Study of the intermediate mixed state of niobium by small-angle neutron scattering*

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A precise study of fluxoid-line lattice (FLL) in the intermediate mixed state (IMS) in a niobium-crystal sphere has been made with a neutron-scattering technique. Accurate FLL unit-cell size determinations are facilitated by the high angular resolution of the technique so that reversibility of the microscopic flux density with applied-field history in and near the IMS can be demonstrated for the first time. Structural changes associated with hysteresis in the IMS are revealed through integrated intensities and full widths at half-maxima of the FLL rocking curves.

This study represents the first quantitative neutron-scattering investigation of the intermediate mixed state (IMS) in a type-II superconductor. The attractive interaction between fluxoid lines gives rise to a limiting maximum flux-line spacing so that the magnetization jumps discontinuously to a specific value at H_{c1}. In a sample having a nonzero demagnetizing factor, the superconducting structure breaks into a mixture of Meissner and fluxoid-line-lattice (FLL) regions.[1] In the structure the lattice parameter is fixed over an applied-field range $(1-n)H_{c1} < H_a < (1-n)H_{c1} + nB_0$, where B_0 is the limiting flux density and n is the demagnetizing factor. A spherical single crystal of niobium was selected for this study to provide a demagnetizing factor equal to $\frac{1}{3}$ while at the same time eliminating influences due to geometrical anisotropy.[2] In these experiments it is shown that, on a microscopic scale, the transition with field from the mixed state into the IMS is sharp and is independent of applied-field history. Furthermore, the observations reported here of constant flux-line spacing in the IMS are made in near-equilibrium conditions and therefore they must be distinguished from trapped-flux situations in previous neutron-scattering[3] studies. In those studies it is certain that an equilibrium IMS could not have been measured since trapped flux was retained into what should have been a Meissner-state regime.

The extremely high resolution required in this small-angle neutron-scattering experiment is achieved by placing the niobium sample between two near-perfect crystals which are arranged in the "parallel" geometry.[4-6] This system is shown schematically in Fig. 1. For the present studies, we have used less-than-perfect Si crystals (~12 arc sec mosaic spread), which provided a good resolution-intensity compromise.

The niobium crystal used for the present studies was float-zone grown from very-high-purity stock, spark machined into a 14.4-mm-diam sphere,[7] and chemically polished and annealed for 50 h at 2150° C in a vacuum of 2.6×10^{-10} Torr. The sample was mounted in the horizontal neutron beam with its ⟨111⟩ crystal axis vertical and parallel to the applied field of a split-coil superconducting magnet. Incident neutrons of mean wavelength $\lambda = 2.398$ Å which are Bragg scattered from a given set of FLL planes are detected by rotating the Si analyzer crystal through an angle 2θ from its position for reflection of the direct beam. The excellent separation of a typical FLL (10) diffraction peak from the direct-beam peak is shown in Fig. 1. The expected hexagonal symmetry[8] of the FLL was confirmed by checking the consistency of three successive (10)-type reflections.

For this symmetry, one may calculate from the Bragg scattering angle 2θ the flux density B, averaged over a unit cell of the FLL,

$$B = \phi_0 \tfrac{1}{2}\sqrt{3}\,(2\theta/\lambda)^2 , \qquad (1)$$

where $\phi_0 = 2.07\times10^{-7}$ G cm^2 is the flux quantum.

The experiments were done at one arbitrary temperature $T = 5.06$ K. Each applied field H_a was established either by increasing the field from the Meissner state, or by decreasing the field from above H_{c2}. The (10) scattering angle and calculated flux densities B as a function of applied-field

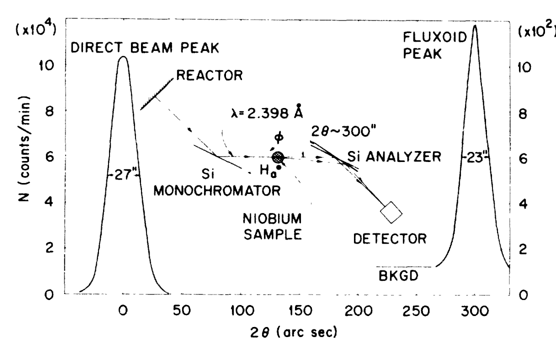

FIG. 1. Schematic representation of the experimental setup in real and reciprocal space. Typical direct-beam and (10) fluxoid Bragg-diffracted peaks.

