Far-infrared transmission studies of a *c*-axis-oriented superconducting MgB_2 thin film

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We have reported far-infrared transmission measurements on a *c*-axis-oriented superconducting MgB_2 thin film in the frequency range 30–250 cm^{-1}. We found that these measurements were sensitive to the scattering rate $1/\tau$ and the superconducting gap 2Δ. By fitting the experimental transmission spectra at 40 K and below, we obtained $1/\tau$ and $2\Delta(0)$ to be (700–1000) cm^{-1} and 42 cm^{-1}, respectively. These two quantities suggest that the MgB_2 film belongs to the dirty limit.

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The surprising discovery of superconductivity in a known compound MgB_2 with a high T_C of about 39 K has attracted a great deal of attention from the solid-state community and initiated a flurry of activity to understand its properties.[1] Most investigations on this binary compound as yet indicate that MgB_2 behaves as a phonon-mediated superconductor.[2,3] However, there is still little consensus on its reported important physical quantities related to electrodynamic and superconducting responses. A wide range of values of the dc resistivity ρ_{dc}, the carrier density *n*, and the superconducting gap 2Δ are reported in the literature. For example, near T_C, the reported values of ρ_{dc} and *n* are in the range from 0.38 (Ref. 4) to 75 $\mu\Omega$ cm (Ref. 1), and from 6.7×10^{22} (Ref. 4) to 1.5×10^{23} cm^3 (Ref. 5), respectively. Moreover, reported values of 2Δ also vary from 3 (Ref. 6) to 16 meV (Ref. 7). Since such physical quantities are closely related to the nature of the superconductivity, correct determination of their values is essential.

It is well known that optical spectroscopy is a powerful tool for measuring important parameters such as scattering rate, $1/\tau$, plasma frequency, 2Δ, and coherence effects.[8] Since the skin depth of light is about 1000 Å in the far-infrared (IR) region, this technique is able to provide information on 2Δ and it is complementary to other surface-sensitive techniques, such as tunneling and photoemission measurements. However, there have been only a few reports on the optical properties of MgB_2. Gorshunov *et al.*[6] reported a lower estimate of $2\Delta(=3–4$ meV) by the grazing incident reflectivity measurement on a polycrystalline MgB_2 pellet. On the other hand, Pronin *et al.*[9] measured the complex optical conductivity on an MgB_2 thin film in the frequency range 4–30 cm^{-1}. Transmission measurement on a superconducting film is a method superior to reflectivity measurement on a superconducting bulk, because the former is much more sensitive to small changes in optical constants than is the latter.[8] However, since the measurements by Pronin *et al.* were limited to 30 cm^{-1} and below, the characteristic features of the gap and its temperature evolution could not be observed.

In this paper, we have reported our investigation of the electrodynamics on a *c*-axis-oriented MgB_2 thin film (T_C ~33 K) using transmission measurements in the frequency range 30–250 cm^{-1}. We found that the transmission spectra $T(\omega)$ were sensitive to changes in the ab-plane sheet resistance $R_\square$ and $1/\tau$. Using the simple Drude model, we have estimated $R_\square$ and $1/\tau$ at 40 K to be 10.3 ± 0.2 $\Omega/\square$ and 700–1000 cm^{-1}, respectively. Below T_C, a peak due to 2Δ appeared and moved towards the higher frequency with decreasing temperature. The ab plane $2\Delta(T)$ seemed to follow the temperature dependence of BCS theory and was estimated to be 42 cm^{-1} (5.2 meV) at 5 K. By comparing the values of $2\Delta(0)$ and $1/\tau$, we suggest that the MgB_2 film belongs to the dirty limit.

A high quality *c*-axis-oriented MgB_2 film was deposited on an Al_2O_3 substrate using a pulsed laser deposition technique. The experimental details were reported elsewhere.[10] X-ray-diffraction measurement showed that most grains were oriented with their *c* axes normal to the substrate, but their ab-plane orientations were random. The dc resistance measurement by the four-probe method showed a sharp T_C near 33 K.[11] Temperature dependent $T(\omega)$ were measured by a Fourier-transform spectrophotometer with a resolution of 5 cm^{-1} in the frequency range 30–250 cm^{-1}.

