The superconducting gap of *in situ* MgB_2 thin films by microwave surface impedance measurements

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Precision measurements of the microwave surface resistance R_s of *in situ* MgB_2 films directly reveal an exponential behavior of R_s at low temperature indicating a fully-gapped order parameter. The entire temperature dependence of R_s is well described by a Mattis–Bardeen formalism, but with a small gap ratio of $\Delta(0)/kT_c = 0.72$, corresponding to $\Delta(0) = 1.9$ meV. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1517181]

Elucidation of the nature of the superconducting state in the binary compound MgB_2[1] is of importance both for fundamental understanding and for applications of this material in microwave communications. Two important issues that are relevant are the nature of the superconducting pairing mechanism and the symmetry of the order parameter. There is mounting experimental evidence from different spectroscopies, tunneling, specific heat, and photoemission measurements that the superconducting order parameter does not follow a BCS s-wave symmetry, and presents a multicomponent gap.[2–5] In the electromagnetic response, the characteristic signature of a gapped superconductor is an exponential behavior of the microwave absorption represented by the surface resistance R_s at low temperatures.

We have measured the microwave properties of high quality MgB_2 films prepared by pulsed laser deposition (PLD) with an *in situ* annealing using an adaptation of a cavity perturbation technique for thin films in the perpendicular geometry. Our results yield a direct measurement of the superconducting gap, and we find a value of $\Delta(0)/kT_c = 0.72$. This leads to a gap value of $\Delta(0) = 1.92$ meV, consistent with the smaller of the two gaps typically observed in tunneling measurements. The entire temperature dependence of the R_s data is well described by a BCS s-wave Mattis–Bardeen calculation with the same small gap ratio.

The films were grown by PLD of MgB_2/Mg mixture at 300 °C followed by an *in situ* annealing at 600 °C.[6] The structural morphology of the films is nanocrystalline mixture of textured MgO and MgB_2 with grain sizes around 50 Å (Fig. 1). Zero-resistance transition temperature of 34 K was obtained in the best samples using this technique, and the film used in this paper has a sharp superconducting transition at 31 K with a width of 1 K.

The measurements were carried out in a Nb superconducting cavity resonant at 10 GHz using the ''hot finger'' technique, which has been well established to yield results on bulk and single crystals.[7] Using an analysis method to be described subsequently, the microwave properties were measured in the perpendicular geometry, that is, $H_\omega \perp$ film plane. The film is placed in the center of the cavity and the measurements are carried out in the TE_{011} mode. The sample temperature can be varied over a wide range from 300 to 4.2 K (or 2 K), while the Nb cavity is maintained at 4.2 K (or 2 K) ensuring a very high Q and correspondingly low background, and the surface impedance $Z_s = R_s + iX_s$ is obtained as described later.

We have adapted the cavity perturbation method that has previously been exhaustively analyzed and used for bulk samples.[7] To take into account the enhancement of the external fields at the sharp edges of the sample, we use a general theory by E. H. Brandt[8] calculating the response to an applied perpendicular ac magnetic field H_ω of the thin film, which is characterized by frequency dependent complex resistivity $\tilde{\rho}_\omega$. In this formalism one is able to calculate the sheet current $J_\omega(y,z)$ by solving a one-dimensional integral equation. The solution can be approximated with high precision by a finite sum and allows to calculate the magnetic moment and the complex magnetic permeability $\tilde{\mu} = \mu' - i\mu''$ of the sample. $\tilde{\mu} = \Sigma_n c_n/(\Lambda_n + \tilde{w})$. The coefficients c_n and the poles Λ_n were calculated by Brandt for both strip and disk geometry.[8] The parameter $\tilde{w}$ is given by the expression $\tilde{w} = i\omega a d \mu_0 \tilde{\sigma}_\omega(\omega)/2\pi$, where $2d$ is the thickness of

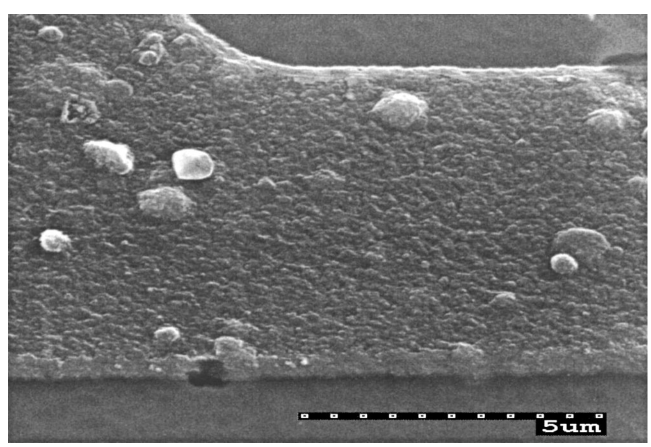

FIG. 1. SEM of *in situ* MgB_2 film.

