}_i \rangle_{\beta_e}], \quad (12)$$

where we have defined

$$\frac{1}{\tau_0} = \frac{2A^2}{\hbar \beta_e} \lim_{\omega \rightarrow 0} \frac{1}{N} \sum_i \frac{\text{Im} \chi_{ii}^{(e)}(\omega)}{\hbar \omega}. \quad (13)$$

For a noninteracting electron gas, Eq. (13) turns into the Korringa relaxation rate $1/\tau_0 = \pi A^2 \rho_F^2 k_B T / \hbar$ using Eq. (10).

The nuclear spin Hamiltonian is of the form

$$H = H_{int} + H_z, \quad (14)$$

where H_{int} consists of the Ruderman-Kittel interaction, expressed as

$$H_{ex} = - \sum_{(i,j)} J_{ij} \mathbf{I}_i \cdot \mathbf{I}_j, \quad (15)$$

and the dipole-dipole interaction between the nuclear spins. The Zeeman energy H_z in the presence of an external field H_0 is given by

$$H_z = -\hbar \gamma H_0 \sum_i I_{iz}. \quad (16)$$

It holds that

$$\sum_i [H_{int}, \mathbf{I}_i] \cdot \mathbf{I}_i = 2H_{int}, \quad (17)$$

and

$$\sum_i [H_z, \mathbf{I}_i] \cdot \mathbf{I}_i = H_z. \quad (18)$$

Inserting Eqs. (17) and (18) into Eq. (12), we obtain

$$\frac{dQ}{dt} = \frac{1}{\tau_0} (2\Delta E_{int} + \Delta E_z), \quad (19)$$

where $\Delta E_{int} = E_{int}(\beta) - E_{int}(\beta_e)$, with $E_{int}(\beta) = \langle H_{int} \rangle_\beta$ and ΔE_z is defined in a similar way. On the other hand, by definition we have

$$\frac{dQ}{dt} = -\frac{d}{dt} (E_{int} + E_z). \quad (20)$$

Combination of Eqs. (19) and (20) yields

$$\frac{d}{dt} (E_{int} + E_z) = -\frac{1}{\tau_0} (2\Delta E_{int} + \Delta E_z). \quad (21)$$

By the assumption that the nuclear spin system is in internal equilibrium at β , E_{int} and E_z are expressed in terms of β . Therefore, we rewrite Eq. (21) in the form

$$\frac{d\beta}{dt} = -\frac{1}{\tau_0} \frac{2\Delta E_{int} + \Delta E_z}{\frac{d}{d\beta} (E_{int} + E_z)}, \quad (22)$$

which determines the relaxation rate of β . Equation (22) is our central result which tells that the inverse temperature approaches the equilibrium in proportion to the differences of interaction and Zeeman energies from their equilibrium values and inversely proportional to the specific heat. In the actual experimental situation, $\beta_e/\beta \approx 10^{-5}$ and, moreover, E decreases linearly with β at high temperatures. Therefore $E_{int,z}(\beta_e) \ll E_{int,z}(\beta)$ and $\Delta E_{int,z}$ can be replaced with $E_{int,z}$ in Eq. (22). In order to integrate Eq. (22), one must know explicitly $E_{int}(\beta)$ and $E_z(\beta)$ as functions of β . For simplicity we discard the dipole-dipole interaction hereafter, so that $E_{int}(\beta)$ is replaced by $E_{ex}(\beta)$.

III. EVALUATION OF THE RELAXATION RATE

In the high-temperature limit, $E_{ex} = -\beta \text{Tr} H_{ex}^2$ and $E_z = -\beta \text{Tr} H_z^2$. Then Eq. (20) is easily solved to give $\beta - \beta_e = (\beta_i - \beta_e) \exp(-t/\tau)$ with

$$\tau^{-1} = \tau_0^{-1} (2 \text{Tr} H_{ex}^2 + \text{Tr} H_z^2) / (\text{Tr} H_{ex}^2 + \text{Tr} H_z^2). \quad (23)$$

