

Muon depolarization and magnetic field penetration depth in superconducting $\text{GdBa}_2\text{Cu}_3\text{O}_x$

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Positive muon spin-rotation (μSR) measurements on $\text{GdBa}_2\text{Cu}_3\text{O}_x$ ($x \approx 7$) have been conducted in the temperature interval 5.8–300 K. A temperature-dependent relaxation, attributable to Korringa relaxation of the Gd ion, as well as vortex-state relaxation are observed. Magnetic field penetration depths of approximately 1550 and 1900 Å (two samples) are extracted from the data. Two muon stopping sites exist for $T \leq 80$ K, one being associated with normal regions of the sample and the other with superconducting regions.

The discovery of superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_x$ ($x \approx 7$) (Ref. 1) was quickly followed by similar observations in rare-earth barium-copper oxides.²⁻⁷ Not only are the transition temperatures in the rare-earth compounds similar to $\text{YBa}_2\text{Cu}_3\text{O}_x$ ($T_c \approx 90$ K) but, remarkably, the superconductivity is unaffected by the magnetic dopants (Gd and Er, for example). Previous research has demonstrated the utility of muon spin rotation (μSR) to probe the magnetism of the nonsuperconducting compound $\text{La}_2\text{CuO}_{4-y}$,⁸ as well as the superconducting state of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ ($T_c \approx 40$ K) (Refs. 9 and 10) and $\text{YBa}_2\text{Cu}_3\text{O}_x$.¹¹ Additionally, $\text{GdBa}_2\text{Cu}_3\text{O}_x$ has been studied by zero-field μSR in the temperature interval 30 mK to 130 K.¹² In this paper we report transverse and zero-field μSR studies of $\text{GdBa}_2\text{Cu}_3\text{O}_x$ in the temperature interval 5.8 to 300 K.

A sintered disk 25.4 mm in diameter and 3.9 mm in thickness ($m = 12.174$ gm) was prepared by the methods of Ref. 13. Assuming the theoretical density ρ_{th} to be 6.94 gm cm^{-3} (Ref. 14), our sample is characterized by a density fraction $\rho/\rho_{\text{th}} = 0.89$. Moreover, magnetization measurements indicate a 30% Meissner effect with 81% shielding (applied field = 100 Oe). The superconducting transition deduced from susceptibility measurements is $T_c \approx 92$ K (sample 1). X-ray diffraction and Rutherford backscattering measurements indicate that the sample is high-quality, single phase, and has the correct stoichiometry. Direct-current resistance and ac eddy-current measurements also suggest that the sample is a high-quality, single-phase superconductor. A second sample with similar properties was also prepared and measured.

Standard μSR techniques¹⁵ were used to obtain positive muon (μ^+) Gaussian depolarization rates $\Lambda(T)$ in $\text{GdBa}_2\text{Cu}_3\text{O}_x$ for both 1 and 5 kOe externally applied

transverse fields and for zero applied field; in the latter case the residual field was less than 20 mOe.

Figures 1 and 2 show the μ^+ Gaussian depolarization rates for two samples of $\text{GdBa}_2\text{Cu}_3\text{O}_x$ taken in a transverse field of 1 kOe. Two relaxation rates (indicated by circles and triangles in Figs. 1 and 2) are observed for $T < T_c$, corresponding to two distinct muon environments. Unlike previous μSR results in $\text{YBa}_2\text{Cu}_3\text{O}_x$,¹¹ where a constant relaxation rate is observed for $T > T_c$, we find a $1/T$ dependence, presumably due to Korringa relaxation of the Gd ion. For $T < T_c$ the μ^+ relaxation rate increases (solid circles of Figs. 1 and 2) due to the magnetic field inhomogeneities produced by the vortex state. Assuming a square Abrikosov lattice for the mixed state ($H_{c1} < H_{\text{app}} < H_{c2}$), the field inhomogeneity is given by¹⁶

$$\langle |\Delta H|^2 \rangle = \frac{H_{\text{app}}\phi}{4\pi\lambda^2} [1 + (4\pi^2\lambda^2 H_{\text{app}})/\phi]^{-1} = \frac{2\Lambda^2}{\gamma_\mu^2}, \quad (1)$$

