Penetration depth, microwave surface resistance, and gap ratio in NbN and $Ba_{1-x}K_xBiO_3$ thin films

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We report values of the zero temperature magnetic penetration depth $\lambda(0)$, microwave surface resistance R_s, and gap ratio $2\Delta(0)/k_BT_c$ in technologically useful thin films of NbN and $Ba_{1-x}K_xBiO_3$. A novel analysis technique was used to extract the absolute magnitude of $\lambda(0)$ and $2\Delta(0)/k_BT_c$ from shifts in resonant frequency of a parallel-plate resonator. For NbN and $Ba_{1-x}K_xBiO_3$ values of $\lambda(0)=3900\pm200$ Å and 3300 ± 200 Å were obtained, respectively. The gap ratios were found to be $2\Delta(0)/k_BT_c=4.1\pm0.1$ and 3.8 ± 0.5, respectively, for $T_c=16.3$ K in NbN and $T_c=17.2$ K in $Ba_{1-x}K_xBiO_3$. The surface resistance measurements on $Ba_{1-x}K_xBiO_3$ represent the lowest values ever reported at microwave frequencies in this material.

The high-T_c bismuthate $Ba_{1-x}K_xBiO_3$ ($T_c\approx15$–26 K) and the granular superconductor NbN ($T_c\approx16$ K) are attractive candidates for microwave devices because they can be cooled below T_c by standard closed-cycle helium refrigerators, and are available in thin film form. In addition, they are both known to be cubic superconductors with coherence lengths suitable for fabrication of tunnel junctions.[1,2] For both microwave and junction applications it is desirable to know values of fundamental parameters such as the magnetic penetration depth $\lambda(T)$, the microwave surface resistance R_s, and the gap ratio $2\Delta/k_BT_c$, since these quantities strongly influence device design and performance. This letter represents an important addition to the literature on $\lambda(T)$ and R_s for these materials, as well as independent confirmation of the value of $2\Delta/k_BT_c$ obtained from tunneling and IR reflectivity measurements.

Samples were studied in thin film form to evaluate their performance and characteristics in the geometry most likely to be encountered in real devices made from these materials. The NbN samples were deposited by dc reactive magnetron sputtering on thermally oxidized Si(100) substrates, in a process detailed elsewhere.[3] Samples with and without a Nb underlayer were measured. This process yielded samples with thickness 8000 Å, $T_c=16.3$ K, and uniform, reproducible, high quality microwave properties as reported below. The $Ba_{1-x}K_xBiO_3$ ($x=0.49$) samples were grown on (100) MgO substrates by molecular beam epitaxy, also described elsewhere.[4] They had a thickness of 3600 Å, and showed a superconducting transition at $T_c=17.2$ K (by ac susceptibility), which, although low compared to bulk values, is typical of films prepared by this method. Two identical films from the same deposition run were used in each parallel-plate measurement.

Microwave measurements were made by placing these two identical samples face-to-face with a Teflon™ dielectric layer (thickness $d=25$ μm$\pm5\%$ or 12.5 μm$\pm10\%$) sandwiched between them, forming a parallel-plate resonator.[5] Excitation of transverse electromagnetic modes between the films was accomplished by two 50 Ω microstrip antennas placed nearby. The position of each antenna could be varied sensitively by a micrometer located on the room-temperature end of the cryostat, so that the capacitive coupling to the resonator could be optimized. All measurements were done in the weak coupling limit, and at lowest possible microwave power, so that $Q_{\text{measured}}\approx Q_{\text{unloaded}}$. The resonator was also enclosed in a vacuum can (pressure ~100 mTorr) to minimize helium vapor effects on the resonant frequency.

The transmission coefficient S_{21} was measured as a function of frequency using an HP 8510C Vector Network Analyzer, and the Q and resonant frequency f_0 of each resonance were found using a complex impedance fitting routine. Using the relations $R_s^{\text{eff}}=\pi\mu_0 f_0 d/Q$ and

$$f_0=f_a\bigg/\sqrt{1+\frac{2\lambda}{d}\coth\left(\frac{t}{\lambda}\right)}, \tag{1}$$

where t is the film thickness and f_a is the geometrical resonant frequency (if $\lambda=0$), both R_s^{eff} and λ were deduced for each sample. The effects of radiative and dielectric loss were determined independently by varying the Teflon thickness from 12.5 to 125 μm.[5] Corrections were then made to the values of R_s^{eff} by subtracting away these extrinsic losses. Finite film thickness corrections were then applied to these corrected R_s values using the fit results for λ and the expression of Klein *et al.*[6] This second correction was necessary because in some cases the condition $t\approx\lambda$ caused screening to occur over a shorter distance than in an infinitely thick sample, increasing the measured losses. At 10 GHz the resolution limit in R_s was found to be 6 $\mu\Omega$ at 25-μm dielectric thickness and 4 $\mu\Omega$ at 12.5-μm thickness.

