Optical conductivity and penetration depth in MgB$_2$

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The complex conductivity of a MgB$_2$ film has been investigated in the frequency range $4$ cm$^{-1} < \nu < 30$ cm$^{-1}$ and for temperatures $2.7$ K $< T < 300$ K. The overall temperature dependence of both components of the complex conductivity is reminiscent of BCS-type behavior, although a detailed analysis reveals a number of discrepancies. No characteristic feature of the isotropic BCS gap temperature evolution is observed in the conductivity spectra in the superconducting state. A peak in the temperature dependence of the real part of the conductivity is detected for frequencies below $9$ cm$^{-1}$. The superconducting penetration depth follows a $T^2$ behavior at low temperatures.

Recent discovery of superconductivity at relatively high temperature ($T_c \approx 39$ K) in a simple binary compound, magnesium boride [1], has stimulated extensive theoretical and experimental studies in this material. Most of these studies are attempting to find the pairing mechanism that leads to the superconductivity in this compound [2]. The strongest evidences for phonon-mediated superconductivity in MgB$_2$ comes from the boron-isotope effect measured by the P. Canfield group [3]. Further confirmation for such a mechanism would be an observation of characteristic features predicted by the BCS-theory for different experimentally measured quantities, such as an s-wave superconducting gap in tunneling and optical experiments, or a coherence peak in NMR and in the real part of the low-frequency conductivity. During several months passed from the discovery of superconductivity in MgB$_2$, large experimental efforts have been undertaken to observe such features. However, the absence of high-quality samples hampers the collection of reliable experimental results. The tunneling data obtained on ceramic samples [4] confirm roughly the BCS predictions for a s-wave gap, although the ratio $2\Delta(0)/kT_c$ varies significantly from one report to another, being between 2.5 to 4. These variations together with some deviations from the temperature dependence of the BCS gap have mostly been explained by imperfections of the sample surface, but alternative explanations e.g. based on multiple gaps have also been proposed [5]. The photoemission data [6] can be best fitted with an isotropic s-wave gap using $2\Delta(0)/kT_c = 3$. The NMR experiments [7] reveal a tiny coherence peak with $2\Delta(0)/kT_c = 5$, which indicates that strong coupling regime dominates in MgB$_2$. The temperature dependence of the penetration depth, which is indicative to the gap symmetry, shows either quadratic (muon spin rotation experiments [8]) or linear ($H_{c2}$ measurements [9]) laws, both being inconsistent with an activated behavior, predicted by the BCS-theory. Only one optical study has been reported till now to our knowledge [10].

The superconducting-to-normal grazing reflectivity ratio demonstrates a gradual increase at frequencies below 70 cm$^{-1}$, which might be considered as a sign of a superconducting gap with $2\Delta(0)/kT_c = 2.6$ [11]. Therefore, the experimental data are rather controversial, and there is no general agreement, whether MgB$_2$ is a BCS-type superconductor or not.

In this paper we report on optical investigations of a high quality MgB$_2$ film in the submillimeter frequency range ($4$ cm$^{-1} < \nu < 30$ cm$^{-1}$). Both components of the complex conductivity $\sigma^*(\nu, T) = \sigma_1 + i\sigma_2$ have been directly measured as function of temperature and frequency. This kind of measurements has been already applied to a variety of high-temperature and conventional superconductors and proven to be a powerful method for studying the electrodynamical properties of these materials [11,12].

The MgB$_2$ film has been grown by two-beam laser ablation [13] on a plane-parallel sapphire substrate $10 \times 10$ mm$^2$ in size. The crystallographic orientation of the substrate was [1102] with a thickness of about 0.4 mm. Magnetization measurements of the film have indicated a sharp superconducting transition at 32 K with a width of 1 K (upper frame of Fig. 1). The details of the growth process will be given elsewhere [14]. The measurements in the submillimeter-wave range have been performed using a coherent source spectrometer [15]. Four backward-wave oscillators (BWO’s) have been employed as monochromatic and continuously tunable sources covering the range from 4 cm$^{-1}$ to 30 cm$^{-1}$. The Mach-Zehnder interferometer arrangement has allowed measuring both the intensity and the phase shift of the wave transmitted through the MgB$_2$ film on the substrate. Using the Fresnel optical formulas for the complex transmission coefficient of the substrate-film system, the complex

