Terahertz thermal spectroscopy of a NbN superconductor

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We report far-infrared optical properties of a NbN superconductor. Transmission through a high-quality NbN film grown on a birefringent sapphire substrate above and below its superconducting transition down to the zero-temperature limit is measured at six different frequencies from 0.4 to 2.5 THz both above and below its optical gap. The experimental results agree with theoretical calculations developed based on utilization and extension of the BCS model of Zimmermann *et al*. [Physica C **183**, 99 (1991)] applied for the NbN film. Full quantitative agreement over the entire ranges of temperature and frequencies is found based solely on the physical properties of this NbN film sample and on the parameters of an identical sapphire substrate as measured in time-domain spectroscopy experiments, without use of any additional fitting parameters.

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I. INTRODUCTION

The exact knowledge of physical properties of conventional superconductors such as NbN is fundamental from the point of view of their future application. At first glance it might seem that all of them are indeed well known and understood. A closer look shows, however, that some of them are still understood rather poorly. The high-frequency conductivity serves as an example—in principle, the Mattis-Bardeen theory[1] can be used to calculate it, but some terahertz frequency experiments still lack a satisfactory explanation. This Brief Report represents our effort to improve this situation.

Time-domain terahertz spectroscopy provides data in a broad frequency region and enables one to determine the complex conductivity; see, e.g., Ikebe *et al.*[2] On the other hand, laser thermal spectroscopy is advantageous in that the transmission measurements are performed quasistatically. It enables precise measurement of transmission at fixed laser frequencies as a function of temperature in the vicinity of the superconducting transition, where a peculiar behavior was observed, and has proved to be a very useful experimental method.[3,4]

Earlier we performed experiments in the terahertz frequency range,[4,5] but a satisfactory theoretical explanation has not been found. Our previous experiments suffered from the fact that the NbN sample we used displayed a rather broad superconducting transition.[5] In order to get better agreement between the experiment and the BCS theory we had to artificially treat it as a mixture of superconductors with different T_c's.

In this Brief Report we present experimental data on a much higher-quality NbN film obtained using our setup,[4] as well as additional supporting experiments.[6] Further, we discuss a full analysis of the thermal and frequency behavior of the NbN superconductor in terms of a developed BCS-based model that takes into account optical properties of the birefringent sapphire substrate. Without using any additional fitting parameters, we demonstrate full quantitative agreement between theory and experiment over entire ranges of temperature and frequency.

II. EXPERIMENT

We have measured far-infrared (FIR) transmission of a superconducting NbN film of nominal thickness $d_1 = 15$ nm grown epitaxially on a sapphire substrate. We estimate the thickness uncertainty of ± 1 nm to be due to the partially oxidized NbN surface layer.[7]

The quality of our NbN sample is very high, as is apparent from the temperature dependence of its dc resistivity displayed in Fig. 1, showing that the resistance drops to zero within an interval of 0.5 K. Measurements of the dc electrical conductivity have been performed using a 0.7×10 mm^2 strip cut for this purpose from our actual sample and measured by the four-probe method, as sketched in the upper inset of Fig. 1. The critical temperature has been determined as $T_c \approx 16$ K. The normal-state dc resistivity has been estimated as $\rho_0 = (2.0 \pm 0.2) \times 10^{-6}\ \Omega$ m, with the accuracy limited mainly by the uncertainty of the film cross section and the size and separation of the potential contacts. The lower inset in Fig. 1 shows that the normal-state resistivity is nearly temperature independent over the entire temperature range up to room temperature with a residual resistivity ratio of 0.97.