This is a well-known result,⁹ where τ^{-1} is independent of β so that no difference appears between $\beta > 0$ and $\beta < 0$. However, when we include the first-order correction

$$E_{ex} = -\beta \text{Tr} H_{ex}^2 + \frac{1}{2} \beta^2 \text{Tr} (H_{ex}^3 + H_{ex} H_z^2), \quad (24)$$

$$E_z = -\beta \text{Tr} H_z^2 + \beta^2 \text{Tr} H_{ex} H_z^2, \quad (25)$$

Eq. (22) becomes

$$\begin{aligned} \frac{d\beta}{dt} = & -\frac{\beta}{\tau_0} \frac{(2 \text{Tr} H_{ex}^2 + \text{Tr} H_z^2)}{(\text{Tr} H_{ex}^2 + \text{Tr} H_z^2)} \\ & \times \left[1 + \beta \frac{(\text{Tr} H_{ex}^3 + 2 \text{Tr} H_{ex} H_z^2) \text{Tr} H_{ex}^2}{(2 \text{Tr} H_{ex}^2 + \text{Tr} H_z^2)(\text{Tr} H_{ex}^2 + \text{Tr} H_z^2)} \right]. \end{aligned} \quad (26)$$

It is obvious from Eq. (26) that β no longer shows exponential decay and that a difference in the relaxation rate between $\beta > 0$ and $\beta < 0$ appears. Furthermore, in vanishing field the last factor on the r.h.s. of Eq. (26) turns to $[1 + (\beta/2) \text{Tr} H_{ex}^3 / \text{Tr} H_{ex}^2]$, which grows with increasing $\beta > 0$ for AF exchange interaction ($J_{ij} < 0$) since $\text{Tr} H_{ex}^3 > 0$. The reverse holds for F exchange interaction ($J_{ij} > 0$). Conversely, for $\beta < 0$ the above relations are reversed. For strong fields, on the other hand, the last factor on the r.h.s. of Eq. (26) becomes unity, so that the relaxation remains just as in the high-temperature limit. We next discuss in general terms the roles of the exchange and Zeeman energies.

First, if the exchange interaction can be neglected, it holds that $E_z(\beta) = -N\hbar \gamma H_0 / 2 \tanh(\beta\hbar \gamma H_0 / 2)$ for $I = 1/2$. Inserting this into Eq. (22), we obtain $d\beta/dt = -(1/\tau_0 \hbar \gamma H_0) \sinh(\beta\hbar \gamma H_0)$ which is integrated as $\tanh(\beta\hbar \gamma H_0 / 2) = c \exp(-t/\tau_0)$, ($c = \text{constant}$). This shows that relaxation takes place equally for $\beta > 0$ and $\beta < 0$. Second, if the external field is absent, Eq. (22) turns to

$$\frac{d\beta}{dt} = -\frac{2}{\tau_0} \frac{1}{\frac{d}{d\beta} \log |E_{ex}|}. \quad (27)$$

That is, the β dependence of $E_{ex}(\beta)$ determines fully the relaxation rate. The steeper is the change of $E_{ex}(\beta)$, the slower is the relaxation rate. The rate is no longer symmetric with respect to $\beta = 0$ in contrast to the case of the Zeeman energy and this appears via the energy spectrum of the exchange interaction. Let $\rho(E)$ be the density of states due to the exchange interaction. Then, from the expression $E_{ex}(\beta) = \int E \rho(E) \exp(-\beta E) dE / \int \rho(E) \exp(-\beta E) dE$, it becomes clear that the larger the density of states is at positive high (negative low) energy, the steeper is $|E_{ex}|$ at $\beta < 0$ ($\beta > 0$). This can be seen to be the case using the results of Monte Carlo simulations as discussed below. With the change of the sign of exchange interaction, the structure of the energy spectrum reverses around $E = 0$ and so the relaxation rate at $\beta > 0$ is replaced with that at $\beta < 0$. Equation (27) tells also about the critical behavior at the nuclear ordering temperature T_C ($\beta_C = 1/k_B T_C$). Let us suppose that $E_{ex} \sim (\beta - \beta_C)^{-a}$ near T_C . Then the r.h.s. of Eq. (27) varies proportionally to $(\beta - \beta_C)$, which shows critical slowing down of the relaxation time. To obtain semiquantitative understanding of the experimental results⁷ we next proceed with estimations based on Monte Carlo simulation and on high-temperature expansion.

