Dynamic conductivity and coherence peak in the submillimeter spectra of superconducting niobium nitride films

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A technique has been developed for measuring the electrodynamic properties of superconducting films in the submillimeter wavelength range. The temperature dependence and frequency dependence of the dynamic conductivity and the dielectric constant of superconducting NbN films have been measured in detail for the first time in the frequency interval 5–39 cm^{-1} at 5–300 K. A coherence peak has been seen for the first time on the temperature dependence of the dynamic conductivity.

1. Among classical (low-temperature) superconductors, niobium nitride is one of the most popular compounds for use in rf electronic devices. The reasons for this popularity are the relatively high transition temperature of NbN (13–18 K), its mechanical strength, and its chemical stability. Still, the literature reveals essentially no experimental data on such important characteristics of NbN as the absorption and dielectric properties at frequencies near and below the superconducting energy gap 2Δ, which falls in the submillimeter region of the spectrum. The data which are available, found by conventional methods (Refs. 1–4, for example), are greatly limited because of difficulties in measuring the absorption of electromagnetic radiation in the superconducting phase.

Our purposes in the present study were accordingly to develop a technique for reliably measuring the electrodynamic properties of superconducting films in the submillimeter wavelength region and to carry out systematic measurements of the submillimeter spectra of the dynamic conductivity and the dielectric constant of superconducting niobium nitride films.

2. The test samples were plane-parallel, isotropic, crystalline sapphire substrates with a thickness of about 0.5 mm on which superconducting films of NbN, 100–500 Å thick, were deposited on one or two sides. The films were grown by magnetron sputtering. Their transition temperature was about 12 K, and the transition width 0.3–0.5 K.

All the submillimeter measurements were carried out on an Épsilon laboratory backward-wave submillimeter spectrometer, which is described in detail in Ref. 5 (the backward-wave tube is a source of electromagnetic radiation). The submillimeter spectra of the transmission coefficient t and the phase shift φ of electromagnetic radiation transmitted through the sample were measured in the experiments. The properties of the NbN films were calculated from the spectra with the help of known equations[6] for the transmission of layered media. The dielectric properties of the sapphire substrates were measured beforehand. The measurements were carried out over the frequency interval $\nu=5$–$39\ \text{cm}^{-1}$ and the temperature interval $T=5$–300 K.

3. We begin by looking at the experimental results found in the conventional experimental layout, which uses a sample consisting of a superconducting film deposited on one side of a substrate. This system is essentially an asymmetric Fabry–Perot cavity, whose mirrors are the test film and the free face of the substrate.

Figure 1a shows submillimeter transmission spectra of a NbN film 115 Å thick on a sapphire substrate 0.43 mm thick. The spectra shown here correspond to temperatures in the normal and superconducting phases of NbN. The oscillations in the spectra are a consequence of an interference of the radiation inside the substrate. The distance between oscillations is determined primarily by the substrate refractive index and thickness, while the peak heights are determined primarily by the transparency of the film. In the normal phase, the transmission spectra change only slightly as the temperature is lowered from room temperature to near T_c. When the film goes superconducting, substantial changes occur in the spectra: The low-frequency transmission decreases by almost an order of magnitude, while the high-frequency transmission increases.

Figure 1b shows the temperature dependence of the dynamic conductivity σ and the dielectric constant ε' of an NbN film as calculated from the transmission spectra in Fig. 1a for a frequency of 5 cm^{-1}. In the normal phase, a lowering of the temperature is accompanied by a slight decrease in the conductivity, while the dielectric constant remains essentially constant at $\varepsilon'\approx 0$. When the sample goes into the superconducting phase, the dielectric constant decreases sharply (to a level on the order of -10^6). This result agrees with the picture of the optical properties of superconductors drawn by the BCS theory.[7–10] On the other hand, although the transmission spectra $t(\nu)$ were measured quite accurately, there is almost nothing definite we can say about the behavior of the dynamic conductivity in the superconducting state. The difficulty lies in the substantial increase in the errors in the determination of σ.