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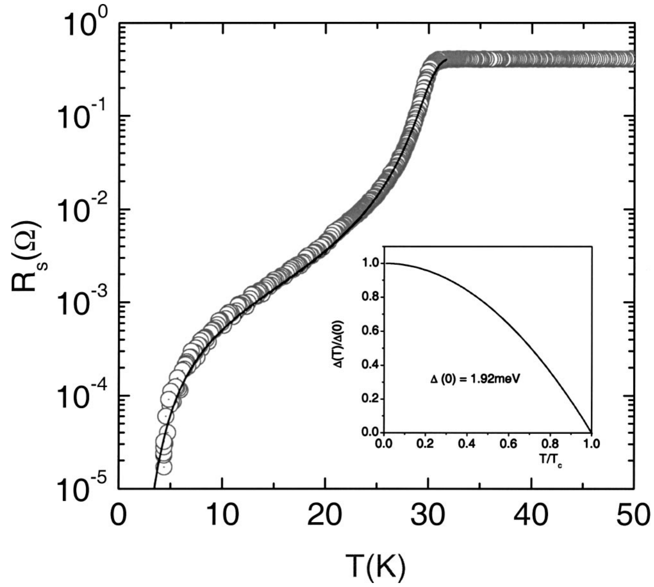

FIG. 2. R_s vs T for MgB_2. The solid line represents the BCS calculation with a gap parameter $\Delta(0)/kT_c=0.70$.

the sample and a represents the dimension of a square strip or the radius for a disk. The frequency dependent complex conductivity is $\tilde{\sigma}_\omega=\sigma_1(\omega)-i\sigma_2(\omega)=1/\tilde{\rho}$.

The methodology of the experiment is similar to the bulk samples microwave measurements. At every temperature T, the cavity resonant frequency $f_0(T)$ and the width $\Delta f(T)$ were measured. The complex cavity frequency $\tilde{f}\equiv f_0+i\Delta f/2$ is related to the sample electromagnetic permeability $\tilde{\mu}_\omega$ by the following equation: $\tilde{f}_s-\tilde{f}_e=g(1-\tilde{\mu}_\omega)$, where $\tilde{f}_e$ and $\tilde{f}_s$ are complex frequencies of the empty cavity and the loaded one, respectively. From $\tilde{\mu}_\omega$, the sample's complex conductivity $\tilde{\sigma}_{s\omega}$ can be determined by inverting the complex frequency shift equation using a MATLAB routine. The geometric factor g was experimentally determined by assuming $g=\alpha[f_{0s}(0\text{ K})-f_{0s}(T\gg T_c)]$, where $f_{0s}(0\text{ K})$ was determined by extrapolating f_{0s} from the lowest measured temperature (usually 2 or 4.2 K) to $T=0$ K. The coefficient α is determined such that the calculated value of $\lambda(0)$ matches the experimental value. Only the very low T value

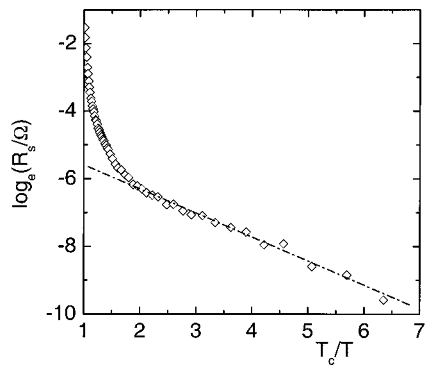

FIG. 3. $\ln R_s$ vs $1/T$. The line represents exponential behavior with a slope characterized by $\Delta(0)/kT_c=0.72$.

of λ, viz., $\lambda(0)$, is sensitive to this choice, overall, the results obtained are extremely robust over a wide range of T. From the measured complex conductivity $\tilde{\sigma}_{s\omega}$ the surface impedance, $\tilde{Z}_s(\omega,T)=R_s(T)+iX_s(T)=(\mu_0 i\omega/\tilde{\sigma}_{s\omega})^{1/2}$, was obtained. Note that this is the bulk impedance of the thin film material, and not the actual thin film impedance which can be calculated using suitable thickness-dependent thin-film expressions.

The bulk superconducting microwave surface resistance $R_s(T)$ of the thin film material is shown in Fig. 2. The normal state surface resistance is rather high $\sim 0.4\Omega$, consistent with a normal state resistivity of $230\mu\Omega$ cm. Although the surface resistance starts at a relatively high value above T_c, it starts to drop rapidly as temperature decreases. At $T_c/2$, $R_s\sim 1$ mΩ, and below approximately 15 K the drop becomes more rapid, reaching values below 50 $\mu\Omega$ as the temperature reaches 4 K. This last rapid decrease is characteristic of an exponential behavior, as will be seen shortly.

