Anomalous behavior of the diamagnetic profile of superconducting niobium near a vacuum boundary

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The profile of a static magnetic field penetrating into superconducting niobium films has been measured by specular reflection of polarized thermal neutrons. At a temperature of 4.9 K, a field of 500 Oe penetrates essentially unweakened to a depth $\xi = 28$ nm at a vacuum interface. The reason is a suppression of the order parameter of the superconductor near the surface. Deeper in the film, the field falls off by a London law with a constant $\Lambda = 45$ nm. The measurements were carried out on the SPN polarized-neutron spectrometer of the IBR-2 pulsed reactor of the Joint Institute for Nucelar Research at Dubna.

As was demonstrated in the pioneering study by Felcher *et al.*,[1] the specular reflection of polarized thermal neutrons can serve as a direct method for measuring the absolute value of the depth (Λ) to which a magnetic field penetrates into a superconductor. As those investigators pointed out, however, after the first measurements on niobium, the situation did not look entirely satisfactory. Another method for determining Λ, and a sensitive one, involves measuring the direct current in Josephson tunnel junctions in a weak magnetic field.[2] This method has yielded a value $\Lambda = 90$ nm (referred to absolute zero) in niobium films. That result is considerably larger than the $\Lambda = 41$ nm found in Ref. 1. That contradiction is cleared up by the present study, which was undertaken in an effort to refine the picture of the penetration of a static magnetic field into a niobium superconductor in the Meissner phase.

We carried out experiments on two niobium films, grown by the same deposition method but differing in thickness and roughness. The experiments were carried out on the polarized-neutron spectrometer at the IBR-2 high-flux reactor at Dubna under neutron-reflectometer conditions.[3] We first adjusted the reflectometer for a glancing angle $\theta = 0.004$ rad with $\delta\theta/\theta = 0.025$. Two measurements were then carried out in succession. First, we measured the spectra of specularly reflected neutrons at room temperature. From these spectra we found the neutron-optics properties (including the thickness and roughness) of the film. The measurements were then repeated at 4.9 K in a magnetic field of 500 Oe, directed parallel to the plane of the film. In analyzing the experimental results we used a new method which we have developed for calculating reflection coefficients. This method, described in Ref. 4, involves replacing the continuous one-dimensional neutron-optics potential of the film by a discrete series of Fermi quasipotentials, with the goal of modeling the reflection of plane waves from a medium which is inhomogeneous in a single dimension. This approach differs from the standard approach[1] in that the roughness is taken into account in the step of calculating the reflection coefficient. This is done by introducing increasing amplitudes of the Fermi potentials in accordance with a Gaussian error function with a variance equal to the square of the roughness depth parameter σ. Separately, we analyzed the equivalence of the results found by these two methods for dealing with the roughness, using purely nuclear potentials. The ranges of applicability of the methods were found to be identical. In the case of an interference of nuclear and magnetic reflection, these approaches for dealing with the roughness are not equivalent. We discussed that situation in Ref. 4. In particular, we found that taking the roughness into account incorrectly can cause the value found for Λ from the experiments to be much too low.

The particular features of oblique deposition have the consequence that the "thin" film of the two used in these measurements, with an area of 28×50 mm, on a silicon substrate, has a smoothly varying thickness. The average thickness over the area found from the neutron measurements is 265 nm; the variations are within ± 15 nm, with $\sigma<0.5$ nm. The "thick" film, on a substrate of Sitall (a glass-ceramic), was grown by oblique deposition in an effort to increase the roughness. For this film we found $\sigma=8$ nm. Its thickness was 700 nm according to Rutherford backscattering of helium ions. The superconducting transition temperature was 8.95 K for both films. Studies of the composition of the starting material for the deposition of the niobium and studies of the films themselves by neutron-activation analysis revealed that, aside from the 0.3% concentration of tantalum, the concentration of other impurities was less than 10^{-5}. Additional information on the film composition was found by resonant backscattering of helium ions by oxygen. No oxygen impurity was found, at the 3% sensitivity of these measurements. The films which we obtained by this deposition method generally exhibited superconducting properties at thicknesses starting at 40 nm at liquid-helium temperature.

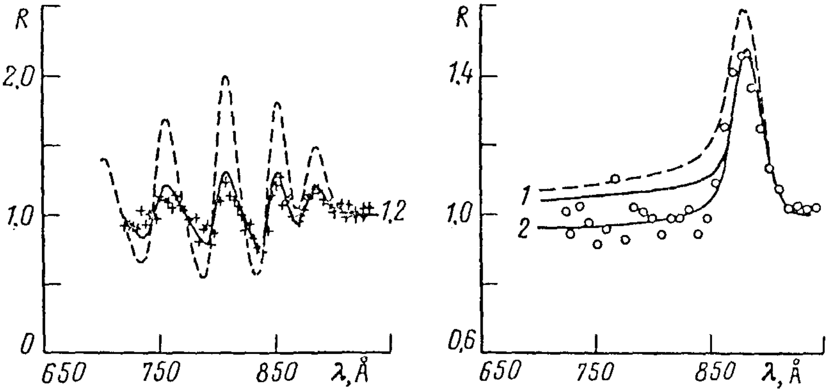

FIG. 1. Experimental ratio (R) of the reflection coefficients for neutrons with opposite polarizations versus the component of the neutron wavelength λ normal to the plane of the film. Plus signs—Thin film; squares—thick film; dashed lines—theoretical, according to the London model with $\Lambda=43$ nm; solids lines *1*—theoretical, for the London model, with a best fit with the value $\Lambda=95$ nm for the thin film and $\Lambda=90$ nm for the thick one; solid lines *2*—model with a dead layer near the film boundaries. This refinement leads to a consistent solution with $\xi=28$ nm and $\Lambda=45$ nm (for both films).

