Penetration depth measurements in MgB_2: Evidence for unconventional superconductivity

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We have measured the magnetic penetration depth of the recently discovered binary superconductor MgB_2 using muon spin rotation and low-field ac susceptibility. From the damping of the muon precession signal we find the penetration depth at zero temperature is ~ 85 nm. The low-temperature penetration depth shows a quadratic temperature dependence, indicating either the presence of nodes in the gap or that the gap minimum must be smaller than the lowest temperature measured (4.2 K) if the pairing has anisotropic *s*-wave symmetry.

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The discovery of superconductivity in the simple binary compound MgB_2 with a remarkably high transition temperature $T_c \sim 39$ K has attracted great interest.[1] MgB_2 is a hexagonal AlB_2-type compound, consisting of alternating hexagonal Mg layers and graphitelike B layers. To explore the mechanism of superconductivity in this material it is important to determine the symmetry of the superconducting order parameter, which governs the behavior of quasiparticle excitations below T_c. Experimentally this can be done by measuring thermodynamic responses of superconducting quasiparticles at low temperatures. In conventional *s*-wave superconductors, there are no quasiparticle excitations at low energies and the thermodynamic and transport coefficients decay exponentially at low temperatures. However, in unconventional superconductors with gap nodes, such as in high-T_c oxides, power law behaviors are expected in thermodynamic coefficients at low temperatures.

Recently, the scanning tunneling conductance,[2–5] the nuclear spin-lattice relaxation rate[6] of MgB_2, and the specific heat[7] have been measured. Although it was claimed that the experimental data are consistent with a conventional *s*-wave pairing gap, the data reported are rather controversial and no consensus can really be reached regarding the pairing symmetry. The gap values obtained from the scanning tunneling measurements vary from 2 meV to 7 meV. The low-temperature dependences of tunneling spectra reported by different experimental groups also behave differently. The difference in both the value of the energy gap and the low-temperature dependence of the tunneling spectrum is probably due to the state of the surface, as well as the inhomogeneity of the samples measured. The spin-lattice relaxation rate measured by Kotegawa *et al.*[6] shows a very small coherence peak just below T_c, followed by an exponential decay in a broad temperature range. However, their measurement was done at relatively high temperatures (above 15 K), and it is not known whether this exponential behavior extends to lower temperatures.

In this paper, we report our experimental data of the magnetic penetration depth λ of MgB_2 in the superconducting state. We have measured λ using the transverse-field muon spin rotation (TF-μSR) and low field ac susceptibility. From both measurements, we find that at low temperatures λ varies quadratically with temperature and does not show the activated exponential behavior expected for a conventional *s*-wave superconductor. The quadratic behavior of λ is evidence for unconventional superconductivity.

The penetration depth is inversely proportional to the square root of the superfluid density. Its temperature dependence is a measure of the low-lying superconducting quasiparticle excitations and no phonon contribution is directly involved. This makes the analysis of penetration depth data simple. Another advantage of the penetration depth measurement is that it allows us to determine whether there are nodes in the superconducting energy gap even with slightly impure samples.[8,9]

The sample measured was commercially available MgB_2 powder (Alfa Aesar). The superconducting transition temperature, as determined by both ac susceptibility and dc superconducting quantum interference device (SQUID) (at 20 G) measurements, is 37.5 K. High-field dc SQUID and electron microscopy investigations showed less than 1% of impurities present. The ac susceptibility measurements were performed at an applied field $H_{ac}=1$ G rms and frequency $f=333$ Hz on fine powder. The absence of weak links was confirmed by checking the linearity of the pickup voltage at 4.2 K for H_{ac} from 1 to 10 G rms and f from 16 to 667 Hz.

TF-μSR is a sensitive technique for measuring λ. In this technique, the field distribution of a flux-line lattice of a type-II superconductor produced by an applied magnetic field is probed by fully polarized positive muons implanted into a specimen. The muon decays with a lifetime 2.2 μs, emitting a positron preferentially in the direction of the muon spin at the time of decay. By accumulating time histograms of the decay positrons the muon polarization can be followed as a function of time. In type-II superconductors, the muon spin precesses about the local field, which is modulated by flux vortices. The time resolved polarization signal is oscillatory with a decreasing amplitude. The damping of the muon precession signal provides a measure of the inhomogeneity of the magnetic field ΔB in the vortex state, hence the magnetic penetration depth λ.[10–12] For polycrystalline samples the envelope of the muon precession signal has approximately a Gaussian form $\exp(-\sigma^2 t^2/2)$ and the depolarization rate σ can be shown to be proportional to the superfluid density $1/\lambda^2$.[11,12] For isotropic type-II superconductors, λ is given by[10]

$$\sigma(\mu s^{-1}) = 7.904\times 10^4 \times \lambda^{-2} \ \ (\text{nm}), \tag{1}$$

whereas for anisotropic superconductors such as the high-T_c cuprates, the in-plane penetration depth can be determined from σ (Ref. 11),

$$\sigma(\mu s^{-1}) = 7.086\times 10^4 \times \lambda^{-2} \ \ (\text{nm}). \tag{2}$$

