Negative-Muon Hyperfine Anomaly in Ferromagnetic Nickel

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Negative-muon spin precession has been investigated in ferromagnetic nickel in zero applied field. A comparison of the hyperfine field for μ^-Ni with that for ^{59}Co in nickel yields a giant hyperfine anomaly of $-2.4(3)\%$ to $-2.8(5)\%$, over the temperature range 23–303 K. The results indicate that the electron spin density near the Ni nucleus decreases with increasing radial distance more steeply than the *s*-state electron charge density, and they are in good agreement with an unrestricted Dirac-Fock calculation by Freeman *et al.*

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The spatial variation of the electron spin density near an atomic nucleus can be investigated by measurement of the hyperfine field seen by different nuclear isotopes (Bohr-Weisskopf effect[1]). However, the range of nuclear radii possible with isotopes is very limited. A potentially more informative probe of the hyperfine field is a bound negative muon whose 1*s* wave function in light elements ($Z < 30$) extends far outside the nuclear surface. To the atomic electrons the muonic "pseudonucleus" appears very similar to one with nuclear charge $Z-1$. Thus, by comparing the hyperfine field seen by the muon and that seen by a $Z-1$ nucleus in the same atomic environment one can obtain new information on the spin-density distribution outside the nucleus. The situation is particularly interesting in the ferromagnetic transition metals where large hyperfine fields are present and where core polarization results in an electron spin density that does not follow the electron charge density near the nucleus.[2] In a previous experiment a large hyperfine anomaly of μ^-Pd relative to Rh in Pd was reported,[3] in disagreement with an unrestricted Dirac-Fock calculation by Freeman *et al.*[4] This suggests that the μ^-Pd was displaced from the normal lattice position as a result of recoil from the emission of a μ^-Pd x-ray. Since the hyperfine field at dilute ^{59}Co in nickel has been measured accurately with NMR, and since the above-mentioned recoil effect is expected to be small for μ^-Ni as a result of the smaller recoil energy, a measurement of the μ^-Ni hyperfine field in ferromagnetic Ni appeared attractive.

We have recently reported the observation of μ^-Ni precession near room temperature using the negative-muon spin-rotation technique.[5] In this Letter we present measurements of the μ^-Ni frequency in zero applied field at three well-defined temperatures between 23 and 303 K. Since the μ^-Ni precession frequency is approximately 1.5 GHz, a special high–timing-resolution apparatus[6] was required (Fig. 1). The experiment was performed at the Swiss Institute for Nuclear Research (SIN) with an 80%-polarized 125-MeV/*c* negative-muon beam. The muons were collimated, degraded, and then stopped in a 40-g high-purity nickel single crystal in a cryostat. A stopped muon was defined by the coincidence $BM_t\bar{F}$, and a decay electron was defined by $FE_t\bar{M}_t$ (see Fig. 1). Fast scintillator material (NE111), short light guides, fast photomultipliers (XP2020), and constant-fraction differential discriminators were used for the timing counters M_t and E_t. The timing resolution was measured to be 140 ps by use of muons which

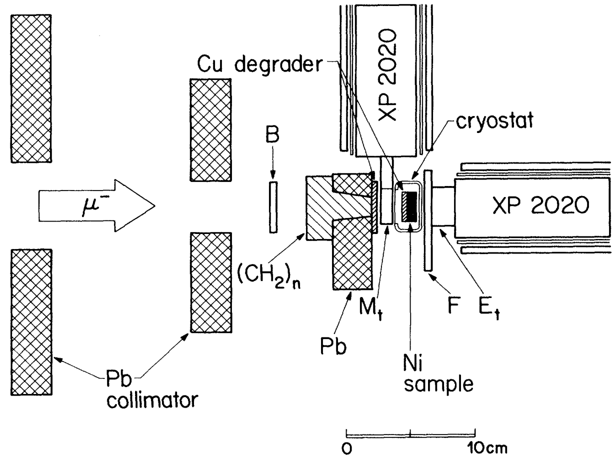

FIG. 1. The high–time-resolution muon spin-rotation apparatus. *B*, M_t, *F*, and E_t are plastic scintillation counters.

passed through all counters. The time spectrum was measured with a time-to-amplitude converter (TAC) which was periodically calibrated with a precision clock. The sample was cooled with a cold-finger cryostat, and the sample temperature was regulated to within 0.5 K. Measurements were performed at 23.1, 113.0, and 303.5 K.

