

## $\mu$ SR studies of superconducting $\text{MgB}_{1.96}\text{C}_{0.04}$

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### Abstract

The superconducting properties of  $\text{MgB}_{1.96}\text{C}_{0.04}$  have been investigated by the transverse-field muon spin rotation (TF- $\mu$ SR) technique. The extracted temperature dependence of the  $\mu^+$  spin depolarization rate,  $\sigma$  at TF = 0.6 T has been analyzed in terms of a two-gap model. Carbon doping affects the size of both superconducting gaps and the doping effect is more pronounced for the smaller gap, which is related to the 3D  $\pi$ -sheets of the Fermi surface. The ‘universal’ correlation between the superconducting transition temperature,  $T_c$  and the effective Fermi temperature,  $T_F$  (Uemura plot) is also discussed.

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### 1. Introduction

$\text{MgB}_2$  exhibits a superconducting transition at  $\sim 39$  K which is by far the highest  $T_c$  for a binary intermetallic compound. It adopts the  $\text{AlB}_2$ -type hexagonal structure (P6/mmm), comprising close packed Mg layers alternating with graphite-like boron layers. The Mg atoms are located at the centers of boron hexagons midway between adjacent B layers. Chemical substitution is an effective way to study the superconducting properties of  $\text{MgB}_2$  as a function of doping. Several

experiments concerning substitution on the Mg sites have been reported [1], invariably leading to a decrease in  $T_c$ . Successful substitution at the boron sites can be achieved by carbon doping [2]. Carbon substitution leads to a significant contraction in the  $a$  lattice parameter ( $\sim 0.5\%$ ), while the  $c$ -axis remains essentially unchanged, indicating that carbon resides in the boron layers without affecting the interlayer separation. The transition temperature  $T_c$  decreases with increasing carbon concentration,  $x$ .

In this work, we report the temperature dependence of the TF- $\mu$ SR depolarization rate,  $\sigma$  at 0.6 T for  $\text{MgB}_{1.96}\text{C}_{0.04}$ . The results have been successfully analyzed in terms of a two-gap model and the low temperature penetration depth, the

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gap sizes, and the relative density-of-states of the two bands have been extracted. Finally, the position of the  $\text{MgB}_{2-x}\text{C}_x$  compounds in the Uemura plot is discussed.

## 2. Experiments

The  $\text{MgB}_{1.96}\text{C}_{0.04}$  sample was synthesized by heating mixed powders of amorphous boron, carbon black and magnesium at  $900^\circ\text{C}$  for 2 h. The powder was placed in stainless steel tubes and sealed inside quartz tubes. The sample was characterized by synchrotron X-ray powder diffraction [3] and dc magnetic susceptibility ( $T_c = 36.1\text{ K}$ ) measurements.

The  $\mu\text{SR}$  measurements were performed at the Paul Scherrer Institute (Switzerland). After cooling the sample at an external field ( $H_{\text{ext}}$ ) of 0.6 T to temperatures below  $T_c$  in order to induce a homogenous flux line lattice, positive muons (100% spin-polarized) with their initial muon spin polarization transverse to the external field were implanted into the solid sample. In type II superconductors, the muon spin precesses about the local field, which is modulated by the flux vortices. The time evolution of the muon spin polarization function,  $P_\mu(t)$  is measured by monitoring the positrons, which are preferentially emitted along the muon spin direction. For polycrystalline samples in the vortex state, the depolarization function is approximately Gaussian,  $P_\mu(t) \sim \exp(-\frac{1}{2}\sigma^2 t^2)$  and the depolarization rate,  $\sigma$  is proportional to the second moment of the field distribution,  $\langle \Delta B^2 \rangle^{1/2}$ .

## 3. Results and discussion

Fig. 1 presents the extracted temperature dependence of the TF- $\mu\text{SR}$  depolarization rate at  $H_{\text{ext}} = 0.6\text{ T}$  for  $\text{MgB}_{1.96}\text{C}_{0.04}$ . The depolarization rate almost vanishes for  $T > 35\text{ K}$ , while as the temperature decreases a monotonic increase in  $\sigma$  is observed. For  $T < 4\text{ K}$ ,  $\sigma(T)$  reaches a plateau and remains almost constant at lower temperatures.  $\text{MgB}_2$  exhibits an analogous behavior but the onset of the plateau is at higher temperature