where Λ is the μ^+ depolarization rate, γ_μ is the muon gyromagnetic ratio, λ is the magnetic field penetration depth, and ϕ is the flux quantum. By subtracting the $a + (b/T)$ contribution (a is the constant relaxation rate at 300 K) from the μ^+ depolarization rate, and extrapolation Λ to 0 K, we find $\lambda(T=0)$ to be 1550 and 1900 Å for the two samples, respectively. These values are similar to those previously reported for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ (Ref. 10) (2000 Å) and $\text{YBa}_2\text{Cu}_3\text{O}_7$ (Ref. 11) (1400 Å). The temperature dependence of the magnetic field penetration depth for an ordinary Bardeen-Cooper-Schrieffer (BCS) superconductor is given by¹⁷

$$\lambda(T) = \lambda(0) [1 - (T/T_c)^4]^{-1/2}. \quad (2)$$

Substitution of Eq. (2) into Eq. (1), with $\lambda(0) = 1900$ Å (1550 Å), yields the solid line shown in the inset of Fig. 1

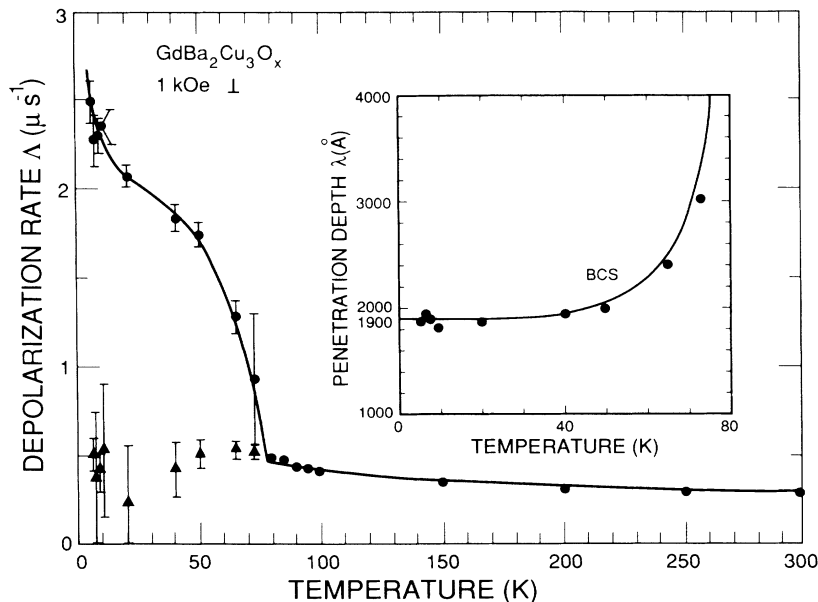


FIG. 1. Muon Gaussian depolarization rates in $\text{GdBa}_2\text{Cu}_3\text{O}_x$ (sample 1) taken in a 1-kOe transverse applied field. The triangles and circles correspond to the muon precessional frequencies identified with the same symbols in Fig. 3. See text for a description of the fit to the data. Inset depicts the temperature dependence of the magnetic field penetration depth.

(Fig. 2). Similar temperature dependences have been previously reported.^{10,11}

The fit to the μ^+ depolarization rate, shown as the solid lines of Figs. 1 and 2, are obtained in the following way. It is assumed that there exists a temperature-dependent component to the relaxation rate determined by the strength G and correlation time τ_s of the Gd-ion/ μ^+ interaction,¹⁸ i.e., $\Lambda \propto G\tau_s$, and that the ion spin-lattice relaxation rate is assumed to have the Korringa form $\tau_s^{-1} \propto T$. Additionally, there exists the vortex-state relax-

ation for $T < T_c$, for which $\Lambda(T)$ has the form $[1 - (T/T_c)^4]$. Thus the solid line fit shown in Fig. 1 is given by $\Lambda(T) = 17/(T+2) + 0.25$ for $T > T_c$; and $\Lambda(T) = 5/(T+2) + 0.25 + 1.6[1 - (T/T_c)^4]$ for $T < T_c$ (Λ is in μs^{-1} and T is in K). Similarly in Fig. 2, $\Lambda(T) = 17/(T+2) + 0.25$, for $T > T_c$; and $\Lambda(T) = 5/(T+2) + 0.25 + 2.3[1 - (T/T_c)^4]$ for $T < T_c$. Note that T_c found from the fit is 80 K for sample 1 (Fig. 1) and 95 K for sample 2 (Fig. 2). Eddy-current measurements indicate that a relatively sharp superconducting transition