In the thin-film geometry, $T(\omega)$ can be written into an approximate form when $\lambda \gg \lambda_p \gg d$, where λ is the wavelength of light, λ_p is the skin depth, and *d* is the film thickness. By taking the multiple reflections inside the substrate into account, $T(\omega)$ can be approximated as

$$T(\omega)=\frac{4n}{|1+\tilde{N}+\tilde{y}|^2}\times\frac{T_s\exp(-\alpha x)}{1-R_sR_f\exp(-2\alpha x)}, \qquad (1)$$

where $\tilde{N}(=n+ik), \alpha(=4\pi\omega k)$, and x are the complex refractive index, absorption coefficient, and thickness of the substrate, respectively. $T_s(=4n/|1+\tilde{N}|^2)$ and $R_s(=|1-\tilde{N}|^2/|1+\tilde{N}|^2)$ are the transmission and reflectivity of the substrate-vacuum boundary, and $R_f(=|\tilde{N}-1+\tilde{y}|^2/|\tilde{N}+1+\tilde{y}|^2)$ is the reflectivity of the substrate-film boundary with $\tilde{y}=4\pi\tilde{\sigma}_\square/c$, where $\tilde{\sigma}_\square=\sigma_{1\square}+i\sigma_{2\square}$ is the complex sheet conductance of the film. Using the transmis-

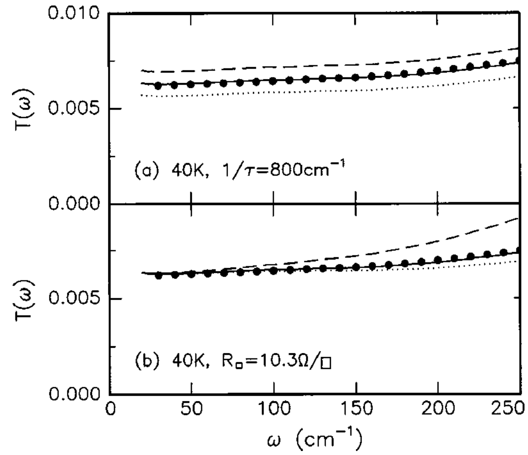

FIG. 1. $T(\omega)$ at the normal state. In (a), the dashed, solid, and dotted lines represent the fitting lines for $R_{\square}=10.8$, 10.3, and 9.6 $\Omega/\square$, respectively. In (b), the dashed, solid, and dotted lines represent the fitting lines for $1/\tau=400$, 800, and 4000 cm^{-1}, respectively.

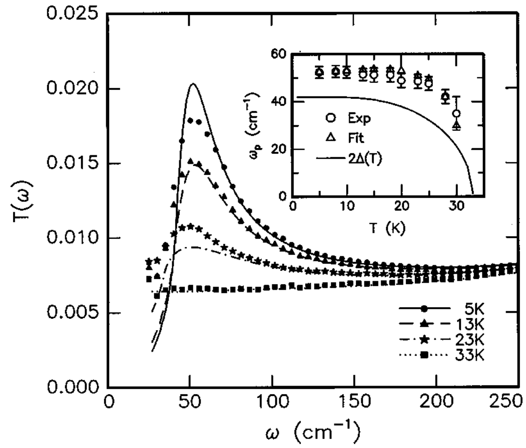

FIG. 2. Experimental and theoretical results of $T(\omega)$ at superconducting states. In the inset, the open circles and open triangles represent ω_P obtained from the experimental and theoretical data, respectively. For comparison, $2\Delta(T)$ is also shown as a solid line.

sion and reflectivity spectra of Al_2O_3, we have obtained $\tilde{N}$ values independently and used these to calculate the theoretical $T(\omega)$.