FIR transmission is strongly influenced by optical interference of multiple internal reflections, especially in a relatively thick substrate. Thus for accurate numerical analysis of transmission it is important to have a well-determined optical thickness of the substrate. Our NbN sample is grown on a birefringent *R*-cut sapphire with dimensions 10×10 mm^2 and thickness $d_2 = 0.333$ mm. Sapphire is an important anisotropic uniaxial material with ordinary, n_o, and extraordinary, n_e, refractive indices that are tabulated.[8] The structure and orientation of our sample in the experiment are shown in Fig. 2. The extraordinary ray axis is oriented diagonally (at an angle $\alpha = 45^\circ$ with respect to the y axis), while $\vartheta = 57.6^\circ$ is the angle between the c axis of the sapphire crystal and the z axis oriented normally to the substrate.

Both the ordinary and extraordinary refractive indices were determined in the supporting independent time-domain terahertz spectroscopy experiment[6] at room temperature, using an identical sapphire substrate, as shown in Fig. 3. While the

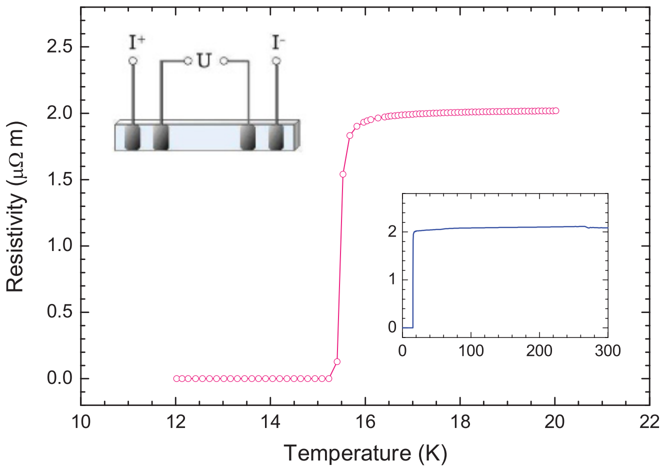

FIG. 1. (Color online) Temperature dependence of the electrical resistivity of the NbN sample in the vicinity of its critical temperature T_c (red). The lower inset shows the extension of the temperature range up to room temperature (blue); the upper inset is a sketch of the sample strip and the four-probe method used in this measurement.

real part of the refractive index increases slightly, approximately linearly with frequency ν, the imaginary part is almost frequency independent. The optical thickness of the substrate, $n_2 d_2$, is to a good approximation temperature independent.[9,10] Absorption by the substrate is nearly negligible at room temperature and is even smaller at low temperature. The values of refractive indices deduced from the time-domain spectroscopy experiment are given in Table I together with those tabulated by Palik:[8] the agreement is excellent. Additionally, these measurements allowed us to determine that the polarization of the ordinary ray is parallel with the diagonal direction of the square sapphire substrate.

Our experimental setup has been described in detail in our previous report.[4] In short, the FIR gas laser can be tuned to various terahertz frequencies. A wire grid polarizer was inserted in the optical path to ensure a horizontal or vertical orientation of the FIR laser beam polarization. The sample is placed in the cryomagnetic system and its temperature tuned and controlled by means of a heater and by flowing He gas.

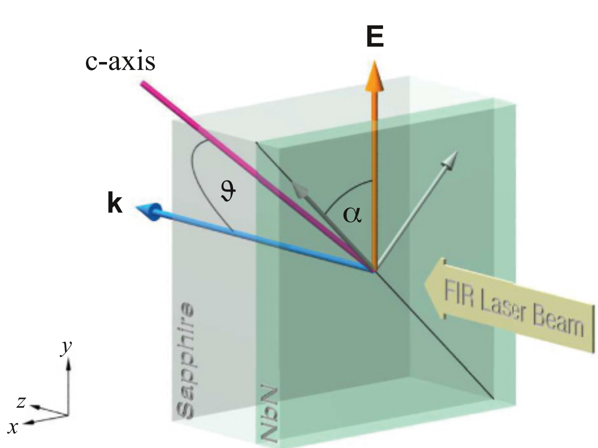

FIG. 2. (Color online) Schematic representation of the double-layer structure of the NbN sample on the sapphire substrate. For notation details, see the text.