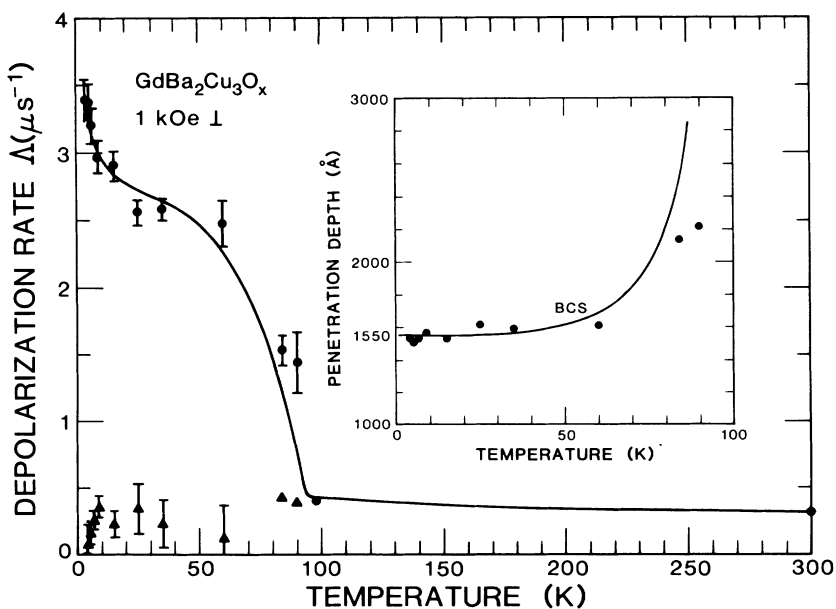


FIG. 2. Muon Gaussian depolarization rates in $\text{GdBa}_2\text{Cu}_3\text{O}_x$ (sample 2) taken in a 1-kOe transverse applied field. See text for a description of the fit to the data. Inset depicts the temperature dependence of the magnetic field penetration depth.

occurs in sample 2, whereas a broad transition is seen in sample 1.¹⁹ The coefficient of the $[1 - (T/T_c)^4]$ term in the relaxation rate expression may be associated with this sharpness, and, accordingly, is larger for sample 2 than sample 1. Moreover, the coefficients a and b , described above, are identical for the two samples, although b changes in magnitude for $T \leq T_c$, perhaps reflecting a change in the μ^+ /Gd ion coupling strength. For selected muon sites a compares favorably with our calculated Van Vleck linewidths in $\text{GdBa}_2\text{Cu}_3\text{O}_x$. The denominator of the first term is $T+2$ rather than T because of the antiferromagnetic ordering that occurs near 2 K.³

Owing to the paucity of data and large error bars in $\Lambda(T)$ for $T < T_c$ (circles in Figs. 1 and 2), the $1/T$ dependence in this interval is somewhat speculative. In analogy with NMR experiments, where it is known²⁰ that the spin-lattice relaxation rate follows an Arrhenius relation for $T \ll T_c$, we attempted to fit the relaxation data in this region with an exponential $\Lambda(T) \propto \exp(\Delta/k_B T)$, where Δ is the gap amplitude, and for the isotropic BCS case $2\Delta/k_B T_c = 3.53$. Although a reasonable fit was achieved for $\Lambda(T)$, the resulting value of 8 K for Δ was unreasonable yielding $2\Delta/k_B T_c \approx 0.2$. Thus we do not believe that the exponential form is the correct description for $\Lambda(T)$ in this interval.