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conductivity has been determined directly from the measured spectra. The optical parameters of the bare substrate have been obtained in a separate experiment. In the present study we have measured the frequency spectra of the transmittance and of the phase shift at several temperatures in the normal (above $T_c$) and in the superconducting states of MgB$_2$. In addition, we have also measured temperature dependences of these quantities at a fixed frequency (4.1 cm$^{-1}$) continuously from 2.7 to 60 K. A more detailed description of the experimental setup and of the data analysis is given in Refs. [11-13].

The lower frame of Fig. 2 represents the temperature dependence of the transmittance and the phase shift of MgB$_2$ film at $\nu = 4.1$ cm$^{-1}$. Above the superconducting transition both quantities are almost temperature independent. This behavior is consistent with the absence of temperature dependence of the dc-resistivity in MgB$_2$ in this temperature region [11]. The onset of the superconductivity is immediately reflected in the transmittance and phase shift. Both start to decrease below $T_c$, corresponding to an increase of $\sigma_2$ in the superconducting phase due to Meissner effect.

Examples of the complex conductivity spectra, directly calculated from the frequency-dependent transmittance and phase shift, are shown in Fig. 3. The normal-state conductivity (35 K curve) demonstrates a typical metallic behavior. The real part of conductivity is essentially frequency independent, while the imaginary part is almost zero and exhibits a small linear increase for increasing frequencies. This indicates a Drude conductivity with a scattering rate above the measured frequency window. Simultaneous fitting of the $\sigma_1$ and $\sigma_2$ spectra with a Drude model gives an estimate for the scattering rate at 35 K: $1/2\pi\tau = 150^{+70}_{-50}$ cm$^{-1}$.

The transition into the superconducting state gives rise to significant changes in the spectra. $\sigma_2$ starts to diverge for $\nu \to 0$ (Fig. 3 upper frame). This divergence increases with decreasing temperature, reflecting a growth of the spectral weight of the superconducting condensate. The frequency dependence of $\sigma_2$ can be well described by the $1/\nu$-dependence, that corresponds to the $\delta$-function in $\sigma_1$ at $\nu = 0$ via the Kramers-Kronig relations. In the superconducting state a pronounced frequency dispersion arises in the $\sigma_1$ spectra as well (Fig. 3 lower frame). At low frequencies ($\nu < 9$ cm$^{-1}$) and starting from the normal state $\sigma_1$ initially increases (by approximately a factor of 1.5 at the lowest frequency) and then decreases, while at higher frequencies, $\nu > 9$ cm$^{-1}$, $\sigma_1$ monotonously decreases upon cooling. The spectra of $\sigma_1$ are quite smooth, and no characteristic BCS behavior with a pronounced minimum at $\nu = 2\Delta(T)$ (a "dip") is detected. This "dip" is predicted by the BCS theory for a dirty-limit superconductor with an isotropic $s$-wave gap at temperatures between $T = 0$ and $T = T_c$ [16] and has been observed in conductivity spectra of conventional superconductors (e.g. Ref. [11]). The absence of this feature in the $\sigma_1(\nu)$ spectra does not allow direct determination of the superconducting gap value, and might indicate an unconventional character of the gap parameter in MgB$_2$. Another possibility would be a fast developing of the energy gap, which removes the characteristic gap-signature from the measured spectra already for $T$ close to $T_c$. At the lowest temperatures ($\sim 3$ K) $\sigma_1$ almost approximates zero at higher frequencies (within the experimental accuracy), indicating the absence of the normal carrier contribution to the electromagnetic response at $T \to 0$, and confirming the high quality of the film.