Far-IR transmission measurements can determine $R_{\square}$ and $1/\tau$ of the film quite precisely. Figures 1(a) and 1(b) show $T(\omega)$ at 40 K. Note that the transmission spectra of the Al_2O_3 substrate are nearly flat and are independent of temperature below 40 K.[12] To understand the effects of $R_{\square}$, we have calculated $T(\omega)$ using Eq. (1) with the simple Drude model. Figure 1(a) shows the variations of $T(\omega)$ at different $R_{\square}$ by keeping the value of $1/\tau$ fixed at 800 cm^{-1}. The dashed, solid, and dotted lines represent the calculated $T(\omega)$ at different $R_{\square}$ such as 10.8, 10.3, and 9.6 $\Omega/\square$, respectively. It is clear that $T(\omega)$ are quite sensitive to $R_{\square}$ and we have found that $R_{\square}$ of our film was 10.3 ± 0.2 $\Omega/\square$ at 40 K.

The frequency dependence of $T(\omega)$ should be dependent on $1/\tau$. At 40 K, $T(\omega)$ gradually increase as the frequency increases, due to the finite value of $1/\tau$. Figure 1(b) shows the variations of $T(\omega)$ at different $1/\tau$ by keeping the value of $R_{\square}$ fixed at 10.3 $\Omega/\square$. The dashed, solid, and dotted lines represent the calculated $T(\omega)$ at different $1/\tau$ such as 400, 800, and 4000 cm^{-1}, respectively. It was found that $1/\tau$ of our film was 800^{+200}_{-100} cm^{-1}.

Figure 2 shows the temperature dependent $T(\omega)$ of the MgB_2 film in its superconducting state, i.e., at temperatures below 33 K. In the superconducting state, $T(\omega)$ show a peaklike structure near 50 cm^{-1}. At 5 K, with an increase in frequency, $T(\omega)$ gradually increase up to 52 cm^{-1} and then approach those at the normal state. As the temperature increases, the peak height decreases and the peak position ω_P shifts towards the lower frequencies.

Similar peak structures have been observed for numerous superconducting thin films, and their 2Δ values were found to be close to ω_P.[13] Such a peak structure can be understood from the complex optical conductivity spectra $\tilde{\sigma}(\omega)$. In the normal state, $\sigma_1(\omega)$ at $\omega\ll1/\tau$ become nearly frequency independent. But in the superconducting state, below $2\Delta,\sigma_1(\omega)$ become suppressed and the missing spectral weight moves towards the zero frequency to form a delta function, representing a superconducting condensate. As a result of the delta function, $\sigma_2(\omega)$ will have a $1/\omega$ dependency. Thus, $T(\omega)\propto\omega^2$ for $\omega\ll2\Delta$, which can be easily seen in Eq. (1). On the other hand, for $\omega\gg2\Delta, T(\omega)$ in the superconducting state will be almost the same as those in the normal state. Near $2\Delta,\sigma_1(\omega)$ are nearly zero and $\sigma_2(\omega)$ drastically decrease, resulting in a minimum value of the denominator in Eq. (1). Thus, there is a peak in transmission.

One important physical quantity in a superconductor is the ratio between $1/\tau$ and $2\Delta(0)$, which will determine the electrodynamic responses.[14,15] For most metal superconductors, such as Al and Pb, $(1/\tau)/2\Delta(0)\gtrsim100$. The optical responses of such BCS superconductors in the extremely dirty limit can be explained by Mattis-Bardeen theory.[16] However, in high-temperature superconductors, the $2\Delta(0)$ values are much larger than those of metal superconductors. Thus the high-temperature superconductors are believed to belong to the clean limit, for which $(1/\tau)/2\Delta(0)\sim1$.[17] Optical properties of superconductors in the clean limit are different from those in the dirty limit.[14,15] Zimmermann *et al.* calculated the optical conductivity of a homogeneous BCS superconductor with arbitrary purity.[18]