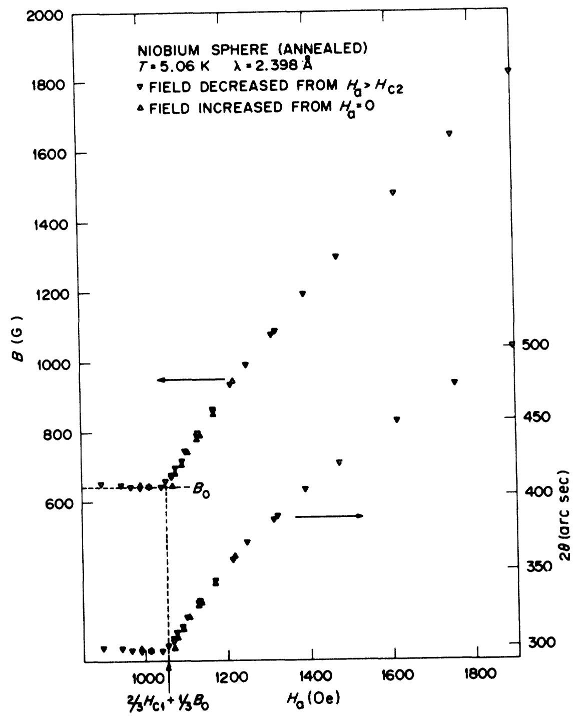

FIG. 2. Scattering angle 2θ for the (10) fluxoid Bragg reflection, and the calculated flux density $B(2\theta)$ vs the applied field H_a at the fixed temperature $T=5.06$ K.

and field history are shown in Fig. 2. It is to be noted that there is negligible dependence of the FLL parameter on field history. This lack of magnetic hysteresis in the FLL unit-cell size is important in view of the well-known surface-state influence on the reversibility of homogeneous superconductors.[9,10] The present results indicate that the observed field history effects can be interpreted in terms of surface-induced bulk defects in the FLL, rather than surface pinning or barriers which would lead to hysteresis in the FLL parameter. We have verified this point by performing a thermal surface treatment,[9,10] which yields a nearly reversible magnetization curve for the sample used in these studies.

The data of Fig. 2 also display an essentially constant lattice parameter for applied fields below $H_a=1059$ Oe. This constant lattice parameter defines $B_0=647\pm 7$ G at this temperature. These observations corroborate the model which interprets the IMS as consisting of constant-parameter FLL domains.[1,3]

Additional details on the behavior of FLL in the IMS were made through integrated intensities from the (10) diffraction peak as a function of applied field and applied-field history in the vicinity of the intermediate mixed state. The integrated intensity, normalized to the incident neutron flux,

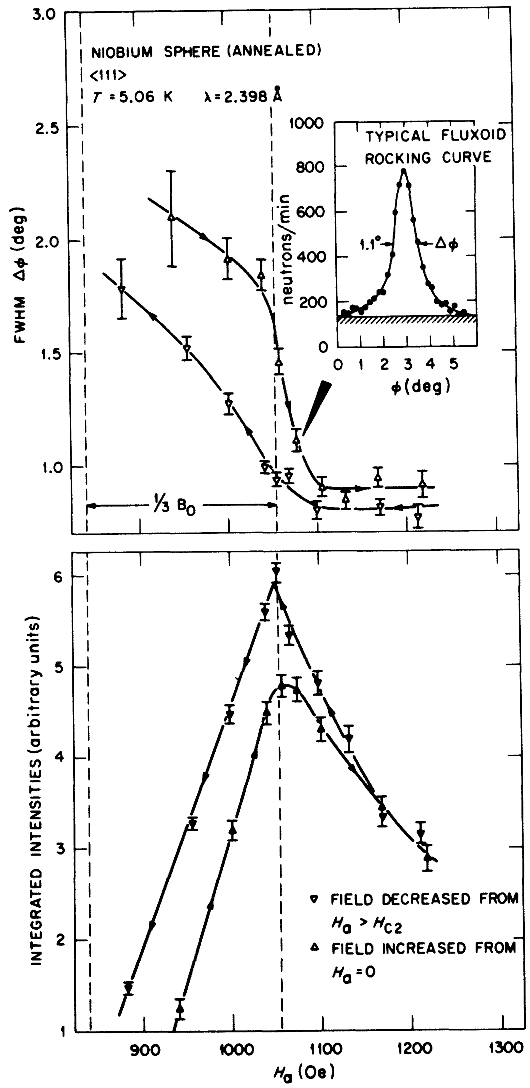

FIG. 3. Fluxoid-lattice rocking-curve integrated intensities and angular full widths at half-maxima vs the applied field H_a at the fixed temperature $T=5.06$ K.

is given by

$$I_{10}(H_a)=\left(\frac{\gamma}{4}\right)^2 \frac{\lambda^2}{\phi_0^2}\frac{2\pi}{G_{10}}\,|h_{10}|^2\eta(H_a)V\,, \qquad (2)$$

where γ is the (magnetic moment)/(nuclear magneton) of the neutron, $\eta(H_a)$ is the volume fraction of constant parameter FLL, V is the sample volume, h_{10} is the form factor for the flux-density distribution within the FLL unit cell,[3] and $\vec{G}_{10}$ is the (10) reciprocal-space vector.

