Penetration depth study of very thin superconducting Nb films

Thomas R. Lemberger,* Iulian Hetel, Jacob W. Knepper, and F. Y. Yang
Department of Physics, The Ohio State University, 191 W. Woodruff Ave., Columbus, Ohio 43210-1117, USA
(Received 22 March 2007; revised manuscript received 29 May 2007; published 20 September 2007)

Using a low-frequency two-coil technique, we measure the magnetic penetration depth $\lambda(T)$ of superconducting Nb films with thicknesses 20 Å $\leq d \leq$ 228 Å sputtered onto oxidized Si substrates. We find a phenomenological dependence of T_c on d, $T_c/8.5\text{ K} \approx \tanh(d/70\text{ Å})$ for films thinner than 250 Å. $\lambda^{-2}(T)/\lambda^{-2}(0)$ is well fitted by weak-coupling dirty-limit theory with a weak-coupling gap, $\Delta(0)=1.8k_BT_c$. $\lambda^{-2}(0)$ agrees with dirty-limit theory, given the experimental values of transition temperature T_c and residual resistivity ρ_0. These results indicate that the suppression of T_c is due to mechanisms that weaken the effective pairing interaction and not due to pair breaking interactions.

DOI: 10.1103/PhysRevB.76.094515 PACS number(s): 74.25.Nf, 74.78.Db, 74.62.−c, 74.25.Fy

I. INTRODUCTION

A. Background

Gubin *et al.*[1] recently reported the first systematic study of the magnetic penetration depth λ of superconducting Nb films. They measured films as thin as 80 Å dc sputtered onto Si substrates. In agreement with numerous previous studies, Gubin *et al.* found that resistivity goes up and superconducting transition temperature goes down as Nb film thickness d decreases. They were able to establish that $\lambda(0)$ increases as d decreases in quantitative agreement with dirty-limit BCS theory,[2] given the measured T_c and resistivity. They detected small amounts of carbon on their Nb film surfaces, and hypothesized that surface contaminants result in thin normal layers on the top and bottom of sputtered Nb films, weakening the average pairing interaction and thereby contributing to the reduction of T_c while preserving the applicability of BCS theory. Another possibility is that dc sputtering produces films with surface or volume impurities that are magnetic, and the observed reduction in T_c is due in part to magnetic pair breaking.[3] Differences between these two possibilities grow as d and T_c decrease, motivating the present study of thinner Nb films, down to 20 Å.

The present work extends Gubin *et al.*[1] by studying thinner films and by providing a more detailed study of the T dependence of λ designed to see whether deviations from dirty-limit BCS theory emerge in films thinner than 80 Å. On a technical level, our work complements Gubin *et al.* in that we measure λ with a different, low-frequency method. The fact that we obtain essentially the same values for $\lambda(0)$ for films of similar thickness bolsters our confidence in the accuracy of our measurements and the lab-to-lab and day-to-day reproducibility of dc sputtered Nb films. There have been a number of studies of thin Nb films. Of particular note is the tunneling study of Park and Geballe[4] of Nb films as thin as 9 Å, which were electron-beam evaporated onto sapphire substrates using a protocol that eliminated carbon contamination. They found a clean BCS-like gap in the electron density of states, indicating the absence of pair breaking. Our sputtered films will be compared with e-beam films below.

B. Theory

In dirty-limit theory for superconductivity, where the elastic scattering rate $1/\tau$ is much larger than the superconducting gap $\Delta(0)/\hbar$, the magnitude and T dependence of λ are simple to express. $\lambda^{-2}(T)$ has the form[2]

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \frac{\Delta(T)}{\Delta(0)} \tanh\left[\frac{\Delta(T)}{2k_BT}\right], \tag{1}$$

where k_B is Boltzmann's constant, and the normalized gap $\Delta(T)/\Delta(0)$ is approximated by[5]

$$\frac{\Delta(T)}{\Delta(0)} \approx \left[\cos\left(\frac{\pi T^2}{2T_c^2}\right)\right]^{1/2}. \tag{2}$$

$\lambda^{-2}(0)$ is reduced by scattering such that its magnitude is inversely proportional to residual resistivity ρ_0,

$$\lambda^{-2}(0)|_{BCS} = \frac{\pi\mu_0\Delta(0)}{\hbar\rho_0}. \tag{3}$$

Here, μ_0 is the permeability of vacuum. We measure $\lambda^{-2}(0)$ and ρ_0 and obtain $\Delta(0)$ from fitting $\lambda^{-2}(T)$, so we can test whether dirty-limit BCS applies to our films.