In order to explain the temperature dependent $T(\omega)$, we have applied the formula developed by Zimmermann *et al.*[18] to fit our experimental data by using the measured values of $1/\tau$ and $R_{\square}$ at 40 K as 800 cm^{-1} and 10.3 $\Omega/\square$, respectively. As shown in Fig. 2, ω_P of the 5 K data is fitted well with $2\Delta(0)\simeq42$ cm^{-1}. Note that the $2\Delta(0)$ value is smaller than the ω_P value by about 10 cm^{-1}. Based on the assumption that the temperature dependence of $2\Delta(T)$ follows BCS prediction, we have calculated $T(\omega)$ at various temperatures. The solid circles (solid line), solid triangles (dashed line), solid stars (dot-dashed line), and solid squares (dotted line) in Fig. 2 represent the experimental (calculated) $T(\omega)$ at 5, 13, 23, and 33 K, respectively. The predicted temperature dependencies of ω_P and $2\Delta(T)$ are shown in the inset as the open triangles and solid line, respectively. Within the experimental error bars, $2\Delta(T)$ seems to follow the prediction of BCS theory quite well.

Although many experimental studies have been performed on the $2\Delta(0)$ value of MgB_2, there is little consensus on its magnitude and symmetry. From the specific-heat measurements, a couple of groups reported that $2\Delta(0)/k_BT_C \sim 2.4$ (Ref. 19) or 4.2 (Ref. 20), and both were explained in terms of the conventional *s*-wave-type BCS model. Using the same technique, Wang *et al.*[21] reported that the values of $2\Delta(0)/k_BT_C$ varied from 1.2 to 4.2 and suggested a *d*-wave superconductor with nodes in the gap. Using photoemission measurements, Takahashi *et al.*[22] found that the superconducting gap was *s*-like with $2\Delta(0)/k_BT_C \sim 3.0$, but Tsuda *et al.*[23] reported spectroscopic evidence for two gaps with $2\Delta(0)=3.4$ and 11.2 meV. More significantly, in tunneling experiments, the values of $2\Delta(0)$ varied from 4 to 16 meV, and both isotropic and anisotropic gap symmetries were suggested.[24] To explain the large variations of $2\Delta(0)$ values, Haas and Maki[25] recently proposed a model of anisotropic *s*-wave superconductivity.

Our measured value of $2\Delta(0)$, i.e., about 42 cm^{-1} (5.2 meV), is significantly smaller than the BCS prediction for the isotropic *s*-wave superconductor. Since our *c*-axis-oriented film has $T_C \sim 33$ K, $2\Delta(0)/k_BT_C$ can be evaluated to be about 1.8. There are at least two possibilities which can explain the small value of $2\Delta(0)/k_BT_C$. The first possibility is inhomogeneities of our film, which may lead to a wide range of T_C and/or $2\Delta(0)$. However, our $T(\omega)$ cannot be explained by introducing a distribution of $2\Delta(0)$ at a higher-frequency region in our calculation. The second possibility is the anisotropic gap symmetry. Since our measurements were made on the *c*-axis-oriented film, the $2\Delta(0)/k_BT_C$ value represents the ab-plane property. The anisotropic *s*-wave superconductivity, suggested by Haas and Maki,[25] is consistent with the small ab-plane value of $2\Delta(0)$. Further experiments of polarization-dependent optical measurements on a single crystal or an epitaxial film with its *c* axis parallel to a substrate will be useful to prove this gap symmetry.

Many workers claimed that the MgB_2 superconductor should be in the clean limit [i.e., $2\Delta(0) \geqslant 1/\tau$]. Canfield *et al.*[4] estimated the Fermi velocity $v_F = 4.8\times10^7$ cm/s and the mean free path $l=600$ Å. By comparing the superconducting coherence length $\xi_0 \sim 52$ Å,[26] they suggested that MgB_2 should belong to the clean limit. According to the relation $l = v_F \times \tau$, we estimated the value of $1/\tau \simeq 42$ cm^{-1} from their quantities, whereas, our experimental data in the normal state as well as in the superconducting state showed that $1/\tau \approx 800$ cm^{-1}.[27] We have calculated $T(\omega)$ at 5 K with various values of $1/\tau$, and the results are shown in Fig. 3. Note that the calculated $T(\omega)$ with $1/\tau \simeq 42$ cm^{-1} predict a very sharp peak structure near $2\Delta(0)$ and a steep increase of $T(\omega)$ in the high-frequency region, which do not agree with our experimental observations. Our experimental $T(\omega)$ seem to be quite close to the predictions

