

Calculation of the diamagnetism of the surface sheath of superconductivity

Roberto Felici and K. E. Gray

Materials Science and Technology Division, Argonne National Laboratory, Argonne, Illinois 60439

(Received 27 December 1983)

The nonlinear Ginzburg-Landau equations are solved for the surface sheath of superconductivity for applied fields H_a between H_{c2} and H_{c3} . The resultant diamagnetism is found to be largest for $\kappa \sim 1/\sqrt{2}$ and $H_a \rightarrow H_{c2}$. We find that the field decrease in the surface sheath is sufficiently large to be observable by low-angle polarized neutron reflections.

INTRODUCTION

A new technique to study surface magnetism using polarized neutrons has been recently proposed^{1,2} and is currently being developed. Any variation in the magnetic induction B close to the surface will alter the relative reflections of neutrons polarized parallel and antiparallel to B . As such, the dependence of these reflectivities on scattering angle or neutron wavelength can provide information on the spatial dependence of B relative to the surface.

In the case of a type-II superconductor in an external magnetic field (below H_{c1}), $B(z)$ is nonzero only close to the surface; numerical calculations² appropriate to niobium show that neutron reflection can determine the penetration length λ to within 10%. Above H_{c1} the neutron scattering is complicated by scattering from the flux-line lattice in the superconductor.

However, another intriguing possibility is to directly observe the diamagnetism (field expulsion) associated with the surface sheath of superconductivity predicted³ to occur between H_{c2} and $H_{c3} \cong 1.7H_{c2}$. The practicality of such an experiment depends on the size of the field reduction associated with this surface sheath. The Ginzburg-Landau (GL) equations⁴ form a suitable basis for calculations, although they are strictly valid only in the high- κ limit ($\kappa = \lambda/\xi$, where ξ is the superconducting coherence length) since the GL theory is a local one and low- κ superconductors are known to be nonlocal.⁵ The nonlinear equations can be linearized³ near H_{c3} where the superconducting order parameter ψ is much less than its value in zero field ψ_∞ . However, for arbitrary fields, one must resort to numerical integrations of the nonlinear equations. Such calculations for the surface sheath have been done previously,⁶ but are inadequate for our current purposes since, for example, $B(z)$ was not obtained explicitly.

PROCEDURE

Consider a semi-infinite half-space with a superconductor on the right and a vacuum on the left. The dimensionless GL equations written⁷ in reduced units are as follows:

$$f'' = \kappa^2 [(b')^2/f^3 + f^3 - f], \quad (1)$$

$$b'' = f^2 b + 2f'b'/f, \quad (2)$$

where $f = \psi/\psi_\infty$ is the local, reduced order parameter, $b = 2\pi\xi^2\kappa B/\phi_0$ is the reduced magnetic induction, ϕ_0 is the flux quantum, and the length scale of the derivatives is the penetration depth λ . Our calculation differs from that of Ref. 6 in several significant ways. Whereas Fink and Presson used an analog computer, our procedure relies on the predictor-corrector algorithm of Gear⁸ on a digital computer using double precision. Additionally, we calculate b explicitly as well as f . Most importantly, the boundary condition on b , deep in the superconducting half-space, is different; we always set it equal to the applied field, thus avoiding the infinite energy which would be associated with diamagnetism in the infinite half-space far away from the boundary (where $f=0$).

The presence of the vacuum interface simplifies the initial conditions since⁹ it implies $f'(0)=0$. Thus we only need to determine $f(0)$ and $b'(0)$ since $b(0)$ is the applied field. The method we have used to find the approximate solution in the $f(0)-b'(0)$ plane begins by our choosing values for $f(0)$ and $b'(0)$ and then determining the first nonphysical behavior of one of the parameters f or b as the integration proceeds from the vacuum interface. Thus, points in the $f(0)-b'(0)$ plane are characterized by whichever of the following that occurs first: $f > 1$, $f < 0$, $b < 0$, or $b > b(0)$, the applied field. As a result, the $f(0)-b'(0)$ plane consists of regions in which a particular nonphysical behavior occurs. For the surface sheath, it is apparent from such plots that the correct solution occurs along the line separating the $f < 0$ and $b > b(0)$ regions. To establish the position along this line, one seeks the lowest Gibb's free energy per unit surface area, ΔG , where

$$\Delta G = \frac{H_c^2}{8\pi} \int_0^\infty \{2[b(0)-b]^2 - f^4\} dz, \quad (3)$$

and H_c is the thermodynamic critical field. Since one can never determine the correct initial conditions precisely, we have calculated the behavior of b and f near the interface for points in the vicinity of the minimum. They are essentially indistinguishable, and thus the solution is not so singular that greater-than-double precision is needed to specify a useful approximation. In addition, there is a discontinuous change in the slope of the line separating the two solutions at the place where ΔG is a minimum.