Now we consider what to expect if most of the suppression of T_c is due to a pair breaking interaction.[3] These interactions cause significant deviations from BCS theory. In particular, the peak in the density of states at $E \approx \Delta$ is broadened, and the gap edge shifts down in energy. The density of states is gapless when the pair breaking energy $\hbar/\tau_{pb}$ exceeds Δ. The pair breaking energy $\hbar/\tau_{pb}$ can be estimated from the suppression of T_c below the 9.288 K transition temperature[6] of bulk Nb. For present purposes, we can use the approximate result,[3]

$$k_B(9.3\text{ K} - T_c) \approx \frac{\pi\hbar}{4\tau_{pb}}. \tag{4}$$

As pair breaking lowers the gap edge in the density of states, $\lambda^{-2}(0)$ decreases below its BCS value [Eq. (3)],[3]

$$\frac{\lambda^{-2}(0)}{\lambda^{-2}(0)|_{BCS}} \approx 1 - 0.42\frac{\hbar}{\tau_{pb}\Delta(0)} \quad [\hbar/\tau_{pb}\Delta(0) \leq 1]. \tag{5}$$

For example, a d=32 Å film has $T_c \approx 3.9$ K. Equation (4) then predicts $\hbar/k_B\tau_{pb} \approx 3.5$ K, assuming that half of the suppression of T_c is due to pair breaking. The order parameter can be estimated as $\Delta(0)/k_B \approx 2T_c \approx 7.8$ K. Equation (5) tells us to expect a 20% deviation below BCS in this case. The T

dependence of λ^{-2} should be affected as well. Pair breaking softens the exponentially flat low-T behavior in BCS theory until, in the gapless limit, $\hbar/\tau_{pb} > \Delta(0)$; $\lambda^{-2}(T)$ is approximately quadratic,

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} \approx 1 - \frac{T^2}{T_c^2} \quad [\hbar/\tau_{pb}\Delta(0) > 1]. \tag{6}$$

The foregoing shows that if a significant portion of the decrease in T_c is due to pair breaking, the thinnest films may exhibit deviations from BCS theory, large enough to be discerned experimentally.

II. EXPERIMENTAL DETAILS

Films are dc sputtered from a 2 in. diameter Nb target onto oxidized Si substrates 2 in. above the target. A protective layer of Ge 200 Å thick is dc sputtered immediately after the Nb deposition. Deposition rates are 1.5 and 2.0 Å/s, respectively. Substrates nominally $18 \times 18 \times 0.4$ mm^3 are placed in a load-locked UHV chamber with a background pressure of 5×10^{-10} torr. Substrates are nominally at room temperature during deposition. Film thickness is calibrated by growing a thick film, measuring its thickness with an atomic force microscope, and thereafter assuming that film thicknesses are proportional to sputtering time. We checked that films with the same nominal thickness grown in separate runs have very similar properties.

The sheet conductivity $Y \equiv \sigma_1 d - i\sigma_2 d$ is measured at 50 kHz using a two-coil mutual inductance technique in which coaxial drive and pickup coils are pressed against opposite sides of the center of the sample film.[7,8] The usual conductivity σ_1-$i\sigma_2$ can be obtained from Y/d if the film thickness d is known. The coils are solenoids nominally 2 mm in diameter and 2 mm in length, designed to be much smaller than the areal dimensions of the films. Induced currents lie in the plane of the sample. The ac magnetic field generated by the 50 kHz current in the drive coil is kept low enough that Y is independent of the ac field amplitude. Reducing the applied ac field by a factor of 10 has no effect on the data. The ambient dc magnetic field is reduced by a double μ-metal shield around the He-4 cryostat.

From the mutual inductance measured with the sample present, we subtract the mutual inductance measured with a thick (0.1 mm) superconducting Pb foil in place of the sample. This adjustment removes coupling due to magnetic flux that goes around the sample film as well as stray coupling from other parts of the circuitry. It permits us to proceed as if the films were infinite in area.[7,8] We analyze the normalized mutual inductance, i.e., the ratio of the adjusted mutual inductance to the mutual inductance above T_c, where the sample is undetectable. Normalization cancels out small run-to-run variations in substrate thickness, alignment of coils, etc. To illustrate how all this plays out, consider the worst case. Our thickest film, d=228 Å, screens the mutual inductance from about 210 nH above T_c to about 300 pH at $T \ll T_c$. We measured that a Pb foil with the same areal dimensions as the sample film screens from 210 nH to 44 ± 10 pH. Thus, we deduce that coupling through the sample film is 256 ± 10 pH at low T. The ± 10 pH uncertainty in the Pb foil measurement contributes an uncertainty of $\pm 4\%$ in Y. At higher temperatures and for thinner films, coupling through the film is larger than 256 pH, so the ± 10 pH uncertainty contributes a proportionally smaller uncertainty to Y.