IV. APPLICATION TO RHODIUM SPINS

Rhodium, as well as silver, has $I = 1/2$ and face-centered-cubic lattice. Following the model for rhodium,⁶ we replace the Ruderman-Kittel interaction with the nearest- and next-nearest-neighbor interactions $J_{NN}/h = -17.1$ Hz and

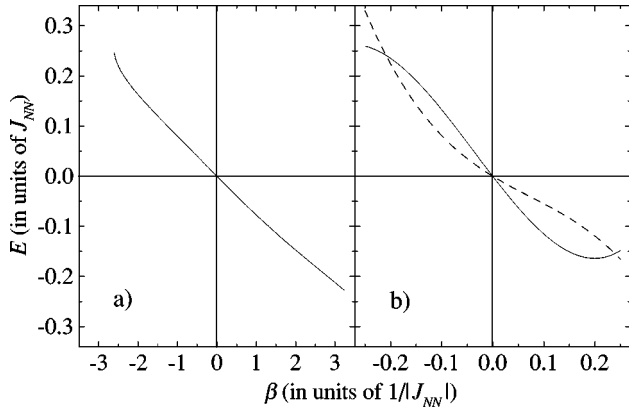


FIG. 1. Energy as a function of inverse temperature β calculated using (a) Monte Carlo simulation at $H_0=0$ (Ref. 13) and (b) high- T expansion at $H_0=20 \mu\text{T}$ (see text for details). Exchange and Zeeman energies are shown by solid and dashed lines, respectively.

$J_{NNN}/h=9.8 \text{ Hz}$, respectively. For this system microcanonical Monte Carlo simulations have been performed to calculate the density of states $\rho(E)$ and the entropy $S(E) \propto \ln \rho(E)$ in zero field by treating the spins as classical.¹³ Figure 1 of Ref. 13 shows the asymmetry with respect to $E=0$ in $\rho(E)$, which is somewhat extended towards the positive, high-energy side. Using this $S(E)$, we have calculated $E_{ex}(\beta)$ which is shown in Fig. 1(a). The asymmetry in $\rho(E)$ between $\beta>0$ and $\beta<0$ is reflected in the energy as well. Note that the slope $dE_{ex}/d\beta$ at $\beta=0$ differs from a quantum-mechanical high- T expansion by an order of magnitude, which is due to the classical treatment of spins having $I=1/2$. By using this $E_{ex}(\beta)$, Eq. (27) is integrated and the resulting $\beta(t)$ is shown in Fig. 2(a). Relaxation at $\beta<0$ is clearly slower than at $\beta>0$. As the employed Monte Carlo simulation treats the system as classical spins and is limited to the case of vanishing field, we next apply the method of high-temperature expansion.^{14,15}

Elaborate calculation of the high-temperature expansion

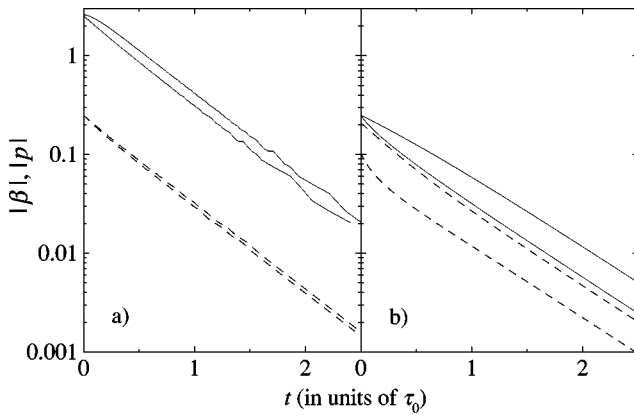


FIG. 2. (a) Relaxation of inverse spin temperature β as a function of time at $H_0=0$ obtained from Eq. (22) using the energy from Monte Carlo simulation (solid line) and high- T expansion (dashed line). In the latter, the initial values for β have been chosen so that the expansion remains convergent. (b) Inverse temperature (solid curve) and spin polarization p (dashed curve) as functions of time calculated using high- T expansion at $H_0=20 \mu\text{T}$. In all cases, the upper and lower traces refer to $\beta<0$ and $\beta>0$, respectively.

