Anisotropies of the Lower and Upper Critical Fields in MgB_2 Single Crystals

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The temperature dependence of the upper (H_{c2}) and lower (H_{c1}) critical fields has been deduced from Hall probe magnetization measurements of high quality MgB_2 single crystals along the two main crystallographic directions. We show that $\Gamma_{H_{c2}} = H_{c2\|ab}/H_{c2\|c}$ and $\Gamma_{H_{c1}} = H_{c1\|c}/H_{c1\|ab}$ differ significantly at low temperature (being $\sim$5 and $\sim$1, respectively) and have opposite temperature dependencies. We suggest that MgB_2 can be described by a single field dependent anisotropy parameter $\gamma(H)$ $(= \lambda_c/\lambda_{ab} = \xi_{ab}/\xi_c)$ that increases from $\Gamma_{H_{c1}}$ at low field to $\Gamma_{H_{c2}}$ at high field.

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MgB_2 belongs to an original class of superconductors in which the electronic system consists of two types of carriers — derived from boron π and σ orbitals — with two superconducting energy gaps [1]. The coexistence of these two gaps [2] with different anisotropies then gives rise to very peculiar physical properties [1,3,4]. Among these, the strong temperature dependence of the anisotropy of the upper critical field $\Gamma_{H_{c2}} = H_{c2\|ab}/H_{c2\|c}$ is now well established [5,6] ($H_{c2\|ab}$ and $H_{c2\|c}$ being the upper critical fields for magnetic fields parallel to the ab planes and c direction, respectively). However, the anisotropy of the *lower* critical field is still to be clarified. Caplin *et al.* [7] suggested that $\Gamma_{H_{c1}} = H_{c1\|c}/H_{c1\|ab} \sim 2$, independent of temperature, but found a temperature independent $\Gamma_{H_{c2}}$ value (above 25 K) in striking contrast with other measurements [5,6]. On the other hand, Zehetmayer *et al.* [8] found that $\Gamma_{H_{c1}}$ decreased with temperature *assuming*, however, that Γ_λ $(= \lambda_c/\lambda_{ab}$, where λ is the penetration depth) is equal to $\Gamma_{H_{c2}}$.

In this Letter, we present magnetization measurements performed on a high quality single crystal [9] ($T_c \approx$ 36.5 K) with flat and shiny surfaces of typical dimensions: $100 \times 100 \times 25\ \mu m^3$. The largest surface (i.e., the ab planes) of the sample has been placed either parallel to or perpendicular to the surface of a Hall probe in order to measure the magnetization for $H \| c$ and $H \| ab$, respectively [see sketches in Fig. 2 (below)]. We show that H_{c1} is almost isotropic at low temperature and that, in contrast to $\Gamma_{H_{c2}}$, $\Gamma_{H_{c1}}$ *increases* with T. This increase is in good agreement with recent calculations of the anisotropy of λ in the weakly coupled two bands superconductor [3].

Typical magnetization loops at $T = 5$ K and $T = 20$ K are presented in Fig. 1 for $H \| c$. The alignment of the external field with the main crystallographic axis has been obtained by slightly rotating the ensemble in order to get the maximum and minimum H_{c2} values for $H \| ab$ and $H \| c$, respectively. In the Bean model (i.e., assuming that the hysteresis mainly arises from bulk pinning), the half-width of the loop is related to the critical current density J through $Jd = (M_{up} - M_{down})/2$, where M_{up} and M_{down} are the magnetization for increasing and decreasing magnetic fields, respectively, and d is a characteristic length scale on the order of the sample dimension. The deduced magnetic field dependence of Jd is displayed in the inset of Fig. 2(b) together with the values deduced from ac-susceptibility measurements [5]. Taking $d \sim$ 50 μm, $J \sim 10^3$–10^4 A/cm^2 at low T and low H with very similar values obtained from the two measurements in the common magnetic field range. Given such small J values (being several orders of magnitude smaller than the ones obtained in thin films [10]), the reversible magnetization can be easily obtained assuming that

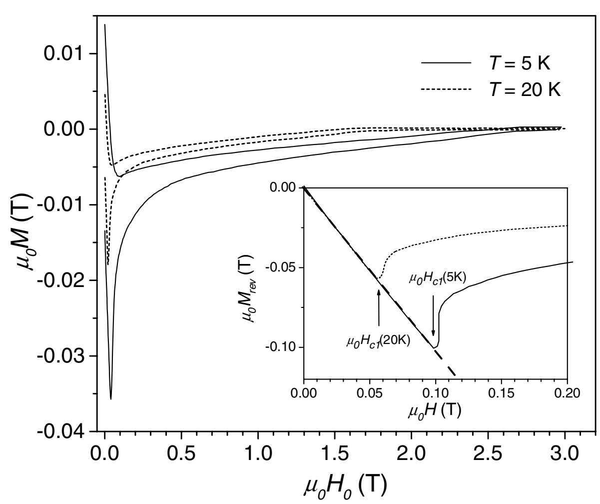

FIG. 1. Magnetization loops at $T = 5$ K and $T = 20$ K for $H \| c$ (H_0 is the applied field). In the inset: Zoom of the magnetic field dependence of the reversible part of the magnetization after correction for demagnetization effects.

