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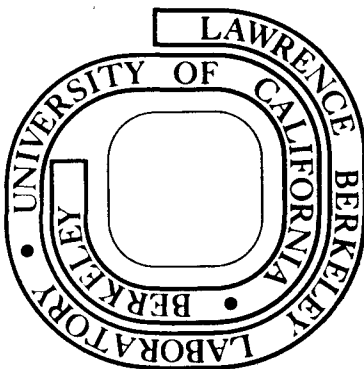
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$\mu^+$  STUDIES OF CRITICAL SPIN FLUCTUATIONS IN Ni\*

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## ABSTRACT

Critical fluctuations above the Curie point in nickel have been studied by observing the relaxation of the free precession of positive muons. This work was prompted by the apparent discrepancy in nickel spin correlation time as determined by neutron scattering and by perturbed angular correlation (PAC) experiments. Up to 20K above the Curie point we obtain a temperature dependence in agreement with that determined from PAC. At higher temperatures the computed nickel spin correlation time becomes independent of temperature. The observed high temperature muon relaxation time of 16  $\mu\text{sec}$  is very much shorter than expected from either the observed contact field at the low temperature muon site or from classical dipolar coupling to the muon nearest neighbors. The increased relaxation rate may arise from an enhanced pseudodipolar interaction.

## INTRODUCTION

In this communication, we report the observation of critical spin fluctuations in nickel using positive muons. The implanted positive muon, as has been described in the literature,<sup>1,2,3</sup> is a convenient probe of local magnetic fields in solids in general and in magnetic materials in particular. In Ni at temperatures just above the Curie temperature ( $T_C = 630\text{K}$ ), the muon spin is relaxed by spin fluctuations; our experiment consists of measuring the relaxation time  $T_2$  as a function of  $(T-T_C)$  and externally applied magnetic field (see Fig. 1). In most of what follows, it will be assumed that the site of the implanted muon is the octahedral interstitial site in the fcc Ni lattice as is indicated from studies below  $T_C$ .<sup>2,3</sup>

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RESULTS AND DISCUSSION

For values of  $(T-T_c)$  greater than 70K, we find that  $1/T_2$  is approximately constant and has the value of  $\sim 10^5 \text{ sec}^{-1}$  (see Fig. 1). Microwave studies have established<sup>4</sup> that at these temperatures effectively all correlation between Ni spins is destroyed and the spins fluctuate with a correlation time  $\tau_c \sim 1/\omega_e \sim \hbar/J \sim 2 \times 10^{-14} \text{ sec}$ . Here  $\omega_e$  is the exchange frequency and J is the exchange energy. If each of N mutually uncorrelated spins produces a magnetic field of strength  $\omega/\gamma_\mu$  at the  $\mu^+$ , fluctuations of correlation time  $\tau_c$  will relax the  $\mu^+$  at a rate given approximately by:

$$\frac{1}{T_2} \approx N\omega^2\tau_c \quad (1)$$

for  $\omega\tau_c \ll 1$ . Two magnetic interactions responsible for  $\omega$  suggest themselves immediately: (1) the isotropic contact interaction of the  $\mu^+$  with its screening cloud of (polarized) electrons, and (2) the classical dipolar interaction with the neighboring Ni cores.

The strength of the contact interaction may be inferred from the measured<sup>2</sup> hyperfine field at the  $\mu^+$  in the ordered state,  $B_{hf} = -.66\text{kG}$ . The octahedral site has N=6 nearest neighbors, and we make the assumption that they share equally in the interaction, each contributing  $-.11\text{kG}$ . Thus,

$$\omega_{\text{cont}} = \frac{\gamma_\mu B_{hf}}{N} \approx 10^7 \text{ sec}^{-1}, \quad (2)$$

where  $\gamma_\mu$  is the gyromagnetic ratio of the muon. This implies a relaxation rate

$$\frac{1}{T_2}_{\text{cont}} \approx N\omega_{\text{cont}}^2\tau_c \approx 10 \text{ sec}^{-1}, \quad (3)$$

which is well below the observed rate. Clearly, the isotropic contact interaction is too weak to account for the observed relaxation.

