Polarized-Neutron Reflections: A New Technique Used to Measure the Magnetic Field Penetration Depth in Superconducting Niobium

G. P. Felcher, R. T. Kampwirth, K. E. Gray, and Roberto Felici
Materials Science and Technology Division, Argonne National Laboratory, Argonne, Illinois 60439
(Received 14 February 1984)

An absolute determination of the superconducting penetration length in niobium has been carried out by a novel technique, in which the reflectivity of a polarized-neutron beam from a polished surface is measured. The result thus obtained, $\Lambda(0)=410\pm40$ Å, is in substantial agreement with the earlier work.

PACS numbers: 74.30.Ci, 61.12.-q, 74.60.-w

The reflection of x rays has been extensively used[1-3] to study the composition of materials close to the surface. The experiments consisted of sending a monochromatic beam onto the sample surface at a grazing incidence angle θ_i and measuring the reflected intensity for a range of θ_i slightly larger than the angle of total reflection θ_c. The angle-dependent reflectivity was shown to be correlated with the variation of the index of refraction as a function of the distance from the surface $n(z)$. In the case of neutron optics,[4] $n(z)$ is sensitive to the magnetic state of the material, and the present experiment shows that the neutrons can indeed[5] measure magnetic perturbations close to the surface. It was chosen to measure the magnetic field penetration Λ into a superconductor, because the typical range of Λ (of a few hundred angstroms) is a convenient size. At the same time the result obtained is of considerable value, since past experiments[6] were unable to determine the absolute value of Λ, despite their accuracy in measuring its temperature variation.

In the experiment, a film of niobium is kept at a temperature lower than the superconducting transition temperature T_c, in a magnetic field H parallel to the surface and less than the critical magnetic-flux entry field H_{c1}. The magnetic induction in the material is $B(z)=H\exp(-z/\Lambda)$, where z is the distance from the surface. Neutrons arriving at the surface are polarized either parallel (+) or antiparallel (−) to H; they are reflected from the plate as from a medium with refractive index

$$n^{\pm}(z)=1-\frac{\lambda^2}{2\pi}\left\{\frac{b}{v}\mp cH\left[1-\exp\left(-\frac{z}{\Lambda}\right)\right]\right\}, \quad (1)$$

where λ is the neutron wavelength, b/v is the nuclear scattering amplitude per unit volume (for niobium, $b/v=3.96\times10^{-6}$ Å^{-2}), and the constant c is[4] $c=2\pi\mu_n m/h^2=2.3\times10^{-10}$ Å^{-2} Oe^{-1}.

The reflectivity is[7] the resultant of the reflection occurring at the surface as well as of the reflections in the interior of the material: The latter are due to the variation of $n(z)$. In general, the reflectivity cannot be given by an exact yet simple analytical expression, and is rather obtained numerically. An approximate expression for the spin-dependent reflectivities of a medium with refractive index given by Eq. (1) is[8]

$$R^{\pm}=|r_1|^2\left[1\mp\frac{2\theta_i}{p_0}\,\frac{cH}{b/v}\,\frac{1}{1+(4\pi p_0\Lambda/\lambda)^2}\right]. \quad (2)$$

In Eq. (2), $r_1=(p_0-\theta_i)/(p_0+\theta_i)$ gives the reflectivity of the material in absence of magnetic terms; $p_0\simeq[\theta_i^2-(\lambda^2/\pi)b/v]^{1/2}$ is the effective angle of the neutron beam in the material and becomes zero at a critical wavelength λ_c. The effect of the magnetic terms is accounted for in Eq. (2) only in the first-order approximation and away from λ_c.

Equation (2) provides a first indication of the sensitivity of the measurements to the penetration depth. Suppose that the niobium surface is inclined at $\theta_i=0.32°$ with respect to the neutron beam and that the reflectivity is measured as a function of the neutron wavelength. For $\lambda\geqslant4.98$ Å the reflectivity is total, while it diminishes rapidly for shorter wavelengths, becoming at the same time spin dependent, as evidenced by taking the "flipping ratio" $F_R=R^+/R^-$. For instance, at 4.7 Å the spin-average reflectivity is $R=0.26$ for an external field $H=500$ Oe and a penetration length $\Lambda=400$ Å, and $F_R=0.93$. For comparison, if at the surface there was a sharp magnetic boundary ($\Lambda=0$), $F_R=0.65$ under the same conditions.

