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DYNAMIC SPIN RELAXATION PROCESSES OF μ^+ IN AN ANTIFERROMAGNET

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A new mechanism for positive muon spin-lattice relaxation (SLR) in an antiferromagnet is described. The temperature dependence of the relaxation rate ($1/T_1$) is calculated in a simplified case, and SLR of muon in MnF_2 is studied in the context of this model.

Whereas nuclear spin-lattice relaxation in an antiferromagnet (AFM) with localized spins is well understood, the study of muon spin relaxation in that environment is yet to be completed. The lack of attention to muon SLR is due to the assumption that both nuclear and muon spin, in an AFM, relax by the same mechanism. The possible existence of an additional source of muon SLR will be examined in this paper. As a result of this mechanism, the muon spin relaxation rate ($1/T_1^\mu$) is expected to exhibit a temperature dependence that is different from that of the spin of the nuclei ($1/T_1^N$). This model is compared with measurements in MnF_2.

The fact that the nuclear SLR mechanism, at temperatures much smaller than the Néel temperature T_N, is dominated by magnon Raman scattering (MRS) was predicted by Van Kranendonk *et al.* [1]. They argued that at such temperatures the phonon contribution to SLR can be ignored, provided that T_N is smaller than the Debye temperature Θ_D. Direct absorption can also be ignored because the typical energy carried by a magnon is much larger than the nuclear Zeeman splitting. An explicit calculation carried out by Moriya supports this assumption [2]. It was later demonstrated quantitatively by Kaplan *et al.* that the MRS model can explain very well the relaxation rate of ^{19}F spin in MnF_2 [3].

However, the implanted muon is coupled to its neighbors in a different manner than a nucleus, and has a much smaller mass. This can result in vibration frequencies of the μ^+ different from those of the nuclei, which in turn allows additional sources of $1/T_1$ to become important. The relaxation mechanism pursued in this paper applies in cases where the muon vibration energy levels' spacings are of the same order of magnitude as the energy carried by a magnon. In these cases muon SLR could proceed via the absorption of a magnon, excitation of the muon to a higher vibrational level and a spin flip. This excitation-assisted spin flip process will coexist with the previously studied MRS process.

When a narrow field distribution at the muon site (at $T \to 0$ in an AFM) is observed, one can assume that there is a muon bound state. An estimate of the energy scale of the muon vibration energy levels in a particular compound can be obtained from muon diffusion. For example, thermally activated muon diffusion with an activation energy of $E_a = 0.39(1)$ eV was recently observed in the ordered state of the so-called Infinite Layer AFM $Ca_{0.86}Sr_{0.14}CuO_2$ [4]. The vibrational energy spacings are only a fraction of E_a, and can be estimated by $\sqrt{2\hbar^2 E_a/mL_{eff}}$, where m =105.66 MeV/c^2 is the muon mass and L_{eff} is some effective length scale of the potential well. For $L_{eff} = 3$ Å (the lattice parameters are $a = 3.8$ and $c = 3.2$ Å) the energy spacings are on the order of 0.05 eV. It so happens that the Néel temperature of this compound is also 0.05 eV (540 K) which is the expected energy scale for magnons.

The model used here to account for the temperature dependence of $1/T_1^\mu$ assumes that the muon, after entering the sample, is trapped instantaneously in a crystallographically unique electrostatic potential well. Once the μ^+ is in this well, the Hamiltonian of the system is given by $\mathcal{H} = \mathcal{H}_{ss} + \mathcal{H}_{mag} + \mathcal{H}_v$ where $\mathcal{H}_{ss}$ describes the AFM spin system, $\mathcal{H}_{mag}$ describes the magnetic interaction between the muon spin and the AFM ionic moment, and the electrostatic interaction of the muon is given by $\mathcal{H}_v$. The magnetic Hamiltonian $\mathcal{H}_{mag}$ can be written in the most general case as

$$\mathcal{H}_{mag} = -\sum_l \sum_{\alpha\beta} \mu^\alpha A_l^{\alpha\beta}(\vec{r}) S_l^\beta \tag{1}$$

where S is the spin of the local moments of the AFM, and $\vec{\mu} = \frac{1}{2}\hbar\gamma_\mu\vec{\sigma}$ is the magnetic moment of the muon; $\vec{\sigma}$ are the Pauli spinors. The Greek letters take the values $(+, -, z)$, and l is taken over the nearest neighbors of the muon. The matrix A depends on the displacement $\vec{r}$ of the μ^+ from its equilibrium position. It is this dependence which couples the magnons to the vibrational levels of the μ^+ in its well.

In order to simplify the problem, the quantization axis of the AFM electronic spins, the direction of the field at the μ^+ site, and the initial muon polarization are taken to align with the $\hat{c}$ axis (as in MnF_2) [5]. The relaxation processes are accounted for by two expansions: A_l is expanded in terms of r around $r = 0$; $\vec{S}_l$ is expressed in terms of creation ($M_{\vec{k}p}^+$) and annihilation ($M_{\vec{k}p}$) operators of magnons, as described in spin wave theory [6]; $\vec{k}$ and p are the wave vector and polarization of the magnon. These expansions give rise to various terms. All terms contributing to the static local field are represented by the observed internal field $\vec{B}_{loc}$. Other terms induce the direct absorption, the MRS, and the excitation assisted spin flip processes. Since the first two processes have been examined elsewhere, only the latter will be considered here.