with nearest- and next-nearest-neighbor interactions has been made for the susceptibility and the zero-field specific heat up to sixth order in β .¹⁵ However, to evaluate Eq. (22) in the presence of an applied field, we must know the susceptibility and the specific heat in a finite applied field. We made an expansion of $E_{int}(\beta)$ and $E_z(\beta)$ up to third order in β which contains the term of H_0^4 in $E_z(\beta)$. As the high-temperature expansion is valid for βJ_{NN} , $\beta g \mu H_0 \ll 1$, it is difficult to compare directly with the experiments done at $H_0=40 \mu\text{T}$ and $T \sim 1 \text{ nK}$ ($=20.8 \text{ Hz} \cdot h$) since this field corresponds to $\gamma H_0/2\pi = 53.6 \text{ Hz}$ in Rh ($\gamma/2\pi = 1.34 \text{ MHz/T}$).⁷ The calculated results for E_{ex} and E_z at $20 \mu\text{T}$ are presented in Fig. 1(b). It can be seen from Fig. 1(b) that $E_{ex}(\beta)$ varies steeper at $\beta<0$ than at $\beta>0$. Using these values of E_{ex} and E_z , we integrate Eq. (22) to obtain the time dependence of β and the nuclear spin polarization $\langle I_z \rangle$, which are displayed in Fig. 2(b). Here we have assumed that the initial β at $t=0$ is ± 0.25 in the units of $|J_{NN}|^{-1}$, in which the critical value is known as $\beta_c=0.498$ for the model with J_{NN} only.¹⁴ Although $\langle I_z \rangle$ is proportional to β in the high-temperature limit, nonlinearity appears with increasing $|\beta|$. As a result, a difference in behavior is seen between β and $\langle I_z \rangle$, as well as in the initial values of $\langle I_z \rangle$ for $\beta = \pm 0.25$. Certainly, one can see both $\beta(t)$ and $\langle I_z(t) \rangle$ to relax slower at $\beta<0$ than at $\beta>0$. This behavior of $\langle I_z \rangle$ is consistent with the experimental result.⁷ For a detailed comparison with the experimental results, the calculation should be done at the experimental value $H_0=40 \mu\text{T}$. However, such an attempt displayed unphysical behavior in the time dependence of $\beta(t)$ in the third-order approximation. It is therefore necessary to go to higher order in the high-temperature expansion, or to use more accurate results for $E_{ex}(\beta)$ and $E_z(\beta)$.

Nuclear spin relaxation at ultralow temperatures has recently been studied in silver by Tuoriniemi *et al.*¹⁶ using neutron transmission techniques. They found that the relaxation time τ depends on nuclear entropy. In zero field and at high entropies, i.e., at high temperatures, the experiment yields $\tau^{-1} = (2.2 \pm 0.5) \tau_0^{-1}$. However, at lower entropies ($S < 0.8 R \ln 2$), $\tau^{-1} = (2.9 \pm 0.2) \tau_0^{-1}$, i.e., the relaxation is considerably faster. Moreover, they observed that the characteristic field at which τ crosses over from low- to high-field regions at small entropies is larger by a factor of about three than that given by Eq. (23). Concerning the low-field relaxation, as lower temperatures correspond to smaller entropies, we have found a qualitative agreement with this experimental result since the exchange interaction is antiferromagnetic in Ag. For further comparison with the experimental result, the exchange and Zeeman energies should be calculated including the ordered state.

V. CONCLUSIONS

In conclusion, to clarify the anomaly that nuclear relaxation at negative temperatures is slower than at positive temperatures, we have derived a formula for the relaxation rate of β . It consists of a product of factors, one of which is governed by the exchange and Zeeman energies for the nuclear spins while the other one is represented by the imaginary part of the conduction electron susceptibility which does also include the effect of magnetic impurities. The former depends on the nuclear spin temperature, in particu-

lar, whether the system is at $\beta > 0$ or $\beta < 0$. Since the AF-dominated exchange interaction increases the density of states at positive energy, and thus enhances $d \ln |E_{ex}| / d\beta$ at $\beta < 0$, it makes relaxation slow at $\beta < 0$ (while the reverse holds for the F-dominated interaction). That is, when $\beta < 0$, the positive energy states contribute to the increase of E_{ex} and suppress the relaxation rate; this just corresponds to the fact that the free energy F is maximized at $\beta < 0$. Critical slowing down has been predicted from the theory, which shows the relaxation rate to turn proportional to $(\beta - \beta_C)$ when β passes through the nuclear ordering temperature T_C ($\beta_C = 1/k_B T_C$). Regarding the effect of magnetic impurities, they act to enhance the relaxation rate equally at $\beta < 0$ and $\beta > 0$ and, therefore, do not affect the difference in the relaxation rate between $T < 0$ and $T > 0$. We have applied the results of Monte Carlo simulations with classical spins as well as high-temperature expansions up to third order in β to

the derived formula and found a qualitative agreement with the experimental results. For a more quantitative comparison, further improvement is necessary in the evaluation of the exchange and Zeeman energies.

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