Evidence about the anisotropy of the superconductivity may be obtained from the form of the distribution of internal fields $P(B)$, which can be derived by Fourier transforming the muon precession signal. Detailed information requires data from single crystals. However in powder samples of anisotropic superconductors it is found that the distribution $P(B)$ has a characteristic shoulder on the low-field side of the central frequency.[13] We used the maximum entropy method to extract $P(B)$ from the TF-μSR data for MgB_2. For temperatures just below T_c the low-field shoulder is clearly evident, so we conclude that MgB_2 is anisotropic.

Our TF-cooled μSR measurements were performed at the ISIS Facility, Rutherford Appleton Laboratory. The sample measured by TF-μSR was a pellet of MgB_2, 4 cm in diameter and 2 mm thick, prepared by cold pressing the powder. The pellet was mounted in a transverse magnetic field H_{app}, which was above the lower critical field but below the upper critical field, $H_{c1} < H_{app} < H_{c2}$. H_{c1} and H_{c2} of MgB_2 are about 300 G and 18 T, respectively.[14] We used a field of 450 G. (Measurements at not too high fields compared to H_{c2} can minimize possible effects of dissipation due to flux motion and allow us to obtain reliable data for the temperature dependence of the relaxation rate σ.[15]) A set of measurements at different fields (up to 600 G) was also done to ensure that the values of σ obtained were independent of the applied field.

The low-field ac susceptibility is also a commonly used technique for measuring λ. It has been successfully applied to high-T_c materials[16] and is particularly suitable for powder samples. The accuracy in the temperature dependence of λ determined from the ac susceptibility technique is significantly higher than that of TF-μSR.[15] In the superconducting state, the effective ac susceptibility χ of a powder sample is related to λ by the equation[17,18]

$$\chi(T) = \chi_0 \left\langle 1 - \frac{3\lambda}{R}\coth\frac{R}{\lambda} + \frac{3\lambda^2}{R^2} \right\rangle, \tag{3}$$

where χ_0 is the susceptibility in an ideal diamagnetic system, R is the radius of a grain, and $\langle \cdots \rangle$ denotes a grain average defined by $\langle x \rangle \equiv \int dR x R^3 g(R)/\int dR R^3 g(R)$ with $g(R)$ the grain-size distribution function. At low temperatures, $\chi(T)$ can be expanded with $\delta\lambda(T) = \lambda(T) - \lambda_0$, where $\lambda_0 = \lambda(0\ \text{K})$. To the leading order in $\delta\lambda$, we find

$$\chi(T) \approx \chi(0) + \alpha\,\delta\lambda(T), \tag{4}$$

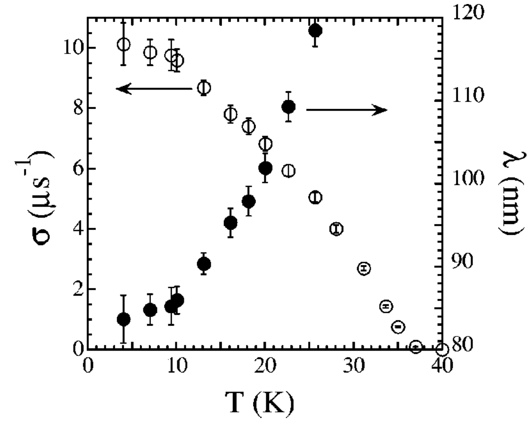

FIG. 1. The muon depolarization rate σ for MgB_2 and the corresponding penetration depth λ as determined from Eq. (2) versus temperature.

where

$$\chi(0) = \chi_0 \left\langle 1 - \frac{3\lambda_0}{R}\coth\frac{R}{\lambda_0} + \frac{3\lambda_0^2}{R^2} \right\rangle, \tag{5}$$

$$\alpha = \chi_0 \left\langle \frac{6\lambda_0}{R} - 3\coth\frac{R}{\lambda_0} - \frac{3R}{\lambda_0 \sinh^2[R/\lambda_0]} \right\rangle < 0. \tag{6}$$

At low temperatures, the change in λ is proportional to the change in χ. Thus from the temperature dependence of $\chi(T)$ (exponential or power law), one can readily determine the temperature dependence of $\lambda(T)$ in the low-temperature regime. However, to determine the absolute value of λ, we need to know accurately the grain distribution function $g(R)$, which is beyond the scope of the present work.

