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Optical measurements of the superconducting gap in MgB₂

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Abstract. Far-infrared reflectivity studies on the polycrystalline intermetallic compound MgB₂ with a superconducting transition temperature $T_c = 39$ K were performed at temperatures 20 K to 300 K. We observe a significant raise of the superconducting-to-normal state reflectivity ratio below 70 cm^{-1} , with a maximum at about $25\text{--}30\text{ cm}^{-1}$, which gives a lower estimate of the superconducting gap of $2\Delta(0) \approx 3\text{--}4\text{ meV}$.

PACS. 74.25.Gz Optical properties – 74.70.Ad Metals; alloys and binary compounds (including A15, Laves phases, etc.)

Recently superconductivity in the binary intermetallic compound MgB₂ with a T_c close to 40 K was reported by Akimitsu *et al.* [1]. Most studies performed on this compound yet indicate that MgB₂ consistently behaves as a phonon mediated superconductor within the framework of the BCS theory, probably in a strong coupling limit [2–5]. Up to now no optical investigations of this material were reported, and this is in part due to the poor quality of the presently available samples, which are sintered polycrystals. It is well known, however, that optical spectroscopy is an extremely powerful method for studying the materials in the superconducting (SC) state and is able of providing information on such important parameters as the SC energy gap, penetration depth, coherence effects, scattering mechanism, etc. [6,7].

In this short note we report on far-infrared reflectivity measurements performed on a sintered pellet of MgB₂. We are fully aware that these experiments bear inherent problems, mainly connected with the surface roughness, which we cannot overcome at this point. Nevertheless, we have attempted to extract important information on the intrinsic properties of MgB₂ from our reflectivity data.

MgB₂ has a hexagonal structure with P6/mmm symmetry. It crystallizes in the so-called A1B₂ structure where the boron atoms are located at a primitive honeycomb lattice, consisting of graphite-type sheets. The structure of MgB₂ is shown in Figure 1; the dimensions of the unit cell are $a = 3.086\text{ \AA}$ and $c = 3.524\text{ \AA}$, according to our X-ray analysis. The borons span hexagonal prisms; the large, almost spherical pores are filled by Mg which acts

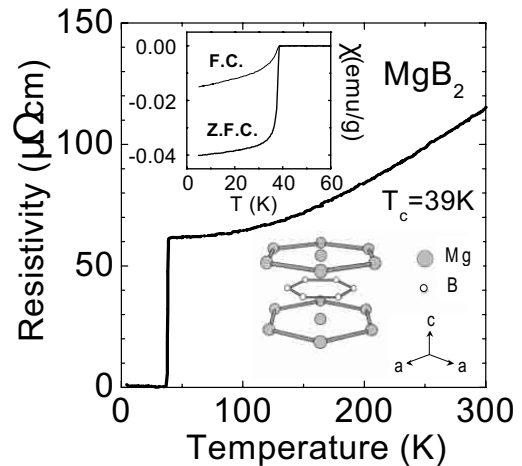


Fig. 1. Temperature dependence of the dc resistivity and of the magnetic susceptibility (field-cooled, zero-field-cooled) of MgB₂ and the unit cell of MgB₂.

as a spacer. Similar to graphite, the distance between the boron planes is larger by a factor of two compared to the intraplanar B-B bonds, and hence the B-B bonding is strongly anisotropic (two-dimensional).

Powder of high purity MgB₂ was pressed in a pellet and treated in Ar atmosphere at $\approx 900\text{ }^\circ\text{C}$; details on the sample preparation are reported in reference [1]. The quality of the sintered samples was checked by X-ray analysis, resistivity, and susceptibility measurements. In Figure 1 the temperature dependence of the dc resistivity and

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of the magnetic susceptibility are plotted. The SC phase transition is characterized by a width of less than 1 K from resistivity and of around 5 K from the susceptibility measurements. At $T = 300$ K we find $\rho = 115 \mu\Omega \text{ cm}$, and just above the SC transition temperature $T_c = 39$ K the resistivity has decreased by a factor of 2. The resistivity in the normal state can best be fitted by a power-law temperature dependence T^α with $\alpha = 2.5$ up to 250 K; behaviors with $\alpha = 2$ and 3 were reported earlier [3, 4]. The susceptibility (inset of Fig. 1) is decreasing rapidly in the temperature range 39–34 K by 0.034 emu/g, whereas for lower temperatures it decreases only slowly to the value -0.04 emu/g. Magnetization measurements allow us to estimate a SC fraction of more than 50%.

For the optical investigations the sintered pellet of MgB_2 was cut to a piece of $5 \times 5 \times 2 \text{ mm}^3$ size and polished. We note that it had a smooth but not shiny surface, with a remaining roughness due to pores. The scanning electron microscopy analysis revealed some traces of oxygen on the sample surface which may correspond to a layer of the MgO on it.

Here we report the far-infrared reflectivity spectra measured in the grazing (80 degrees) incidence geometry using a FT-IR spectrometer. The grazing incidence geometry has been chosen because in this case the reflectivity is more sensitive to changes of optical conductivity due to opening of the superconducting gap. This experimental technique has been previously used to measure the s -wave gap of NbN and the d -wave gap of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ [10]. In addition, normal incidence spectra were collected in the same frequency range, which were qualitatively consistent with the grazing reflectance data reported in this paper.