An interesting feature of our relaxation data is the appearance of two μ^+ precession frequencies for $T < T_c$, as shown in Fig. 3. Both samples exhibit qualitatively similar behavior so only one data set is shown (sample 1). Recall that the μ^+ precessional frequency in a 1 kOe field is 13.55 MHz. Of the two observed frequencies, one (filled triangles) remains approximately constant, whereas the other decreases with decreasing temperature. One explanation for this behavior is the existence of two distinctly different μ^+ stopping sites; one associated with normal regions and the other with superconducting regions of the sample. A reduction in the magnitude of the local field seen by the μ^+ as T decreases is consistent with flux expulsion in the superconducting state; however, note that at

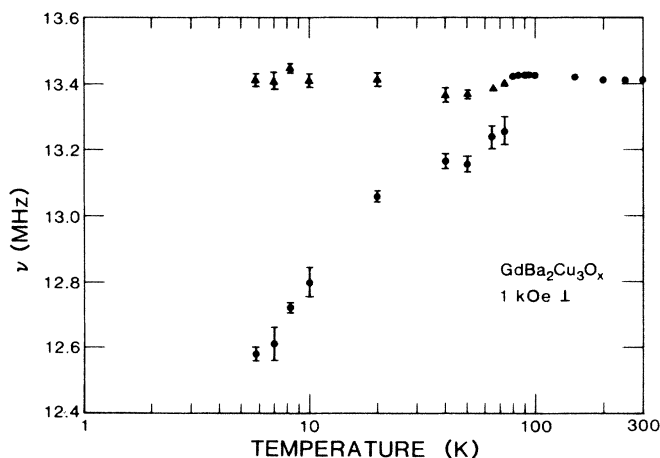


FIG. 3. Positive muon spin precessional frequencies in $\text{GdBa}_2\text{Cu}_3\text{O}_x$ taken in a 1-kOe transverse applied field. The corresponding depolarization rates are shown by the same symbols in Fig. 1.

$T = 5.8$ K the muon frequency corresponds to a local magnetic field of 930 Oe. Thus the sample expels only 7% of the external field. Taken together with the measured relaxation rate (filled circles of Fig. 1), the temperature dependence of ν is associated with muons stopping in superconducting regions of the sample. We note, however, that the expected frequency spectrum, as predicted by Abrikosov,²¹ is not realized. Of course flux pinning is expected to smear this theoretical distribution and may account for our experimental results. Because our μSR data are taken over a 10- μs interval, we cannot resolve two frequencies closer than ~ 0.1 MHz. To minimize flux pinning we measured relaxation rates by field cooling the sample from RT.

Relaxation rates for those muons stopping in normal regions of the sample at low temperature are shown as filled triangles in Figs. 1 and 2. Unfortunately the error bars are too large to allow elucidation of any functional form for $\Lambda(T)$; however, it is clear that these rates are much smaller than would be expected if one extrapolated the value of Λ at RT ($0.25 \mu\text{s}^{-1}$) to 5.8 K, assuming a $1/T$ dependence. Thus the normal-state μ^+ environment at low T is quite different than the one at high T . Qualitatively similar results were obtained for an applied field of 5 kOe, with the relaxation rates being proportionately higher. Previous transverse-field μSR studies¹² of $\text{GdBa}_2\text{Cu}_3\text{O}_7$ were confined to the interval $30 \text{ mK} < T < 130 \text{ K}$. Only one muon stopping site was reported for T_N (2.24 K) $< T < 130 \text{ K}$. However, the sample was multiphased, exhibiting both 90 and 60 K superconducting transitions with a measured muon depolarization rate approximately three times that of the present study. It is likely that the large depolarization rate is attributable, in part, to spurious phases. A broad frequency spectrum

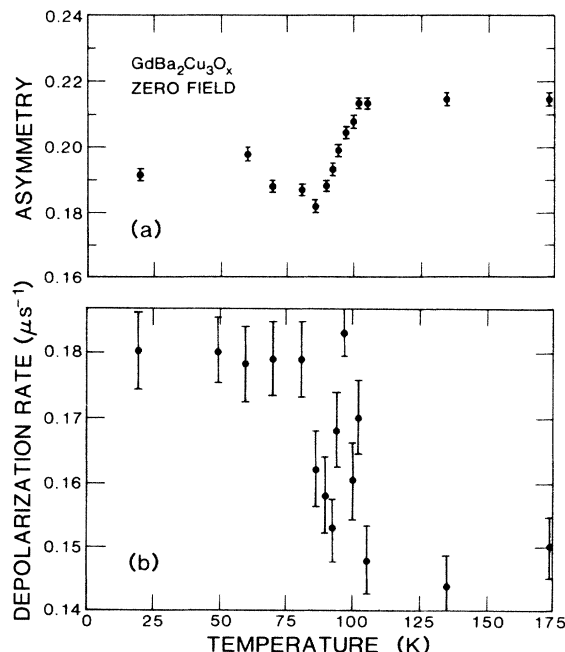


FIG. 4. Muon exponential relaxation rates and asymmetries for $\text{GdBa}_2\text{Cu}_3\text{O}_x$ measured in zero applied field.