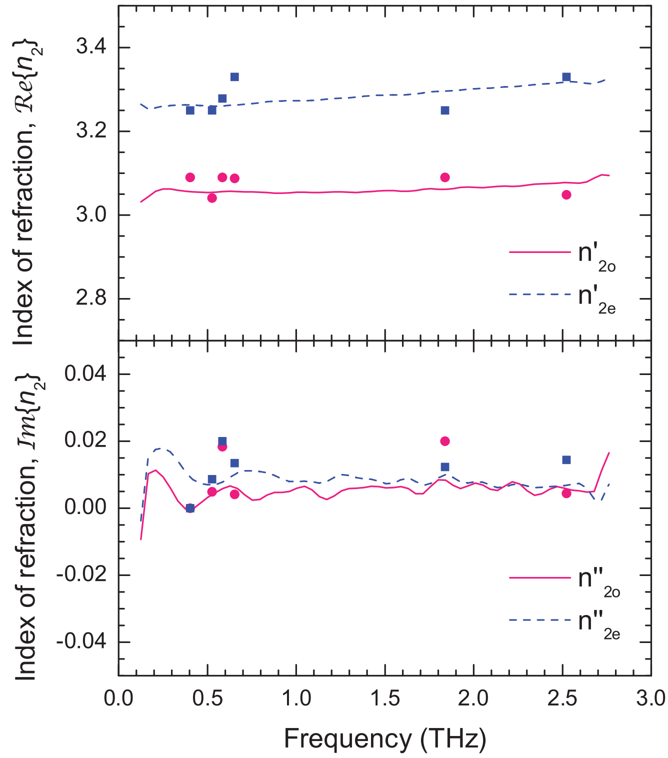

FIG. 3. (Color online) Real and imaginary parts of the refractive indices of the *R*-cut sapphire substrate. Lines, data from terahertz time-domain spectroscopy; single points, obtained from the best fit of the FIR transmission. The full (red) and dashed (blue) lines represent the ordinary and the extraordinary ray, respectively.

To eliminate a possible temperature difference between the sensor and the true temperature of the sample, the dc resistivity is monitored by the four-probe method simultaneously with the transmission measurement. This allows, in particular, determination of the superconducting transition temperature T_c with sufficient precision.

Since NbN is an isotropic superconductor, the only anisotropy introduced in the system is due to the birefringent sapphire substrate. In the special case of a 45° angle between the electric vector of the incident beam and the extraordinary ray axis of the sapphire substrate (and only in this case), the relative transmission of the horizontally and vertically polarized radiation should be equal. Transmission was measured in both polarizations and the observed difference was less than 5%, which can most likely be attributed to a slight misalignment of the sample. Even for the 0.4037 THz line, for which the

TABLE I. Parameters of the sapphire substrate.

			Palik, Ref. 8		Ref. 17
ν (THz)	n_{2o}	$n_{2e}(\vartheta)$	n_o	n_e	$n_e(\vartheta)$
0.4037	3.06	3.26	3.06	3.39	3.29
0.5254	3.05	3.26	3.06	3.40	3.29
0.5843	3.06	3.26	3.06	3.40	3.29
0.6538	3.06	3.26	3.06	3.40	3.29
1.8191	3.06	3.30	3.08	3.44	3.33
2.5228	3.08	3.32	3.10	3.47	3.35