In our measurements, we have observed that the copper block on which the sample is mounted in the cryostat and the sample surface had quite different temperatures. In order to get a precise surface temperature, an additional Pt thermoresistor was later attached to the sample face. It turned out that the temperature difference is strongly enhanced below 40 K, suggesting that the large thermal gradient is due to strong lowering of the sample thermal conductivity in the SC state. In particular, the lowest surface temperature we could achieve was only about 20 K, while the cryostat cold finger was at liquid helium temperature. This additional temperature calibration has been used in the analysis presented in this paper. However, we should note that the systematic error of this measurement on this rather thick sample is about 2–3 K.

Figure 2 presents the intensity reflected from the sample at various temperatures normalized to the intensity reflected at 45 K, *i.e.*, slightly above T_c . We clearly see a rise of the reflectivity ratio (RR) above 1, starting below T_c . This is seen also in Figure 3, where the temperature dependences of the RR is plotted for fixed frequencies. The frequency below which the RR starts to increase above 1 shifts to higher values with decreasing T ; for the lowest temperature $T = 20$ K the increase starts at around $\nu = 70 \text{ cm}^{-1}$. As expected, the RR spectra in the SC state reveal maxima, meaning that $RR = 1$ at $\nu = 0$.

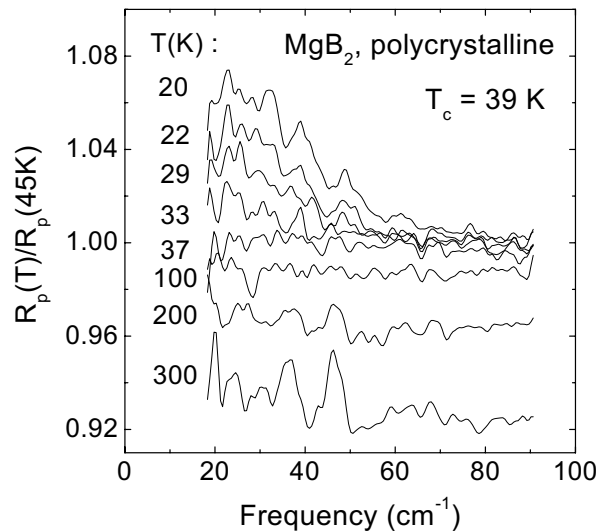


Fig. 2. Frequency dependence of the ratio of the reflectivity of MgB_2 measured in grazing incidence geometry (p -polarization) at various temperatures T (above and below T_c) to the grazing incidence reflectivity at $T = 4$ K.

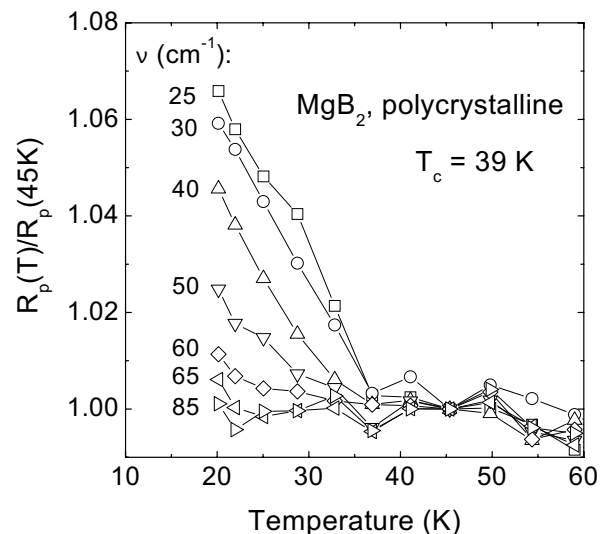


Fig. 3. Temperature dependence of the grazing incidence (p -polarization) reflectivity ratio of MgB_2 .

The observed variation of the reflectivity is reminiscent of the behavior found in conventional superconductors and, to a certain extent, in the high-temperature cuprates. There, the condensation of carriers into pairs and the opening of the energy gap 2Δ in the density of states reveal themselves as a decrease of the conductivity starting around the corresponding frequency $\nu = 2\Delta/hc$, and as the response in the dielectric constant of $\epsilon \propto -1/\nu^2$, both leading to pronounced changes of reflectivity and a maximum in the RR at around $\nu = 2\Delta/hc$ [6, 7]. Accordingly,

we associate the changes we observe for MgB_2 by entering the SC state with the response of condensed superconducting pairs, which is only in part obscured by the surface scattering. (We note that the rise in reflection for an essentially two dimensional system is not always an indication of the SC gap; for instance, the gap-like feature in the reflectivity spectrum of polycrystalline $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ [8] was later explained by the manifestation of the c -axis Josephson plasmon [9].)

A straight-forward assignment based on an isotropic s -wave gap would associate the gap energy 2Δ with the frequency where the RR reaches its maximum (around $25\text{--}30\text{ cm}^{-1}$, according to Fig. 2), providing an unrealistically small estimate of $2\Delta = 3\text{--}4\text{ meV}$. On the other hand it has been anticipated that superconducting gap is *not* uniform and has different values for different four sheets of the Fermi surface [11]. With this interpretation the RR maximum should correspond to the minimum value of the anisotropic gap. This is also consistent with a large variation of 2Δ values obtained from tunneling, Raman, photoemission, and NMR techniques [5,12–17] ranging from 4 to 17 meV

In conclusion, we have investigated the optical properties of sintered MgB_2 in the normal and superconducting state by measuring the ratio of the reflected intensity in the superconducting state to that in the normal state. An increase of the ratio in the superconducting state is reliably detected and interpreted as the signature of the superconducting gap with the lowest value of $2\Delta(0) \approx 3\text{--}4\text{ meV}$.

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