(large Λ) may mask weak secondary peaks and render them unobservable, thus explaining the absence of two muon stopping sites in that study.¹²

Zero-field μ^+ exponential relaxation rates and asymmetries for $\text{GdBa}_2\text{Cu}_3\text{O}_x$ are shown in Fig. 4. With decreasing temperature there is a drop in the asymmetry and concomitant increase in relaxation rate near T_c . Typically one only observes a loss of asymmetry with decreasing temperature if there is a rapid depolarization of some fraction of the stopped muons, usually associated with magnetic or structural phase transitions. To our knowledge neither of these types of transitions has been observed near T_c for this compound; however, position-annihilation studies have shown that there is a change in the electronic structure as the material becomes superconducting.²² If this effect is accompanied by a change in the local magnetic field at the muon site, one would expect some change in the muon depolarization rate at T_c . Although conventional superconductors do not show any change in zero-field muon relaxation rate as the superconducting state is entered, it has been shown that the heavy-fermion superconductor UPt_3 does exhibit a 5%–10% increase at T_c .²³ Perhaps the electronic structure modification experienced in high- T_c materials is

sufficiently large to cause a measurable alteration of the local magnetic field sensed by the muon. This does not, however, explain the observed change in asymmetry. Further work will be required to understand this effect.

In summary, transverse-field μSR in superconducting $\text{GdBa}_2\text{Cu}_3\text{O}_x$ demonstrates temperature-dependent relaxation, presumably due to Korringa relaxation of the Gd ion, in addition to the expected vortex-state relaxation associated with superconductivity. Magnetic field penetration depths of 1900 Å (sample 1) and 1550 Å (sample 2) ($T=0$ K) are derived from the data. Evidence for the existence of both normal and superconducting regions of the sample for $T < T_c$ is given. It is suggested that a measured shift in zero-field relaxation rate at T_c may be associated with a change in the electronic structure, which produces a concomitant change in the local magnetic field sensed by the muon.