In the IMS, h_{10} can be assumed to be fixed, since the FLL parameter is constant. Therefore, integrated intensities in the IMS are proportional to $\eta(H_a)$. The integrated intensity was measured for the scattering angle fixed at the peak position while the sample was rotated about the applied-

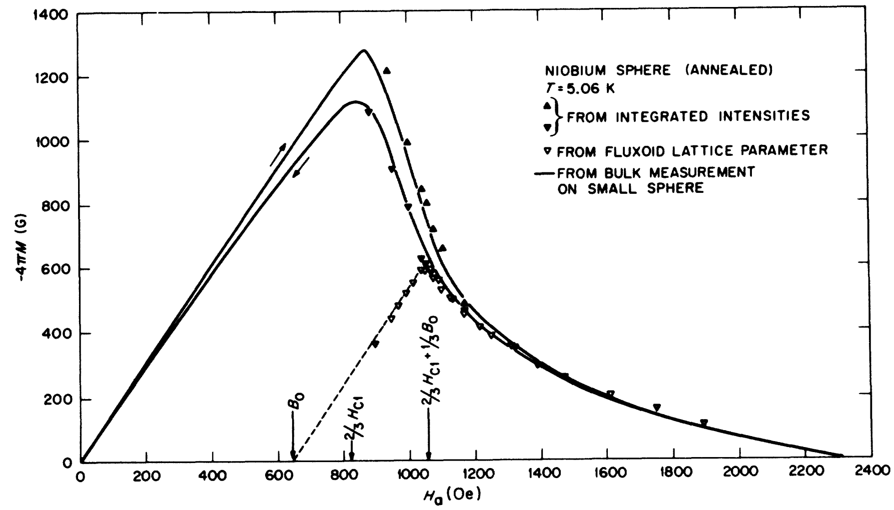

FIG. 4. Magnetization $-4\pi M$ vs the applied field H_a, derived from diffracted-neutron intensities and measured fluxoid-lattice parameters. The full curves are results of magnetometer measurements on a smaller niobium sphere.

field axis. In Fig. 3 the intensity and the full width at half-maximum (FWHM) rocking-curve breadths are shown.

Intensities for field-decreased history are always higher than for field-increased intensity in the IMS. A 20% difference is observed for applied field equal to $\frac{2}{3}H_{c1}+\frac{1}{3}B_0$ (the upper field limit for the IMS). The maximum intensity is found at the upper IMS limit when the field is decreased. Furthermore, extrapolation to zero intensity of this curve yields an applied field $H_a=\frac{2}{3}H_{c1}$. This self-consistency on decreasing field implies $\eta=1$ at the upper limit of the IMS.

In Fig. 3 an abrupt variation in the rocking-curve FWHM can be seen. At a field just below the IMS limit, the FWHM on increasing field drops sharply. The FWHM of the rocking curve is assumed to be due to particle-size broadening and misorientation of subregions on FLL. As the applied field is increased from $\frac{2}{3}H_{c1}$, flux lines enter the sample to form "islands" of FLL structure which nucleate and grow. Hence, the intensity increases and particle-size broadening decreases as shown. At a field just below the IMS limit, we suggest that the "islands" achieve connectivity so that a coherent FLL is formed in the entire sample volume with residual regions of Meissner flux-free voids. Particle-size broadening is then cancelled, and only a void scattering and normal FLL mosaic contribute to broadening. If the field is reversed, we suggest that the FLL retains coherent connectivity well into the intermediate state, with Meissner voids growing within the FLL, with the result that the FWHM is smaller for the decreased-field IMS.

Bulk magnetization measurements have been made for comparison with the neutron-diffraction results. It is seen in Fig. 4 that the decreased-field data in the mixed state are in essential agreement with magnetizations calculated from the neutron results according to

$$-4\pi M=\tfrac{3}{2}(H_a-\overline{B}) , \tag{3}$$

where the average bulk-flux density $\overline{B}$ is just that obtained from Eq. (1). For the IMS and increased-field mixed-state regions, $\overline{B}$ is calculated using Equation (1) in conjunction with the integrated intensity measurements

$$\overline{B}=B(2\theta)(I_{10}/I_{max})=B(2\theta)\eta . \tag{4}$$

I_{max} is the intensity obtained at $\frac{2}{3}H_{c1}+\frac{1}{3}B_0$ in the field-decreased case, where it is seen that the specimen is entirely filled with FLL. The agreement demonstrates that the bulk-magnetization hysteresis is well described by a microscopic interpretation of diffraction intensity results.

Reversibility of the microscopic flux density of a niobium sphere has been demonstrated in precise lattice-parameter determinations along with the sharp transition between mixed and intermediate mixed states. Magnetization irreversibilities measured at a macroscopic scale are well interpreted in terms of gross FLL imperfections which depend on surface entrance and exit of flux, rather than irreversibility at the level of the FLL unit cell. These investigations are being continued.

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