# Temperature and magnetic field dependent optical properties of superconducting MgB<sub>2</sub> thin film

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# 초전도 $MgB_2$ 박막의 온도와 자기장의 변화에 따른 광학적 성질

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#### Abstract

We investigated the temperature and magnetic field dependent optical properties of a MgB<sub>2</sub> thin film in the far-infrared region. In the superconducting state, i.e. 5 K, we obtained the values of superconducting gap  $2\Delta \sim 5.2$  meV and  $2\Delta/k_BT_C \sim 1.8$ . Although the value of  $2\Delta/k_BT_C$  was nearly half of the BCS value, the  $2\Delta$  seemed to follow the temperature dependence of the BCS formula. Under the magnetic field (*H*), the superconducting state became suppressed. Interestingly, we found that the normal state area fraction abruptly increased at low field but slowly increased at high field. It did not follow the *H*-dependences predicted for a *s*-wave superconductor (i.e. a linear dependence) nor for a *d*-wave one (i.e.  $H^{1/2}$  dependence). We discussed the complex gap nature of MgB<sub>2</sub> in comparison with two gap and anisotropic *s*-wave scenarios.

Keywords: optical properties, MgB2 thin film, complex gap

## I. Introduction

The surprising discovery of superconductivity in  $MgB_2$  with  $T_C=39$  K [1], has attracted a great deal of

attention from the solid-state community and initiated a flurry of activity to understand its properties. Many theoretical and experimental efforts have been paid to understand its superconductivity and suggested the phonon-mediated BCS mechanism [2]. However, there is still little consensus on its reported superconducting gap 2Δ. It varied from 3 meV [3] to 16 meV [4]. Also, the experimental

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evidences for the deviation from an isotropic s-wave gap symmetry, have been increased and they suggested a multiple gap [5] or an anisotropic s-wave gap symmetry [6].

Optical measurements have been revealed to be a powerful method to investigate important physical quantities, such as  $2\Delta$ , the scattering rate  $1/\tau$ , penetration depth  $\lambda$  [7]. Since the skin depth of light is about 1000 Å in the far-infrared region, optical measurements have an important advantage to obtain bulk properties compared to other surface sensitive techniques such as tunneling and photoemission measurements. Moreover, the optical measurement at high magnetic field can provide fruitful information, such as vortex dynamics, quasi-particle excitation, and gap symmetry [8].

In this paper, we investigated the optical properties of a MgB<sub>2</sub> thin film in the far-infrared region with changing temperature (T) and the magnetic field (H). By the T-dependent optical measurement, we obtained  $2\Delta(0) \sim 5.2$  meV and found the BCS T-dependences of  $2\Delta(T)$ . On the other hand, by the H-dependent optical measurement, we found that the area fraction of the normal metallic region followed neither H- nor  $H^{1/2}$  dependences expected for s-wave and d-wave superconductors, respectively. We discussed the complex gap nature of newly discovered MgB<sub>2</sub> superconductor in comparison with the existing theoretical scenarios.

## II. Experiment

A high quality c-axis oriented MgB<sub>2</sub> film was deposited on an Al<sub>2</sub>O<sub>3</sub> substrate using a pulsed laser deposition technique [9]. X-ray diffraction measurement showed that most grains were oriented with their c-axis normal to the substrate. The dc resistivity measurement by the four-probe method showed a sharp  $T_{\rm C}$  near 33 K. The T- (5 ~ 40 K) and H-dependent (0 ~ 17 T) transmission  $T(\omega)$  and reflectivity  $R(\omega)$  spectra were measured for the range  $30 \sim 200 \text{ cm}^{-1}$ using the Fourier transform spectrophotometer.

#### III. Results

Fig. 1 shows the T-dependent  $T(\omega)$  of MgB<sub>2</sub> thin film at H = 0 T. In the superconducting state, i.e.