The reason for the latter circumstance is that when the sample goes into its superconducting state there is a sharp

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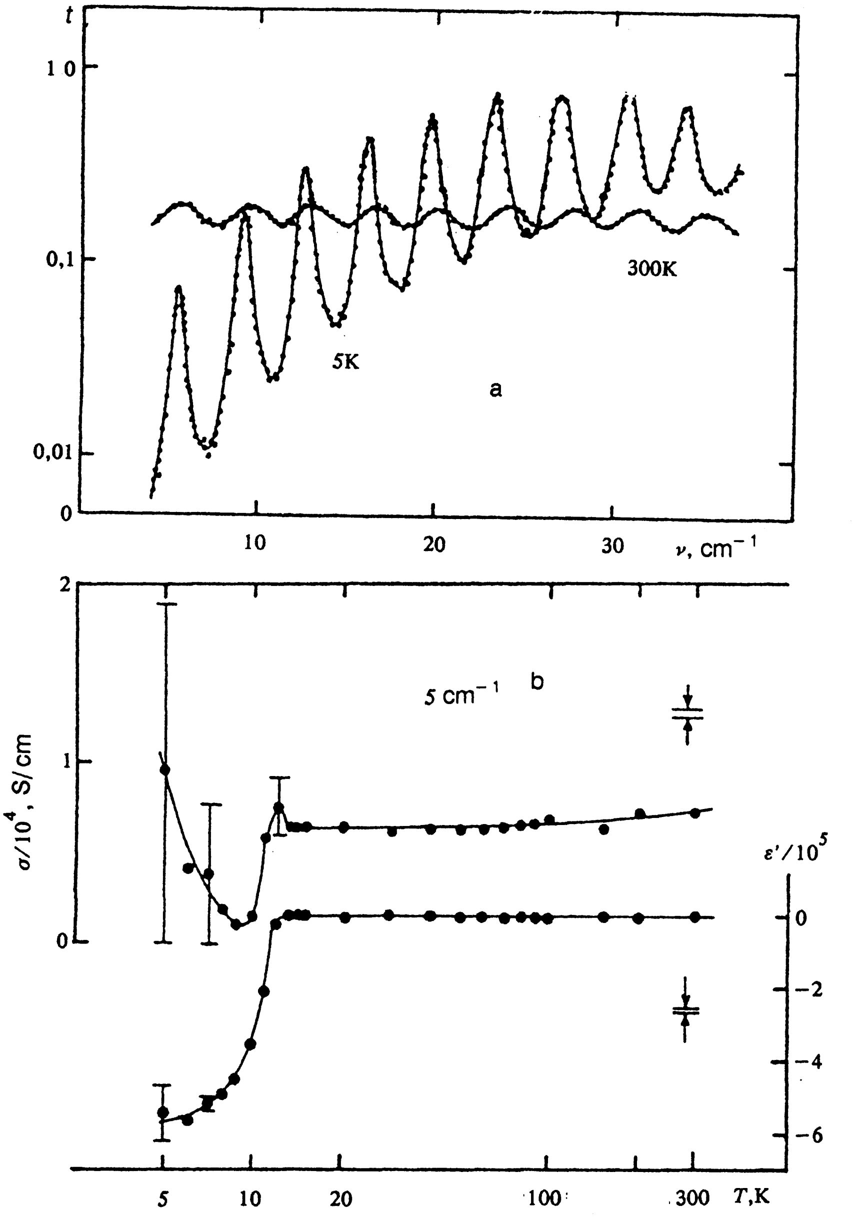

FIG. 1. a) Spectra of the transmission coefficient of an asymmetric Fabry–Perot cavity formed by a NbN film (115 Å) on a sapphire substrate (0.43 mm). These spectra were measured at two temperatures, above and below the transition temperature $T_c \approx 12$ K. b) Temperature dependence of the dynamic conductivity and the dielectric constant of a NbN film at the frequency 5 cm^{-1}, as calculated from the transmission spectra in part a of this figure.

decrease[7–10] in the absorption of the electromagnetic radiation in the material at frequencies below the superconducting energy gap 2Δ (i.e., decreases occur in the dynamic conductivity $\sigma = nk\nu$; in the real part of the surface impedance, $R_s = X_0 n(n^2+k^2)^{-1}$, where $X_0 = 376.7\ \Omega$; and in the refractive index n), along with a significant increase in the absolute values of the attenuation coefficient $k \gg 1$ and the dielectric constant $|\varepsilon'| \gg 1$, $\varepsilon' = n^2 - k^2 < 0$. Analysis has shown[11,12] that the properties T and φ measured in such a situation are relatively insensitive to properties characterizing the absorption of electromagnetic radiation. This is the reason for the sharp increase in the errors in the determination of σ, R_s, and n in the superconducting phase.