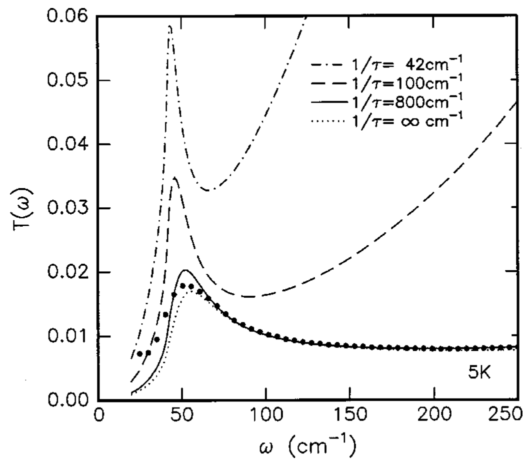

FIG. 3. Calculated $T(\omega)$ at 5 K for various $1/\tau$. The solid circles represent the experimental data. The dot-dashed, dashed, solid, and dotted lines represent $T(\omega)$ at $1/\tau=42$, 100, 800, and ∞ cm^{-1}, respectively.

of Mattis-Bardeen theory in the extremely dirty limit, i.e., $1/\tau=\infty$ cm^{-1}. Although Mattis-Bardeen theory is slightly better than Zimmermann's formula with $1/\tau=800$ cm^{-1} to explain the 5 K data, the temperature dependence of the peak can be explained better by the latter method. Moreover, our 40 K transmission data suggested that $1/\tau \approx 800$ cm^{-1}.

In the literature, there exists a wide range of values for ρ_{dc}. Our measured value of $1/\tau \approx 800$ cm^{-1} provides a certain limitation on the electrodynamic quantities. Using the reported values of $v_F = 4.8\times10^7$ cm/s and $n = 6.7\times10^{22}$ cm^3,[4] we have estimated values of l and ρ_{dc} to be 32 Å and 24 $\mu\Omega$ cm,[28] respectively. Note that our estimated value of l is somewhat smaller than the reported $\xi_0 \sim 52$ Å and that of ρ_{dc} is much larger than the reported 0.38 $\mu\Omega$ cm for an MgB_2 wire.[4] It is clear that further investigations are necessary to determine the physical quantities, such as ρ_{dc} and n.

In summary, we have investigated the *c*-axis-oriented superconducting MgB_2 thin film using far-infrared transmission measurements. By fitting the transmission spectra at normal and superconducting states, we found that the scattering rate was (700–1000) cm^{-1} at 40 K and the zero-temperature superconducting gap was about 42 cm^{-1}. These electrodynamic quantities suggested that the MgB_2 film should belong to the dirty limit.

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Note added in proof. Since this paper was submitted, Kaindl *et al.*[29] reported the far-infrared optical conductivity using terahertz time-domain spectroscopy and drew the same conclusions as ours regarding the size of the superconducting gap and the dirty limit.

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[11] When we deposited 3000 Å thick films using our film fabrication technique, we obtained high quality *c*-axis-oriented MgB_2 films with $T_C \sim 39$ K (Ref. 10). However, their transmissions were too small to be measured.

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[27] Using transmission measurement, Pronin *et al.* (Ref. 9) reported the different values of $1/\tau$, i.e., (628–1382) cm^{-1} at the normal state and 377 cm^{-1} at the superconducting state.

[28] The thickness of the film *d* was estimated to be 500 ± 70 Å by spectroscopic ellipsometry measurement under the assumption that the film was homogeneous. However, due to the possible existence of dead layers at the film surface and the film/substrate interface, we did not directly compare the estimated value of $\rho_{dc} = 24$ $\mu\Omega$ cm with the value of 52 $\mu\Omega$ cm obtained from the measured values of $R_{\square}$ and *d*. Note that if the reduced $T_C \sim 33$ K in our film was due to the reduced carrier density, then the agreement between two values of ρ_{dc} would be better.

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