III. EXPERIMENTAL RESULTS

We measure resistivity by cutting the substrate into an approximately 2×12 mm^2 rectangle, then attaching wires via pressed indium pads. Within the crude experimental uncertainty of about 20%, the residual resistivities of our films agree with the phenomenological expression of Gubin *et al.*,[1]

$$\rho_0 \approx \left(3.7 + \frac{1500 \text{ Å}}{d}\right) \mu\Omega \text{ cm}, \tag{7}$$

down to d=32 Å. The dependence of ρ_0 on d is due to surface scattering. Residual resistivity rises faster than this expression predicts when $d < 32$ Å. A similar deviation is observed in e-beam deposited films.[4] To minimize "noise," we use the phenomenological expression for $d \geq 32$ Å and our measured value for $d < 32$ Å.

Our films are in the dirty limit, $\ell \ll \xi_0$, where $\ell = v_F\tau$ is the electron mean free path for elastic scattering, and $\xi_0 = \hbar v_F/\pi\Delta(0)$ is the superconducting coherence length, as we now show. v_F is the Fermi velocity. The value[9] $\rho_0\ell = 3.72 \times 10^{-6}$ $\mu\Omega$ cm^2 is commonly used[1,4] to estimate the electron mean free path of thin Nb films, although its use relies on certain assumptions regarding the similarity of band structure of thin films and bulk Nb. With Eq. (7), ℓ is less than 100 Å,

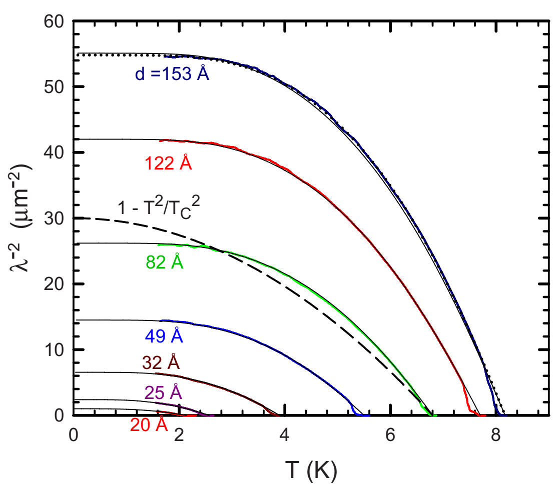

FIG. 1. (Color online) $\lambda^{-2}(T)$ vs T for Nb films with various thicknesses. Dirty-limit BCS theory (thin solid black curves) fits the data well. Best fit values of $\alpha \equiv \Delta(0)/k_B T_c$ are 1.80 for thinner films and perhaps 1.90 for thicker films, $d \geq 153$ Å. Dashed curve is the quadratic predicted by pair breaking theory for gapless superconductors.

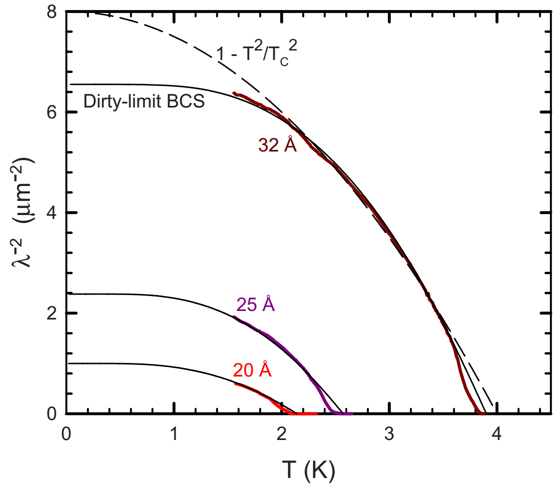

FIG. 2. (Color online) $\lambda^{-2}(T)$ vs T for three thinnest Nb films. BCS theory (thin solid curves) fits data for $d=32$ Å, whereas a quadratic (thin dashed curve) does not.

$$\ell \approx \frac{100\ \text{Å}}{1+400\ \text{Å}/d}. \tag{8}$$

With $v_F \approx 6\times10^5$ m/s,[9] and $\Delta(0)\approx 2k_BT_c$, we estimate a clean-limit coherence length, $\xi_0=\hbar v_F/\pi\Delta(0) > 1000$ Å. Hence, we are confident that our films are well into the dirty limit. The disorder-shortened coherence length, $\xi\approx(\xi_0\ell)^{1/2}$, is larger than d, so our films are two-dimensional superconductors.

