PHYSICAL REVIEW B, VOLUME 65, 020506(R)

Magnetic penetration depth measurements in MgB_2 sintered pellets and thin films

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(Received 4 July 2001; published 19 December 2001)

We have performed accurate measurements of the temperature dependence of the magnetic penetration depth of the new intermetallic superconductor MgB_2 using a high resolution single coil mutual inductance technique. Both sintered pellets and thin films exhibiting critical temperature values ranging between 37 and 38.4 K have been studied. In the case of the film, for $T<T_c/2$ a clear exponential behavior is observed, which can be explained by a simple BCS *s*-wave model with a reduced value of the energy gap. On the contrary, pellets show no evidence of saturation, and the experimental results strictly follow a quadratic dependence down to the lowest temperatures. The experimental data on sintered samples can be explained in the framework of a conventional *s*-wave BCS theory using a two-band model.

DOI: 10.1103/PhysRevB.65.020506 PACS number(s): 74.25.Nf, 74.80.−g

Great interest has been raised on the simple binary compound MgB_2 after the observation that it becomes a superconductor at a relatively high temperature ($\sim$39 K).[1] An impressive number of theoretical and experimental studies has been performed to understand the origin of the pairing mechanism in this system. Experimental results have provided evidence for a phonon mediated superconductivity. The boron-isotope effect,[2] the observation of a tiny coherence peak just below the critical temperature T_c, and the spin lattice relaxation rate in the superconducting state in NMR,[3] Raman spectroscopy,[4] tunneling measurements,[5–7] and optical conductivity data[8] are all consistent with the existence of a conventional mechanism of superconductivity. The effect of impurities has been also investigated: chemical substitution of magnetic ions such as Mn^{2+}, Fe^{2+}, Co^{2+}, and Ni^{2+} suppresses the critical temperature T_c (Ref. 9) while nonmagnetic Zn substitution increases it.[10] The majority of spectroscopic measurements have shown so far a finite but widely variable value for the energy gap, ranging from 2 to 7 meV.

Although it seems that the body of the experimental data can be explained in a BCS conventional framework, no consensus has been reached on the pairing symmetry of this system yet. The magnetic penetration depth λ is known to be a very sensitive probe of the low-lying quasiparticles energy, and it is capable of giving information which is significant on a $\lambda(0)$ scale rather than on a $\xi(0)$ scale. This corresponds, in the case of a thin film, to probing the true "bulk" properties of the superconductor. Experiments performed on pellets using μSR (Ref. 11) and ac susceptibility[11,12] showed a quadratic behavior of $\Delta\lambda$ at low temperatures, that authors interpreted as the signature of the presence of nodes in the gap. However, a more careful analysis of μSR data showed[13] results consistent with a BCS *s*-wave framework, while the peculiar exponential behavior expected for a fully gapped superconductor was indeed observed in polycrystalline samples,[14] in wires,[15] and very recently in single crystals.[16]

In this paper we present experimental results on the temperature change of the magnetic penetration depth for MgB_2 polycrystalline samples in bulk and thin film form.

We studied two high density sintered pellets (No. 1A and No. 2A) and one film (No. B) deposited on a *r*-plane sapphire substrate. Bulk samples were obtained by reaction sintering of elemental B and Mg for 3 h at 950 °C in a sealed stainless steel container, lined by a Nb foil. Details about the preparation technique are described elsewhere.[17,18] The resulting high density ($\sim$2.4 g/cm^3) MgB_2 cylinders had metallic appearance and very high hardness. Two samples ($10\times10\times3$ mm^3) were cut from the bulk material with a diamond circular saw. The mechanically polished surface of these samples was observed by optical and atomic force microscopy (AFM), revealing the presence of large single crystal grains (up to 200 μm large), embedded in a more fine-grained metalliclike background, as it was found in previous studies.[19–22] The inductively measured critical temperatures are 38.4 K ($\Delta T_c\approx1.3$ K) and 37 K ($\Delta T_c\approx2.3$ K) for No. 1A and No. 2A, respectively. The film was obtained by appropriate postannealing of an electron-beam-evaporated B precursor (500 nm) grown directly on a Al_2O_3 single-crystal substrate. After deposition the boron film was subsequently

