as does the itinerant model used here.

We have reported optical-conductivity data which provide an unambiguous first observation of the 5*f* states in an actinide metal. The itinerant model provides distinctly better agreement with experiment than does the localized model. Thus, we conclude that the 5*f* electrons found in the thorium excitation spectrum 1-5 eV above the Fermi energy are itinerant in nature. This result provides strong confirming evidence for the prediction[2,3] that the occupied 5*f* states in the lighter actinide metals are itinerant.

*Work supported by the U. S. Atomic Energy Commission, the National Science Foundation, and the U. S. Air Force Office of Scientific Research.

[1]T. L. Loucks, Phys. Rev. 139, A1333 (1965).

[2]A. J. Freeman and D. D. Koelling, J. Phys. (Paris), Colloq. 33, C3-57 (1972).

[3]D. D. Koelling and A. J. Freeman, Solid State Commun. 9, 1369 (1971).

[4]D. D. Koelling and A. J. Freeman, in Proceedings of the Eighteenth Annual Conference on Magnetism and Magnetic Materials, Denver, Colorado, 1972 (to be published).

[5]A. C. Thorsen, A. S. Joseph, and L. E. Valby, Phys. Rev. 162, 574 (1967); D. J. Boyle and A. V. Gold, Phys. Rev. Lett. 22, 461 (1969).

[6]S. C. Keeton and T. Loucks, Phys. Rev. 146, 429 (1966).

[7]R. P. Gupta and T. L. Loucks, Phys. Rev. Lett. 22, 458 (1969), and Phys. Rev. B 3, 1834 (1971).

[8]B. Veal, F. M. Mueller, R. Afshar, and J. Shaffer, in *Magnetism and Magnetic Materials—1971*, AIP Conference Proceedings No. 5, edited by C. D. Graham and J. J. Rhyne (American Institute of Physics, New York, 1971), p. 1285; B. W. Veal, to be published.

[9]C. B. Haselgrove, Math. Comput. 15, 323 (1961).

[10]A. R. Williams, J. F. Janak, and V. Moruzzi, Phys. Rev. Lett. 28, 671 (1972).

Precession of Positive Muons in Nickel and Iron

M. L. G. Foy, Neil Heiman, and W. J. Kossler
College of William and Mary, Williamsburg, Virginia 23185

and

C. E. Stronach
Virginia State College, Petersburg, Virginia 23803

(Received 15 February 1973)

Polarized μ^+ were stopped in Ni and Fe. The magnetic fields at the site of the muons, the initial polarization, and the depolarization time constant were obtained from 300 to 700°K, i.e., through the Curie temperature of Ni. This experiment demonstrated that precession of muons stopped in ferromagnetic material can be observed and, and it determined what the magnitude of the internal fields are: ≈150 G for Ni and 4000 G for Fe.

We implanted polarized positive muons in nickel and iron. The magnetic fields at the sites of the muons (B_μ) were measured by observing the precession of the angular distribution of the decay positrons. The same data yielded the initial polarization (P) of the stopped muons and the time constant (τ) of the slow depolarization. Data were collected as a function of temperature from room temperature to 700°K so that we observed the shift from the ferromagnetic to the paramagnetic state in nickel. This experiment demonstrated for the first time that the precession of muons stopped in ferromagnetic material can be observed, and it further determined what the magnitude and direction of the internal fields are.

That the muon can be a fundamentally important tool for condensed-matter physics has recently begun to be recognized.[1] The work of Schenck and Crowe[2] and Brewer *et al.*[3] has shown that the implanted positive muon behaves very much like a hydrogen nucleus, and that muon studies can provide information about lattice structure and chemical reactions. The present Letter provides new evidence of the usefulness of implanted muons for the study of metals and internal magnetic fields. The behavior of hydrogen in metals is of intrinsic interest as one of the simplest alloy problems.[4] It is also a problem of considerable technological importance.[5] The behavior of the hydrogenlike positive muons in ferromagnetic metals is of additional interest because of the interactions with the magnetic medium. Implanted muons cause minimal radiation damage, occur in infinitesimal concentration, leave no residual contamination, have no nuclear or quadrupole interactions, and do not possess a complicated ion core. Consequently the use of implanted muons cannot only yield fundamental solid-state information, but also contribute to the understanding of radioactive ion implantation, particularly in the case of ferromagnetic targets.

