Temperature dependence of the microwave surface impedance measured on different kinds of MgB$_2$ samples.

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In this paper we present the results of measurements of the microwave surface impedance of a powder sample and two films of MgB$_2$. One film has $T_c = 30$ K and is not textured, the other is partially c-axis orientated with $T_c = 38$ K. These samples show different types of temperature dependence of the field penetration depth: linear for the powder sample, exponential with $\Delta/kT_c < 1.76$ (film with $T_c = 30$ K) and strong coupling behavior with $\Delta/kT_c \sim 2.25$ (film with $T_c = 38$ K). The results are well described in terms of an anisotropic gap model or by the presence of an Mg deficient phase.

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Since the discovery of superconductivity in MgB$_2$ [1] a number of papers have been dedicated to the measurement of the energy gap. Determining whether MgB$_2$ resembles conventional superconductors is of primary importance. A variety of techniques have been employed such as tunnelling, Raman, specific heat, etc. (see Table I). The first tunnelling measurements were performed on bulk polycrystalline material with values of the gap varying from 2 meV [2] to 8 meV [3]. In the latest tunneling [2] and point contacts [9] experiments coexistence of two-gap structure ($\Delta_L(T)$ and $\Delta_S(T)$) was observed up to the transition temperature. A two-gap structure was also observed in photoemission [10]. Specific heat measurements of bulk polycrystalline material [11] show that it is necessary to involve either two gaps or a single anisotropic gap $\Delta(\vec{k})$ [12] ($\Delta_S = min(\Delta(\vec{k}))$, $\Delta_L = max(\Delta(\vec{k}))$, where $\vec{k}$ belongs to the Fermi surface) to describe the data. The sign of the anisotropic gap was obtained from tunnelling [13] into a c-axis oriented film [14]. The size of the smaller gap $\Delta_{ab} \approx 5$ meV was supported by far infrared conductivity (FIR) [15] where a single gap $\Delta = 5.2$ meV was observed on the same type of film. There are no reported observations of the two gap structure in tunnelling or point contact measurements made in films, as far as we know. At this point the key question is whether the material has an isotropic, an anisotropic gap or multiple gap structure.

The value of the superconducting gap can be determined by measuring the temperature dependence of the field penetration depth ($\lambda(T)$). $\lambda(T)$ is connected to the concentration of the superconducting carriers and thus to the structure of the superconducting gap. Most of the reported data can be fitted with an analytical formula $\lambda(T) = \lambda_0/(1 - (T/T_c)^n)^{1/2}$ with different values of $n$ [11,12,13]. For the reported values of $n$ and details of the experiments see Table II, all experiments were performed on polycrystalline samples. These temperature behaviors do not contradict the two-gap model with s-wave order parameter, and values of $n = 1 \div 2$ can be interpreted as the presence of the small gap ($\Delta_S/kT_c < 1.76$) [13]. The value of $\Delta_S = 2.8$ meV was extracted from AC susceptibility of especially prepared powder samples [13].

Measurements of the temperature dependence of the real part of the microwave surface impedance $Z_s(T) = R_s(T) + i\omega\mu_0\lambda(T)$ [16,17] demonstrated that $\Delta R_s(T) \propto T$ at low temperatures. This behavior is associated with the presence of the small gap also.

In this paper we report measurements of $Z_s(T)$ performed on three types of MgB$_2$ sample: a powder sample, a randomly oriented polycrystalline film with $T_c = 30$K (Film I) and c-axis orientated film with $T_c = 38$K (Film II). We would like to stress that as well as determining $\lambda(T)$, the microwave measurement checks the quality of the sample under investigation by using as a parameter the value of residual surface resistance $R_{res} = R_s(T = 0)$. Linear temperature behavior of the deviation of the field penetration depth $\Delta\lambda(T)$ on the powder sample was observed. $\Delta\lambda(T)$ measured on the films
demonstrates activated behavior with $\Delta/kT_c < 1.76$ for the Film I and $\Delta/kT_c \approx 2.25$ for the Film II.

In our measurements we used a multilayered powder (Alfa Aesar Co., 98% purity) sample fixed in wax (as described in ref. [24] sample 3). Film I (4mm×6mm×300nm) was prepared by pulsed laser deposition (PLD) on c-plane oriented Al$_2$O$_3$ substrate with post annealing in ArH$_2$ atmosphere and has $T_c = 30$ K (see [24], film CAM 6). Film II partially c-axis orientated (4.5mm×10mm×750nm) was prepared by e-beam evaporation on R-plane Al$_2$O$_3$ and then annealed in Mg vapor for 1 hour (see [23] and reference therein, film A type). DC resistivity measurements result $\rho(300$ K) = 250$\mu\Omega$-cm and $\rho(T_c) = 250$ $\mu\Omega$-cm for the Film I. The width of the superconducting transition of this film is $\delta T_c = 5$ K. For the Film II these values are $\rho(300$ K) = 16$\mu\Omega$-cm, $\rho(T_c) = 6$ $\mu\Omega$-cm and $\delta T_c = 0.3$ K [24]. For the powder sample $\rho(T_c) = 25$ $\mu\Omega$-cm was calculated from the surface resistance measurement. A crude evaluation can be made of the mean free path for Film II using data from the Hall measurements ($R_H(T_c)$ = 5.5$\cdot 10^{-11}$m$^3$/C) performed on the same type of film [25] and from the calculated mean value of the Fermi velocity $v_F = 4.8$ $\cdot 10^5$m/s [26], such that $l(T_c,Film II) \approx 3$ (nm) $\sim \xi$. Other samples are in dirty limit because of the higher values of the resistivity.