From $\sigma_2 d$, we obtain the ratio of areal superfluid density $n_S d$ to effective mass of superconducting electrons: $n_S d/m^* = \omega\sigma_2 d/e^2$, where e is the electronic charge. It is conventional to present the results in terms of the inverse magnetic penetration depth squared $\lambda^{-2}(T)$, which is proportional to n_S/m^*, $d/\lambda^2 \equiv \mu_0\omega\sigma_2 d$. We believe that our method determines d/λ^2 to <3% accuracy for films thinner than 200 Å, and $1/\lambda^2$ to about ±5% accuracy, with the extra uncertainty arising from the roughly 4% uncertainty in d.

TABLE I. Nb film parameters. Residual resistivity ρ_0 is calculated from Eq. (7) for $d\geq 32$ Å and measured for $d=20$ and 25 Å. Residual sheet resistance, $R_N\equiv\rho_0/d$. We could not determine a meaningful value for $\Delta(0)/k_BT_c$ for the 20 Å thick film. $\lambda^{-2}(0)|_{BCS}$ is calculated from Eq. (3) with experimental values for $\Delta(0)$ and ρ_0.

d (Å)	T_c (K) (±0.1 K)	$2\Delta(0)/k_BT_c$ (±0.05)	ρ_0 ($\mu\Omega$ cm) (±10%)	R_N (Ω)	$\lambda^{-2}(0)$ (μm^{-2}) (±5%)	$\lambda^{-2}(0)\vert_{BCS}$ (μm^{-2})
20	2.14	⋯	153	760	1.0±0.25	1.2
25	2.57	1.80	100	400	2.38	2.4
32	3.90	1.80	51	160	6.55	7.1
49	5.50	1.80	34	69	14.3	15
82	6.80	1.80	22	27	26.2	29
122	7.70	1.80	16	13	42.0	45
153	8.17	1.90	13.5	9	54.8	56
228	8.45	1.90	10.3	4.5	73.0	76

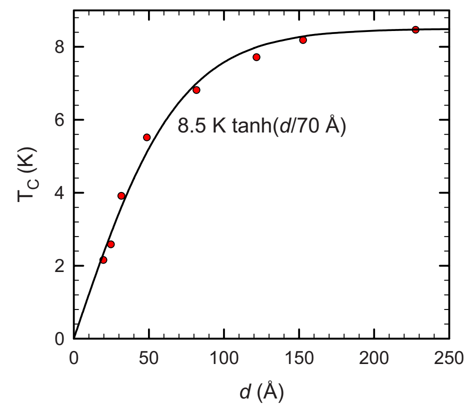

FIG. 3. (Color online) T_c vs d for sputtered Nb films. Black curve is an empirical fit valid for $d<250$ Å.

The main contribution of this paper is the set of $\lambda^{-2}(T)$ vs T curves in Figs. 1 and 2, together with the film parameters in Table I. The thin solid curves in Figs. 1 and 2 are dirty-limit fits with a weak-coupling value of $\alpha\equiv\Delta(0)/k_BT_c = 1.80$. The fits are good, and they yield values for $\lambda^{-2}(0)$ and the mean-field transition temperature T_c. Figure 3 shows the dependence of T_c on d. For the thickest films, it appears that a slightly larger value, $\alpha=1.90$ (dotted curve), fits a little better, suggesting that electron-phonon coupling is stronger in films thicker than 150 Å or so. For comparison, quadratic "fits" (dashed) are shown for the 82 and 32 Å films. The 32 Å film is the thinnest film for which we have data over a wide enough temperature range to distinguish that the BCS fit is better than the quadratic. However, even though the T dependence of $\lambda^{-2}(T)$ for the 20 and 25 Å films can be fitted equally well by either theory, the value of $\lambda^{-2}(0)$ obtained from the BCS fit is consistent with the BCS theory [Eq. (3) and Table I], whereas the value of $\lambda^{-2}(0)$ obtained from the quadratic fit is larger than the BCS prediction, and it would have to be smaller to be consistent with pair breaking theory.

