

Fluctuation conductivity in superconducting MgB₂

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According to the crystal structure of MgB₂ and band structure calculations quasi-two-dimensional (2D) boron planes are responsible for the superconductivity. We report on critical field and resistance measurements of 5.6 μm thick MgB₂ films grown on a sapphire single crystalline substrate. Resistivity measurements yield a temperature dependence of the fluctuation conductivity above the critical temperature which agrees with the Aslamazov-Larkin and Maki-Thompson theory of fluctuations in layered superconductors, indicating a quasi-two-dimensional nucleation of superconductivity in MgB₂.

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Recent discovery [1] of superconductivity in magnesium diboride (MgB₂) raised questions about the origin and properties of superconductivity in this compound. MgB₂ has a hexagonal crystal structure with boron layers interleaved by magnesium layers. Band structure calculations [2, 3] indicate that electrons at the Fermi level are predominantly derived from boron atoms. MgB₂ may be regarded as a layered compound having sheets of metallic boron with strong covalent intralayer bonding, separated by Mg layers with ionic interlayer B-Mg bonding. The strong B-B bonding induces enhanced electron-phonon interaction, so that the superconductivity in MgB₂ is mainly due to the charge carriers in the boron planes.

Experimental investigations on single crystals and *c*-oriented epitaxial and textured films (see, e.g., the review [4] and references therein) give evidence for a highly anisotropic superconducting gap. Measured critical magnetic fields usually show a pronounced anisotropy for *c*-oriented films and single crystals [4]. Applying the anisotropic Ginzburg-Landau model to these measurements, authors derive an effective mass anisotropy for the charge carriers of $\gamma = \sqrt{m_{ab}/m_c} \approx 0.15 - 0.3$. Thus, the band structure calculations and experimental measurements strongly suggest that superconductivity nucleates at the quasi-two-dimensional (2D) boron planes, and then extends through the magnesium layers by a

nanoscale proximity effect forming an anisotropic 3D superconducting state in the material.

In this Letter we present experimental evidence for quasi-2D nucleation of superconductivity in a 3D magnesium diboride film. To demonstrate this, we measured the temperature dependence of the excess conductivity caused by fluctuations above the critical temperature, T_c . If quasi-2D boron planes are responsible for the superconductivity, then the excess conductivity should exhibit 2D-like behavior, although measured in a 3D sample. We found that the temperature dependence of the excess conductivity agrees with the Aslamazov-Larkin (AL) [5] and Maki-Thompson (MT) [6] theory of superconducting fluctuations in layered superconductors [7].

The MgB₂ films were prepared by DC magnetron sputtering on single crystalline (100) oriented sapphire substrate according to the procedure described in [8]. To compensate losses of magnesium due to its oxidation in plasma, a composite target was used which contained MgB₂ and metallic magnesium in approximately equal amounts. The Mg-MgB₂ target was sputtered in a 99.999% purity argon atmosphere at a pressure of 3 Pa. The substrate temperature during sputtering was held at 200 °C and then raised to 600 °C for several seconds at the final stage. At this final *in situ* annealing the plasma discharge was not switched off. Next, the films were annealed *ex situ* in a saturated Mg vapor atmosphere during 1 hour at 850 °C. X-ray studies revealed a textured (101)-oriented structure of our polycrystalline films. The MgB₂ film thickness was

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about $5.6 \mu\text{m}$. For the resistance and critical field measurements 1.5 mm wide stripes were cut by a diamond cutter.

The superconducting transition temperature, T_c^{MP} , taken as the mid-point of the transition curve obtained by a conventional four-terminal resistive method in the absence of an applied magnetic field, was about 35.7 K . The upper critical field perpendicular to the film plane has been measured using the 7 T superconducting magnet of a "Quantum Design MPMS-7" SQUID magnetometer.

The resistive transition, $R(T)$, in zero magnetic field for one of the investigated samples is plotted in Fig.1. The transition width according to a $(10-90)\%$ R_n cri-