The situation here is typical of not only this particular geometry for submillimeter-range measurements (a film on a substrate; see also Refs. 11 and 12) but also of the conventional methods in the neighboring microwave and far-IR regions (Refs. 2 and 13, for example).

4. Our study has shown that the sensitivity of submillimeter-range measurements can be raised dramatically by using a technique based on a sample made up of two identical test films on a dielectric substrate. This test sample is essentially a symmetric Fabry–Perot cavity. The advantages of this approach result from the sharp increase in the Q of the system or, equivalently, the increase in the effective number of reflections of a wave inside the cavity.

Figure 2a shows submillimeter transmission spectra of a symmetric Fabry–Perot cavity formed by two NbN films (540 Å) on a sapphire substrate (0.39 mm). Spectra are shown for 300 K (in the normal phase) and 5 K (the superconducting phase). As in the case of the asymmetric cavity (Fig. 1a), the basic changes in the spectra occur when the film goes superconducting. The heights of the interference peaks, $\tau_{\max} = (1+A/T)^{-2}$, and their quality factors Q increase sharply [for two films in free space we would have $\tau_{\max}$ and $Q = \pi R^{1/2}(1-R)^{-1}$; Ref. 6]. The increase in Q and $\tau_{\max}$ implies a decrease in the loss A and an