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- ¹M. K. Wu, J. R. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Gao, Z. J. Huang, Y. Q. Wang, and C. W. Chu, *Phys. Rev. Lett.* **58**, 908 (1987); P. H. Hor, L. Gao, R. L. Meng, Z. J. Huang, Y. Z. Wang, K. Forster, J. Vassiliou, and C. W. Chu, *ibid.* **58**, 911 (1987).
- ²Z. Fisk, J. D. Thompson, E. Zirngiebl, J. L. Smith, and S-W. Cheong, *Solid State Commun.* **62**, 743 (1987).
- ³J. O. Willis, Z. Fisk, J. D. Thompson, S-W. Cheong, R. M. Aikin, J. L. Smith, and E. Zirngiebl, *J. Magn. Magn. Mater.* **67** L139 (1987).
- ⁴P. H. Hor, R. L. Meng, Y. Q. Wang, L. Gao, Z. J. Huang, J. Bechtold, K. Forster, and C. W. Chu, *Phys. Rev. Lett.* **58**, 1891 (1987).
- ⁵J. P. Golbin, S-I. Lee, S. Y. Lee, Y. Song, T. W. Noh, X-D. Chen, J. R. Gaines, and R. T. Tettenhorst, *Phys. Rev. B* **35**, 8705 (1987).
- ⁶S-W. Cheong, S. E. Brown, J. R. Cooper, Z. Fisk, R. S. Kwok, D. E. Peterson, J. D. Thompson, G. L. Wells, E. Zirngiebl, and G. Gruner, *Phys. Rev. B* **36**, 3913 (1987).
- ⁷J. W. Lynn, W-H. Li, Q. Li, H. C. Ku, H. D. Yang, and R. N. Shelton, *Phys. Rev. B* **36**, 2374 (1987).
- ⁸Y. J. Uemura, W. J. Kossler, X. H. Yu, J. R. Kempton, H. E. Schone, D. Opie, C. E. Stronach, D. C. Johnston, M. S. Alvarez, and D. P. Goshorn, *Phys. Rev. Lett.* **59**, 1045 (1987); J. I. Budnick, A. Golnik, Ch. Niedermayer, E. Recknagel, M. Rossmannith, A. Weidinger, B. Chamberland, M. Filipkowski, and D. P. Yang, *Phys. Lett. A* **124**, 103 (1987).
- ⁹F. N. Gygax, B. Hitti, E. Lippelt, A. Schenck, D. Cattani, J. Cors, M. Decroux, Ø. Fischer, and S. Barth, *Europhys. Lett.* **4**, 473 (1987).
- ¹⁰W. J. Kossler, J. R. Kempton, X. H. Yu, H. E. Schone, Y. J. Uemura, A. R. Moodenbaugh, M. Suenaga, and C. E. Stronach, *Phys. Rev. B* **35**, 7133 (1987); G. Aeppli, R. J. Cava, E. J. Ansaldo, J. H. Brewer, S. R. Kreitzman, G. M. Luke, D. R. Noakes, and R. F. Kiefl, *ibid.* **35**, 7129 (1987).
- ¹¹D. R. Harshman, G. Aeppli, E. J. Ansaldo, B. Batlogg, J. H. Brewer, J. F. Carolan, R. J. Cava, M. Celio, A. C. D. Chaklader, W. N. Hardy, S. R. Kreitzman, G. M. Luke, D. R. Noakes, and M. Senba, *Phys. Rev. B* **36**, 2386 (1987).
- ¹²A. Golnik, Ch. Niedermayer, E. Recknagel, M. Rossmannith, A. Wiedinger, J. Budnick, B. Chamberland, M. Filipkowski, Y. Zhang, D. Yang, L. Lynds, F. Otter, and C. Baines, *Phys. Lett. A* **125**, 71 (1987).
- ¹³J. L. Smith, Z. Fisk, J. D. Thompson, J. O. Willis, and H. A. Borges, *Physica B* **148**, 14 (1987).
- ¹⁴Y. Le Page, T. Siegrist, S. Sunshine, L. Schneemeyer, D. Murphy, S. Zahurak, J. Waszczak, W. McKinnon, J. Tarascon, G. Hull, and L. Greene, *Phys. Rev. B* **36**, 3617 (1987).
- ¹⁵See, for example, A. Schenck, *Muon Spin Rotation Spectroscopy* (Hilger, Bristol, England, 1985), pp. 7–59, and references therein.
- ¹⁶P. Pincus, A. C. Gossard, V. Jaccarino, and J. Wernick, *Phys. Lett.* **13**, 21 (1964).
- ¹⁷P. D. De Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966), p. 26.
- ¹⁸J. A. Brown, R. H. Heffner, R. L. Hutson, S. Kohn, M. Leon, C. E. Olsen, M. E. Schillaci, S. A. Dodds, T. L. Estle, D. A. Vanderwater, P. M. Richards, and O. D. McMasters, *Phys. Rev. Lett.* **47**, 261 (1981).
- ¹⁹J. D. Doss, D. W. Cooke, C. W. McCabe, and M. Maez, *Rev. Sci. Instrum.* **59**, 659 (1988).
- ²⁰M. Lee, M. Yudkowsky, W. P. Halperin, J. Thiel, S.-J. Hwu, and K. R. Popelmeier, *Phys. Rev. B* **36**, 2378 (1987).
- ²¹A. A. Abrikosov, *Zh. Eksp. Teor. Fiz.* **32**, 1442 (1957) [*Sov. Phys. JETP* **5**, 1175 (1957)].
- ²²Y. C. Jean, S. Wang, H. Nakanishi, W. Hardy, M. Hayden, R. Kiefl, R. Meng, H. Hor, J. Huang, and C. Chu, *Phys. Rev. B* **36**, 3994 (1987).
- ²³D. W. Cooke, R. H. Heffner, M. E. Schillaci, J. L. Smith, J. O. Willis, D. E. MacLaughlin, C. Boekema, R. L. Lichti, A. B. Denison, and J. Oostens, *Hyperfine Interact.* **31**, 425 (1986).