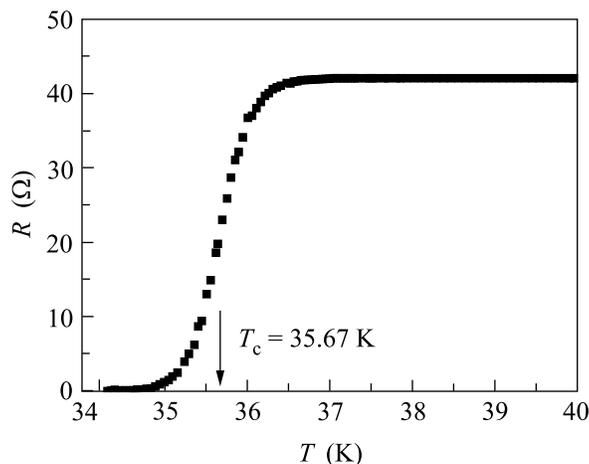


Fig.1. Resistive transition, $R(T)$, for a $5.6 \mu\text{m}$ thick MgB_2 film in zero magnetic field

terion, is about 0.8 K and slightly increases in stronger magnetic field to about 1.5 K at 6 T . The temperature dependence of the critical field perpendicular to the film plane is displayed in Fig.2. The temperature dependence of $B_{c2}(T)$ is linear except for temperatures very close to the critical temperature where a slight positive curvature is observed. Several reasons may be responsible for the positive curvature, such as: anisotropy of the energy gap, proximity effect due to the weakly superconducting Mg interlayers in the MgB_2 compound [9, 10]. This nonlinear behavior of the perpendicular critical field is commonly observed in MgB_2 (see Ref. [4]).

Usually, a linear behavior of $B_{c2}(T)$ oriented parallel to the planes of a layered structure is used as indicator of 3D superconductivity [11, 12], whereas a square root behavior indicates 2D superconductivity. Since the MgB_2 film according to X-ray data is (101)-textured, the boron planes are inclined to the direction of the applied magnetic field by roughly 40° . Therefore, we have two components of the magnetic field of comparable mag-

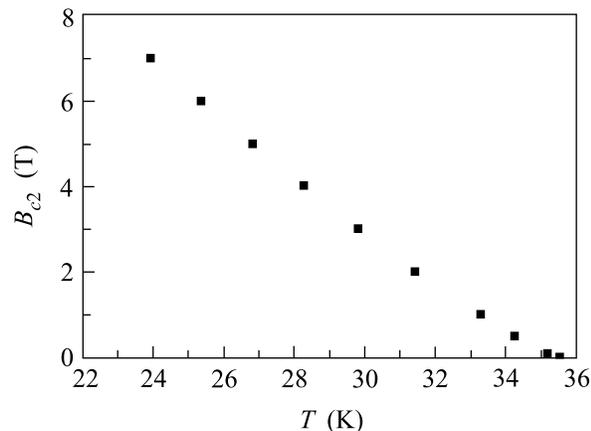


Fig.2. The temperature dependence of the critical magnetic field $B_{c2}(T)$ perpendicular to the film plane obtained from the midpoints of $R(T)$

nitude, one perpendicular to the boron planes and the second one parallel. The parallel component seems to show no square root behavior. Otherwise this should be visible in the temperature dependence of the measured $B_{c2}(T)$. Thus, the linear behavior of $B_{c2}(T)$ shown in Fig.3 indicates the three dimensional nature of the superconducting state in our specimens.

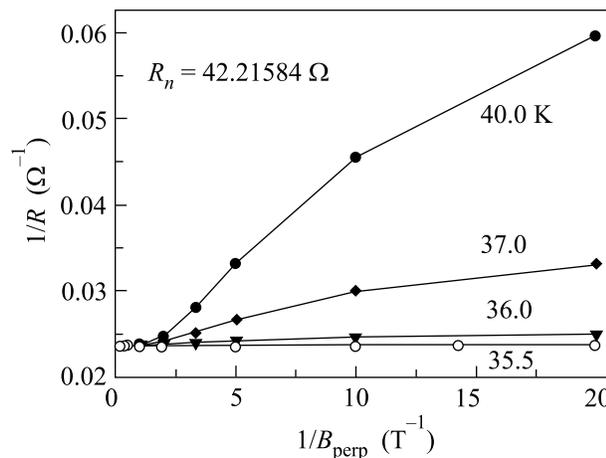


Fig.3. Illustration of the procedure to determine the normal state resistance of the MgB_2 film. Temperatures of measurements are indicated near the curves. Further reduction of the temperature gives results coinciding with the 35.5 K data

To obtain the Ginzburg-Landau coherence length, $\xi_{GL}(0)$, from the slope of $B_{c2}(T)$ close to the critical temperature [13], we use

$$\xi_{GL}(0) = [-(dB_{c2}(T)/dT)(2\pi T_c/\phi_0)]^{-1/2}, \quad (1)$$

where ϕ_0 is the magnetic flux quantum, resulting in $\xi_{GL}(0) = 3.0 \text{ nm}$. According to the discussion on $B_{c2}(T)$

given above, this value may be regarded as a rough estimate.