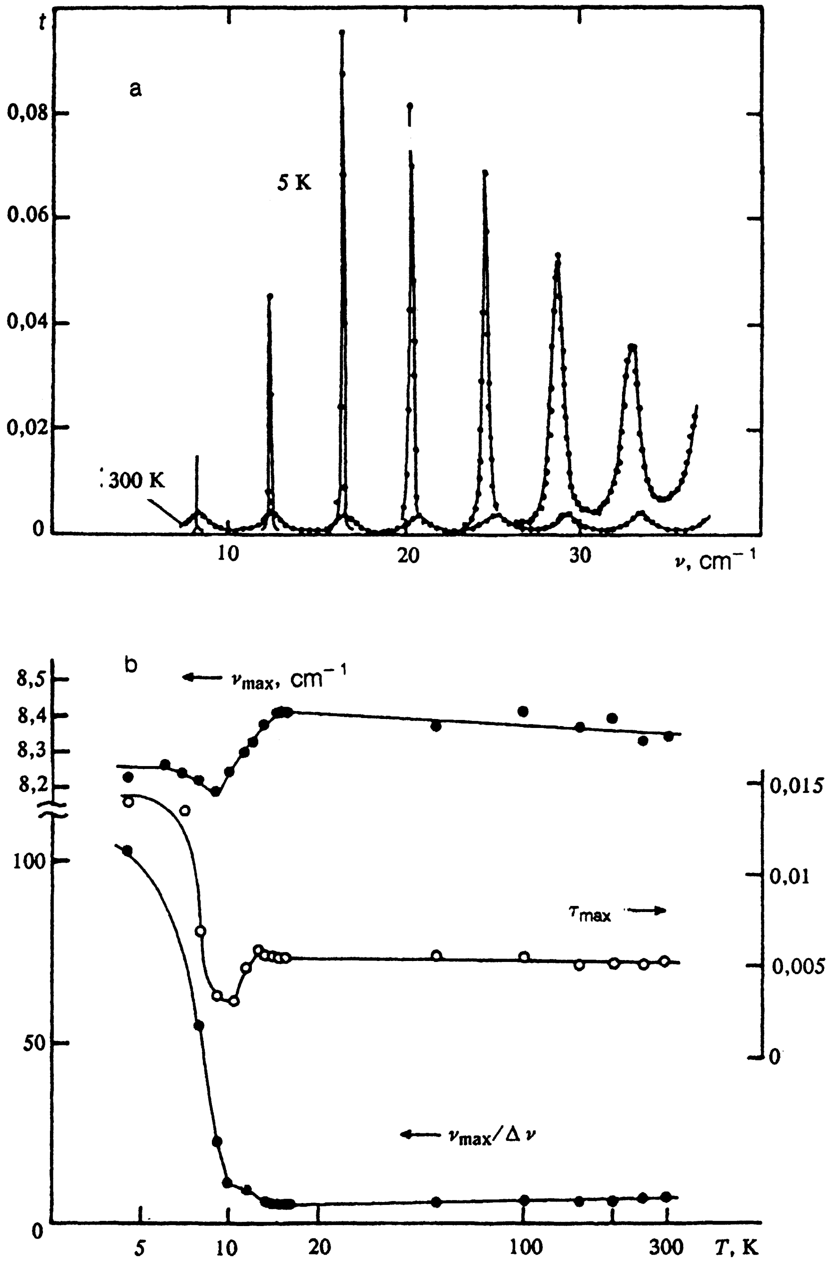

FIG. 2. a) Spectra of the transmission coefficient of a symmetry Fabry-Perot cavity formed by two identical NbN films (540 Å) on a sapphire substrate (0.39 mm). These spectra were measured at two temperatures, above and below the transition temperature $T_c \approx 12$ K. b) Temperature dependence of the properties of the interference peak (its frequency, height, and Q) of a Fabry-Perot cavity (as in part a of this figure).

increase in the reflection coefficients R of the cavity "mirrors." For the peak at the frequency of 8 cm^{-1}, at $T = 5$ K, the value of $\tau_{\max}$ nearly triples, and the corresponding quality factor increases by more than an order of magnitude (Fig. 2b). The temperature dependence of the characteristics of the interference peak near the transition temperature is not monotonic.

Figures 3–6 show the temperature dependence and the frequency dependence of the electrodynamic properties of NbN as calculated from the transmission spectra of the symmetric Fabry-Perot cavity. We see from these figures that the symmetric measurement layout permits a fully reliable determination of the electrodynamic properties (within $\pm 20\%$), in particular, of the conductivity of the NbN in both the normal and superconducting phases.

The temperature dependence and frequency dependence of the submillimeter properties of NbN films according to the present experiments agree qualitatively with the data of Refs. 11 and 12. There is some difference in the absolute values of σ, apparently because of a difference in the grain size in the films.

In the normal phase the dynamic submillimeter properties of NbN correspond to the Drude conductivity model with noninteracting free carriers[14] in the low-frequency limit $\nu \ll \gamma$ (γ is the carrier relaxation frequency): At no temperature $T > T_c$ is there dispersion in the submillimeter spectra of the dynamic conductivity or the dielectric constant (spectra 1 in Fig. 3), and we have $n \approx k \approx (\sigma/\nu)^{1/2}$ (Fig. 4a). The conductivity does decrease somewhat with decreasing temperature, possibly because of localization effects associated with the granular structure of the films.[2]

When the NbN films go superconducting, the temperature and frequency dependence of the electrodynamic properties of these films corresponds to the picture of the