Analysis of the measurements resulted in the plots shown in Figs. 1–3, which show $\lambda(T)$ and $R_s(T)$ for both materials. Shifts in resonant frequency $f_0(T)$ are related to

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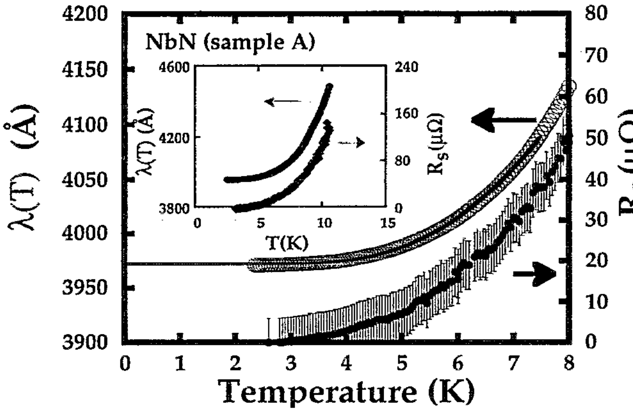

FIG. 1. Surface resistance and penetration depth of 8000-Å-thick NbN thin films on Si (100) substrates without Nb underlayer, measured at 10.54 GHz using a 25-μm dielectric spacer. Solid line is BCS fit. Inset shows data over entire measurement range. R_s has been corrected for finite film thickness, radiation and dielectric losses. Error bars on λ are ± 200 Å (not shown).

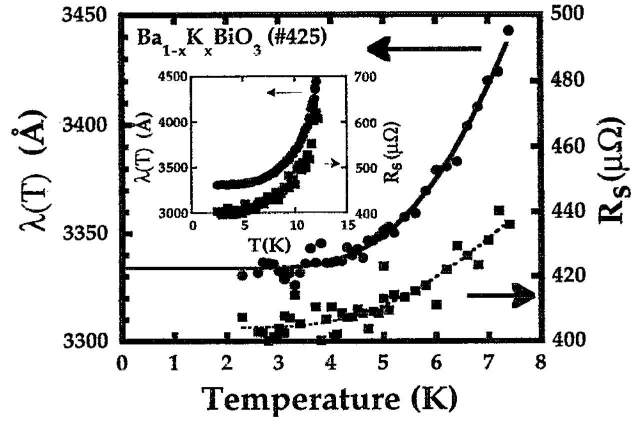

FIG. 3. Surface resistance and penetration depth of 3600-Å-thick $Ba_{1-x}K_xBiO_3$ thin films on MgO (100) substrates, measured at 6.46 GHz using a 25-μm spacer. Solid and dashed lines are BCS fits. Inset shows data over entire measurement range. Surface resistance has been corrected for finite film thickness; radiation and dielectric losses are negligible, accounting for 20 $\mu\Omega$ at most.

shifts in $\lambda(T)$, as given by Eq. (1). However, since the quantity f_a appearing in that equation is unknown, the absolute magnitude of $\lambda(T)$ must be inferred by fitting to an assumed temperature dependence.[7] Our high resolution in frequency shift made small changes in λ (of order 1 Å) readily measurable, so it was straightforward to fit those changes in λ to an assumed dependence on temperature and deduce the absolute penetration depth.

A multitude of tunneling measurements[2,8–12] have shown both NbN and $Ba_{1-x}K_xBiO_3$ to be well described by the Bardeen–Cooper–Schrieffer (BCS) theory of superconductivity, with well-defined energy gaps Δ. This implies that their penetration depths should have the s-wave BCS asymptotic temperature dependence,[13]

$$\frac{\lambda(T)}{\lambda(0)}=1+\sqrt{\frac{\pi\Delta(0)}{2k_BT}}\exp\left(-\frac{\Delta(0)}{k_BT}\right), \quad T<\frac{T_c}{2} \tag{2}$$

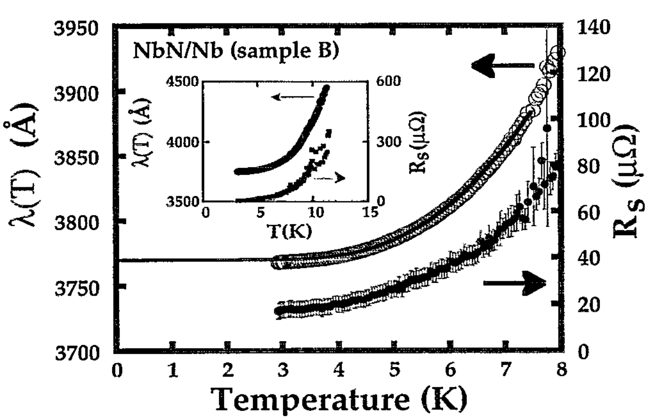

FIG. 2. Surface resistance and penetration depth of 8000-Å-thick NbN thin films on Si (100) substrates with 200-Å Nb underlayer, measured at 11.39 GHz using a 12.5-μm dielectric spacer. Solid line is BCS fit. Inset shows data over entire measurement range. R_s has been corrected for finite film thickness, radiation and dielectric losses. Error bars on λ are ± 400 Å (not shown).