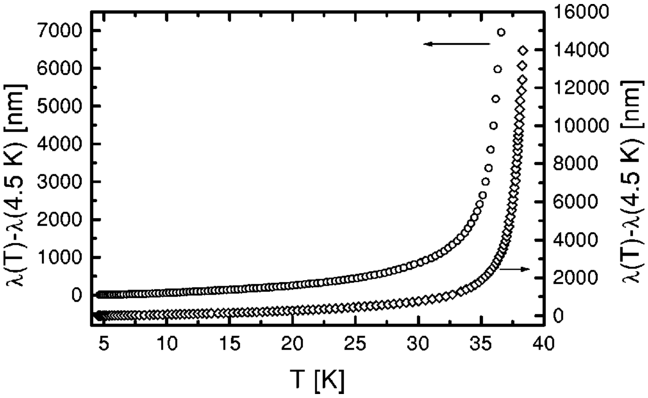

FIG. 1. Variation of the magnetic penetration depth up to T_c for samples No. 1A (○) and No. 2A (◇). Note the different scales.

sandwiched between cold pressed MgB_2 pellets, along with excess Mg turnings, packed inside a crimped Ta cylinder and finally annealed at 890 °C for 20–25 min.[23] A typical θ-2θ scan revealed the presence of a c-axis aligned film while the pole figure showed a random in-plane texture. AFM study showed the presence of a polycrystalline-type surface with grains up to 100 nm of diameter. The inductive critical temperature and critical current density (at 4 K) are 37.9 K ($\Delta T_c \approx 1$ K) and 6.4 MA/cm^2, respectively. In spite of not optimized structural properties, these high values of T_c and J_c point to a sample of very good quality.

$\lambda(T)$ is investigated employing a single coil-mutual inductance technique.[24] This is a radio frequency technique (4 MHz) that measures the change of inductance ΔL of a pancake coil located in the proximity of the sample and connected in parallel with a low-loss capacitor. Such LC circuit is connected at the input of a marginal oscillator. A change of impedance of the LC circuit is detected as a change of resonant frequency f of the oscillating signal:

$$f=\frac{1}{2\pi}\sqrt{\frac{1}{L(T)C(4.2K)}-\left[\frac{R(T)}{L(T)}\right]^2}, \qquad (1)$$

where L is the total inductance of the system formed by the coil and superconducting sample, C is the capacitor, and R a function of the coil resistance and of the surface losses of the sample. In the London regime, where the electromagnetic response of the sample is mostly diamagnetic and the losses are negligible, the explicit expression of the variation of the coil inductance as a function of the magnetic penetration depth is

$$\Delta L = L_0 - L = \pi\mu_0 \int_0^\infty \frac{M(\gamma)d\gamma}{1+2\gamma\lambda\coth(d/\lambda)}, \qquad (2)$$

where $M(\gamma)$ is a geometrical factor that depends on the sample-coil distance and d is the sample thickness. The details of this technique are described elsewhere.[24]

In Figs. 1 and 2 we show the change $\Delta\lambda(T)$ in the effective magnetic penetration depth for the three samples in the overall temperature range. A much larger variation is observed in the case of pellets with respect to the film. At low temperature (Fig. 3) this feature is even more evident: sample No. B displays a large flat region up to 6 K whereas samples No. 1A and No. 2A show no evidence of saturation. In the case of the film a simple BCS conventional s-wave calculation fits very well the low-temperature data using as fitting parameters $\Delta = 3.6$ meV and $\lambda(0) = 120$ nm. We estimate the error on these two quantities to be about 20%. $\lambda(0)$ is of the same order of magnitude of other measurements reported in literature on bulk samples[11,14] and it is nearly half of the value obtained in thin films[25] from far-infrared conductivity. The value of the gap is a bit larger than the one obtained by optical measurements performed on films grown by the same technique and it is in the range of values obtained by tunneling spectroscopy.[5–7] The strong coupling ratio $2\Delta_0/k_BT_c$ is evaluated to be 2.3. The penetration depth behavior remains the same removing by ion milling a (nominal) layer of 60 nm from the sample surface. This rules out the proximity effect eventually associated to the presence of a metallic Mg overlayer.[23] Therefore, if a normal or poorly superconducting layer exists on the film surface, its effect on the temperature dependence of the penetration

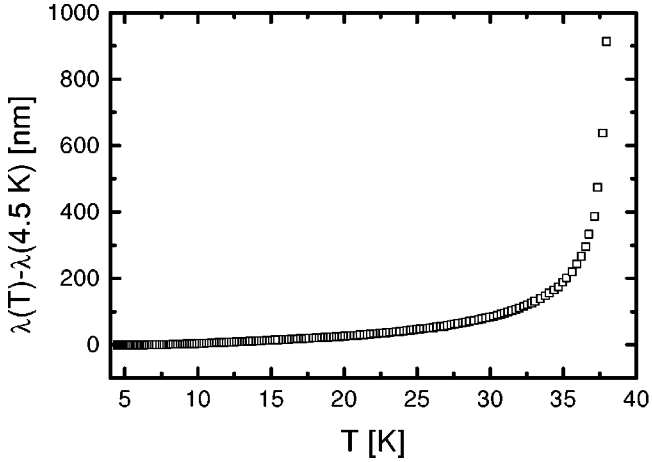

FIG. 2. Variation of the magnetic penetration depth up to T_c for sample No. B (□).