The field penetration below H_{c1} can also be calculated to check the overall procedure. The results shown below are entirely consistent with expectations—an approxi-

mately exponential decay of the field over a distance λ . These calculations also provide a better comparison with polarized-neutron-reflection results at such fields,¹⁰ since if f is significantly less than 1 at the surface, there will be deviations from a precise exponential field penetration.

RESULTS

Not surprisingly, the diamagnetism decreases as the applied field increases from H_{c2} to H_{c3} . The diamagnetism at $H_a = 1.1H_{c2}$ is plotted in Fig. 1 against distance from the surface for several values of κ , the only other free parameter in the GL equations. Note that the surface sheath is energetically favorable for all $\kappa > 0.418$, even though the superconductor is type I for $\kappa < 1/\sqrt{2} \approx 0.707$. In this latter case the applied field for the calculation shown in Fig. 1 is 1.1 times the thermodynamic critical field H_c . The Gibbs free energies per unit area for these solutions are shown as the circles in Fig. 2 as a function of κ . Note that the negative of the Gibbs energy as well as the diamagnetism shown in Fig. 1 is maximum for $\kappa = 1/\sqrt{2}$. This is an artifact coming about because $H_c > H_{c2}$ in the type-I regime, and thus the applied field is closer to H_{c3} . The dashed line of Fig. 2 shows the behavior for $\kappa < 1/\sqrt{2}$ when the ratio H_a/H_{c2} is maintained constant at 1.3 for all κ values.

The behavior of b and f as a function of distance from the surface for various applied-field values is shown in Figs. 3–5, in each case with a different value of κ . The value 0.726 represents pure bulk niobium ($\sim 10 \mu\text{m}$) sputtered onto room-temperature silicon substrates, and the value of 0.6 shows the effect of a type-I film as could be achieved, for example, with a dilute Bi in Pb alloy.

The implication for type-I superconductors of the data in Fig. 5 for $H_a = H_c$ is interesting. At this field, the free

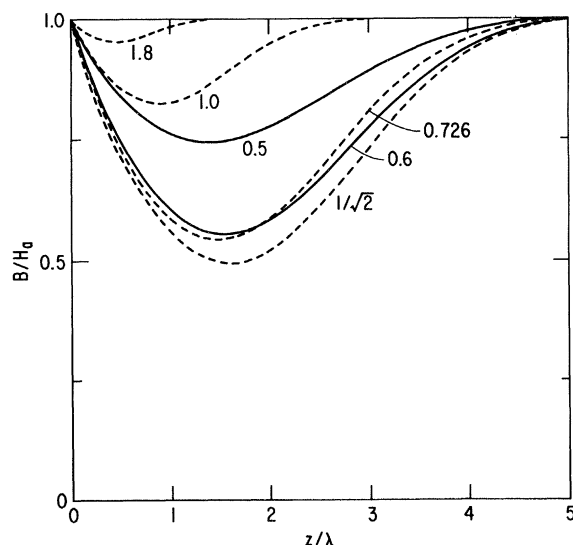


FIG. 1. Magnetic induction in the surface sheath is shown for various values of the Ginzburg-Landau parameter κ as a function of distance from the surface of the superconductor. The applied field H_a is $1.1H_{c2}$ for $\kappa > 1/\sqrt{2}$ (dashed lines) and $1.1H_c$ for $\kappa < 1/\sqrt{2}$ (solid lines).