The first step in the modeling of the experiment was to use the idea of the profile of the diamagnetic response of a film which follows from the local London electrodynamics of superconductors. Figure 1 shows the ratio (R) of the reflection coefficients for neutrons with opposite polarizations versus the component of the neutron wavelength λ normal to the plane of the film. Solid curves *1* were found by fitting a London model, with $\Lambda = 95$ nm for the thin film and $\Lambda = 90$ nm for the thick one, to the experimental data. These figures correspond to the best fit of the experimental curves by this model. The results of an analysis of the experimental data by this model have been published previously.[5] (The value $\Lambda = 145$ nm which we reported in Ref. 5 for the "thin" film was too high because of a technical error made in the calculations.) The dashed curves in Fig. 1 correspond to a calculation with the value $\Lambda = 43$ nm, which was found in Ref. 1.

It might seem at first glance that this analysis of the data by the London model confirms the result of Ref. 2 for Λ. However, a closer look is necessary. The experimental values of the spectral R function for the thick film at wavelengths λ less than 850 Å are systematically lower than theoretical curve *1*, indicating a value $R < 1$ in this interval, on the average. A corresponding property of the R function can be seen in Ref. 1 at the edge of the spectral interval at one experimental point, but this circumstance was not mentioned by the authors. In a later study,[6] of the superconductor $Pb_{0.992}Bi_{0.008}$, the corresponding property of the R function was detected more reliably. This aspect of the behavior of the R function is thus more general in nature—not peculiar to superconducting niobium. The analysis below shows that this behavior reflects an unusual state of the diamagnetic response of the film, which is not the state predicted by London electrodynamics.

Anomalies in measurements of direct tunneling currents in Josephson junctions similar to the anomaly for vanadium have periodically been mentioned in the literature.[2] These anomalies have been attributed to a suppression of the order parameter near the surface of the niobium.[7] Let us take a closer look at this hypothesis as it applies to our own results. We assume that there may be a suppression of the order parameter at the actual interface between niobium and vacuum and that this suppression gives rise to an effective "dead layer" down to a depth ξ, at both boundaries. The reason for this behavior of the superconductor may be a fundamental one, reflecting a property of a real surface. This hypothesis does not fit into the phenomenological picture of either London electrodynamics or the Ginzburg–Landau theory. It apparently requires the construction of a microscopic model to explain the field penetration across a real vacuum interface. The Pippard version of electrodynamics leads to a correction on the order of 20% to the penetration depth, so it would not improve the description of the experimental results. So far, there has been a stronger case for assuming a dead layer near the substrate, resulting from a proximity effect or some other phsyicochemical factor, as was done in Ref. 7.

The introduction of a dead layer at the vacuum interface in an effort to interpret the data on the thick film proves to be of decisive importance, since the state of the film–substrate interface has essentially no effect on the R function in this case. One reason is the large film thickness. Another is that there is no film-versus-substrate contrast in the neutron-optics potential. At this stage of the study, we are quite content with a crude model which qualitatively reflects a suppression of the order parameter at both boundaries of the film. We use the following description of the diamagnetic profile: The field penetrates completely to a depth ξ at the film boundaries. Deeper in the film, it obeys a London law with a corresponding penetration depth. After calculating spectral curves of the effect for both films, we found the consistent parameters $\xi = 28 \pm 5$ nm and $\Lambda = 45 \pm 15$ nm. In the process we significantly improved the fit by the theoretical curve of R for the thick film (line *2* in Fig. 1) at $R < 1$. The estimate of χ^2 decreased from 7.5 for line *1* to 1.4 for line *2*. On this basis, we selected the model with a dead layer ξ. The experimental spectral R function for the thin film has the characteristic oscillatory shape discussed in Ref. 4. For the thin film, the introduction of the parameter ξ affects the oscillation amplitude, requiring the introduction of a compensatory decrease in Λ as ξ increases. The noncontradictory description of the data on the two films, with their very different surface roughnesses, adds to our confidence in this modification of the London model. At this stage of the research we cannot assert that the parameter ξ introduced here is related in any way to the Ginzburg–Landau correlation length. Nevertheless, the properties of the films which we used are serious arguments against ξ having a chemical nature (an oxide situated at the surface) and against an interpretation based on a masking of a region of this size by the roughness. The value $\xi = 28$ nm which we found is considerably larger than the roughness parameter of both the thick film ($\sigma = 8$ nm) and, especially, the thin one ($\sigma < 0.5$ nm). A fit of experimental data by the London model in Ref. 1 resulted in the underestimate $\Lambda = 43$ nm, because the roughness was dealt with incorrectly, in our opinion. The structural feature which we found in the R function in this region was not discussed in that earlier study. The agreement between the value found for L in Ref. 1 and estimates which have been found for the London depth by rf methods (see Ref. 1 for citations of that work) looks a bit on the fortuitous side.

Our experimental data thus directly confirm an increase in the order parameter not only at the superconductor–substrate interface, as has been seen indirectly in the anomalies mentioned in Refs. 2 and 7, but also at the vacuum interface.

This effect might be studied in more detail in an experiment much like that reported here but involving measurements of the dependence of the effect on the strength of the external magnetic filed. We intend to carry out such measurements in the next stage of research on this effect.

We note in conclusion that our measurements of the magnetic penetration depth by the method of polarized-neutron reflection have revealed an anomalous behavior of the diamagnetic profile of the superconductor. This behavior can be linked with a suppression of the order parameter at the superconductor–vacuum interface. There is no other way to explain this anomaly in interpreting the experimental data. The observation of this anomaly constitutes a direct observation of the effect.

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