The positive muon is also an excellent probe from the standpoint of experimental simplicity. High count rates are available with polarization approaching 100%. The muon mean life (2.2 μsec) is long enough for easy timing and short enough for high count rates. The decay positrons are easily detected and the angular distribution of these positrons is highly anisotropic $[dN/d\theta \simeq N(1+0.33\cos\theta)]$. The muon's magnetic moment has been determined to a few parts per million allowing high-precision measurement of B_μ.

Positive muons from the Space Radiation Effects Laboratory synchrocyclotron were implanted in large ($20\times20\times1$ cm^3) targets of nickel and iron because the experiments were performed without the benefit of the meson channel. For measurements on paramagnetic nickel, the external magnetic field was supplied by a large Helmholtz pair capable of providing about 100 G with a homogeneity of about 2%. The external field for ferromagnetic targets was supplied by an iron-core electromagnet. The ferromagnetic sample almost completely closed the magnetic flux path. A small air gap (≈ 0.25 cm) allowed us to enclose the sample in a furnace and to measure the field in the gap.

Scintillators 1, 2, and 3 were in front of the sample, 4 behind, and 5 at 90°. A muon which stopped in the sample would traverse scintillators 1, 2, and 3 but not 4. Approximately 90% of the incident muons were stopped. A stopped muon signal, a $123\bar{4}$ (meaning a coincidence between counters 1, 2, and 3 but not 4), was used to start a time-to-amplitude converter (TAC). A decay positron signal, either a $4\bar{1}$ or a $5\bar{1}$, stopped the TAC. (The $\bar{1}$ anticoincidence requirement ensured that the signal in 4 or 5 was not due to a particle from the incident beam.) The output of the TAC was fed to a pulse-height analyzer which directly produced a plot of the number of decay positrons as a function of time after a muon stop.

Figure 1 provides examples of data for para-

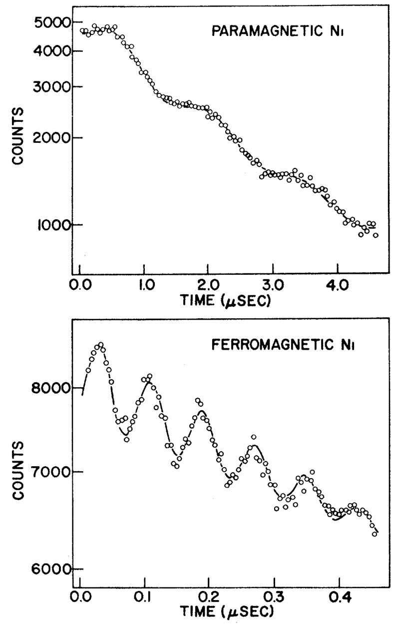

FIG. 1. Data for paramagnetic nickel at about 670°K and ferromagnetic nickel at 551°K. The data shown are semilog plots of the number of decay positrons detected by scintillator number 5 as a function of time after a muon stop. Note that the time scales differ by a factor of 10.

magnetic nickel (at 670°K) and ferromagnetic nickel (at 551°K). Each set of data is the result of approximately 8 h of counting. Note that the time scales differ by about a factor of 10. The data were fitted by the function[1,2,6]

$$N(t) = N_0 \exp(-\lambda t)[1 + aP\exp(-t/\tau)\cos(\omega t + \varphi)] + \text{background}.$$

N_0 is for normalization. The term $\exp(-\lambda t)$ accounts for the fact that the muon decays into a positron with a 2.2-μsec mean life. P is the initial polarization of the stopped muons and a is the positron anisotropy. τ is the time constant characterizing the relaxation of the transverse polarization of the precessing muons. The angular precession frequency $\omega = 2\mu_\mu B_\mu/\hbar$, where μ_μ is the magnetic moment of the muon, provides direct measurement of the magnetic field at the muon site, B_μ. φ is the initial phase angle. The solid lines in Fig. 1 are the fitted functions.