A frequency analysis of the time spectra between 27 and 155 ns is shown in Fig. 2. Prominent μ^-Ni precession signals are evident at all three temperatures, and the frequency decreases with increasing temperature as expected from the ^{59}Co NMR. After correction for a small nonlinearity in the TAC time spectrum, the data were fitted by the function

$$N(t)=N_0\{\exp(-t/\tau_\mu)[1+A_\mu\exp(-\lambda_\mu t)\cos(2\pi f_\mu t+\phi)]+B\},$$

where N_0 is the normalization, τ_μ is the μ^-Ni lifetime, A_μ is the initial precession amplitude, λ_μ is the muon depolarization rate, f_μ is the μ^-Ni frequency, ϕ is the initial phase, and B is a small time-dependent background. The negative-muon lifetime τ_μ is shorter than the 2.2-μs free-muon lifetime as a result of nuclear capture. Our fitted value for τ_μ is 173(3) ns. The fitted values of the initial amplitude and the depolarization rate are summarized in Table I. The initial amplitude is equal to a product of factors resulting from (1) incomplete beam polarization, (2) fraction of muons stopping in the sample, (3) depolarization during muonic atom formation and cascade,[7] (4) fraction of domains magnetized transverse to the muon polarization, (5) electron asymmetry from μ^- decay, (6) finite solid angle, and (7) finite time resolution.[6] By measuring the positive-muon spin-rotation amplitude, we determined the product of all factors except (3) and (7) to be 12%. For spinless nuclei the factor (3) is expected to be 15%. The amplitude reduction due to the 140-ps timing resolution (factor 7) is 85%. Thus the expected amplitude is 1.5%, which is approximately twice that observed (see Table I). This discrepancy may be due to a disruption of the local environment for some fraction of the μ^-Ni, which is also usually the case in the perturbed angular distribution of recoil nuclei after reactions.

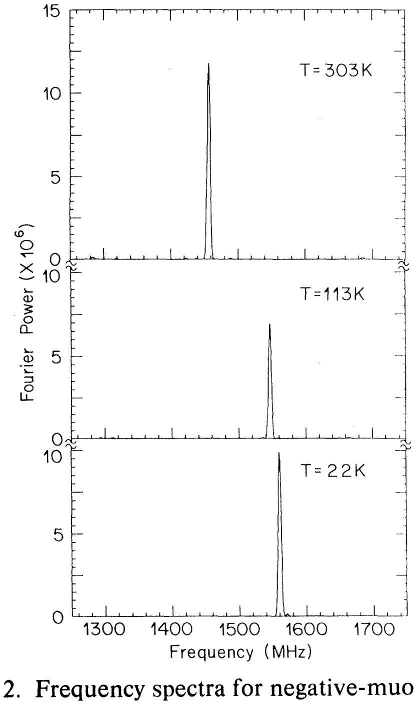

FIG. 2. Frequency spectra for negative-muon spin precession in single-crystal Ni in zero applied field as a function of temperature.

It has been pointed out that the emission of an x ray during the cascade may eject the muonic atom from its lattice site. The $2p$-$1s$ μ^-Ni x ray has an energy of 1.34 MeV implying a recoil energy of only 16 eV, which is three times smaller than the recoil energy from a $2p$-$1s$ x ray from μ^-Pd. Since this is also less than the displacement energy for Ni (34 eV),[8] a large fraction of μ^-Ni should occupy undisrupted sites. From the facts that the observed signal has a narrow width and an amplitude as much as 50% and that there is no other significant signal, we conclude that the observed signal arises from μ^-Ni at the normal lattice position. This is supported by the fact the frequency is close to that expected from the ^{59}Co hyperfine field in Ni; in a ferromagnetic phase any signal other than that from a normal site should be far apart and broadened in the frequency spectrum.