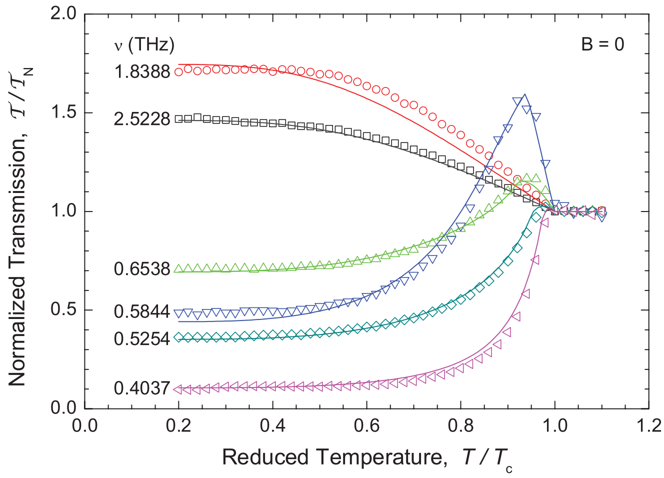

FIG. 4. (Color online) Normalized transmission as measured (individual data points) and calculated (solid lines) as described in the text for six different frequencies as indicated, plotted versus relative temperature T/T_c.

transmission in the zero-temperature limit is very weak, the relative difference is still within 10%.

Our experimental data for the horizontally polarized beam are displayed in Fig. 4 as individual data points. The sample temperature was swept up and down at a low rate of about 1 K/min, without observable temperature hysteresis. The far-infrared transmission, $\mathcal{T}$, is related to that in the normal state, $\mathcal{T}_N$, taken slightly above T_c.

The transmission of photons with energies far below the optical gap displays a monotonic increase toward the critical temperature. As the frequency approaches the optical gap, the characteristic BCS thermal peak starts to appear close below T_c. As the frequency increases further, the peak becomes wider and its center shifts toward a lower temperature. Above the optical gap, the transmission displays a monotonic decrease and in the high-frequency limit approaches the normal-state transmission.

III. THEORETICAL MODEL

The theoretical calculation and subsequent numerical evaluation of transmission requires precise values of the refractive indices of both the NbN film and the sapphire substrate. Additionally, precise knowledge of the relative permittivity of the superconductor film for angular frequency ω is also needed, which is given by[11]

$$\varepsilon(\omega,T) = \varepsilon_\infty + \frac{i\sigma(\omega,T)}{\omega\varepsilon_0}, \tag{1}$$

where $\varepsilon_\infty \approx 1$ is the core permittivity, ε_0 denotes the permittivity of vacuum, and $\sigma(\omega,T)$ is the optical complex conductivity. Other contributions, such as that from phonons, are negligible in our spectral range, as proved by Ikebe *et al.*[2] and Semenov *et al.*[7] The refractive index of the superconducting film $n_1 = \sqrt{\varepsilon(\omega,T)}$, and the indices of the substrate n_{2o}, n_{2e} enter the recurrence relations[12] for the transmission coefficients t_{03} of the extraordinary and ordinary rays. The electric vector of a monochromatic wave incident normally on the double-layered system shown in Fig. 2 can be decomposed in the directions parallel and perpendicular to the plane containing the **k** wave vector and the c axis of the sapphire crystal. The extraordinary and ordinary rays do not interfere with each other and travel through the system independently, resulting in summation of their intensities at the output. Due to the geometry of the system, the transmissivities are the same for both horizontally and vertically polarized beams:

$$\mathcal{T} = |t_{03}(n_{2e})|^2 \cos^2\alpha + |t_{03}(n_{2o})|^2 \sin^2\alpha\,. \tag{2}$$

The transmission in the normal state, $\mathcal{T}_N$, can be calculated in the same way, replacing the BCS conductivity by the Drude formula for a normal metal:

$$\sigma_N(\omega) = \frac{\sigma_0}{1 - i\omega\tau}, \tag{3}$$

where σ_0 and τ are the dc conductivity and the relaxation rate of the NbN film, respectively.

The optical conductivity $\sigma(\omega,T)$ of a BCS superconductor is well described by the theoretical approach of Zimmermann *et al.*[13] We utilize their approach, but instead of their simple approximation for the gap temperature dependence we use values obtained by numerically solving the BCS gap equation.[14,15] The model parameters T_c and σ_0 are sufficiently well established and their variations within experimental errors do not appreciably influence the calculated transmission. The values of the optical gap $2\Delta_0 = 4.15 k_B T_c$ and the relaxation rate τ as low as 3.86 fs have been taken from the literature.[7] In our frequency range, however, $\omega\tau < 0.1$, and the dirty limit $\tau \to 0$ seems appropriate.