Rather sophisticated equipment for neutron reflection has been designed[9] in the past in order to determine accurately the nuclear scattering amplitudes. For this initial experiment, a preliminary instrument was set up at the intense pulsed neutron source at Argonne National Laboratory, and is described in Fig. 1. For each neutron pulse, the reflected intensities of all the neutrons of wavelengths between 3.8 and 8 Å were measured, with the surface at a fixed angle. These intensities were normalized to the incident neutron spectrum to give the reflectivities. The beam divergence was of the order of 0.02°; the wavelength resolution was 0.1 Å. The polarized intensities had to be corrected for the polarization efficiency of the Co mirror (75%) and, for one spin state, for the efficiency of the flipper (95%).

The preparation of the sample received considerable attention. The reflecting surface dimensions were 15×50 mm to fit into the cryostat/superconducting-magnet assembly. Initially the surface was prepared by polishing a polycrystalline plate of niobium. However, much better reflectivity was obtained by sputtering a 5-μm-thick layer of niobium onto an unheated, polished single crystal of silicon, (111) oriented. The reflectivity of a film of such a thickness—on a proper support—is not very different from that of a surface of bulk material. For a neutron plane wave, the film reflectivity is[7]

$$R=\frac{r_l^2+r_2^2+r_1r_2\cos(b_1z_0p_0/\lambda)}{1+r_1^2r_2^2+2r_1r_2\cos(4\pi z_0p_0/\lambda)}, \tag{3}$$

where r_1 and r_2 are due to the reflections at the first and second boundaries, and z_0 is the film thickness. When z_0 is of the order of a few micrometers, the interference terms oscillate quite rapidly with the angle of incidence θ_i, so that Eq. (3) integrated over the experimental angular divergence simplifies to the incoherent limit. The reflectivity of the support can then be easily taken into account, provided it does not overshadow that of the film (and this is certainly true for a silicon support, with $b/v=2.02\times10^{-6}$ Å^{-2}). The superconducting properties of this film were also found to be satisfactory, with $H_{c1}=1.0$ kOe at $T=5$ K, the Ginzburg-Landau parameter $\kappa=1.8$, and $T_c=9.2$ K.

The reflectivity of niobium at $T=10$ K and in a field of 500 Oe was found to be independent of the neutron spin. These data are presented in Fig. 2, together with the reflectivity obtained by solving numerically the optical equations.[7] It can be noticed that the resolution-averaged reflectivity

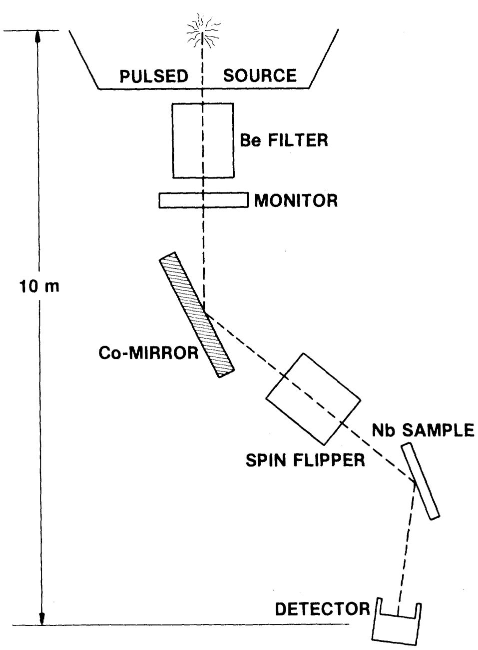

FIG. 1. Layout of the polarized-neutron reflection experiment. The source emits short pulses of neutrons with a thermal spectrum. Only the neutrons with wavelength $\lambda > 3.8$ Å pass through the berillium filter, and are polarized by the magnetized cobalt mirror. The polarized beam is partially reflected by the niobium surface in a magnetic field and detected in time of flight by the counter. The magnetic fields (and the quantization axis of the neutron spins) are normal to the flow plane of the page. Subsequent measurements are taken for the two states of the neutron spin: this is reversed by energizing the flipper.