Although the dipolar fields from neighboring Ni cores vanish by symmetry at the octahedral site in the ordered state, dipolar fields from fluctuating spins can cause relaxation when cubic symmetry is destroyed by the disappearance of spin correlation. In this case,  $\omega$  is given by (for nearest neighbors):

$$\omega_{\text{dip}} = \frac{\gamma \mu_{\text{Ni}}}{(a/2)^3} \approx 10^8 \text{ sec}^{-1}, \quad (4)$$

where  $\mu_{\text{Ni}} = .6 \mu_{\text{B}}$  is the magnetic moment of a Ni core and  $a$  is the lattice constant. Since  $\omega_{\text{dip}}^2$  for neighbors further removed from the  $\mu^+$  drops off rapidly with distance, we consider only the nearest neighbor contribution:

$$\frac{1}{T_2} \approx N \omega_{\text{dip}}^2 \tau_c \approx 10^3 \text{ sec}^{-1} \quad (5)$$

which is too small by a factor of  $\sim 100$ . This implies that the dipolar interaction is too weak by a factor of 10.

It is often the case that in addition to the isotropic contact interaction between the spins of a magnetic material, there exists a contact interaction with dipolar symmetry. This "pseudo-dipolar" enhancement of the classical dipolar field is known<sup>4</sup> to be the source of the ferromagnetic resonance line-width and the magnetic anisotropy in Ni. We believe that pseudo-dipolar enhancement by a factor of 10 may not be unreasonable for the case of  $\mu^+$  in Ni.

Figure 2 compares the muon data with existing<sup>5</sup> perturbed angular correlation and neutron scattering data. From each experiment, a spin correlation time  $\tau_c$  is extracted and plotted versus  $(T-T_c)/T_c$ . For the PAC and muon techniques,  $\tau_c$  is derived from  $T_2$  via the relation  $1/T_2 \propto \omega^2 \tau_c$ . The known (hyperfine) interaction strength is used for PAC data, and for purposes of illustration, the dipolar field (without pseudo-dipolar enhancement) is used for the muon data. The extraction of  $\tau_c$  from the neutron data is more involved and requires fairly restrictive assumptions<sup>5</sup>. We see that for all three probes, as the temperature is lowered toward  $T_c$ , the correlation time increases according to the power law:

$$\tau_c \propto \left[ \frac{T-T_c}{T_c} \right]^{-n} \quad (6)$$

where  $n$  for PAC and  $\mu^+$  is .7 and is 1.4 for neutrons. The fact that the neutron data disagree with both the PAC and the  $\mu^+$  data may be due to a failure of the assumptions made<sup>5</sup> in extracting  $\tau_c$  from the neutron data, or it may possibly be inherent in the manner in which the different probes sample the momentum spectrum of the critical fluctuations.

As  $T$  approaches  $T_C$  several things happen: (1) the product  $\omega\tau_C$  is no longer small compared to one, and the above treatment breaks down. It is believed that this will happen only in a temperature region inaccessibly close to  $T_C$ . (2) The nearest neighbors to the muon will begin to correlate. When this occurs, the dipolar (and pseudo-dipolar) fields will cancel by symmetry, and the interaction will be solely via the weaker contact term. It is interesting to note that the shift from a predominantly dipolar interaction to a contact interaction would change the way in which the muon samples spin fluctuations. This is because the  $\mu^+$  preferentially sees fluctuations of small wave number via the contact interaction while the dipole interaction weights fluctuations of higher wave number---as they are more likely to destroy the cubic symmetry near the  $\mu^+$ .

