Far-infrared transmission of a superconducting NbN film

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We report far-infrared optical properties of a thin NbN superconductor in magnetic field, B, up to 10 T. Transmission, $Tr(T,B)$, of monochromatic linearly polarized laser beam with frequency below and above an optical gap is measured both below and above T_c. $Tr(T,B=0)$ is well described by the BCS-based model that approximates the sample as a mixture of superconductors with different T_c. $Tr(T,B\neq 0)$ appears qualitatively different for Voigt and Faraday geometry. To calculate optical properties, we use Bruggeman's approach and present a phenomenological model accounting for both field orientations. The model captures all observed features of the $Tr(T,B)$ data.

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I. INTRODUCTION

Far-infrared spectroscopy serves as a sensitive tool for investigating physical properties of superconductors, as their optical gap $2\Delta(0)$ is comparable with the energy of radiation quanta. Photons with energy higher than the gap can break Cooper pairs, which strongly influences high-frequency conductivity. Measurements of optical properties of superconductors thus enable deduction of important physical phenomena. Most authors present infrared transmission spectra measured at fixed temperatures using a continuous source of radiation[1,2] or a set of many monochromatic laser lines[3] while we report a complementary method using monochromatic source of radiation while sweeping the temperature.[4,5] In gigahertz frequency region, Oates *et al.*[6] reported both approaches.

Superconductors of the second type in magnetic field larger than the lower critical field $B > B_{c1}$ are in Abrikosov state. Superconductivity in the vortex core region is suppressed and its conductivity approaches normal-state value. Electric currents flowing through the material interact with the vortices, cause their motion, and the nondissipative regime is lost. Motion of the vortex lattice thus also influences conductivity. Controlled vortex motion—*fluxonics*—currently represents a hot topic of investigation. High-frequency vortex dynamics constitutes an essential tool for these aspirations, however, it is not yet properly understood. One of the most successful theoretical models is that by Coffey and Clem.[7] At present, there is aspiration to understand vortex dynamics by means of the time-dependent Ginzburg-Landau theory.[8] This paper represents our experimental contribution to this effort.

II. EXPERIMENT

We report far-infrared transmission of a thin superconducting (sc) NbN film deposited on silicon substrate in magnetic field B up to 10 T for both Faraday ($\boldsymbol{B}^{\perp}s$) and Voigt ($\boldsymbol{B}^{\parallel}s$) orientations with respect to the sample surface s. Our far-infrared gas laser can be tuned to various frequencies including 0.4037 THz (3.3 meV) and 2.5228 THz (10.4 meV) reported in our previous paper.[5] By using a beam splitter, the laser output is monitored by a pyroelectric detector and the transmitted intensity is measured by a helium-cooled bolometer. Transmission is measured at normal incidence in a constant magnetic field while temperature is swept continuously from above T_c to minimum attainable and back. The temperature of the sample monitored by a Cernox thermometer (error in T due to magnetic field less than 3.2%) is controlled by adjusting He-gas flow rate and by a resistive heater. To keep measurement times reasonable, sweep rate of 1K/min was chosen [sweep rates (0.5–2) K/min as well as cool-down and heat-up data series showed no appreciable differences]. Our experimental method was described in more detail in previous papers.[4,5]

The investigated high-quality polycrystalline NbN sample on Si substrate with a very thin SiO_2 interlayer[9] was prepared by Benačka, who measured its basic properties listed in Table I. It is important to characterize each NbN sample individually, since various NbN films[10] can display different T_c.