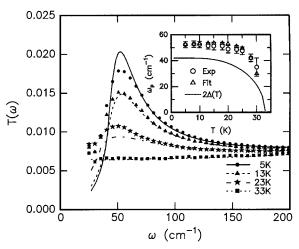


Fig. 1. Experimental (symbols) and theoretical (lines) results of  $T(\omega)$  at superconducting states without H. In the inset, the open circles and open triangles represent  $\omega_P$  obtained from experimental and theoretical  $T(\omega)$ , respectively. The solid line represents the  $2\Delta(T)$ .

below 33K,  $T(\omega)$  show a peak-like structure near 50 cm<sup>-1</sup>. At 5 K, with increase of frequency,  $T(\omega)$  gradually increase up to 52 cm<sup>-1</sup> and then approach those at the normal state, i.e.  $T(\omega)$  at 33 K. As T increases, the peak height decreases and the peak position  $\omega_P$  shifts towards the lower frequencies.

Similar peak structures have been observed for numerous superconductors, and their  $2\Delta$  values were found to be close to  $\omega_P$  [10]. In order to explain the T-dependent  $T(\omega)$ , we have applied the formula developed by Zimmerman et al [11], which calculated the optical conductivity of a homogeneous BCS superconductor with arbitrary purity. Based on the assumption that the T-dependence of  $2\Delta(T)$  follow the BCS prediction, we have calculated  $T(\omega)$  at various temperature and found that the experimental  $T(\omega)$  were explained quite well.

The predicted T-dependences of  $\omega_P$  and  $2\Delta(T)$  are shown in the inset as the open triangles and solid line, respectively.  $\omega_P$  of 5 K data is fitted well with  $2\Delta \sim 42~\text{cm}^{-1}$  (5.2 meV). Within the experimental error bars,  $2\Delta(T)$  seemed to follow the prediction of the BCS theory quite well.

Note that our measured value of  $2\Delta \sim 5.2$  meV is significantly smaller than the BCS prediction for the isotropic s-wave superconductor. Since our c-axis oriented thin film has  $T_{\rm C} \sim 33$  K,  $2\Delta/k_{\rm B}T_{\rm C}$  can be

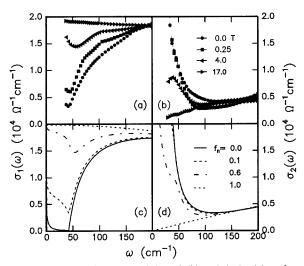


Fig. 2. *H*-dependent (a)  $\sigma_1(\omega)$  and (b)  $\sigma_2(\omega)$ . In (c) and (d), the calculated  $\sigma_1(\omega)$  and  $\sigma_2(\omega)$  using Maxwell Garnett Theory are shown. Here  $f_n$  represents the area fraction of normal metallic region.

evaluated to be about 1.8, which is nearly half of the weakly coupled BCS value of 3.5.

To obtain further information on the gap nature of MgB<sub>2</sub>, independently, we measured  $T(\omega)$  and  $R(\omega)$  at 4.2 K with changing H. [Note that the H was applied along the c-axis.] Using the Intensity Transfer Matrix Method [12], we obtained the complex optical conductivity spectra  $\sigma(\omega)$  (= $\sigma_1(\omega)$ +i $\sigma_2(\omega)$ ).

In Fig. 2(a), we show the *H*-dependent  $\sigma_1(\omega)$ . At 0 T, there is a deep structure around 44 cm<sup>-1</sup> related with  $2\Delta$  and a sharp increasing behavior above  $2\Delta$ . With increasing H, the deep structure becomes smooth and  $\sigma_1(\omega)$  at 17 T slowly decreases with increase in frequency. In Fig. 2(b), we show the *H*-dependent  $\sigma_2(\omega)$ . At 0 T,  $\sigma_2(\omega)$  show a  $1/\omega$ -dependence due to the delta-function of  $\sigma_1(0)$ .  $\sigma_2(\omega)$  at 4 T show a peak-like feature around 50 cm<sup>-1</sup> and  $\sigma_2(\omega)$  at 17 T slowly increase with increase in frequency. Note that  $\sigma(\omega)$  at 0 T and 17 T are similar to the behaviors at the superconducting and the normal metallic states, respectively. From Fig. 2(a) and 2(b), we obtained  $\sigma_1(0) \sim 20000 \ \Omega^{-1} \text{cm}^{-1}$  and  $1/\tau$  $\sim 800 \text{ cm}^{-1}$  in the normal state, and  $2\Delta \sim 44 \text{ cm}^{-1}$  and  $\lambda \sim 2000 \text{ Å}$  in the superconducting state.