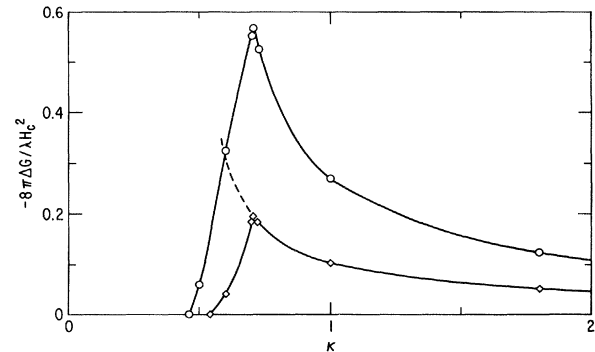


FIG. 2. Gibbs free energies per unit area for the solutions shown in Fig. 1 are plotted as a function of κ (circles). Also included are the results (diamonds) for a larger applied field ($\times 1.18$). The points at $\Delta G = 0$ are determined from the linearized GL equations.³ See text for dashed curve.

energies per unit volume of the fully superconducting and fully normal states are, by definition, equal in the absence of surfaces. Therefore the surface sheath, which has a negative free energy with respect to the normal state, is thermodynamically stable. For the sample considered here, which fills the infinite half-space, the fully superconducting state becomes energetically favorable when H_a is infinitesimally less than H_c , resulting in a first-order phase transition. This is because an arbitrarily small

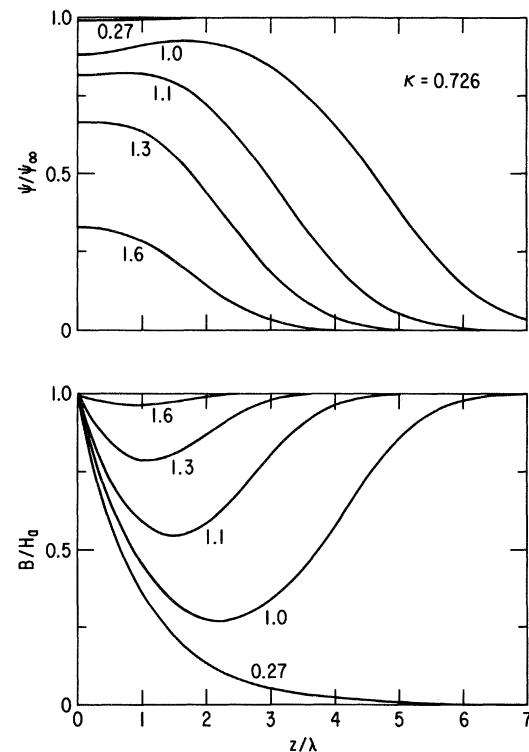
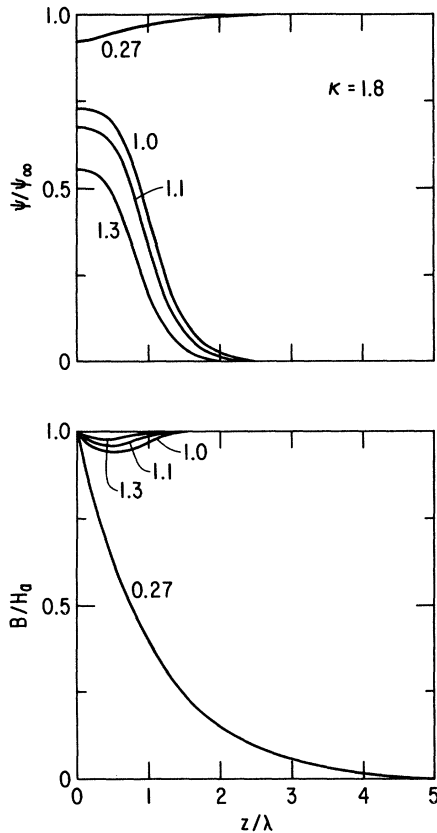