Using our data on the temperature dependence of the resistance at zero magnetic field we calculate the fluctuation conductivity in the Lawrence-Doniach (LD) model [15] for layered superconductors (see Ref. [7], Ch. 1.7):

$$\sigma^{AL}(T) = \frac{e^2}{16\hbar d} \frac{1}{[\varepsilon(\varepsilon + r)]^{1/2}}, \quad (2)$$

$$\sigma^{MT}(T) = \frac{e^2}{4\hbar d(\varepsilon - \delta)} \ln \left[\frac{\varepsilon^{1/2} + (\varepsilon + r)^{1/2}}{\delta^{1/2} + (\delta + r)^{1/2}} \right], \quad (3)$$

where d is the interlayer spacing, $\varepsilon = (T - T_c^{AL})/T_c^{AL}$, and T_c^{AL} is the Aslamazov-Larkin critical temperature [5]. We use the simple estimation for the pair-breaking parameter $\delta = (T_{c0} - T_c^{AL})/T_{c0}$, where T_c^{AL} is the real superconducting transition temperature of the sample (but not the midpoint T_c^{MP} quoted above), and T_{c0} is the transition temperature for negligible pair-breaking [14]. The Lawrence-Doniach anisotropy parameter, r , is given by ([7], § 1.2.4, Eq. (1.63)):

$$r(T) = 4\xi_z^2/d^2 = \frac{J^2}{k_B T} \begin{cases} \frac{\pi\tau}{4\hbar}, & k_B T\tau/\hbar \ll 1, \text{ "dirty" limit,} \\ \frac{7\xi(3)}{8\pi^2 k_B T}, & k_B T\tau/\hbar \gg 1, \text{ "clean" limit.} \end{cases} \quad (4)$$

It characterizes the strength of interlayer coupling. Here ξ_z is the interlayer tunneling coherence length, $J = \hbar W$ is the energy of interplanar tunneling with W being the electron tunneling rate, τ is the in-plane electron mean free time. We note that for $T \approx T_c$, $k_B T_c\tau/\hbar = 0.18l/\xi_{BCS}$, where ξ_{BCS} is the BCS-coherence length and l is the electron mean free path.

The interlayer coupling strength r , according to Eq. (1), is strongly temperature dependent. When the temperature decreases, the interlayer coupling $r(T)$ increases, so that the effective dimensionality of fluctuations changes from 2D towards 3D.

Taking into account that in Eqs. (2) and (3) the parameter $\varepsilon \ll 1$, and that our samples correspond to the "dirty" case, we used the first line in Eq. (1) in the form

$$r(T) = r_0 (1 - \varepsilon) \quad (5)$$

with $r_0 = r(T_c^{AL})$.

The excess conductance due to superconducting fluctuations has to be calculated from the temperature dependence of the resistance by the relation

$$\sigma'(T) = \frac{1}{R(T)} - \frac{1}{R_n}. \quad (6)$$

For this purpose the normal state resistance, R_n , of our samples was determined with high accuracy as the asymptotic convergence point of the $R^{-1}(1/B)$ dependence at $B \rightarrow \infty$, according to the procedure given in Ref. [14], as shown in Fig.3. Figure 4 shows the temperature dependence of the inverse excess conductance σ' normalized by the normal state conductance $\sigma_n = R_n^{-1}$.

We fit the experimental data with the sum of the Aslamazov-Larkin, Eq. (2), and Maki-Thompson, Eq. (3), terms. As fitting parameters we used T_c^{AL} , δ , r_0 , and the amplitude, $a = 4\hbar d L/e^2 R_n S$, where S is the cross-section of the stripe shaped sample and L is the distance between the voltage probes. The result of the free fitting with unconstrained range of parameters is shown in Fig.4 by the solid line. The fitting shows

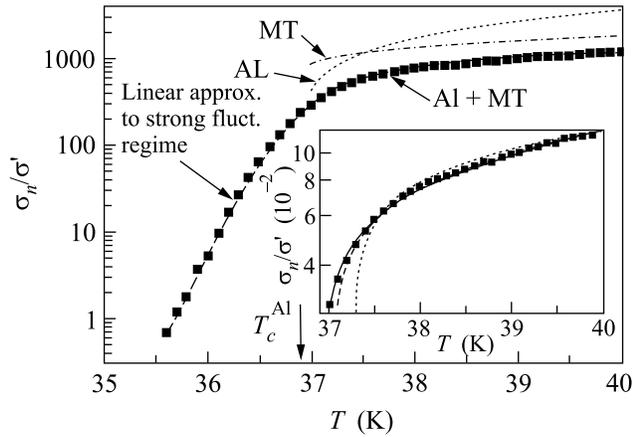


Fig.4. The temperature dependence of the inverse fluctuation conductance, $[\sigma'(T)]^{-1}$, normalized by the normal state value, σ_n , for the resistive transition in zero field (data of the curve from Fig.1). The solid line is the fit of experimental data by the sum of Eqs. (2) and (3) valid for layered superconductors. At about 37.5 K the AL and MT terms contributions become equal. In the Inset the dashed line shows the result of a free fit of the 3D model to the experiment. The dotted line shows the fit of the 3D model with fixed parameter $\delta = 0.09$