Paramagnetic nickel results.—In this region $B_\mu \approx B_{ext}$, the externally applied field, approximately 50 G. The paramagnetic Knight shift was small, and significant variation of B_μ with temperature was not detected. The initial polarization P was equal to $0.8P_C$, where P_C was the initial polarization observed in a carbon target of similar dimensions. Muons stopped in carbon retain virtually 100% of their polarization.[7] Therefore carbon is useful for calibration. The depolarization time constant τ was 4 μsec. The parameters B_μ, P, and τ did not vary significantly over the temperature range 630 to 705°K ($T_C \approx 630$°K). Paramagnetic nickel thus behaves as a normal metal in that little or no muonium appears to have formed. Muonium formation would have resulted in reduced polarization and the appearance of frequency components about 100 times higher than the frequency observed. Our resolution was sufficient to detect such high-frequency components, and none were present.

Ferromagnetic nickel results.—In this region we were able to fit B_μ with a Brillouin function (see Fig. 2) with a saturation field of 1550 G. The circular data points were taken with a current of 1.5 A in the electromagnet. This corresponded to a field in the small gap between the magnet and target of 1100 G at room temperature. Increasing the field to 2200 G in the gap or lowering the field to 500 G or even to zero had little effect on B_μ. In the runs with nickel below 385°K and in an external field of about 1100 G, we were not able to detect the precession of the angular distribution. On increasing the magnetic field in the gap to approximately 2200 G, we were able to observe the precession (the square datum point in Fig. 2). The internal field is in the same direction as the external field. This was determined by noting the relative phase of the data from detectors 4 and 5 and also by noting that the initial phase for each detector was maintained during the transition from paramagnetic to ferromagnetic behavior.

Figure 3 shows the behavior of P and τ. P/P_C increases from about 20% at room temperature to near 40% as T_C is approached. We believe this is due to domain alignment. A muon will not precess if it stops in a domain where the magnetization or internal field is in the same direction as the polarization. The amplitude of the precession signal thus depends upon the degree to which the domains are aligned transverse to the muon polarization. The solid line in Fig. 3 is a measure of this domain alignment extracted by dividing the measured permeability by the magnetization and normalizing to P. We were unable to saturate the sample, but increasing the magnet current from 1.5 to 3.0 A increased P only slightly, indicating that the initial polarization even at saturation may be less in ferromagnetic than in paramagnetic nickel.

τ was on the order of 0.25 μsec and increased slightly with temperature and dropped sharply as T_C was approached. The dashed line in Fig. 3 is intended only to illustrate this trend. Varying B_μ by changing temperature or varying the external field seemed to have little effect on τ, implying that the depolarization was not arising from field inhomogeneity.

Ferromagnetic iron results.—In the temperature range from room temperature to 675°K, B_μ decreased from approximately 4100 to 3700 G. P/P_C was about 10% and τ was on the order of 0.5 μsec. It is interesting that the ratio of the field in Fe to that in Ni is not much different from the ratio of the magnetizations.

There is evidence that hydrogen atoms are implanted in the octahedral or body-centered site in nickel.[8] It is reasonable to assume that the muon stops in the same site although it may not be as well localized due to its smaller mass. It

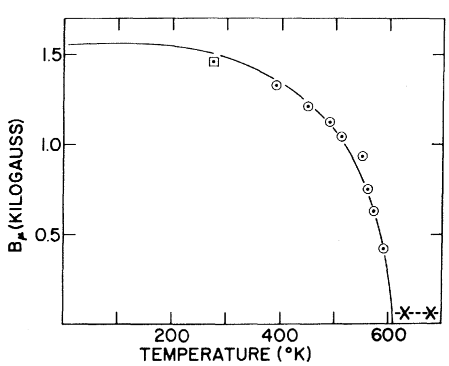

FIG. 2. B_μ as a function of temperature. Circular data points were obtained with 1.5 A in the electromagnet, the square datum point with 3.0 A.