A. Zero magnetic field data

The T-dependent transmission, shown in Fig. 1, is in good quantitative agreement with our previous experiments.[4] Transmission in the normal state $Tr_n(T_c < T)$ is constant and serves as a suitable reference for transmission of the sc state Tr_{sc}; all measured transmission dependencies are expressed in a normalized dimensionless form as $Tr = Tr_{sc}/Tr_n$. There are two important cases: (i) $\hbar\omega > 2\Delta(0)$, represented by 2.5228 THz line with the photon energy safely above the optical gap and (ii) $\hbar\omega < 2\Delta(0)$ for 0.4037 THz line, which is more complicated. The quantity $2\Delta(T)$ is almost constant at low temperature, approximately equal to the $T\to 0$ value, but with increasing temperature it falls toward zero at T_c. For each frequency there is a temperature where the energy of radiation exceeds $2\Delta(T)$ and Cooper pairs can absorb photons. In case (i) Tr monotonously decreases with temperature until it reaches its normal-state value and remains constant up to 16 K and in case (ii) a well-pronounced peak is observed. The T_c onset can easily be determined from $Tr(T)$ data.

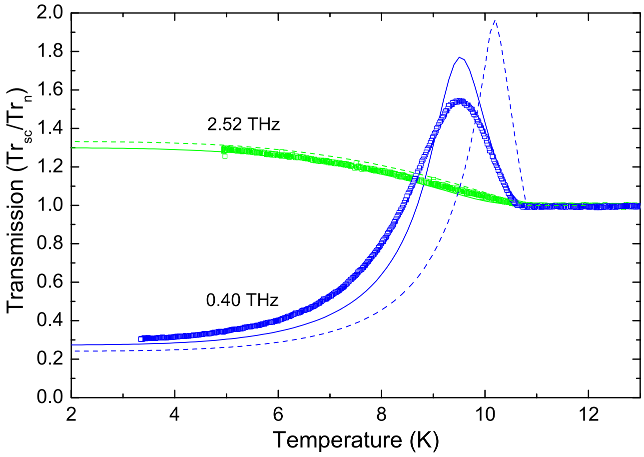

FIG. 1. (Color online) The observed zero-field transmissions for two frequencies are plotted versus temperature. Symbols represent experimental data, dashed lines are calculated curves for BCS superconductor with the sharp transition and solid lines are calculated using modified conductivity as described in the text.

B. Finite magnetic field data

In $B>0$ the optical response of material depends on mutual orientation of $\boldsymbol{B}$ and electric component of light $\boldsymbol{E}$. $\boldsymbol{B}$ determines orientation of the vortex lattice, therefore it is useful to measure in both ($\boldsymbol{B}^{\perp}$) and ($\boldsymbol{B}^{\parallel}$) geometries. In both cases, $T_c(B)$ is easily determined since transmission of sc state deviates from constant normal-state value at T_c.

In Faraday orientation (Fig. 2), *Tr* in small *B* exhibits qualitatively similar behavior as in $B=0$. With increasing *B* the transmission peak decreases and shifts to lower temperatures while low-*T* transmission slowly approaches its normal-state level. For highest attainable fields *Tr* becomes nearly the same as in the normal state over entire *T* interval.

III. PHENOMENOLOGICAL MODEL

Let us attempt to interpret the family of measured *Tr* curves phenomenologically. If our model is successful, it ought to shed light on optical properties of second type sc in the terahertz range. In order to interpret these data one has to calculate relevant material properties of superconductor and only then to evaluate *Tr* and compare it with the experimental data sets.

In order to evaluate transmission of our double-layer system, the refractive index and the thickness of each layer have to be known. Refractive index of NbN is given as $\tilde{n}_1=\sqrt{\tilde{\varepsilon}}$. As a first step, we evaluate the complex permittivity of our NbN superconductor

$$\tilde{\varepsilon} = 1 + i\tilde{\sigma}(\omega)/(\varepsilon_0\omega) + \chi_0, \tag{1}$$

where the contribution from conductive electrons is described by complex conductivity ($\tilde{\sigma}=\sigma_1+i\sigma_2$) and the contribution from all other effects via χ_0. This last term can be neglected, since contribution from conductive electrons in our frequency range dominates. Complex conductivity calculation will be described later.