valid well below T_c. This dependence was combined with Eq. (1) to fit the resonant frequency shift data $f_0(T)$ in a manner similar to that described in the literature.[7]

The NbN samples studied were of two types: those deposited directly onto the oxidized Si substrate without a Nb underlayer and those deposited with a 200-Å Nb underlayer. An investigation of the microwave properties of these two samples was of interest because of recent work indicating that the Nb underlayer improves the crystalline orientation and decreases the dc resistive transition width of the NbN film.[3] One would expect improvement in microwave properties over samples on bare Si, which have already shown extremely low values of $R_s=3.6$ $\mu\Omega$ at 10 GHz, 4.2 K.[14]

Both deposition methods produced films of extremely good quality, with surface resistance values of $R_s<6$ $\mu\Omega$ at 4.2 K, 10.54 GHz and $R_s=22\pm 4$ $\mu\Omega$ at 4.2 K, 11.39 GHz for samples on bare Si and with an underlayer of Nb, respectively. BCS fits to the frequency shift data for both types of samples yielded $\lambda(0)=3900\pm 200$ Å, indicating that the NbN films were thicker than their penetration depth in both cases. This means that the thin 200-Å Nb layer underneath was exposed to a reduced rf magnetic field equal to less than 2% of its surface value. Still, if the thin Nb layer were nonsuperconducting it may have caused large enough losses to account for the increased surface resistance. This increased loss was clearly visible over a range of frequencies, as shown in Table I, even though the penetration depth remained essentially unaltered.

Moreover, the BCS gap ratio for both NbN samples was found to be $2\Delta(0)/k_BT_c=4.1\pm 0.1$, in agreement with results from tunneling in thin film junctions.[2,12] As with recent results on $Nd_{1.85}Ce_{0.15}CuO_4$ thin films and crystals,[15] this provided confirmation that NbN is intrinsically a strong-coupled BCS material with a well-known, well-defined gap, implying that the differences in R_s found in this work are likely related to conductor losses in the Nb underlayer.

For the $Ba_{1-x}K_xBiO_3$ films, R_s was found to be 400 ± 35 $\mu\Omega$ at 6.46 GHz and 2.2 K, uncorrected for radiation and

TABLE I. Penetration depth[a] and surface resistance of 8000-Å-thick NbN films on Si(100) substrates without Nb underlayer (sample A) and with 200-Å Nb underlayer (sample B).

	Frequency (GHz)	R_s at 4.2 K ($\mu\Omega$)	$\lambda(0,\omega)$ (Å)[a]
Sample A	10.54	<6	3970±200
	13.08	17±8	3910±200
	16.97	28±10	3850±200
Sample B	11.39	22±4	3770±400
	13.87	30±8	3810±200
	18.20	130±15	3900±200

[a]Extracted from BCS fits (see Ref. 13).

dielectric losses (presumed negligible). When scaled to 10 GHz with an ω^2 dependence, this yielded 958±85 $\mu\Omega$. This value is lower than other published values on laser-ablated films[16] and much less than measurements on melt fragments,[17] and represents a significant improvement in technological value for this material. Since the parallel-plate resonator technique requires no lithographic processing, conditions were favorable for observing low R_s in $Ba_{1-x}K_xBiO_3$.

The magnetic penetration depth and energy gap for $Ba_{1-x}K_xBiO_3$ were found by BCS fitting to be $\lambda(0)=3300 \pm 200$ Å and $\Delta(0)=2.82\pm0.35$ meV. This yielded a gap ratio of $2\Delta(0)/k_BT_c=3.8\pm0.5$, using $T_c=17.2$ K. Fitting $R_s(T)$ to BCS theory[13] yielded a similar gap ratio (3.5±0.5). This result is completely consistent with results of tunneling measurements on polycrystalline pellets,[8] single crystals, and thin films in $Ba_{1-x}K_xBiO_3$/Au junctions,[9] sputtered films in $Ba_{1-x}K_xBiO_3$/I/Au S-I-N junctions,[10] all-$Ba_{1-x}K_xBiO_3$ thin film junctions,[11] and with infrared reflectivity results.[18] Our value of $\lambda(0)$ also compares favorably to the value of 3400 Å reported from μ on-spin-relaxation (μSR) measurements on high purity polycrystalline bulk samples.[19] Our results for $\lambda(0)$ and $2\Delta(0)/k_BT_c$ are therefore in agreement with those obtained by completely different methods.