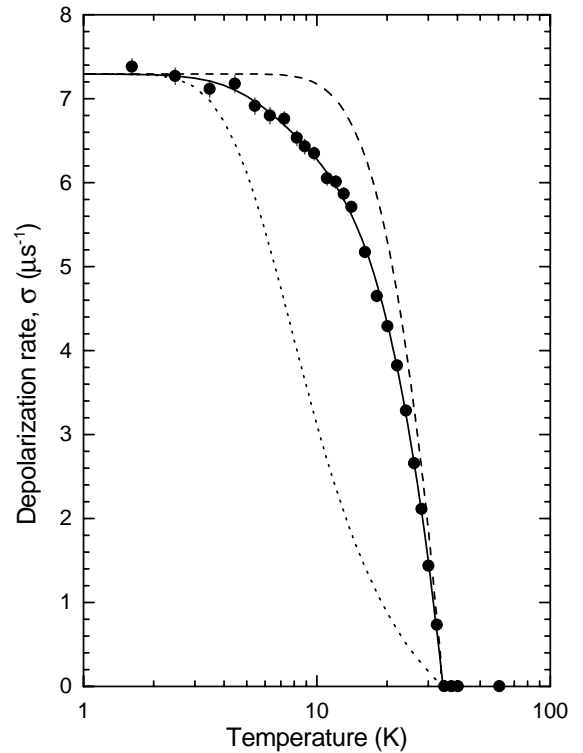


Fig. 1. Temperature dependence of the depolarization rate,  $\sigma$  at  $H_{\text{ext}} = 0.6\text{ T}$  for  $\text{MgB}_{1.96}\text{C}_{0.04}$ . The fit to the two-gap model of Eq. (1) is shown as a solid line. The contributions from the two gaps,  $\Delta_1$  (dashed line) and  $\Delta_2$  (dotted line) are also included.

( $T \approx 5\text{ K}$ ), while its extrapolated low temperature value,  $\sigma(0)$  is larger [4].

Although there is still a debate concerning the applicability of a multi-band description to  $\text{MgB}_2$  (in particular tunneling measurements [5] show only a single gap), recent experiments including scanning tunneling microscopy, point-contact spectroscopy, specific heat measurements,  $\mu\text{SR}$ , optical and Raman spectroscopy [4,6] point towards the existence of two distinct gaps. Furthermore, a significant confirmation of the two-gap model for  $\text{MgB}_2$  comes from the solution of the Eliashberg equations for the gap distribution on the Fermi surface [7,8]. According to this, the gap on the four Fermi surface sheets of this material has two sharp maxima,  $\Delta_1 \approx 6.8\text{ meV}$  at the two 2D  $\sigma$ -bands and  $\Delta_2 \approx 1.8\text{ meV}$  at the two 3D  $\pi$ -bands. Our experimental  $\sigma(T)$  dependence

for  $\text{MgB}_{1.96}\text{C}_{0.04}$  can be also reproduced well (vide infra) by means of a two-gap model. Attempts to fit the experimental data with an isotropic gap model led to unsatisfactory results.

The two-gap model is based on the existence of two discrete superconducting gaps,  $\Delta_1$  and  $\Delta_2$  at  $T = 0$  K, both closing at  $T_c$ . By assuming that the coupling between the 2D  $\sigma$ - and the 3D  $\pi$ -bands, e.g. due to impurity or phonon scattering, is sufficiently weak, the measured  $\sigma(T)$  can be considered as the sum of the contributions from each band. In this case,  $\sigma(T)$  can be expressed as [4,9]

$$\sigma(T) = \sigma(0) - w \cdot \delta\sigma(\Delta_1, T) - (1 - w) \cdot \delta\sigma(\Delta_2, T), \quad (1)$$

where

$$\delta\sigma(\Delta, T) = \frac{2\sigma(0)}{kT} \int_0^\infty f(\varepsilon, T) \cdot [1 - f(\varepsilon, T)] d\varepsilon \quad (2)$$

and  $f(\varepsilon, T) = [1 + \exp(\sqrt{\varepsilon^2 + \Delta(T)^2}/k_B T)]^{-1}$  is the Fermi distribution of quasiparticles. Each band is characterized by partial Sommerfeld constants  $\gamma_1$  and  $\gamma_2$  ( $\gamma_1 + \gamma_2 = \gamma_n$ , where  $\gamma_n$  is the total Sommerfeld constant). The fitting parameter  $w$  in Eq. (1) is equal to the relative weight,  $\gamma_1/\gamma_n$  (with  $\gamma_2/\gamma_n = 1 - w$ ). As the Sommerfeld constant is proportional to the density-of-states at the Fermi level, the ratio  $w/(1 - w)$  determines the ratio of the densities-of-states of the two bands at the Fermi level. For  $\Delta(T)$ , the BCS values tabulated by Mühlischlegel have been used [10]. The fit of the experimental  $\sigma(T)$  data to the two-gap model (solid line) together with the individual contributions of the two superconducting gaps,  $\Delta_1$  (dashed line) and  $\Delta_2$  (dotted line) are shown in Fig. 1. The obtained fitting parameter values for  $\text{MgB}_{1.96}\text{C}_{0.04}$  are  $\Delta_1 = 5.2(2)$  meV,  $\Delta_2 = 1.5(1)$  meV,  $w = 0.8(1)$ , and  $\sigma(0) = 7.3(1) \mu\text{s}^{-1}$ . The parameter  $w/(1 - w)$  is 4(2). For pure  $\text{MgB}_2$  [4], the two-gap model gives the parameter values  $\Delta_1 = 6.0(2)$  meV,  $\Delta_2 = 2.6(1)$  meV,  $w/(1 - w) = 1.8(4)$  ( $w = 0.7(2)$ ), and  $\sigma(0) = 7.9 \mu\text{s}^{-1}$ . In the case of anisotropic type II superconductors and in the absence of pinning-induced distortions in the vortex lattice, the depolarization rate is related to the in-plane penetration depth,  $\lambda_{ab}$  by  $\sigma(\mu\text{s}^{-1}) = 7.086 \times 10^4 \lambda_{ab}^{-2}$  ( $\text{nm}^{-2}$ ). By using this equation, we derive the