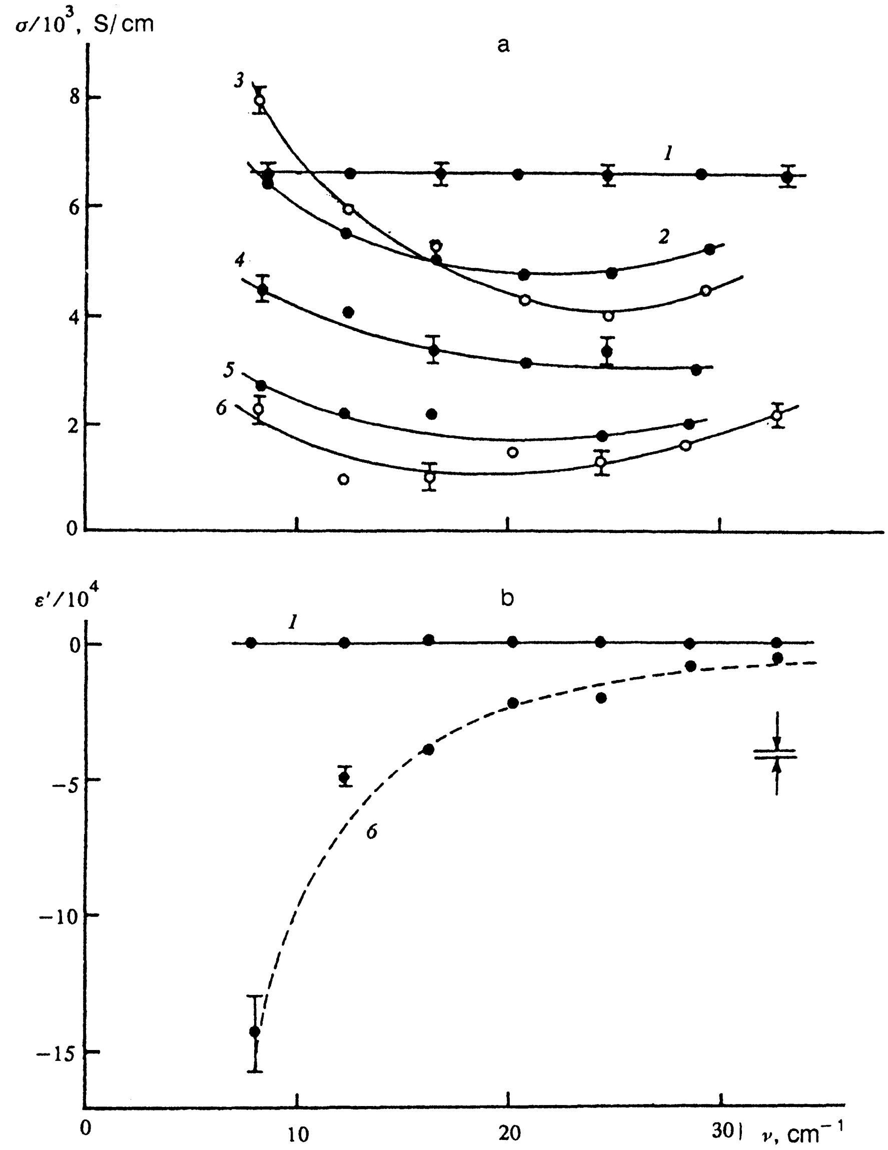

FIG. 3. Submillimeter spectra of the conductivity and dielectric constant of a NbN film at various temperatures. *1*—150–300 K; *2*—11 K; *3*—10 K; *4*—8 K; *5*—6 K; *6*—5 K. The dashed line shows a calculation from the two-fluid model of superconductivity.

optical properties of superconductors at low frequencies $\nu < 2\Delta$ drawn in the BCS model.[7-10]

When the sample goes into the superconducting phase, we see a monotonic decrease in the conductivity σ at relatively high frequencies (on the order of 30 cm^{-1}; Figs. 3a and 4b). This result corresponds to the opening of an energy gap in the spectrum of excitations. At lower frequencies (on the order of 8 cm^{-1}) the behavior is more complex: The conductivity at first increases and then decreases as the temperature is lowered below the transition temperature. This behavior of the dynamic conductivity is known as a "coherence peak." The presence of a coherence peak in NbN is evidence that the ground state is a singlet state.[7] The presence of a peak on the temperature dependence of the conductivity corresponds to nonmonotonic temperature dependences of the refractive index n and of the attenuation coefficient k (Fig. 4a). It also corresponds to peaks on the temperature dependences of the real and imaginary parts of the surface impedance (Fig. 5).

The dynamic dielectric constant ε' of NbN decreases sharply, from zero to values on the order of -10^5, upon the transition to the superconducting phase (at a frequency of 8 cm^{-1}; Fig. 4c). The frequency dependence of the dielectric constant in the superconducting state is described by the London law (the dashed line in Fig. 3b)

$$\varepsilon' = -c^2(4\pi^2\nu^2\lambda_L^2)^{-1},$$

where λ_L is the London penetration depth.