A potential complication would arise if the muon were diffusing through the Ni lattice instead of being well localized at the octahedral site. It is known that hydrogen is diffusing rapidly in Ni at these temperatures<sup>6</sup>. Evidence so far<sup>2</sup> points to the fact that if the muon is diffusing, it spends by far the greatest amount of time in one type of site (presumably the octahedral site). The question then becomes, under what conditions does a muon diffuse a distance of the order of a fluctuation wavelength in a time short compared to the correlation time of that fluctuation. Rough considerations suggest that this criterion is difficult to satisfy, and that the motion of the muon may be ignored.

As seen in Figure 2, the effect of an external field is to increase the correlation time. This is the anticipated dependence since an external field is expected to stabilize the ordered state and hence to increase spin correlation. Since the PAC and neutron scattering experiments were performed in zero external field, a quantitative comparison with  $\mu^+$  measurements requires an extension of this work to zero field.

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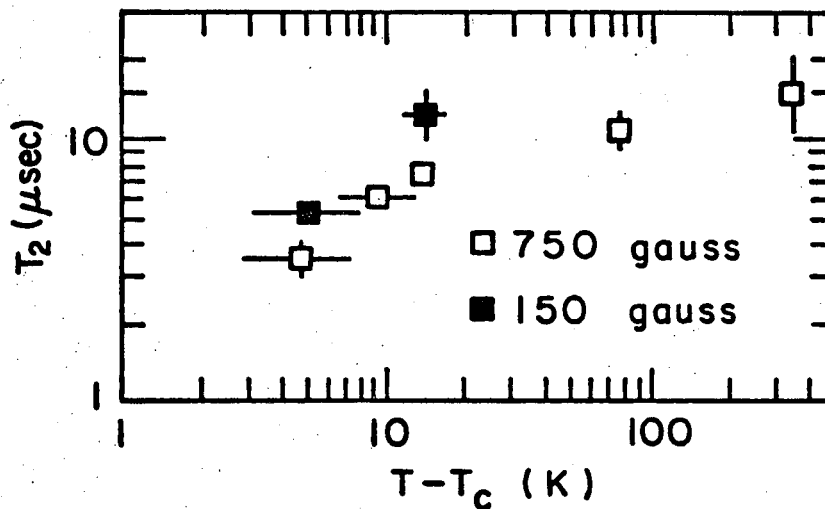


Fig. 1: The experimental dependence of  $T_2$  on temperature and external field.

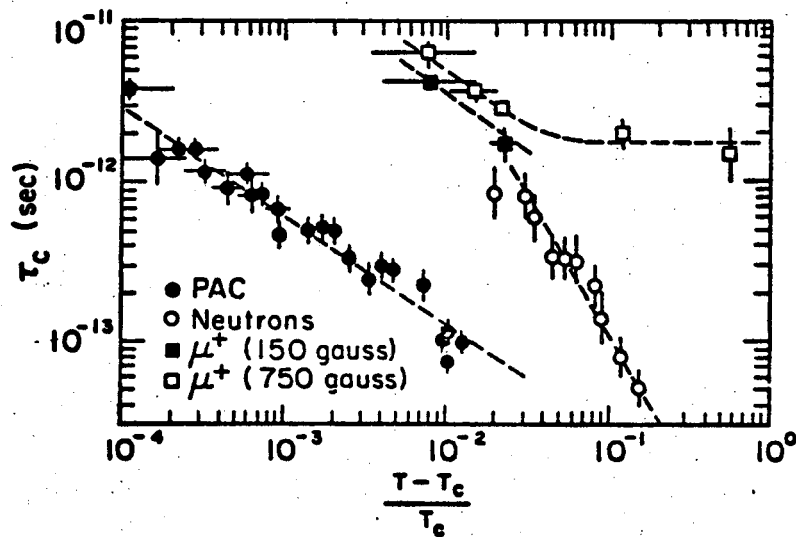


Fig. 2: A comparison of  $\tau_c$  as measured by various techniques.



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