Figure 1 shows the temperature dependence of the muon depolarization rate σ. At zero temperature we find that $\sigma(0\ \text{K}) \sim 10\ \ \mu s^{-1}$. The corresponding in-plane penetration depth at 0 K is $\lambda_0 \sim 85$ nm. The temperature dependence of λ at low temperature is clearly stronger than the exponentially activated temperature dependence expected for an *s*-wave superconductor. In fact, we find that σ varies approximately quadratically with T in the whole temperature range below T_c (Fig. 2). This is supported by our low-

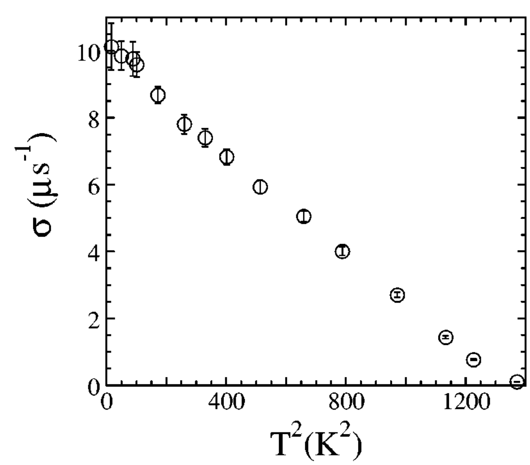

FIG. 2. The muon depolarization rate σ vs T^2.

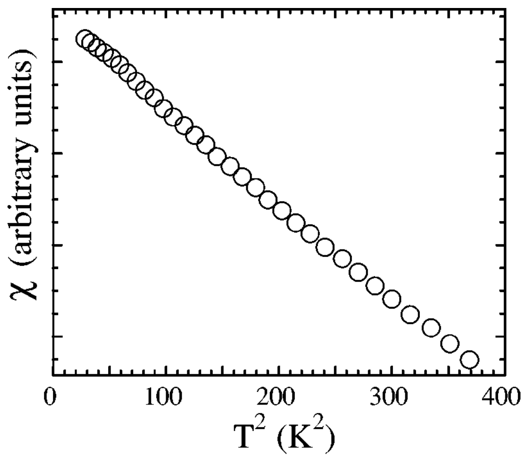

FIG. 3. The ac susceptibility χ as a function of T^2 at low temperatures. The measurement was performed at 1 G and 333 Hz.

temperature ac susceptibility data shown in Fig. 3, which is related to the penetration depth by Eq. (4). The T^2 behavior of λ at low temperatures suggests that either there are nodes in the superconducting energy gap[9] or the minimum gap is significantly less than 4.2 K if the pairing has anisotropic *s*-wave symmetry.

The λ values shown in Fig. 1 have been estimated using Eq. (2). If the electromagnetic response of MgB_2 is more isotropic, the values of λ reported here need to be multiplied by a constant factor c of order 1. In the extremely isotropic case $c=1.06$. However, the T^2 dependence of λ is unchanged.

The T^2 behavior of λ at low temperatures is a typical feature of disordered superconductors with line nodes, such as the Zn-doped high-T_c superconductor $YBa_2Cu_3O_7$.[8,19] In fact as little as 0.31% Zn substitution can cause a crossover from a linear temperature dependence to T^2 without affecting T_c.[8] This has been interpreted as the effect of impurity scattering on a superconductor with line nodes in the energy gap and offers a natural explanation for the T^2 behavior of λ we found in MgB_2. The T^2 dependence of λ in disordered high-T_c oxides is robust against Zn doping.[19]

In conclusion, we have measured the magnetic penetration depth of the newly discovered superconductor MgB_2 using the transverse-field μSR and low-field ac susceptibility techniques. The value of λ at 0 K is about 85 nm. λ shows a T^2 dependence at low temperatures. This is strong evidence for unconventional superconducting pairing in MgB_2.

Note added in proof. After submission of the manuscript, we became aware of penetration depth measurements by Li *et al.*[20] where similar conclusions were drawn.

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