Such strong H-dependent  $\sigma_1(\omega)$  and  $\sigma_2(\omega)$  should be related to the evolution of vortex in a type-II superconductor, such as MgB<sub>2</sub>. Above  $H_{c1}$ , the

magnetic flux starts to penetrate into the superconductor, forming a vortex. Inside the vortex, the superconducting regions become suppressed and turn into a normal metallic regions. With increasing H, the number of vortices increases. Above  $H_{\rm c2}$ , the superconducting properties are totally destroyed [13]. Note that the values of  $H_{\rm c1}$  and  $H_{\rm c2}$  for MgB<sub>2</sub> were reported to be around 450 Oe and 20 T [14], respectively.

To explain the H-dependent optical responses, we used the composite medium theory, called the Maxwell Garnett theory (MGT) [15]. In the long wavelength limit, physical properties of the composite can be described in terms of an effective complex dielectric constant  $\varepsilon^{eff}$ . Since the vortices are well isolated from each other due to the inter-vortex Coulomb repulsion, we can approximately consider our film as a composite system composed of normal metal islands embedded in a superconductor. Then, such geometry can be approximated quite well by the two-dimensional MGT;  $\varepsilon^{eff}$  (= $4\pi i\sigma/\omega$ ) can be written as

$$\varepsilon^{eff} = \varepsilon^{s} \frac{(1 - f_{n})\varepsilon^{s} + (1 + f_{n})\varepsilon^{n}}{(1 + f_{n})\varepsilon^{s} + (1 - f_{n})\varepsilon^{n}},$$
 (1)

where  $\varepsilon^s$  and  $\varepsilon^n$  represent complex dielectric constant of superconducting and normal metallic states, respectively. And,  $f_n$  represents the area fraction of the metallic region. For  $\varepsilon^s$ , we used the Zimmerman formula [11] with  $2\Delta = 44$  cm<sup>-1</sup> and  $1/\tau = 800$  cm<sup>-1</sup>, and for  $\varepsilon^n$ , we used the simple Drude model with  $\sigma_1(0) = 20000 \ \Omega^{-1}$ cm<sup>-1</sup> and  $1/\tau = 800$  cm<sup>-1</sup>.

In Fig. 2(c) and 2(d), we show the calculated  $\sigma_1(\omega)$  and  $\sigma_2(\omega)$ , respectively. It is clear that the calculated  $\sigma_1(\omega)$  and  $\sigma_2(\omega)$  are quite similar to the measured spectra. These results suggest the validity of the MGT in describing the electrodynamic responses of a type-II superconductor under high field.

In Fig. 3, we plot the H-dependent  $f_n$ , estimated by comparing the experimental data with the calculated ones, with error bars. Clearly,  $f_n$  increases sharply at low field and slowly at high field. It is known that the values of  $f_n$  can be obtained from a heat capacity study by measuring the coefficient  $\gamma$ . For a s-wave,  $\gamma$  is known to be proportional to H [16]. On the other hand, for a superconductor with a node on the gap, for example a d-wave symmetry,  $\gamma$  should be proportional to  $H^{1/2}$  [17]. The dashed and solid lines

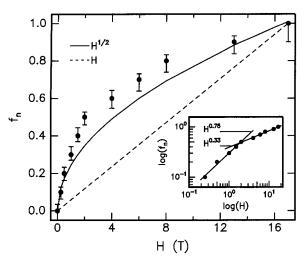


Fig. 3. *H*-dependent  $f_n$ . The dashed and solid lines represent the power law dependences of H and  $H^{1/2}$ , respectively. In inset,  $\log(f_n)$  vs.  $\log(H)$  is shown.

in Fig. 3 show the *H*-dependent  $f_n$  for *s*- and for *d*-waves, respectively. Neither of such *H*-dependences can explain our experimental data.