Experiments were performed in two similar dielectric puck resonator and copper housing systems operated at different frequencies. Measurements of $R_s(T)$ of the two films are made using a TiO$_2$ dielectric puck resonator at 3.5 GHz. Measurements of the $\Delta \lambda(T)$ of all samples are made using an alumina dielectric puck resonator at 8.8 GHz to minimize errors in temperature dependence of the frequency shift. The fundamental TE$_{011}$ mode was used for all measurements, details of experimental set up are given elsewhere [22]. Film and powder samples were attached directly to the puck or to the quartz spacer with Apiezon N grease. Absolute values of $Z_e$ were extracted using the relation $R_s(T) + i\omega\mu_0\lambda(T) = G_s(1/Q(T) - 1/Q_0(T)) + i[f_0(T) - f(T)]/2f$, where 1/Q(T), 1/Q(T), $f_0(T)$ and $f(T)$ are quality factors and frequency shifts of unloaded resonator and resonator with the sample. Calculations of the geometry factor ($G_s$) were performed with MAFIA (commercial available software) [28].

Fig. 3 presents $R_s(T)$ data at low temperatures of all three samples (powder (solid circles), Film I (open circles) and Film II (solid squares)) rescaled to the 3.5 GHz assuming an $\omega^2$ law (measurements of the $R_s(T)$ of these films at different frequencies using dielectric puck and parallel-plane resonators will be published elsewhere [22]). Two distinct behaviors of $R_s(T)$ are seen on the graph. Powder and Film I demonstrate quite high residual losses $R_{res} = R_s(T = 10$K$) = 550\mu\Omega$ and 570$\mu\Omega$, and subsequently show much steeper slopes in comparison with Film II ($R_{res} = 110\mu\Omega$). Such a low residual surface resistance of the Film II is comparable to the values obtained in the MgB$_2$ wire $R_s(20$K$) = 57\mu\Omega$ [30] and an ion milled film $R_s(20$K$) = 92\mu\Omega$ [31]. Values are converted to 3.5 GHz assuming an $\omega^2$ law. As was mentioned in our previous paper, the steep slope of $R_s(T)$ can be attributed to the presence of low energy excitations caused by the small superconducting gap.

Fig. 4 shows the temperature dependence of the deviation of the field penetration depth ($\Delta\lambda(T)$) of the powder sample (open circles) and Film I (solid circles). Linear temperature dependence of $\Delta\lambda(T)$ of powder sample from $T = 10$ K is observed. Temperature dependence of the $\Delta\lambda$ of the Film I (solid line) can be fitted well by the formula

$$\Delta\lambda(T) = \frac{\lambda_0}{\left[1 - \left(\frac{T}{T_c}\right)^{n_0}\right]^{n_0/2}} \coth \left[\frac{d}{\lambda_c}\right]$$

where $d = 300$ nm is the thickness of the film and $n = 3 - T/T_c$ describes the weak coupling regime in the clean limit. The extracted value $\lambda_0 = 300 \pm 20$ nm is associated with changes in screening originating only from Copper pairs belonging to the part of Fermi surface with the "smaller gap" and cannot be associated with London penetration depth $\lambda_L(0)$. The value of the gap obtained from the weak coupling fit is an overestimation and it is definitely less than the BCS value $1.76kT_c$ because Film I is in dirty limit. To extract the exact value of the smaller gap observed on Film I additional calculations are necessary. The form of $\Delta\lambda(T)$ in the strong coupling limit ($n = 4$) with the same values of $d$ and $\lambda_0$ demonstrates disagreement with the data in the whole range of temperatures (dashed line).

Fig. 5 shows $\Delta\lambda(T)$ measured on Film II (dots). The solid line shown on the graph is the fit to the equation (1) with $n = 4$ and $d = 750$ nm. From the fit the extracted value of $\lambda_0(0) = 110 \pm 10$ nm. This value is less than most reported ($140$ nm [33], $180$ nm [4], $132.5$ nm [13], $110$ nm [34], $85$ nm [10], $160$ nm [11]). The extracted value of $\Delta(0)$ is $7.4 \pm 0.25$ meV, where the dash-dotted line is the fit $\Delta\lambda(T) = \lambda_0(2.25\pi T_c/2T)^{0.8} exp(-2.25T/T_c)$, which is in good agreement with our data up to 22.5 K$\approx T_c/2$. Fitting curve of the weak coupling regime (dashed line) shows clear disagreement with our data. We do not think that the smaller gap could be suppressed or masked in Film II. In addition the low $R_{res}$ means that we can exclude the coexistence of two separate gaps in this film, with some confidence.