The bulk surface impedance of a BCS s-wave superconductor can be derived using the relation between the impedance $Z_s=(-\mu_0 i\omega/\tilde{\sigma}_s)^{1/2}$ and the Mattis–Bardeen[9] complex conductivity $\tilde{\sigma}_s=\sigma_1-i\sigma_2$. The normalized conductivity can be calculated using

$$\sigma_1/\sigma_n=2/\hbar\omega\int_\Delta^\infty g(E)[f(E)-f(E+\hbar\omega)]dE,$$

and

$$\sigma_2/\sigma_n=1/\hbar\omega\int_{\Delta-\hbar\omega}^{\Delta}[1-2f(E+\hbar\omega)](E^2+\Delta^2+\hbar\omega E)/\{(\Delta^2-E^2)[(E+\hbar\omega)^2-\Delta^2]\}^{1/2}dE,$$

where $g(E)$ is the appropriate factor incorporating the density of states and BCS coherence factors, and $f(E)=[\exp(E/kT)+1]^{-1}$ is the Fermi function. The gap temperature dependence of the form $\Delta(T)=\Delta(0)[1-(T/T_c)^2]$ is shown in the inset of Fig. 2. We have used experimental parameters $\omega=2\pi 10^{10}(\text{s})^{-1}$ and $T_c=31$ K. Using a small gap ratio, $\Delta(0)/kT_c=0.70\pm 0.02$, the calculated $R_s(T)$ is in excellent agreement with the measured data over the entire temperature range (Fig. 2).

The low temperature data shows an exponential temperature dependence according to $R_s\propto\exp[-\Delta(0)/kT]$. To measure the gap we plotted $\ln R_s$ versus T_c/T, as shown in Fig. 3. A straight line behavior is clearly seen at low T, and is an unambiguous and direct signature of the superconducting gap. A fit for $T\leqslant 15$ K gives a value of the gap $\Delta(0)/kT_c=0.72$, resulting in a gap value of $\Delta(0)=1.92$ meV. (We stress that Fig. 3 represents the actual data, and that a residual resistance was not subtracted, attesting to the high film quality.) The data represented in Fig. 3 represent the clearest evidence of a fully gapped superconductor from microwave measurements.

Although the gap ratio in the present work is smaller than the mean field value of 1.76, it is in good agreement with a number of reports on this material.[2–4] It appears fairly certain that MgB_2 is not a single gap s-wave superconductor; a tentative consensus appears to favor a two-gap superconductor characterized by $\Delta=\Delta_{small}+\Delta_{large}$, or even a strongly anisotropic s-wave superconductor with $\Delta=\Delta_{min}(1+k\cos^2\theta)$.[5,10] Values for the smaller gap reported in the literature vary between $\Delta_{small}(0)=1.9$–2.8 meV, while for the larger gap Δ_{large} the values vary between 6.2–9 meV. The microwave measurements are sensitive to the smallest energy scale in the material and hence we are clearly measuring the smallest gap Δ_{small} or Δ_{min}. The good agreement with the calculations over the entire temperature range indicates that the smaller gap ''turns on'' at T_c along with a possibly larger gap that is not directly seen. This is consistent with tunneling data, which report that the small gap also appears at T_c as the temperature is lowered. The temperature dependence of the gap indicated by our measurements is close to the mean-field temperature dependence.

MgB_2 is of great interest as a superconductor with moderately high T_c that might be suitable for electronic applications at 20 K and below. The R_s values of the *in situ* film are quite low and competitive with the best values of Y-Ba-Cu-O compounds at 4.2 K and approach Nb values. We have measured other *ex situ* films which had much higher values of R_s, likely due to their polycrystallinity and poor texture. Thus far, the present *in situ* films have yielded low R_s, and this may be attributed to their nanocrystalline and highly smooth texture. The nanocrystalline structure with MgO contaminants may cause lower residual dc resistance ratios of typically 1.4 for such films than in the good bulk samples, and may also result in the lower superconducting transition temperature than the bulk since the grain size is comparable to the coherence length. At the same time, the nanocrystallinity appears to lead to some of the lowest microwave losses observed.[11–14]

The gap reported here is largest among the metallic and bimetallic superconductors such as Nb or Nb_3Sn that are candidate materials for microwave applications. If the normal state values are reduced, as is implied by very low resistivities, $<1\,\mu\Omega$ cm, observed in bulk materials, then combined with the corresponding exponential drop in R_s due to the fully gapped nature, dramatically low microwave loss (10^{-9}–$10^{-6}\,\Omega$) may be feasible at elevated temperatures (20 K and below) not achievable with other high T_c superconductors. The present results show great promise for applications of MgB_2 in microwave electronics at 20 K and lower temperatures.

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