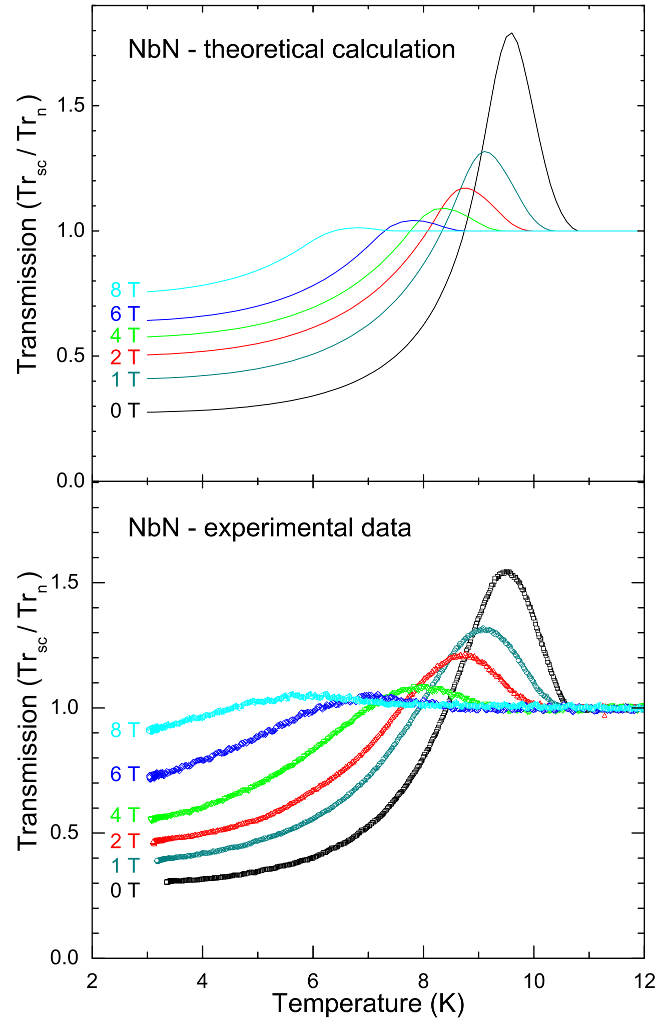

FIG. 2. (Color online) Temperature-dependent transmission in Faraday orientation ($B^{\perp}$). Experimental data are points and theoretical calculations are lines.

We take into account interference effects. Assuming ideally parallel and flat surfaces, relevant quantities describing transmission through interface are transmission and refractive amplitude coefficients

$$t_{ij} = \frac{\tilde{n}_i - \tilde{n}_j}{\tilde{n}_i + \tilde{n}_j}; \quad r_{ij} = \frac{2\tilde{n}_i}{\tilde{n}_i + \tilde{n}_j}. \tag{2}$$

Propagation through material is proportional to $e^{i\delta_i}$, where

TABLE I. Parameters of the sample.

Layer	d_1 (nm)	T_c (K)	ΔT_c (K)	$\sigma_N(0)$ (Ω^{-1} m^{-1})	Substrate	d_2 (mm)	n_2
NbN	80	10.8	0.4	0.45×10^6	Si/SiO_2	0.25	3.5

$$\delta_i = \frac{n_i \omega}{c} d_i. \quad (3)$$

Indices i and j relate to material layers and c is the speed of light. Indices 0 and 3 describe surrounding medium (gaseous helium in this case, $n_0 = n_3 = 1$), index 1 relates to the superconducting layer and index 2 to the substrate. Finally, for the whole multilayer structure the transmission coefficient can be explicitly written as[11]

$$t = \frac{t_{01}t_{12}t_{23}e^{i(\delta_1+\delta_2)}}{1 + r_{12}r_{23}e^{i2\delta_2}}\left(1 + r_{01}\frac{r_{12} + r_{23}e^{2i\delta_2}}{1 + r_{12}r_{23}e^{2i\delta_2}}e^{i2\delta_1}\right)^{-1}, \quad (4)$$

and the transmitted intensity Tr becomes

$$Tr = \frac{n_0}{n_3}|t|^2. \quad (5)$$

In our case it reduces to $Tr = |t|^2$.