The internal magnetic field at the muon, B_μ, is calculated from f_μ by use of the relativistic bound-muon g factor. Ford, Hughes, and Willis have calculated g factors for several elements[9]; an interpolation for Ni yields a g factor which is reduced by 0.95(5)% from the vacuum value. The resulting values for B_μ are given in Table I. The negative sign was not determined experimentally but chosen because the field for ^{59}Co in nickel is known to be negative. The internal field can be decomposed as $B_\mu=B_{\rm L}+B_\mu^{\rm hf}$, where $B_{\rm L}=(4\pi/3)M_s$ is the Lorentz field. The hyperfine field $B_\mu^{\rm hf}$ is determined by use of the literature value of the saturation magnetization M_x.

The internal field at an impurity ^{59}Co in Ni has

TABLE I. Comparison of μ^-Ni spin rotation and Co NMR in Ni. The hyperfine anomaly is defined as $\Delta = (B_\mu^{\text{hf}} - B_N^{\text{hf}})/B_N^{\text{hf}}$.

Temperature T (K)	23.1(3)	113.0(5)	303.5(5)
μ^-Ni frequency f_μ (MHz)	1561.0(3)	1546.9(5)	1458.0(5)
μ^-Ni amplitude A_μ (%)	0.81(14)	0.58(16)	0.56(16)
μ^-Ni depolarization rate λ_μ (μs^{-1})	4.7(21)	1.4(31)	0.7(31)
μ^-Ni local field B_μ (T)[a]	$-11.627(8)$	$-11.522(10)$	$-10.860(10)$
Lorentz field B_{L} (T)[b]	0.214(1)	0.212(1)	0.200(1)
μ^-Ni hyperfine field B_μ^{hf} (T)	$-11.841(8)$	$-11.734(10)$	$-11.060(10)$
^{59}CoNi NMR frequency f_N (MHz)	122.1[c]	119.2(2)[d]	111.953(5)[e]
^{59}Co local field B_N (T)[f]	$-11.92(2)$[c]	$-11.86(4)$	$-11.135(22)$[e]
^{59}Co hyperfine field B_N^{hf} (T)	$-12.13(2)$	$-12.07(4)$	$-11.335(10)$[e]
^{60}Co hyperfine field B_N^{hf} (T)	$-12.09(1)$[g]	...	...
Hyperfine anomaly Δ (%): ^{59}Co	$-2.4(3)$	$-2.8(5)$	$-2.4(3)$
^{60}Co	$-2.1(1)$	...	...

[a] $B_\mu = f_\mu/[134.25(7)\ \text{MHz/T}]$ by interpolating Ford, Hughes, and Willis (Ref. 9).

[b] $B_{\text{L}} = (4\pi/3)M_s$ where M_s is the saturation magnetization.

[c] Yasuoka and Takigawa (Ref. 11), value for 4.2 K.

[d] Bennett and Streever (Ref. 12).

[e] Enokiya and Kawakami (Ref. 10), value for 303.1 K.

[f] $B_N = f_N/[10.054(20)\ \text{MHz/T}]$ from Walstedt, Wernick, and Jaccarino (Ref. 16).