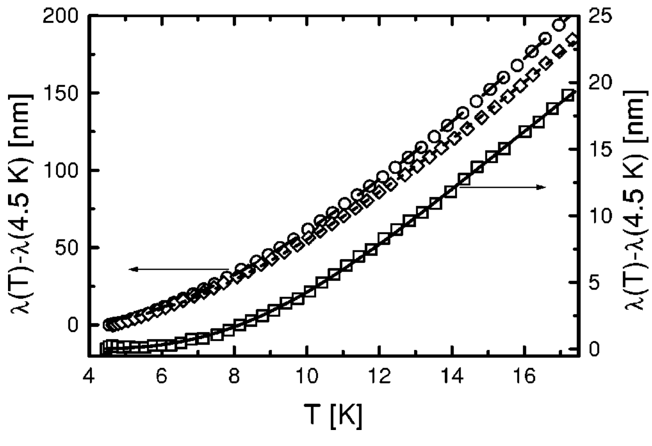

FIG. 3. Variation of the magnetic penetration depth at low temperature for samples No. 1A (○), No. 2A (◇), and No. B (□). The dashed and short-dashed lines represent the two-gap BCS fit for No. 1A and No. 2A, respectively. The solid line represents the single gap BCS fit for No. B. Note the different scales.

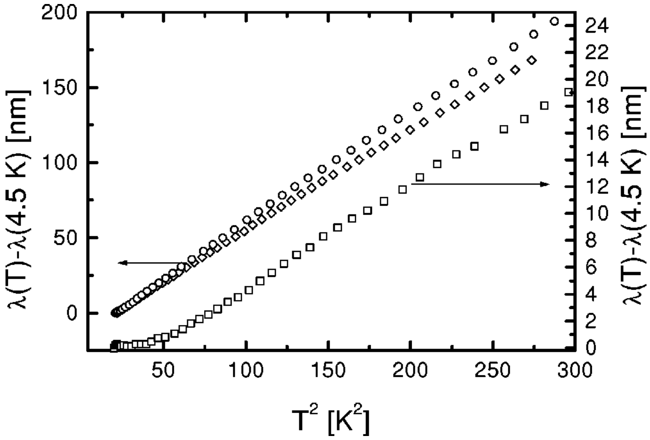

FIG. 4. Variation of the magnetic penetration depth as a function of T^2 at low temperature for samples No. 1A (○), No. 2A (◇), and No. B (□). Note the different scales.

depth is negligible in our measurements. Consequently the rather low value of the energy gap determined in the film seems, to our understanding, intrinsic to MgB_2. Since in the experiment the ab plane penetration depth of a c-axis oriented film is probed, this finding can be well explained by the anisotropy of the order parameter in this material reported by other studies, such as point-contact spectroscopy,[26] and penetration depth measurements in single crystals.[16] The result is also in agreement with the recent observation[15] that the removal of magnesium by chemical etching in a MgB_2 wire results in an exponential temperature dependence of the magnetic penetration depth with a similar value of the energy gap.

The analysis of the low-temperature data for the bulk samples is more complicated. In Fig. 4 we show $\Delta\lambda$ as a function of T^2 for all samples: in the case of No. 1A and No. 2A we found a quadratic behavior of $\Delta\lambda$ down to the lowest temperature. It has been claimed that such gapless behavior would be the signature of nonconventional superconducting pairing.[11] It is worthwhile to mention, however, that a T^2 behavior of the magnetic penetration depth is not sufficient to rule out a BCS conventional s-wave pairing symmetry. In the case of strongly inhomogeneous samples, it has been shown that a spread of superconducting gap values leads to a power-law dependence at low temperature for both the thermodynamic and transport properties.[27]

We note that the critical temperatures of samples No. 1A and No. 2A, made in the same batch, differ by almost 1 K. The values of $\lambda(0)$, determined by a two fluid model fit,[28] are larger than in sample No. B: 2.4 μm for No. 1A and 1.6 μm for No. 2A. Finally, the behavior of $\Delta\lambda(T)$ near T_c is very different for the two bulk samples. These features indicate the presence of strong inhomogeneities in the bulk samples. In this case a Gaussian distribution of gap values in the framework of an s-wave pairing model forcefully fits the experimental data.