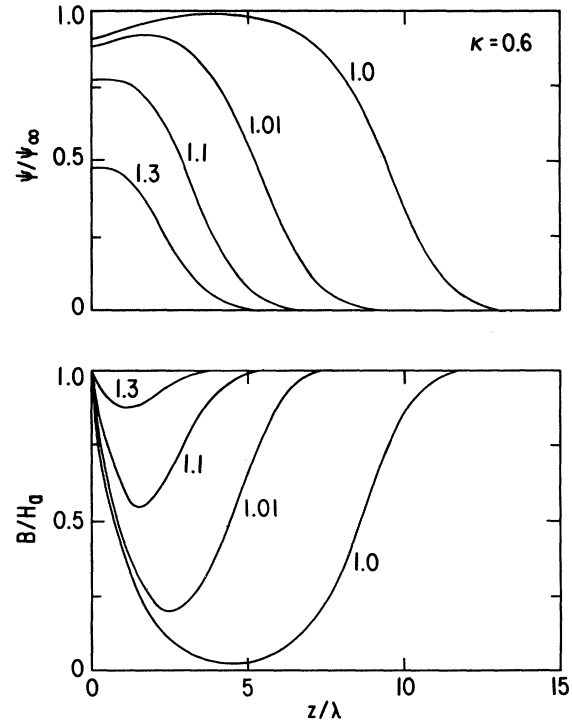
FIG. 3. Magnetic induction (bottom) and the superconducting order parameter (top) are plotted as a function of distance from the surface of the superconductor for $\kappa = 0.726$ and for various indicated values of the reduced applied field H_a/H_{c2} . The case of $H_a = 0.27H_{c2}$ corresponds to the usual field penetration below H_{c1} , while the others are for the surface sheath.

FIG. 4. Same as Fig. 3 except $\kappa=1.8$.

free-energy difference per unit volume will overwhelm the finite free energy per unit area of the surface sheath. Using the same reasoning, it might be expected that for finite-size samples the fully superconducting state only becomes energetically favorable when H_a is a finite amount smaller than H_c . However, within the double precision of our computation, there does not appear to be an acceptable surface-sheath solution in a field of $0.999H_c$, thus indicating that the transition will be first order at H_c , rather than at a lower field or by a continuous increase in the size of the surface sheath.

SUMMARY

The nonlinear Ginzburg-Landau equations have been integrated to find the spatial variation of the magnetic field in the surface sheath of superconductivity for $H_{c2} < H_a < H_{c3}$. The diamagnetism is largest for low κ values and drops off significantly at $\kappa=1.8$. The

FIG. 5. Same as Fig. 3 except $\kappa=0.6$, and the indicated values of reduced applied field correspond to H_a/H_c .

diamagnetism can be large enough to be observable by polarized neutron reflections,^{1,2} and thus it may be possible to directly detect for the first time the surface sheath of superconductivity and determine its spatial extent. However, a low value of κ may be a practical necessity for its observation. It should be reiterated that the results of our GL approach will be only approximate for small κ values because such superconductors are nonlocal. However, these calculations should provide a basis for planning experiments, and it will be quite interesting to see how important the corrections are for nonlocality.

ACKNOWLEDGMENTS

The authors would like to acknowledge the interest and support of Gian Felcher. One of us (K.E.G.) thanks Chia-Ren Hu for useful discussions and interest in the problem. These calculations were carried out on the Intense Pulsed Neutron Source computer at Argonne. We are grateful for its use. This work was supported by the U.S. Department of Energy.

¹G. P. Felcher, Phys. Rev. B **24**, 1595 (1981).

²S. D. Bader, G. P. Felcher, K. E. Gray, and R. T. Kampwirth, Physica **120B**, 219 (1983).

³D. Saint-James and P. G. deGennes, Phys. Lett. **7**, 306 (1963).

⁴V. L. Ginzburg and L. D. Landau, Zh. Eksp. Teor. Fiz. **20**, 1064 (1950).

⁵A. B. Pippard, Proc. R. Soc. London, Ser. A **216**, 547 (1953).

⁶H. J. Fink and A. G. Presson, Phys. Rev. B **1**, 1091 (1970).

⁷K. E. Gray, Phys. Rev. B **27**, 4157 (1983).

⁸C. W. Gear, Argonne National Laboratory Report No. 7126, Argonne National Laboratory (unpublished); *Numerical Initial Value Problems in Ordinary Differential Equations* (Prentice-Hall, Englewood Cliffs, N.J., 1971).

⁹M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 130.

¹⁰G. P. Felcher, R. T. Kampwirth, K. E. Gray, and R. Felici, Phys. Rev. Lett. **52**, 1539 (1984).