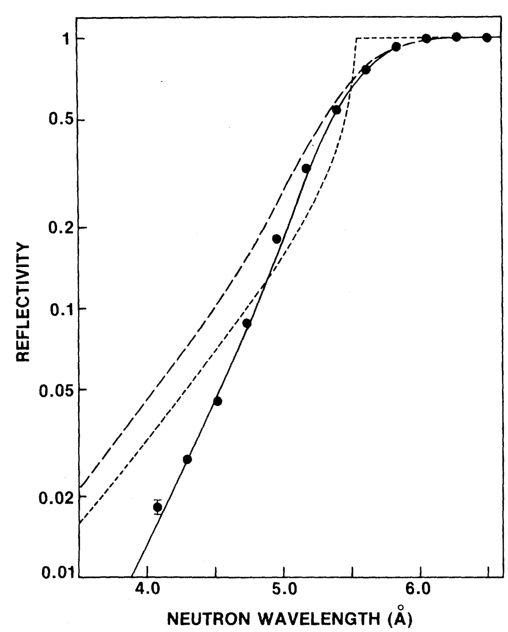

FIG. 2. Reflectivity of niobium (5-μm film on silicon) at 10 K. Dotted line, calculated reflectivity for an angle of incidence $\theta_i=0.36°$. Dashed line, the same, but with a Gaussian spread of angles $\Delta\theta_i=0.02°$. Continuous line, the effect of a surface roughness ($\langle z_D^2\rangle^{1/2}=70$ Å) has been added.

(dashed curve) is consistently higher than the data at the shorter wavelengths. This effect, often encountered in x-ray reflectivity measurements, has been attributed to surface roughness, which modifies the reflectivity[2] like a Debye-Waller factor:

$$I(\lambda)=I_0(\lambda)\exp\left[-\frac{16\pi^2}{\lambda^2}\theta_i p_0\langle z_D^2\rangle\right]. \quad (4)$$

The least-squares fitting to our data gives an effective roughness $\langle z_D^2\rangle^{1/2}=70$ Å, and is shown as the solid curve in Fig. 2.

The spin dependence of the reflectivity measured at $T=4.6$ K, $H=500$ Oe, and $\theta_i=0.34°$ is presented in Fig. 3 in the form of the flipping ratio. The calculated value of the F_R for various values of Λ with the exponential field profile are also shown. The data indicate $\Lambda=430\pm40$ Å, a value consistent with similar runs at $H=700$ Oe or at a different angle of incidence ($\theta_i=0.30°$). At 7 K, the fit gives $\Lambda=600\pm80$ Å, showing the expected increase of Λ with T. From these data, the zero-temperature value $\Lambda(0)$ can be estimated, by use of the Gorter-Casimir or BCS theories, to be 420 ± 40 and 400 ± 40 Å, respectively. Results of others[10,11] indicate that the temperature dependence for niobium lies between these theoretical limits, implying that $\Lambda=410\pm40$ Å.

Several methods have been used previously to determine $\Lambda(0)$. Maxfield and McLean[10] have made high-precision measurements of $d\Lambda/dT$ using

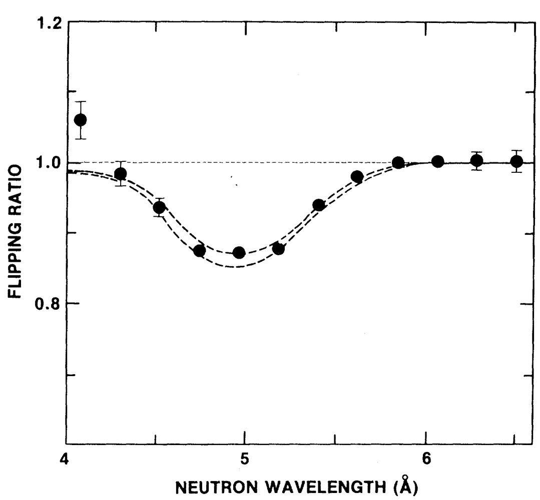

FIG. 3. The flipping ratio for the niobium film at 4.6 K and 500 Oe. $\theta_i=0.34\pm0.02°$. The lines drawn are calculated, in ascending order, for $\Lambda=380$ and 480 Å. The minimum (resolution broadened) for an exponential field decay occurs close to the critical wavelength $\lambda_c^-=5.1$ Å.