fairly good agreement between the experiment and the theory of weak superconducting fluctuations in layered superconductors (see, [7] and references therein) with anisotropy parameter $r_0 \sim 0.06$, parameter $\delta \simeq 0.09$ and $T_c^{AL} \simeq 36.9$ K.

To discuss the above values of parameters it should be remarked that the Lawrence-Doniach model of layered superconductors, Eqs. (2) and (3), used for the fitting, exactly reproduces the limit of a 2D superconductor for $r = 0$ and an isotropic 3D-superconductor for $r \gg 1$ [5, 6]. Our value $r_0 = 0.064$ indicates a highly anisotropic superconductor with weak interlayer

coupling, because, according to Eq. (1), in this case the interplanar coherence length ξ_z is much smaller than the interlayer spacing d .

In comparison, an analysis of fluctuation conductivity for the YBCO superconductor [16] yielded the anisotropy parameter $r_0 \sim 0.06 - 0.09$ for well oxygenated samples. Recent analysis of fluctuation-induced diamagnetism in optimally doped YBCO [17] gave the value $r_0 \simeq 0.1$. From these data we conclude that at least concerning fluctuations the anisotropy of superconducting properties of MgB₂ is very close to those of YBCO.

To evidentialize the highly anisotropic nature of MgB₂ we fitted our experimental data also using pure-3D expressions for the fluctuation conductivity (*i.e.* the $r \gg \varepsilon, \delta$ limit of Eqs. (2) and (3)). The result of free fitting of all parameters is shown in the inset of Fig.4 by the dashed line. The fit by a 3D model seems to look reasonable. However, careful observation reveals two inconsistencies.

First, due to Eq. (5), $r(T)$ increases upon lowering the temperature, shifting the theoretical description of the fluctuation conductivity towards the 3D limit. Therefore, the 3D model should fit better the low-temperature part of the σ_n/σ' measurements than the high-temperature regime. However, the 3D model deviates systematically from the experimental points between 37–37.3K.

Second, the pair-breaking parameter $\delta \simeq 0.0053$ from the 3D fit is too small to meet physical expectations. From the transition temperature of our samples and the bulk value $T_c^{MP} \simeq 39.5$ K [4], both determined using the same mid-point criteria we get $\delta^{MP} = (T_{c0}^{MP} - T_c^{MP})/T_c^{MP} \simeq 0.09$. The parameter δ^{MP} estimated from the T_c -suppression agrees with $\delta = 0.089$ as got from our fit of the LD model. If we fix the parameter δ at $\delta = 0.09$, the fit of our experimental data by the 3D model (the dotted curve in the inset of Fig.4) shows obvious discrepancy between the theory and the experiment.

Finally, the fitting parameter $T_c^{AL} \simeq 36.91$ K (for the free fit of the 3D model it is $T_c^{AL} \simeq 37.03$ K) is the transition temperature which may be treated as the true value of the superconducting transition temperature of our sample. It does not coincide with T_c^{MP} determined by the mid-point transition criteria. The mid-point T_c^{MP} may be regarded as a “technical” value of the superconducting transition temperature, which contains all extrinsic and the intrinsic shift of the critical temperature by fluctuations. Consistent free fitting of fluctuation conductivity provides much more sophisticated and refined determination of the superconducting

transition temperature as a true physical characteristics of a material.

Below T_c^{AL} the regime of fluctuations changes to strong fluctuations with exponential temperature behavior of resistance similar to that observed in thin film superconductors [18].

In summary, we have measured the resistance and critical fields of MgB₂ films prepared on sapphire substrates. From the linear temperature dependence of the critical magnetic field we established three-dimensional superconductivity of our films. From resistivity measurements we showed that the temperature dependence of fluctuation conductivity above the critical temperature agrees with the Lawrence-Doniach theory for layered superconductors. According to the resulting anisotropy parameter, an experimental indication of almost two-dimensional nucleation of superconductivity in MgB₂ was established.

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