Surface roughness might influence transmission, especially when surface imperfections are comparable with, or bigger than, the wavelength λ. In our case of submillimeter waves, however, this should not lead to significant difficulties. Our calculations show that slight deviations (less than 7°) from the normal incidence can be neglected; our experiment is performed at more precise orientation of the sample.

A. Zero magnetic field

Permittivity in the terahertz region is almost entirely governed by the response of conductive electrons that justifies neglecting other contributions. We used BCS-theory-based formula for calculating complex conductivity following Zimmermann *et al.*[12] The essential input parameter is normal-state high-frequency conductivity, which is well described by Drude model: $\tilde{\sigma}(\omega) = \sigma_N(0)/(1 - i\omega\tau)$. Reasonable estimation for momentum scattering time[1,10] is $\tau \approx 5$ fs. The crucial parameter is the optical gap. BCS theory predicts its value $2\Delta(0) = 3.53 k_B T_c$, but many materials, including NbN, exhibit deviations from this formula.[13] For our sample we estimate[1,10] $2\Delta(0) = 4.16 k_B T_c$ (3.87 meV). Temperature dependence of the gap is well approximated by[14]

$$\frac{\Delta(T)}{\Delta(0)} = \sqrt{\cos\left[\frac{\pi}{2}\left(\frac{T}{T_c}\right)^2\right]}. \quad (6)$$

Together with other sample parameters given in Table I, the calculation gives the dashed line in Fig. 1. Although it qualitatively describes the character of the data, quantitative agreement is rather poor.

Better agreement can be achieved by treating our real sample not as a homogeneous sc with sharp transition but as a mixture of superconductors with different T_c and corresponding optical gaps $2\Delta(0) = 4.16 k_B T_c$. This view is supported by the dc electric-resistance measurement showing $\Delta T_c = 0.4$ K. Spread of T_c in the mixture will be even larger than the spread of T_c suggested by resistivity measurements because zero dc resistance does not exclude presence of non-superconducting regions. Since there is no reason to assume any specific distribution function of T_c in such a mixture, we assume that all T_c within suitably chosen T interval are represented with the same probability. For convenience, we approximate our NbN sample as a mixture of ten ideal sc materials with $9.8 \leq T_c \leq 10.8$ K. Conductivity computed this way (solid lines in Fig. 1) agrees notably better with the experimental data.

On a qualitative level, transmission in case of $\hbar\omega > 2\Delta(0)$ is influenced by the absorption caused by breaking of Cooper pairs and by presence of thermally activated quasiparticles. Absorption would be higher at lower temperatures, since Cooper pair density is higher. It leads to a decrease in reflection and, consequently, to an increase in transmission.

In case of $\hbar\omega < 2\Delta(T)$ energy of photons is not high enough for breaking Cooper pairs and the absorption occurs due to thermally excited quasiparticles only. With increasing T the optical gap gradually decreases until at some T optical gap reaches energy of incident photons. For higher T the Cooper pair-breaking mechanism is set on contributing thus to absorption. As a result absorption exhibits a peak. Although it may seem confusing, the highest transmission occurs at the highest absorption. It is because the increase in absorption leads to a decrease in reflection, which governs optical properties.

B. Finite magnetic field

We assume that conductivity of normal state does not depend on magnetic field. This assumption is supported experimentally, as at 16 K in magnetic field up to 10 T transmission within experimental accuracy remains constant.