From the temperature evolution of the conductivity spectra it becomes clear that a maximum in the temperature dependence of $\sigma_1$ should exist below $T_c$ and at low frequencies. The temperature dependences of $\sigma_1$ and $\sigma_2$ for several frequencies are shown in Fig. 4. The temperature dependence of $\sigma_1$ for $\nu = 4.1$ cm$^{-1}$ is calculated from the transmittance and phase-shift measurements, presented in Fig. 2. (For better representation the data for $\nu = 4.1$ cm$^{-1}$ are shown in a separate frame.) The data in the two other frames were taken from the frequency spectra (Fig. 3). Thick lines in Fig. 4 show the weak-coupling BCS calculations for the frequencies indicated. Taking the ratio $2\Delta/kT_c = 3.53$, the scattering rate $1/\tau$ remains the only free parameter for these calculations. We have chosen $1/2\pi\tau = 60$ cm$^{-1}$ that the calculations at the lowest temperatures match the experimental values of $\sigma_2$. It has to be noted already at this point that this scattering rate disagrees with $1/2\pi\tau = 150$ cm$^{-1}$ obtained from the normal-state conductivity. Taking $1/2\pi\tau = 150$ cm$^{-1}$ in the BCS-calculations substantially overestimates the low-temperature values of $\sigma_2$. The BCS curves, even with the lowered $1/2\pi\tau = 60$ cm$^{-1}$, do not perfectly fit to the experimental points, and we were not able to significantly improve the simultaneous fit of $\sigma_1$ and $\sigma_2$ data neither by taking different ratios of $2\Delta/kT_c$, nor by introducing a distribution of superconducting transition temperatures in our calculations.

The peak in the temperature dependence of $\sigma_1$ vanishes at frequencies above 9 cm$^{-1}$, and the temperature dependence of $\sigma_1$ at higher frequencies basically follows the behavior shown in Fig. 2 for $\nu = 16$ cm$^{-1}$. The observed peak in $\sigma_1(T)$ is reminiscent of the coherence peak, predicted for a BCS superconductor in the dirty limit. However, the interpretation of this peak in terms of the BCS theory might contradict to the absence of the characteristic BCS gap signature in $\sigma_1(\nu)$ spectra discussed above. Both features (the coherence peak and the gap in $\sigma_1(\nu)$) should appear in the dirty BCS limit, and none of them is expected in the clean limit.

The measured $\sigma_1(T)$ dependencies for all frequencies are higher then the BCS calculations. This disagreement could be caused by an additional absorption. This additional absorption may smear out the BCS gap signature, or may indicate an unconventional pairing mechanism. It is difficult to say, whether this absorption is of intrin-
sir or extrinsic origin. However, the absorption tends to disappear at $T \to 0$ (see also the lower panel of Fig. 3), which seems to be in favor of an intrinsic nature of the absorption.

The temperature dependence of the imaginary part of the complex conductivity (Fig. 3, lower panel) deviates significantly from the BCS curves. Qualitatively the same deviations from the BCS calculations have been found for all measured frequencies: the slope of the experimental $\sigma_2(T)$ curves is more gradual at temperatures just above $T_c$, and at $T \to 0$ it is steeper than the BCS prediction. Since the superconducting penetration depth is directly connected to the imaginary part of conductivity via $\lambda = (\mu_0 \omega \sigma_2)^{-1/2}$ ($\mu_0$ is the vacuum permeability and $\omega = 2\pi\nu$ is the angular frequency), for the further discussion we focus on the temperature dependence of the penetration depth (Fig. 4).

Fig. 4 compares the penetration depth in MgB$_2$ with the predictions of different models. The experimental data (open triangles) have been calculated from the temperature dependence of the transmittance and phase shift measured at $\nu = 4.1$ cm$^{-1}$ (Fig. 3). The solid line represents the weak-coupling BCS calculations with the parameters used in Fig. 3. The dotted line shows the result of the two-fluid model $[1 - (T/T_c)^{4}]$. The dashed line represents the $[1 - (T/T_c)^{2}]$ dependence. The first two dependencies reproduce the temperature variation of the penetration depth in conventional superconductors $[14]$, and the last one often reasonably fits the experimental penetration depth in the high-$T_c$ cuprates $[17]$. The experimental data in Fig. 4 clearly deviate from all these model curves in the whole temperature range below $T_c$.