The model is further simplified by the following two assumptions: the potential well is taken to be isotropic, and the muon falls down to its vibrational ground state before its spin relaxes. The second assumption is a consequence of the short stopping time 10^{-10} sec, and the expected population of higher vibrational energy levels. Since in our model these energy levels are on the order of T_N, at $T < T_N$ the population of the higher level decreases as $\exp(-T_N/T)$ with decreasing temperature. The focus is therefore on the terms leading to transitions between an initial state of a μ^+ in the vibrational ground state, up spin, and a magnon configuration $\{n(\vec{k},p)\}$, and a final state with the μ^+ in the first excited vibrational state, down spin, and one less magnon.

If ρ_m^+ is the raising operator of the vibrational levels in the m'th spatial direction, then the magnetic Hamiltonian is given by

$$\mathcal{H}_{mag} = -\vec{\mu} \cdot \vec{B}_{loc} + \sum_{p,m} \sum_{k} d_{pm}(\vec{k}) \sigma^- \rho_m^+ M_{\vec{k}p}, \tag{2}$$

where

$$d_{1m}(\vec{k}) = \hbar\gamma_\mu \left(\frac{s\hbar}{2m\omega_v N}\right)^{1/2} \left(v_k \sum_i \frac{dA_i^{--}}{dr^m} e^{ikR_i} + u_k \sum_j \frac{dA_j^{--}}{dr^m} e^{-ikR_j} \right)$$

$$d_{2m}(\vec{k}) = \hbar\gamma_\mu \left(\frac{s\hbar}{2m\omega_v N}\right)^{1/2} \left(u_k \sum_i \frac{dA_i^{-+}}{dr^m} e^{-ikR_i} + v_k \sum_j \frac{dA_j^{-+}}{dr^m} e^{ikR_j} \right).$$

The functions u_k and v_k are given in Ref. [6], $\hbar\omega_v$ is the energy difference between the ground state and the first excited state of $\mathcal{H}_v$, and the indexes i and j are taken over the different sub-lattices. In principle, the local field $\vec{B}_{loc}$ will cause an energy splitting between the up spin and down spin states, which will result in different relaxation rates on the two sub-lattices. However, the splitting (~ 0.01 K) is much smaller than the typical magnon energy and can be ignored.

The relaxation rate is evaluated by the thermally weighted Fermi golden rule. It is easy to see that the temperature dependence comes only from the average number of magnons ($\langle n \rangle$) with energy $\hbar\omega_v$. The relaxation rate is given explicitly by

$$\frac{1}{T_1} = \frac{2\pi}{\hbar} D\langle n(\hbar\omega_v)\rangle, \qquad D = \frac{V}{(2\pi)^3} \sum_{p,m} \int \mathrm{d}^3k \mid d_{pm}(\vec{k}) \mid^2 \delta(E_p(\vec{k}) - \hbar\omega_v), \quad (3)$$

and $\langle n(E)\rangle = 1/(\exp(E/k_BT) - 1)$; $E_p(\vec{k})$ is the energy of a magnon with wave vector $\vec{k}$ and polarization p.

The order of magnitude of this mechanism can be estimated at low temperatures, $T \ll T_N$, by the long wavelength approximation. In the $k \to 0$ limit of an ideal Heisenberg antiferromagnet, the following relations are valid [1]:

$$E(\vec{k}) = \theta_N bk, \text{ and } \sum_{p,m} \mid d_{pm}(k) \mid^2 = \frac{s\gamma^2\hbar^2}{2Nbk} F^2 \qquad (4)$$

where θ_N is of the order of k_BT_N. b depends only on the geometry of the lattice, and is equal to $\sqrt{1/3}a$ for the three cubic structures; a is the nearest neighbors' separation. The factor $F \sim (\hbar/2m\omega_v)^{1/2}(dA/dr)$ has units of magnetic field, and is of the order of the field fluctuations seen by the muon as it oscillates in the well. F can be estimated from the observed static local field B_{loc}. The relaxation rate calculated from Eq. 3 at the long wave length limit obeys an Arrhenius law

$$\frac{1}{T_1} = \frac{Sv}{2\pi b^3} \left(\frac{\hbar\omega_0}{\theta_N}\right)^2 \omega_v \exp(-\hbar\omega_v/k_BT), \qquad (5)$$

where $\omega_0 = \gamma_\mu F$, and v is the volume of the cell. This result is in contrast to the T^3 law at low temperatures of the MRS [1]. The excitation-assisted spin flip relaxation mechanism will be important when $\hbar\omega_0/\theta_N$ is large, and the signature of this mechanism is an activation energy of the order of the Néel temperature.