Thus in three different kind of samples we observed two different kind of $\Delta\lambda(T)$. Bulk material with $T_c = 39$ K shows linear temperature dependence associated with the presence of the small gap. This data is in accordance with measurements performed on polycrystalline material (see Table I). $\Delta\lambda(T)$ measured on Film I with $T_c = 30$ K shows similar behavior demonstrating weak coupling regime at low temperatures and steep slope in $R_s(T)$. These observations support the hypothesis of the presence of the small gap in this film.
In contrast, Film II shows no sign of the smaller gap in the ab-plane. Extracted value $\Delta(0) = 7.4 \pm 0.25$ meV is in good agreement with reported values of the larger gap (see Table I) and maximum gap measured in a c-axis orientated film with $T_c = 38$ K [1].

There are two possible explanations. The first is the presence of an anisotropic gap $\Delta(k) = \Delta_{ab} \cos(\theta) + \Delta_c \sin(\theta)$ that comes from the 2D Fermi surface, where $\Delta_{ab} \sim 7.5$ meV and $\Delta_c \sim 3$ meV. The presence of anisotropic gap in MgB$_2$ is discussed widely [2]. In the c-axis orientated film only the large gap $\Delta_{ab}$ determines the microwave properties whereas on unaligned samples the smaller gap $\Delta_c$ defines the transport behavior. Differences in the form of $\lambda(T)$ for the powder sample and Film I are probably related to the differences properties of the surface (where the gap becomes isotropic over the directions in the momentum space [3]), and the difference of mean free path. The second peak at $2.8 \pm 3.9$ meV in the (DOS) measured in the tunnel and point contact experiments comes from a surface reduced gap. Different kinds of surface region were observed with an STM recently. One type of region was found which showed the large gap $\Delta = 6$ meV and another region with the small gap $\Delta = 3$ meV only. This could be associated with different crystallographic orientations in each type of region. The scenario of an anisotropic gap with $\Delta_{ab} > \Delta_c$ contradicts the tunnelling measurements of Chen et al., [1] and the high Coulomb repulsion in ab-plane suggested in [4].

A second explanation is that some samples contain an Mg deficient phase (due to MgO formation [5]) that has the smaller gap seen in the bulk samples and in the films with $T_c < 38$ K. In order to explain the heat capacity data the volume of second phase would need to be comparable to the primary phase. This scenario implies that Film II is single phase only.

In conclusion, the results of the temperature dependence of the microwave surface impedance measured on three different samples are reported. Powder sample and Film I with $T_c = 30$ K show strong temperature dependence in both components of $Z\omega$ which associated with presence of the small gap $\Delta < 1.76kT_c$. Film II (c-axis aligned, $T_c = 38$ K) shows strong coupling behavior in the $\lambda(T)$. Extracted values of the London penetration depth and superconducting gap are $\lambda_L(0) = 110 \pm 10$ nm and $\Delta(0) = 7.4 \pm 0.25$ meV. Two possible explanations have been introduced in terms of either the existence of an anisotropic superconducting gap or the presence of second phase with lower gap in some MgB$_2$ samples. In the light of our current results the former scenario looks much more plausible. These experimental results show that microwave measurements on single crystals of MgB$_2$ for the current flowing in ab and c directions are highly desirable.

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FIG. 1. Temperature dependence of the microwave surface resistance. Powder (solid circles), Film I (open circles) and Film II (solid squares).

FIG. 2. Temperature dependence of the field penetration depth of the powder sample (open circles) and Film I (solid circles). Lines are the fit using equation (1) \( n = 3 - T/T_c \) (solid one) and \( n = 4 \) dashed one.

FIG. 3. Temperature dependence of the field penetration depth of the Film II (dots). Lines are the fit using equation (1) \( n = 4 \) (solid one) and \( n = 3 - T/T_c \) (dashed one). Dash-dotted line is the fit of exponential behaviour with \( \Delta/kT_c = 2.25 \).

TABLE I. Measurements of the double or anisotropic superconducting gap in MgB\textsubscript{2} with different techniques.

| Ref. | Methods          | \( \Delta_L \) (meV) | \( \Delta_S \) (meV) |
|------|------------------|----------------------|----------------------|
| 11   | tunnelling       | 8                    | 5                    |
| 12   | tunnelling       | 7.5                  | 3.9                  |
| 13   | tunnelling       | 7.8                  | 3.8                  |
| 14   | point contacts   | 7                    | 2.8                  |
| 15   | specific heat    | 6.4                  | 2.1                  |
| 16   | specific heat    | 7.2                  | 2.0                  |
| 17   | photoemission    | 5.6                  | 1.7                  |

TABLE II. Measurements of the temperature dependence of the field penetration depth on MgB\textsubscript{2} with different techniques.

| Ref. | Methods          | n |
|------|------------------|---|
| 14   | AC, M(T)         | 2.8|
| 15   | M(T) (VSM,SQUID) | 1 |
| 16   | AC, \( \mu \)SR  | 2 |
| 17   | Optic            | 2 |