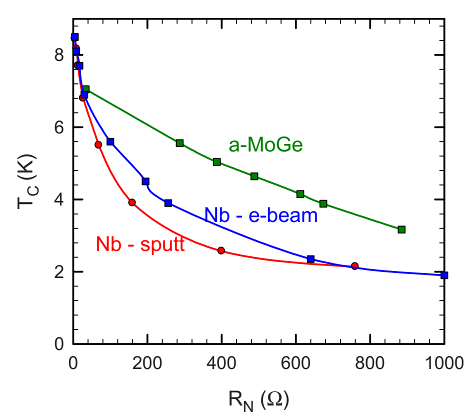

FIG. 4. (Color online) T_c vs sheet resistance R_N for sputtered Nb films, e-beam evaporated Nb films (Ref. 4), and amorphous MoGe films (Ref. 13). Curves are guides to the eye. The six a-MoGe films with $200\ \Omega < R_N < 1000\ \Omega$ have thicknesses from 21.5 to 61 Å, comparable to the five thinnest sputtered Nb films.

Hence, BCS dirty-limit theory provides a consistent description of our data.

As an aside, we note that $\lambda^{-2}(T)$ consistently shows an abrupt downturn as T approaches T_c. We believe that this is due to inhomogeneity in the films, although we have worked very hard to optimize film homogeneity. The downturn is not the vortex-pair unbinding transition expected in two-dimensional superconductors because it occurs at a superfluid density at least a factor of 10 higher than the theory predicts. Slight inhomogeneity of the films does not affect conclusions of this paper.

A comparison between Nb films grown by dc sputtering and e-beam evaporation is interesting. E-beam films consistently have higher T_c's for a given film thickness. A large part of the difference can attributed to the lower resistivities of e-beam films. When we plot T_c vs residual sheet resistance, $R_N = \rho_0/d$ (Fig. 4), we find that the two types of films are not so different. This particular plot is motivated by the fact that in strongly disordered thin films, T_c is expected to decrease roughly linearly with R_N due to diminished screening of the Coulomb interaction between electrons.[10] For context, Fig. 4 shows that amorphous MoGe films display the predicted behavior.[11–14]

IV. CONCLUSION

We examined the magnetic penetration depth of thin superconducting Nb films dc sputtered onto oxidized Si substrates at room temperature. T_c is approximately linear in film thickness d for 20 Å $\leq d \leq$ 70 Å. The magnitude and T dependence of $\lambda^{-2}(T)$ indicate that these films are weak-coupling dirty-limit BCS superconductors, implying that T_c decreases due to non-pair-breaking mechanisms, e.g., an increase in Coulomb pseudopotential.[4] These results are consistent with tunneling measurements of electron-beam deposited Nb films,[4] when films are compared on the basis of sheet resistance rather than thickness.

ACKNOWLEDGMENT

I.H. gratefully acknowledges support from OSU.

*Author to whom correspondence should be addressed.

[1] A. I. Gubin, K. S. Il'in, S. A. Vitusevich, M. Siegel, and N. Klein, Phys. Rev. B **72**, 064503 (2005).

[2] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).

[3] See the review by K. Maki, in *Superconductivity*, edited by R. D. Parks (Dekker, New York, 1969), Vol. 2, Chap 18.

[4] S. I. Park and T. H. Geballe, Phys. Rev. Lett. **57**, 901 (1986).

[5] T. P. Sheahan, Phys. Rev. **149**, 368 (1966).

[6] B. Fellmuth, H. Maas, and D. Elefant, Metrologia **21**, 169 (1985).

[7] S. J. Turneaure, E. R. Ulm, and T. R. Lemberger, J. Appl. Phys. **79**, 4221 (1996).

[8] S. J. Turneaure, A. A. Pesetski, and T. R. Lemberger, J. Appl. Phys. **83**, 4334 (1998).

[9] A. F. Mayadas, R. B. Laibowitz, and J. J. Cuomo, J. Appl. Phys. **43**, 1287 (1972).

[10] A. Finkel'shtein, JETP Lett. **45**, 46 (1987); Physica B **197**, 636 (1994).

[11] J. M. Graybeal, Physica B & C **135**, 113 (1985).

[12] J. M. Graybeal and M. R. Beasley, Phys. Rev. B **29**, 4167 (1984).

[13] S. J. Turneaure, T. R. Lemberger, and J. M. Graybeal, Phys. Rev. Lett. **84**, 987 (2000).

[14] S. J. Turneaure, T. R. Lemberger, and J. M. Graybeal, Phys. Rev. B **63**, 174505 (2001).