The inset of Fig. 4 shows the temperature variation of the penetration depth as a function of $T^2$ and at low temperatures. The experimental points below approximately 15 K closely follow a straight line, indicating that a $T^2$ behavior is a good approximation to the low-temperature data (in agreement with the muon-spin-rotation measurements $[15]$). The power-law dependence of the penetration depth in the zero-temperature limit has been observed in various high-$T_c$ superconductors and interpreted as an evidence for the existence of nodes in the gap function. However, in the case of a strongly anisotropic s-wave energy gap the transition to the BCS-like exponential behavior may take place at even lower temperatures than obtained in the present study ($\sim 2.7$ K). Therefore, the results of Fig. 4 suggest the strong gap anisotropy, or may even indicate the existence of the nodes in the gap function.

In conclusion, we have measured the complex conductivity of MgB$_2$ film in the frequency range $4$ cm$^{-1} < \nu < 30$ cm$^{-1}$ and for temperatures $2.7$ K $< T < 300$ K. The temperature dependence of the real part of the low-frequency conductivity shows a peak below the superconducting transition, which qualitatively agrees with the predictions of the BCS theory. However, detailed comparison of the frequency and temperature behavior of the complex conductivity with the BCS model calculations shows significant discrepancies between the experiment and the theory. In addition, we observe no indications of the temperature evolution of the gap in $\sigma_2(\nu)$ for frequencies below $30$ cm$^{-1}$. The temperature dependence of the penetration depth is consistent with the $\delta \lambda \propto T^2$ behavior below $\sim 15$ K, which implies significant anisotropy in the gap function.

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FIG. 1. Upper panel: Temperature dependence of the magnetic susceptibility of MgB$_2$ film on Al$_2$O$_3$ substrate. Inset shows the data around $T = T_c$ on an enlarged scale. Lower panel: Temperature dependence of the transmittance at a fixed frequency $\nu = 4.1 \text{ cm}^{-1}$ (left scale) and temperature dependence of the phase shift at the same frequency (right scale).

FIG. 2. Frequency dependence of the complex conductivity of MgB$_2$ film above and below $T_c$. Upper panel: imaginary part $\sigma_2$. Solid line represents the $\sigma_2 \propto 1/\nu$ dependence. Lower panel: real part $\sigma_1$. Lines are drawn to guide the eye. Scattering of the experimental points and the mismatch between different BWO’s represent the experimental accuracy.

FIG. 3. Temperature dependence of the complex conductivity of MgB$_2$ film for several frequencies. Upper panel: $\sigma_1$ at 4.1 cm$^{-1}$. Solid line represents the BCS model calculation with $2\Delta/k_BT_c = 3.53$, $1/2\pi\tau = 60 \text{ cm}^{-1}$. Lower panels: temperature dependencies of the real (middle) and imaginary (bottom) parts of the complex conductivity at 8.2 cm$^{-1}$ (full squares) and at 16 cm$^{-1}$ (open circles). Thick lines represent the BCS calculations with the same parameters as in the upper panel, solid line: $\nu = 8.2 \text{ cm}^{-1}$, dashed line $\nu = 16 \text{ cm}^{-1}$. Thin dotted lines are drawn to guide the eye. The error bars are estimated from the scattering of the experimental points in the frequency spectra (Fig. 2).

FIG. 4. Temperature dependence of the low-frequency penetration depth of MgB$_2$ film. Symbols: experimental data as obtained from the temperature-dependent transmittance and phase shift shown in Fig. 1. Lines represent various model calculations, solid: weak-coupling BCS, dotted: two-fluid $[1 - (T/T_c)^4]$, dashed: $[1 - (T/T_c)^2]$. Inset shows the temperature variation of the penetration depth below 20 K as a function of $T^2$. 