The presence of regions of highly suppressed superconductivity, induced by unreacted Mg, can cause either a spread of gap values (from 1.7 to 7 meV as reported in literature) or a simple overlapping of two gaps.[29] The magnetic penetration depth is a bulk measurement and consequently provides an average of the electrodynamic response of the tested sample region. Thus it is sensitive only to the minimum and the maximum value of the gap. For this reason we consider a model of a two-band BCS s-wave gap. In the framework of this model, the magnetic penetration depth is a function of two s-wave gaps of different magnitude. One can write[30]

$$\frac{1}{\lambda_{tot}^2}=\frac{1}{\lambda_{min}^2}+\frac{1}{\lambda_{max}^2}, \tag{3}$$

where for $T<T_c/2$

$$\triangle\lambda_i(T)\approx\lambda_i(0)\sqrt{\frac{\pi\Delta_i(0)}{2k_BT}}e^{-\Delta_i(0)/k_BT} \tag{4}$$

($i=$min,max).

Using this approach we calculate the best fit curves to the experimental data as shown in Fig. 3 for the following fitting parameters: $\Delta_{min}^{1A}=2.45$ meV, $\Delta_{min}^{2A}=2.4$ meV, $\Delta_{max}^{1A}=6.8$ meV, and $\Delta_{max}^{2A}=6$ meV, respectively. We note that the fitting parameters Δ_{min} and Δ_{max} encompass the ones found by photoemission spectra,[31] tunneling spectroscopy[5–7,29,32,33] and specific heat measurements.[34] The strong coupling ratio is 1.5 and 4.1 for No. 1A, 1.5 and 3.7 for No. 2A, respectively.

To our understanding, the results observed on the thin film sample rule out the possibility of an unconventional order parameter in this material. An s-wave single gap model fits the film experimental data very well, even if with a reduced energy gap value. The striking contrast observed in the $\lambda(T)$ behavior between the thin film and the sintered samples may have different origins. It can be ascribed to the anisotropy of the superconducting properties between the ab and c directions, as was recently emphasized in single crystals.[35] Since the film is c-oriented, we probe mainly the ab-plane electrodynamic response, whereas this is not true for the bulk samples. However, the values found for $\lambda^{film}(0)$ and $\lambda^{pellet}(0)$ are not consistent with an anisotropy factor $\gamma=2.6$.[35] The interesting coexistence of a two-dimensional Fermi surface (p_{x-y} orbitals) perpendicular to the z direction and a three-dimensional one (p_z bonding and antibonding bands) reported in Ref. 36 may be also of help in explaining the electrodynamic response of the pellets, which is not inconsistent with the existence of two order parameters. However, it would be difficult to explain the experimental data on the thin film using the same picture. More likely, with the high values of $\lambda(0)$, the difference observed in the critical temperature values and in the temperature dependence $\Delta\lambda(T)$ close to T_c between the two pellets made from the same batch is a strong evidence for the presence of inhomogeneities: residual flakes of unreacted Mg induce a depression in the gap by the proximity effect in the region near the inclusions. In this case the presence of regions of different critical temperature depends strongly on the ratio of the mean free path to metal layer thickness and on the transmittivity properties of the N-S interface.[32] Thus the proximity effect can induce the presence of regions with a weakened gap and with a critical temperature close or even equal to the bulk critical temperature. A spread or a simple superposition of two gap values is equally possible.

In conclusion we have reported on the magnetic penetration depth measurements on MgB_2 sintered and film samples by a single coil technique. A simple model of two different size *s*-wave gaps in the case of pellets and a single finite but reduced *s*-wave gap in the case of film can account for the low-temperature experimental data. The results are to our understanding a further evidence of the conventional nature of the MgB_2 superconductor.

We thank G. Ausanio for the AFM analysis and F. Lombardi for the ion beam etching of the film surface. Many thanks are due to A. Barone and R. Vaglio for useful discussions. We are grateful to N. Bontemps and J. Bok for fruitful discussions and for letting us use the scientific equipment for the single coil inductive measurements. Edison acknowledges the Lecco Laboratory of the CNR-Tempe for making available its facilities for the pellets preparation.

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