Figure 6 shows the frequency and temperature dependence of the penetration depth for electromagnetic radiation, $\lambda = (2\pi k\nu)^{-1}$, calculated from the experimental data. At room temperature, the penetration depth has a $\nu^{-1/2}$ frequency dependence, in agreement with the Drude conductivity model. When the sample goes superconducting, the penetration depth decreases and becomes frequency-independent, in agreement with model-based predictions.[7-10] The sharp decrease in the penetration depth upon the transition to the superconducting state is described well by the expression from the two-fluid superconductivity model:[8]

$$\lambda(T) = \lambda(0)[1-(T/T_c)^4]^{-1/2}.$$

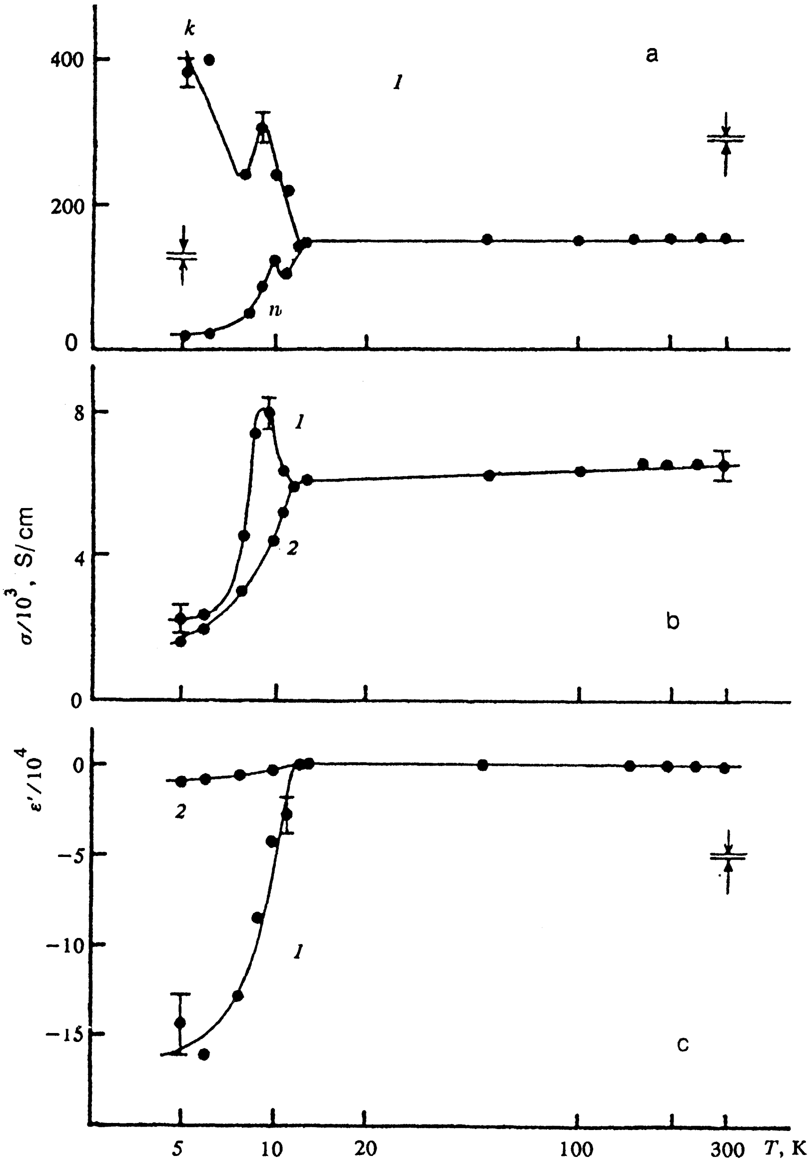

FIG. 4. Temperature dependence of submillimeter optical properties of a NbN film calculated for two frequencies. *1*—8; *2*—20 cm^{-1}.

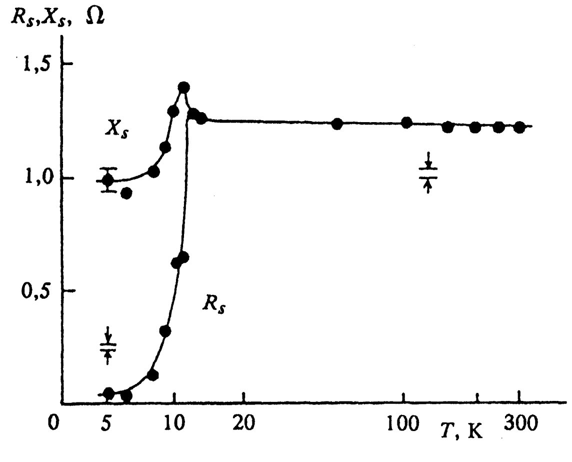

FIG. 5. Temperature dependence of the real and imaginary parts of the surface impedance of a NbN film at 8 cm^{-1}.