In summary, we have presented values of the surface resistance, penetration depth, and BCS gap ratio in NbN and $Ba_{1-x}K_xBiO_3$ thin films. NbN samples on bare Si had extremely low R_s (4.2 K)<6 $\mu\Omega$ at 10.54 GHz. It was found that a Nb underlayer, while improving the crystalline orientation of the sample, caused R_s to increase. The penetration depth was unaltered by the use of the underlayer, within experimental error. The $Ba_{1-x}K_xBiO_3$ samples had R_s (2.2 K)=400 $\mu\Omega$ at 6.46 GHz, which represents the lowest value yet reported for that material. It should be noted that the fabrication of $Ba_{1-x}K_xBiO_3$ is still in its infancy compared to NbN. Since early samples of NbN also exhibited R_s values comparable to those presented here for $Ba_{1-x}K_xBiO_3$,[20] there is every reason to believe that the surface resistance of $Ba_{1-x}K_xBiO_3$ will be substantially reduced with further materials development. Both materials showed BCS gap ratios fully consistent with tunneling measurements.

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[1] B. Batlogg, R. J. Cava, L. W. Rupp, Jr., A. M. Mujsce, J. J. Krajewski, J. P. Remeika, W. F. Peck, A. S. Cooper, and G. P. Espinosa, Phys. Rev. Lett. **61**, 1670 (1988).
[2] S. Kashiwaya, M. Koyanagi, A. Shoji, M. Matsuda, and H. Shibata, IEEE Trans. Magn. **27**, 837 (1991).
[3] S. Y. Lee, M. Bruns, and R. D. Glenn, IEEE Trans. Appl. Supercond. **3**, 2953 (1993).
[4] E. S. Hellman, E. H. Hartford, and E. M. Gyorgy, Appl. Phys. Lett. **58**, 1335 (1991).
[5] R. C. Taber, Rev. Sci. Instrum. **61**, 2200 (1990).
[6] N. Klein, H. Chaloupka, G. Müller, S. Orbach, H. Piel, B. Roas, L. Schultz, U. Klein, and M. Peiniger, J. Appl. Phys. **67**, 6940 (1990).
[7] B. W. Langley, S. M. Anlage, R. F. W. Pease, and M. R. Beasley, Rev. Sci. Instrum. **62**, 1801 (1991).
[8] J. F. Zasadzinski, N. Tralshawala, Q. Huang, K. E. Gray, and D. G. Hinks, IEEE Trans. Magn. **27**, 833 (1991).
[9] F. Sharifi, A. Pargellis, R. C. Dynes, B. Miller, E. S. Hellman, J. Rosamilia, and E. H. Hartford, Jr., Phys. Rev. B **44**, 12 521 (1991).
[10] H. Sato, H. Takagi, and S. Uchida, Physica C **169**, 391 (1990).
[11] A. N. Pargellis, F. Sharifi, R. C. Dynes, B. Miller, E. S. Hellman, J. M. Rosamilia, and E. H. Hartford, Jr., Appl. Phys. Lett. **58**, 95 (1991).
[12] M. Hikita, K. Takei, and M. Igarashi, J. Appl. Phys. **54**, 7066 (1983).
[13] J. P. Turneaure, J. Halbritter, and H. A. Schwettman, J. Supercond. **4**, 341 (1991).
[14] M. A. Bruns, R. D. Glenn, and S. Y. Lee, Physica B (to be published).
[15] D. H. Wu, J. Mao, S. N. Mao, J. L. Peng, X. X. Xi, T. Venkatesan, R. L. Greene, and S. M. Anlage, Phys. Rev. Lett. **70**, 85 (1993).
[16] C. E. Platt, M. R. Teepe, C. Ciofi, H. Zhang, V. P. Dravid, R. A. Schweinfurth, D. J. Van Harlingen, J. A. Eades, C. H. Lin, D. Strothers, and R. Hammond, Mater. Res. Soc. Symp. Proc. **275**, 807 (1992).
[17] J. R. Delayen, C. L. Bohn, and C. T. Roche, IEEE Trans. Magn. **27**, 1532 (1991).
[18] Z. Schlesinger, R. T. Collins, J. A. Calise, D. G. Hinks, A. W. Mitchell, Y. Zheng, B. Dabrowski, N. E. Bickers, and D. J. Scalapino, Phys. Rev. B **40**, 6862 (1989).
[19] E. J. Ansaldo, Z. R. Wang, J. H. Cho, D. C. Johnston, and T. M. Riseman, Physica C **185-189**, 1889 (1991).
[20] L. H. Allen, M. R. Beasley, R. H. Hammond, and J. P. Turneaure, IEEE Trans. Magn. **MAG-23**, 1405 (1987).