low temperature penetration depth value,  $\lambda_{ab}(0) = 98.5(7)$  nm, while for pure  $\text{MgB}_2$  it is 94.7 nm. Hence at  $x = 0.04$ ,  $\sigma(0)$  decreases by  $\sim 7.6\%$ , implying an increase of  $\sim 4\%$  for the in-plane penetration depth.

Our experimental data indicate that carbon doping leads to a reduction of both superconducting gaps.  $\Delta_1$  is reduced by  $\sim 13\%$ , while  $\Delta_2$  by  $\sim 42\%$ , revealing that the doping is somewhat more pronounced for the smaller gap ( $\Delta_2$ ) associated with the 3D  $\pi$ -sheets. The error in the  $w/(1 - w)$  parameter is quite large and hence it is difficult to extract reliably the dependence of this parameter on doping. However, it appears that there may be a tendency for  $w/(1 - w)$  to increase with increasing doping level.

Carbon substitution is expected to lead to an increase in the interband impurity scattering, which should consequently lead to the size of the  $\sigma$ - and  $\pi$ -band gaps converging. However, theoretical calculations [8] have shown that the particular electronic structure of  $\text{MgB}_2$  results in extremely weak  $\sigma\pi$  impurity scattering which is also preserved in cases, like the presence of Mg vacancies, Mg-substitutional impurities, and B-site substitutions by N or C. The dominant mechanism for impurity scattering is due to intraband scattering of the  $\sigma$  and  $\pi$  bands with the scattering rate inside the  $\pi$  bands greater than that of the  $\sigma$  bands [8]. Intraband scattering does not change  $T_c$  and the gap values but influences the penetration depth [11]. Our experimental data can be described adequately with a two-gap model in which impurity scattering is essentially ignored. This indicates that interband impurity scattering is relatively weak at least for  $x = 0.04$ .

The position of the  $\text{MgB}_{2-x}\text{C}_x$  ( $x = 0, 0.02, 0.04, 0.06$ ) superconductors in the so-called Uemura plot [12] is indicated in Fig. 2, which includes the ‘universal’ correlations between  $T_c$  and the effective Fermi temperature,  $T_F$  for various superconductors. For two-dimensional systems,  $T_F$  is proportional to  $\sigma(0) c_{\text{axis}}$ , where  $c_{\text{axis}}$  is the interlayer distance between superconducting planes. By considering the anisotropic nature of superconductivity in  $\text{MgB}_{2-x}\text{C}_x$  and the expressions in Ref. [12], we estimate that  $T_F$  varies between  $\sim 3200$  K for pure  $\text{MgB}_2$  and  $\sim 2700$  K

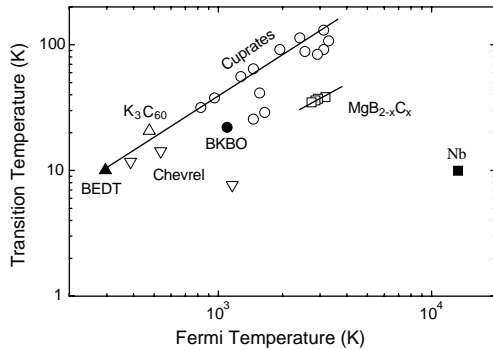


Fig. 2.  $T_c$  versus effective Fermi temperature  $T_F$  in various superconductors, including the  $\text{MgB}_{2-x}\text{C}_x$  ( $x = 0, 0.02, 0.04, 0.06$ ) compounds. Other data are taken from Ref. [12].

for  $\text{MgB}_{1.94}\text{C}_{0.06}$ . In contrast to conventional BCS superconductors in which  $T_c/T_F \ll 0.01$ , the behavior of  $\text{MgB}_{2-x}\text{C}_x$  in which  $T_c/T_F \sim 0.01$ , resembles that of the high- $T_c$  cuprates and other exotic superconductors. This suggests that the superconducting behavior of  $\text{MgB}_{2-x}\text{C}_x$  ( $x = 0, 0.02, 0.04, 0.06$ ) compounds is more complicated than expected in a simple BCS model.

In conclusion, the analysis of the temperature dependence of the TF  $\mu^+$  spin depolarization rate for  $\text{MgB}_{1.96}\text{C}_{0.04}$  shows that the superconducting gap sizes are reduced while the interband scattering remains relatively weak at  $x = 0.04$ . The smaller gap is affected more by carbon doping. Finally, the position of  $\text{MgB}_{2-x}\text{C}_x$  superconductors in the Uemura plot is not in agreement with the expectation from BCS theory.

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