The difference between the spin-lattice relaxation rates of μ^+ [5] and ^{19}F [3] in MnF_2 was studied in an attempt to demonstrate the existence of the excitation-assisted spin flip process in a physical system. Although it was not possible to demonstrate the existence of this process in MnF_2, the results of the study are shown in the rest of this paper as an example of the application of this model to a real system. MnF_2 was chosen for the following reasons:

a) In MnF_2 the muon has two distinct relaxation rates, one much faster than

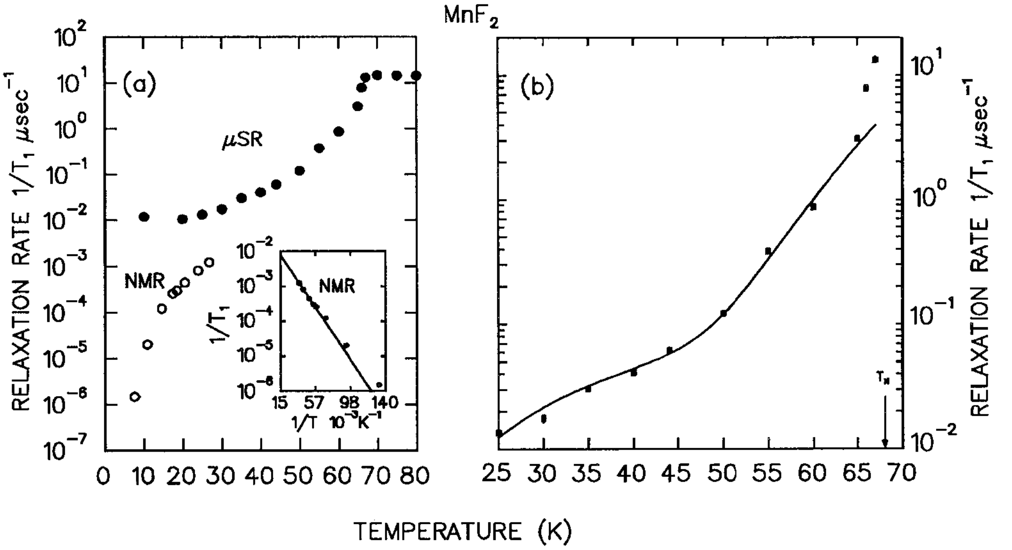

Fig. 1. (a) μSR and NMR relaxation rate in MnF_2. (b) The μ^+ relaxation rate is fitted to Eq. 6.

the other [5]. The fast $1/T_1^{\mu}$ and $1/T_1^N$ of ^{19}F , as a function of temperature, are shown in Fig. 1a. As the temperature increases towards T_N, $1/T_1^N$ seems to curve down on the logarithmic scale while $1/T_1^{\mu}$ curves up. This discrepancy between the two relaxation rates might be due to an additional source of muon spin relaxation mechanism. It should be pointed out that the discrepancy is only based on extrapolation of the NMR data, and it is not clear how does $1/T_1^N$ depend on the temperature closer to T_N.
b) A very good agreement between the NMR data and the magnon Raman scattering theory was achieved [3], and the contribution from MRS can be easily accounted for.
c) The local field, at the muon site with fast relaxation, is 94 kG ($\omega_0 = 2\pi\times1.3$ GHz) [5]. If the assumption $\theta_N \sim \hbar\omega_v$ is valid, the expected relaxation rate is of the order of 1 μsec^{-1} at $T \sim \theta_N$. This is roughly the observed rate.

The NMR data is used to represent the MRS contribution and all the other possible sources of dynamics originated in the AFM system. The NMR data is extrapolated to higher temperatures using the line in the insert of Fig. 1a. $1/T_1^N$ is then scaled up by the ratio of the interaction strength of the two different particles. This ratio is given roughly by ω_μ^2/ω_N^2, where ω_μ and ω_N are the fields seen by the μ^+ and ^{19}F respectively. We then fit the

μSR data with the formula

$$\frac{1}{T_1^\mu}(T) = c_1 \frac{\omega_\mu^2}{\omega_N^2} \frac{1}{T_1^N}(T) + c_2 \langle n(E) \rangle \quad (6)$$

between 25 and 67 K, as shown in Fig. 1b. The best fit obtains for $c_1 = 0.80(1)$ and $c_2 = 1.0(2) \times 10^6$ μsec^{-1}. The fit agrees with the data up to 65 K. However, the activation energy is found to be 0.072(1) meV. Since no magnon with such high energy exists in MnF_2 [7], the model is not applicable in this case.

In conclusion it is suggested that the vibration of μ^+ in AFM can cause significant field fluctuations and provide a strong mechanism for spin relaxation. The relaxation rate $1/T_1^\mu$, caused by the absorption of a magnon by exciting the muon from its vibrational ground state to the first excited state, accompanied by a spin flip, is expected to follow an Arrhenius law at low temperatures. The activation energy is equal to the vibrational energy spacing. It was demonstrated that when the extrapolated MRS relaxation rate is subtracted from the muon spin relaxation rate in MnF_2, the residue is activated with $E = 0.072(1)$ meV.

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