In a sufficiently high magnetic field the superconductor is in Abrikosov state, inhomogeneous at nanoscale level. Each vortex has a complex structure, consisting of cylindrical core area with diameter equal to coherence length ξ in which the order parameter is suppressed. In this paper, we do not aim to describe all complexity of the vortex lattice. Following Clem,[15] we assume fully normal vortex cores surrounded by fully superconducting bulk material. Normal-state volume $f_n = V_N/V$ is proportional to both the core area $\pi\xi(T)^2$ and the vortex density. Temperature dependence of f_n is given by temperature dependence of coherence length $\xi(T)^2 = \xi_0^2\{1 - [T/T_c(0)]^4\}^{-1}$ and must reach unity at experimentally determined $T_c(B)$. It is satisfied by the following formula:

$$f_n = \frac{1 - T_c(B)^4/T_{c0}^4}{1 - T^4/T_{c0}^4}. \quad (7)$$

Superconducting phase $f_{sc} = 1 - f_n$ is assumed to behave in a similar way as a superconductor without magnetic field. Thus our sample is treated as an effective mixture of ideal BCS superconductors with sharp transitions, with critical temperatures equally distributed over temperature range $(T_c - 1\ \text{K}, T_c)$. Additionally, we assume that $2\Delta(0)$ has zero field value and for its temperature dependence in Eq. (6) we use $T_c(B)$ instead of $T_c(B=0)$.

For NbN, ξ is few nanometers (Ikebe[1] estimated 5 nm), several orders of magnitude smaller than the wavelength inside the sample, so we can use the long-wavelength limit, i.e., consider the material as a composite system with some effective high-frequency permittivity $\tilde{\varepsilon}_{eff}$. Optical properties of such composite systems can be described by various effective medium theories. We have to consider a theory that will distinguish between Faraday ($B^{\perp}$) and Voigt ($B^{\parallel}$) sample orientations. Generalized Bruggeman's theory for ellipsoidal inclusions[16] describes two-composite system for any fraction ratios and respects the geometry by considering polarization effects due to local fields when it calculates the effective permittivity of the composite system.

In ($B^{\parallel}$) case we use linear polarization having electric field component parallel with axes of vortex cores. Therefore local charges inside the sample are not induced and Bruggeman's theory gives a simple result for permittivity

$$\tilde{\varepsilon}_{eff} = f_n \tilde{\varepsilon}_n + (1 - f_n)\tilde{\varepsilon}_{sc}, \tag{8}$$

where $\tilde{\varepsilon}_n$ and $\tilde{\varepsilon}_{sc}$ are permittivities of normal and superconducting phases, respectively.

In ($B^{\perp}$) case, electric field of the laser beam acts in the direction perpendicular to the vortex core axis and creates local field that is different from the applied field. It can be shown that the following formula applies:[16]

$$\tilde{\varepsilon}_{eff} = \frac{1}{2}(\beta + \sqrt{\beta^2 + 4\tilde{\varepsilon}_n \tilde{\varepsilon}_{sc}}), \tag{9}$$

where $\beta = (2f_n - 1)\tilde{\varepsilon}_{sc} + (1 - 2f_n)\tilde{\varepsilon}_n$. The theoretical lines shown in Figs. 2 and 3 were calculated using this approach. Despite the simplicity of the model, the experimental data sets are described surprisingly well, including the qualitative difference between Faraday and Voigt sample orientations.

We emphasize that our model does not take into account any vortex motion. In principle, the laser beam interacts with vortices also via the Magnus force; this effect is neglected in our analysis. Good agreement between calculated and observed temperature-dependent transmission suggests that this term is either small or the vortex lattice in our NbN sample is strongly pinned.