The parameters for sapphire—the ordinary and extraordinary refractive indices n_{2o} and n_{2e} and the substrate thickness $d_2 = 0.333$ mm—have been determined from our time domain spectroscopy experimental data. As the optical system is anisotropic, calculation of the transmission is rather complicated. While variations of the film parameters (within their experimental errors) hardly affect the calculated transmission, we found that the results are much more sensitive to the ordinary and extraordinary indices of the substrate. This is understandable, since the optical thickness of the substrate is responsible for interference effects.

To achieve the best agreement between calculated and experimental transmission values, we have modified the substrate parameters slightly, within their experimental errors. This procedure yields optimum indices of refraction that are displayed in Fig. 3 as individual data points. Their scatter can most likely be attributed to small experimental errors in transmission measurements, which in turn could be caused by standing waves in the optical path and/or by small misalignment of the sample with respect to the polarization vector. The optimal calculated values of transmission of light for six different frequencies from 0.4 up to 2.5 THz plotted versus temperature as solid lines, together with the experimental data points, are displayed in Fig. 4.

IV. DISCUSSION AND CONCLUSIONS

The temperature dependencies of FIR transmissions at six frequencies $h\nu$ close to $2\Delta_0$ have been measured on a high-quality NbN epitaxial film deposited on a birefringent sapphire substrate. Linearly polarized laser lines with photon energies in the vicinity of the optical gap revealed typical BCS behavior. The transmission of photons with energies far below/above the optical gap showed monotonic decrease/increase toward the zero-temperature limit. The transmissions for photon energies close to the optical gap follow a complex behavior with a characteristic BCS thermal peak. Let us stress that our approach, unlike that of Ikebe *et al.*,[2] enables the study of quasistatic FIR transmission over a full range of temperature.

The experimental results are found to agree with theoretical calculations developed based on utilization of the BCS model of Zimmermann *et al.*[13] applied for a NbN film with the numerical solution of the gap equation[14] replacing the original simple approximation for the temperature dependence of the gap. Full quantitative agreement within error bars over the entire range of temperature and frequency is found based solely on physical properties of this NbN film sample and an identical sapphire substrate as measured in additional experiments. This important quantitative step was not possible to achieve with our previously investigated NbN samples, where a more or less satisfactory agreement was possible only when the superconducting transition temperature was artificially smeared out over a temperature interval of order of 1 K. In addition, we have found that transmission is notably affected by interference effects in the substrate, which acts as a Fabry-Pérot étalon possessing temperature-dependent surface reflectivity.

In conclusion, the reported results represent a textbook example of thermal spectroscopy of a conventional BCS superconductor: a full quantitative agreement between the experimental data spanning broad ranges of temperature and frequency and the fundamental BCS-based microscopic theory with no free parameters. They constitute therefore a valuable basis for a further important extension: a detailed quantitative investigation of the terahertz properties of superconducting thin films in a magnetic field and in a nonequilibrium regime.

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[11] The convention $E(t) = E_0 \exp(-i\omega t)$ leads to notation for the complex conductivity: $\sigma = \sigma' + i\sigma''$, the complex permitivity: $\varepsilon = \varepsilon' + i\varepsilon''$, and the complex index of refraction: $n = n' + in''$.

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[17] For *R*-cut sapphire the magnitude of the extraordinary index $n_e(\vartheta)$ has to be recalculated as (see also Ref. 16)

$$\frac{1}{n_e(\vartheta)^2} = \frac{\cos^2\vartheta}{n_o^2} + \frac{\sin^2\vartheta}{n_e^2}.$$