rf techniques. Although there is no rigorous way to determine the constant of integration $\Lambda(0)$ from $d\Lambda/dT$, they made a model-dependent estimate of 470 ± 50 Å. A second method relies on a result of the local Ginzburg-Landau theory[11]:

$$\Lambda(T)=[H_{c2}(T)\phi_0/4\pi]^{1/2}/H_c(T). \quad (5)$$

For suitably reversible samples (i.e., negligible flux pinning) both the thermodynamic critical field H_c and the upper critical field H_{c2} can be determined by the magnetization. Measurements by Finnemore, Stromberg, and Swenson[11] and French[12] were extrapolated to zero temperature yielding 410 ± 10 and 397 ± 10 Å, respectively for $\Lambda(0)$. Auer and Ullmaier[13] prefer to perform magnetization measurements closer to T_{c1} in a region where nonlocal corrections to Λ can be avoided, and use Eq. (5) in that limit to obtain the London penetration depth, $\Lambda_L(0)=315$ Å. The effect of nonlocality for pure niobium has been estimated[14] to increase $\Lambda(0)$ over $\Lambda_L(0)$ by about 20%. On the theoretical side $\Lambda_L(0)=325$ Å has been calculated[15] from the band structure of niobium. Our *direct* measurement of Λ is in reasonable agreement with the theoretical value as well as those obtained through the use of Eq. (5). Thus it appears that Eq. (5) is a good approximation even when not in the extreme local limit.[16]

The experiment described here shows that the new technique of polarized-neutron reflection is indeed quite promising for detecting magnetic perturbations close to a surface. The perturbations that can be observed are for thicknesses between 10 and 5000 Å, the lower boundary being due to the neutron counting statistics, and the upper boundary due to the angular resolution. More detailed analysis of the surface (in parallel with the x-ray research such as the surface diffraction recently developed at synchrotron sources[17]) has been thoroughly considered[18-21] and should become rapidly feasible, especially with the advent of the next generation of neutron sources.

This work was supported by the U. S. Department of Energy.

[1] L. G. Parrat, Phys. Rev. **95**, 359 (1954).

[2] L. Névot and P. Croce, Rev. Phys. Appl. **15**, 761 (1980).

[3] D. H. Bilderback, Proc. SPIE Int. Soc. Opt. Eng. **315**, 90 (1981).

[4] A. G. Klein and S. A. Werner, Rep. Prog. Phys. **46**, 259 (1983).

[5]G. P. Felcher, Phys. Rev. B **24**, 1595 (1981).

[6]M. Tinkham, *Introduction to Superconductivity* (McGraw- Hill, New York, 1975), pp. 87-88.

[7]M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1975), Chap. 1.

[8]R. Jacobsson, in *Progress in Optics,* edited by E. Wolf (North-Holland, Amsterdam, 1966), Vol. 5. The expression in this paper was derived by use of Eq. (2.42) appearing on p. 258.

[9]L. Koester, in *Neutron Physics,* Springer Tracts in Modern Physics Vol. 80 (Springer, Berlin, 1977).

[10]B. W. Maxfield and W. L. McLean, Phys. Rev. **139**A, 1515 (1965).

[11]D. K. Finnemore, T. F. Stromberg, and C. A. Swenson, Phys. Rev. **149**, 231 (1966).

[12]R. A. French, Cryogenics **7**, 52 (1967).

[13]J. Auer and H. Ullmaier, Phys. Rev. B **7**, 136 (1973).

[14]P. B. Miller, Phys. Rev. **113**, 1209 (1959).

[15]R. Blaschke, J. Ashkenazi, O. Pictet, D. D. Koelling, A. T. van Kessel, and F. M. Mueller, J. Phys. F **14**, 175 (1984).

[16]A different (and potentially direct) measurement of Λ is provided by the diffraction pattern seen in Josephson tunnel junctions in a small parallel magnetic field. However, R. F. Broom, J. Appl. Phys. **47**, 5432 (1976); and A. M. Cucolo, S. Pace, R. Vaglio, A. DiChiara, G. Peluso, and M. Russo, J. Low Temp. Phys. **50**, 301 (1983), obtained for Nb the rather surprising result $\Lambda(0) = 900$ Å.

[17]P. Eisenberger and W. C. Marra, Phys. Rev. Lett. **46**, 1081 (1981).

[18]G. Vineyard, Phys. Rev. B **26**, 4146 (1982).

[19]P. Mazur and D. L. Mills, Phys. Rev. B **26**, 5175 (1982).

[20]A. Zeilinger and T. J. Beatty, Phys. Rev. B **27**, 7239 (1983).

[21]S. Dietrich and H. Wagner, Phys. Rev. Lett. **51** , 1469 (1983).