[g] Barclay (Ref. 13), value for low temperature.

been reported by several authors (see Table I). In zero applied field the NMR signal from Co nuclei in the domain walls dominates. This signal may differ from the bulk value with which the μ^-Ni results should be compared. Recently, Enokiya and Kawakami[10] performed precision NMR on ^{59}Co in the bulk in single-crystal Ni (0.1–0.2 at.% Co) at 303.1 K (see Table I). The resulting hyperfine field B_N^{hf} can be directly compared with our negative-muon spin-rotation result at 303.5 K. Yasuoka and Takigawa[11] have recently observed ^{59}Co NMR (0.5 at.%) in finely powdered Ni at 4.2 and 300 K. By extrapolation of the field dependence to zero field, B_N^{hf} at 4.2 K is determined to be $-12.14(2)$ T. After making a small temperature correction (0.01 T) this can be compared with our μ^-Ni result at 23 K. Their result for B_N^{hf} in Ni fine powder at 300 K is in agreement with the result from Ref. 10 obtained for single-crystal Ni at 303.1 K. Bennett and Streever[12] have reported less precise measurements on ^{59}Co in Ni in the temperature range 80–450 K. An interpolated B_N^{hf} was used for comparison with our μ^-Ni result at 113 K. The dilute limit with a different Co isotope has been studied with use of ^{60}Co in Ni at low temperature.[13] The resulting hyperfine anomalies Δ, defined as the relative difference between the μ^-Ni and Co hyperfine fields, are given in Table I. We find that Δ is temperature independent to within experimental uncertainty and ranges between $-2.4(3)$% and $-2.8(5)$%. These anomalies are an order of magnitude larger than a typical Bohr-Weisskopf effect for a finite nucleus compared to a point nucleus.

In order to interpret the results in terms of the electron spin density around the nucleus, we use the extended Bohr-Weisskopf model[1-3] in which the muonic hyperfine anomaly relative to a point nucleus is expressed as

$$\epsilon_\mu = \int m(r)\{\rho(r)/\rho(0)\}\,d\tau - 1,$$

where $m(r)$ and $\rho(r)$ are the muon and electron spin densities, respectively. If the electron spin density follows the s-electron charge density ($= |\psi(r)|^2$), then ϵ_μ can be expressed as [2]

$$\epsilon_\mu = -(ZR_0/a_0)\int_0^{R_0} m(r)\,r^2/R_0^2\,d\tau - (ZR_0/a_0)\int_{R_0}^{\infty} m(r)(2r/R_0 - 1)\,d\tau,$$

where a_0 and R_0 are the Bohr and nuclear radii, respectively. For light elements ($Z < 30$) the muonic radius $\cong 260/Z$ fm is much larger than R_0, and thus the second term is dominant. Taking an asymptotic form for $\rho(r)$ near the nucleus,

$$\rho(r) \cong 1 - 2Zr/a_0,$$

and using the hydrogen-like 1s-muonic wave function for $m(r)$, we obtain a simple estimate of ϵ_μ irrespective to Z:

$$\epsilon_\mu^0 = -3m_e/m_\mu = -1.45\%.$$

The nuclear Bohr-Weisskopf estimate for a Co nucleus gives an anomaly relative to a pointlike nucleus:

$$\epsilon_N^0 \cong -\tfrac{3}{5}ZR_0/a_0$$
$$\cong -1.4\times 10^{-5}\ ZA^{1/3} = -0.15\%.$$

Thus on this basis, one would expect an anomaly for μ^-Ni relative to the ^{59}Co result:

$$\Delta = \epsilon_\mu^0 - \epsilon_N^0 = -1.3\%,$$

which is significantly less in magnitude than the observed effect of $-2.4(3)\%$ at 23 K.

The hyperfine anomaly is given by the overlap of the muonic and electronic spin densities. In the above estimation, we have assumed that the electronic spin and charge densities are proportional to one another. A larger anomaly results if the spin density decreases more rapidly than the charge density with increasing radial distance from the nucleus. Such behavior is a consequence of core polarization.[14] Mallow, Freeman, and Desclaux[15] have evaluated the hyperfine anomaly for μ^-Ni, taking account of this effect in the Dirac-Fock formalism. Their result $\Delta = (-2.3$ to $-2.7)\%$ (the spread reflecting an ambiguity in the electronic wave function) is in agreement with our experimental result. This is clear evidence of core polarization. It also demonstrates the usefulness of the negative muon as a probe of the spin density well away from the nucleus in a region not accessible with other techniques.

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