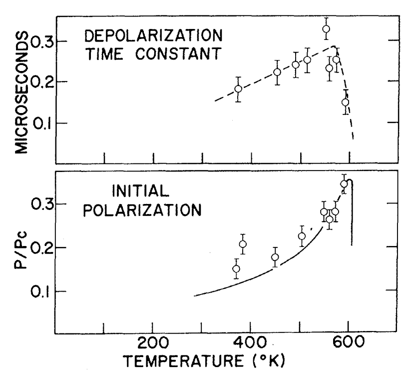

FIG. 3. Initial polarization P and depolarization time constant τ as functions of temperature. The solid line is a measure of domain alignment: the measured sample permeability divided by B_μ. The dashed line is only to guide the eye.

is interesting to compare B_μ for ferromagnetic nickel, 1550 G, to a number of possible contributions. Because of the site symmetry, the classical dipolar field due to the nickel ions is zero. Further, because of sample geometry, the demagnetizing field is negligible. The hyperfine field at the muon in free muonium is on the order of 160 000 G. The Lorentz field $4\pi M/3$ for nickel is about 2800 G, nearly double B_μ. The contributions to B_μ from core polarization and conduction-electron polarization cannot be estimated using conventional techniques[9,10] because the muon does not possess a core in the usual sense and its interstitial location makes extrapolation of conduction-electron polarization from previously existing data questionable. Nevertheless the major contribution to the internal field should arise from the Fermi contact interaction with the conduction electrons, provided the muon exists as a nonmagnetic impurity. Except for unusual circumstances, hydrogenlike impurities in metals should be nonmagnetic and in fact the present result that B_μ follows the host magnetization confirms the nonmagnet character of the implanted muon.[11,12] Under these conditions, one can approximate the conduction-electron contribution to B_μ by[13]

$$B_\mu{}^{CE} \simeq (16\pi/3)\mu_B\,\delta\rho(0),$$

where $\delta\rho(0)$ is the net spin density at the muon. If we neglect other contributions, the observed B_μ of 1550 G implies a net spin density of approximately 1.0% of that of free muonium. If we include the Lorentz field, the spin density is of the same order but of opposite sign. The smallness of this number may reflect a cancelation between the contributions from the d-band and s-band electrons to $\delta\rho(0)$ at the interstitial site. In fact Friedel[4] shows that the electronic density attracted to an interstitial hydrogen nucleus is roughly similar to that of a hydrogen atom in vacuum, and that the screening in transition-metal hosts is primarily by electrons from the d band, although the s-band contribution is not negligible. It seems clear that the problem of calculating the field at the muon site lacks many of the obscuring complications associated with other implanted impurities and therefore provides a sensitive and intrinsically interesting test for the theory of internal fields.

The experiment will be continued shortly using the prime muon beam, allowing us to use much smaller targets, much more intense and homogeneous magnetic fields, and a more uniform sample temperature, permitting more accurate measurements near T_C and in the paramagnetic region. This will also allow us to magnetically saturate our samples. From these improved data we should be able to examine critical behavior near T_C, the depolarization mechanism, the temperature dependence of the paramagnetic Knight shift, the relative d- and s-band electron contribution to the screening charge, the true fast depolarization, and the apparent shielding of the external field from the muon site.

---

[1] Meeting on Muons in Solid State Physics, Bürgenstock, 1-3 September 1971 (unpublished).

[2] A. Schenck and K. M. Crowe, Phys. Rev. Lett. 26, 57 (1971).

[3] J. H. Brewer, K. M. Crowe, R. F. Johnson, A. Schenck, and R. W. Williams, Phys. Rev. Lett. 27, 297 (1971).

[4] J. Friedel, Ber. Bunsenges. Phys. Chem. 76, 828 (1972).

[5] Ber. Bunsenges. Phys. Chem. 76, No. 8 (1972).

[6] A. O. Weissenberg, *Muons* (North-Holland, Amsterdam, 1967), Chaps. IV and VI.

[7] R. A. Swanson, Phys. Rev. 112, 580 (1958).

[8] F. Brotzen, private communication.

[9] R. E. Watson and A. J. Freeman, Phys. Rev. 123, 2027 (1961).

[10] D. A. Shirley and G. A. Westenbarger, Phys. Rev. 138, A170 (1964).

[11] V. Jaccarino, L. R. Walker, and G. K. Wertheim, Phys. Rev. Lett. 13, 752 (1964).

[12] I. A. Campbell, J. Phys. C: Proc. Phys. Soc., London 3, 2151 (1970).

[13] E. Daniel and J. Friedel, J. Phys. Chem. Solids 24, 1601 (1963).