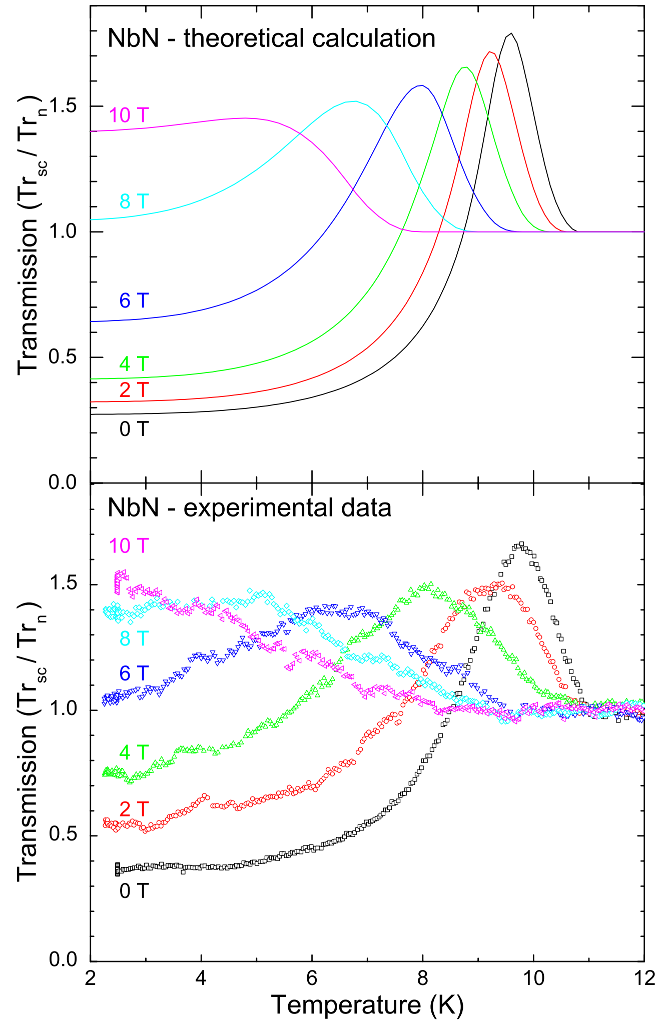

FIG. 3. (Color online) Temperature-dependent transmission in Voigt orientation ($B^{\parallel}$). Experimental data are points and theoretical calculations are lines.

IV. CONCLUSIONS

We have investigated far-infrared magneto-optical properties of the thin-film NbN sample (B up to 10 T) deposited on a Si substrate using our new experimental apparatus. As for T-dependent transmission in $B=0$, presented measurements agree with previous data obtained with the same sample using the old experimental equipment.[4] Two cases are probed by laser beams of two different frequencies—(i) energy above the optical gap and (ii) energy below the optical gap (its zero-temperature value), displaying qualitatively different behavior. The above-mentioned model describes a behavior of transmission for both cases qualitatively well, but better quantitative agreement is achieved by modeling our imperfect real sample as a mixture of superconductors with different T_c and corresponding optical gaps $2\Delta = 4.16 k_B T_c$.

We have probed the Abrikosov state of our sample using 0.4037 THz laser beam by measuring $Tr(T, B_{c1} < B \leq 10\text{ T})$ curves in both ($B^{\perp}$) and ($B^{\parallel}$) geometries, giving qualitatively different results. For clarity, in this work we present only the $B \neq 0$ data obtained with one sample using one particular laser line as a representative data set, although data obtained with several laser lines on this sample as well as on additional NbN samples are available. Further detailed investigations are under progress and will be reported elsewhere.

We are not aware of any relevant microscopic theory that would describe transmission in magnetic field. We have therefore developed a phenomenological theoretical model, based on the following assumptions. Vortex lattice was simplified according to Clem model with entirely normal-state vortex cores and a fully superconducting surrounding bulk—a mixture of ideal superconductors with different T_c spread over about 1 K. The corresponding $Tr(T,B)$ curves have been calculated by means of Bruggeman's approach, allowing to take into account various mutual orientations of $\boldsymbol{B}$ and electric component $\boldsymbol{E}$ of the polarized terahertz radiation, leading to different induced local fields. Good semiquantitative agreement with the observed data suggests that our phenomenological model is capable of capturing essential physics and can be used for further investigations aiming at determining the role of vortex motion that would lead to better understanding and eventually to practical use of superconductors in fluxonics.

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