Inset of Fig. 3 shows the plot of  $\log(f_n)$  vs.  $\log(H)$ . The two H regions with different slopes are clearly seen. Interestingly,  $f_n$  below and above 2 T seems to follow the power law dependences of  $H^{0.75}$  and  $H^{0.33}$ , respectively.

#### IV. Discussion

Although many experimental studies have been performed on the  $2\Delta$  value of MgB<sub>2</sub>, there is little consensus on its magnitude and symmetry. In tunneling measurement, for example, the values of  $2\Delta$  varied from 3 to 16 meV, and both isotropic and anisotropic gap symmetries were suggested [18].

To explain the large variations of  $2\Delta$ , a model of two gap [5] and anisotropic s-wave gap [6] have been suggested. Szabo et al. [19] reported the existence of two superconducting energy gaps using point-contact spectroscopy measurement. Value of the small gap  $2\Delta_S$  and large gap  $2\Delta_L$  were reported to be 5.6 and 14 meV, respectively. Both of them were shown to follow the *T*-dependence of BCS formula. Also,  $2\Delta_S$  became strongly suppressed with *H* at low field. Seneor et al. [4] reported experimental result for anisotropic s-wave pairing symmetry using scanning

tunneling spectroscopic measurement. Values of  $2\Delta$  for ab-plane and c-axis were reported to be 10 and 16 meV, respectively.

Our optical results suggest that the gap nature of MgB<sub>2</sub> should be complex. The T- and H-dependent optical results on  $2\Delta$  seem to agree with the model of two gap; the value of  $2\Delta$  (~ 5.2 meV), BCS T-dependences, and strong H-dependence seem to be consistent with the characteristics of  $2\Delta_S$  within the two gap scenario. Also, the existence of two exponents in Fig. 3 might suggest that the superconducting properties correspond to  $2\Delta_S$ destroyed at low field and those correspond to  $2\Delta_L$ destroyed at high field. However, we cannot observe any features due to  $2\Delta_L$  in Fig. 2. The  $\sigma_1(\omega)$  and  $\sigma_2(\omega)$  are well described by the single gap not by the two gaps. Also, when we simulate the  $T(\omega)$  and  $R(\omega)$ in terms of two independent carriers with different gap values with  $2\Delta_S$ =5.6 meV and  $2\Delta_L$ =14 meV, we cannot observe any features due to  $2\Delta_L$ .

On the other hand, in the anisotropic s-wave scenario, the value of  $2\Delta$  for ab-plane is smaller than for c-axis. Since our measurements were made on the c-axis oriented film, the  $2\Delta$  represents the ab-plane value. Therefore, the small value of  $2\Delta$  might be consistent with their scenario. However, it is highly questionable whether the strong suppression of  $f_n$  under low H and the existence of two exponents could be explained within this scenario. More studies, especially the polarization dependent optical measurement on a single crystal would be useful to clarify the gap nature.

# V. Summary

We have investigated the gap nature of  $MgB_2$  using the temperature and magnetic field dependent optical conductivity spectra. Although the value of  $2\Delta/k_BT_C$  is half of the BCS value, superconducting gap  $2\Delta$  follows the temperature dependences of BCS formula. On the other hand, the area fraction of normal metallic region under external magnetic field, does not follow the *s*-wave and *d*-wave symmetry, but increases rapidly at low field and slowly at high field. These results suggest the complex gap nature of  $MgB_2$ .

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