Using this expression, we find the penetration depth for electromagnetic radiation in the limit $T\rightarrow 0$ to be $\lambda_0=0.5$ μm, and we find a plasma frequency $\nu_p=c/\lambda_0=3200$ cm^{-1}. These results agree with data in the literature.[1-4]

5. In summary, a technique has been developed for quantitative measurements of the electrodynamic properties of superconducting films in the submillimeter wavelength region. This technique is based on the use of a Fabry–Perot interferometer as the test sample. The interferometer is formed by two test films, deposited on two sides of a dielectric substrate.

With the help of this technique, we have carried out the first systematic measurements of the electrodynamic properties of superconducting films of niobium nitride, NbN, at frequencies in the interval 5–39 K and at temperatures of 5–300 K. A coherence peak has been seen for the first time on the temperature dependence of the dynamic conductivity of NbN. This peak indicates that the ground state in this compound is a singlet state. The temperature and frequency dependences of the properties of the films

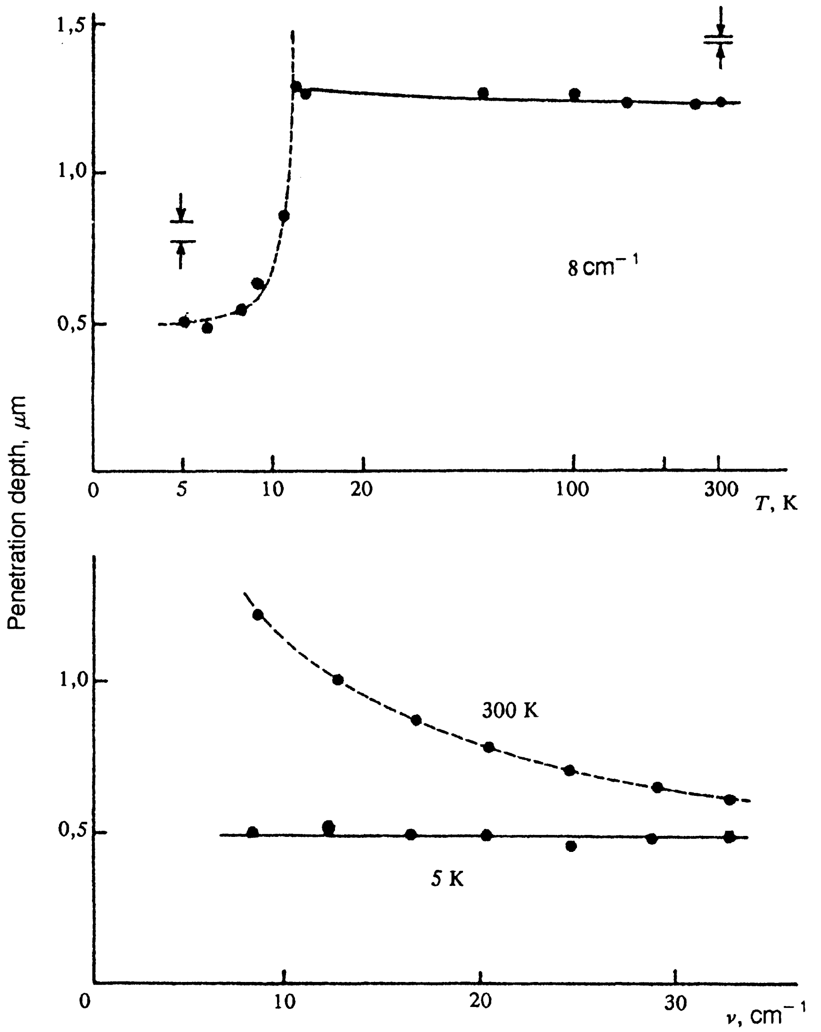

FIG. 6. Temperature and frequency dependence of the penetration depth of submillimeter radiation in a NbN film (for two temperatures, above and below T_c). The dashed lines show calculations from the two-fluid model of superconductivity (top) and from the Drude conductivity model (bottom).

agree with the current understanding of the optical properties of classical (low-temperature) superconductors at frequencies below the energy of the superconducting gap.

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