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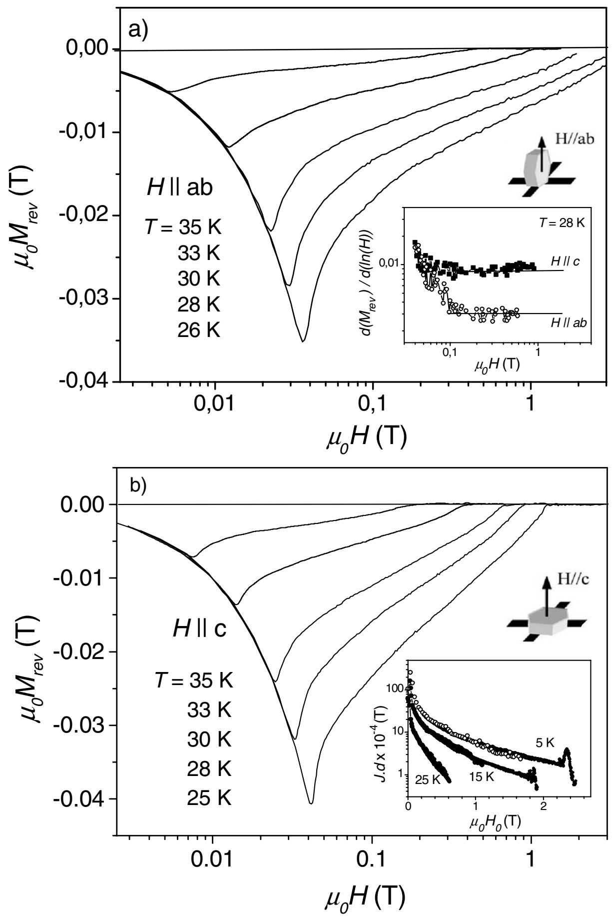

FIG. 2. Reversible magnetization as a function of the internal field H for $H \parallel c$ (a) and $H \parallel ab$ (b) at various temperatures. The schematical drawings show the geometry used for both directions. In the inset of (a), $d\mu_0 M_{rev}/d\ln(H)$ as a function of H at 28 K in both directions, and (b) critical current density as a function of the applied field H_0 deduced from the width of the magnetization loop (open circles) and from ac-susceptibility measurement (closed circles [5]). A peak effect is clearly visible at low temperature close to H_{c2}.

$M_{rev} = (M_{up} + M_{down})/2$. Typical curves are displayed in Fig. 2 for $T > 26$ K for fields along the two main crystallographic directions.

For $H \parallel c$, i.e., perpendicular to the platelet, important demagnetizing effects come into play and H has been rescaled to $H = H_0 - N_c M_{rev}$, where H_0 is the external field and N_c is the demagnetizing factor. N_c can be estimated assuming that the sample is an ellipsoid of thickness $t \sim 25$ μm and width $w \sim 100$ μm giving $N_c = 1 - \pi t/2w \approx 0.6$ [N_{ab} has then been set to $N_{ab} \approx (1 - N_c)/2$]. This value is consistent with the fact that, after correction for the demagnetization effect, the magnetization curves present a nearly vertical slope for $H = H_{c1}$ [11] (see inset of Fig. 1; the y axis has been slightly rescaled in order to get a -1 slope in the Meissner state to account for the fact that the sample did not completely cover the surface of the Hall probe, especially for $H \parallel ab$). Note that, as pointed out by Zeldov *et al.* [12], the absolute value of $H_{c1\parallel c}$ might be overestimated by a factor on the order of $\sqrt{w/t} \sim 2$ due to geometrical barriers. Similarly, for $H \parallel ab$, the effect of Bean-Livingston (BL) [13] barriers could lead to an overestimation of $H_{c1\parallel ab}$ up to a factor $\kappa/\ln(\kappa)$ (where κ is the Ginzburg-Landau parameter). This overestimation is difficult to estimate in MgB_2 as κ may be field dependent (see below), but BL barriers are expected to give rise to very asymmetric magnetization loops (M being close to zero in the descending branch of the loop; see, for instance [14]), which we did not observe for our samples.

In the following, we have thus assumed that pinning mainly arises from bulk defects and that the lower critical magnetic field is equal to the first penetration field. H_{c1} could then be easily determined from the well-defined minima in the $M_{rev}(H)$ curves (see arrows in the inset of Fig. 1). The temperature dependence of $H_{c1\parallel c}$ and $H_{c1\parallel ab}$ is displayed in Fig. 3. The H_{c1} values determined this way are about 5% larger than those estimated from the point where the reversible magnetization deviates from linearity, but we did not observe any significant difference in the temperature dependence of H_{c1} deduced from either of the two criteria. It is important to note that, even though the absolute values of H_{c1} depend on demagnetization effects, possible surface barriers, and/or on the determination criterion, the T and H dependence of the anisotropy parameters discussed below, and therefor our conclusion, is not affected by such uncertainties.

The temperature dependence of H_{c2} defined as the onset of the diamagnetic response at $M_{rev}(H_{c2}) = 0$ is also presented (see inset of Fig. 3). For $H \parallel ab$, the values deduced from our magnetization measurements above

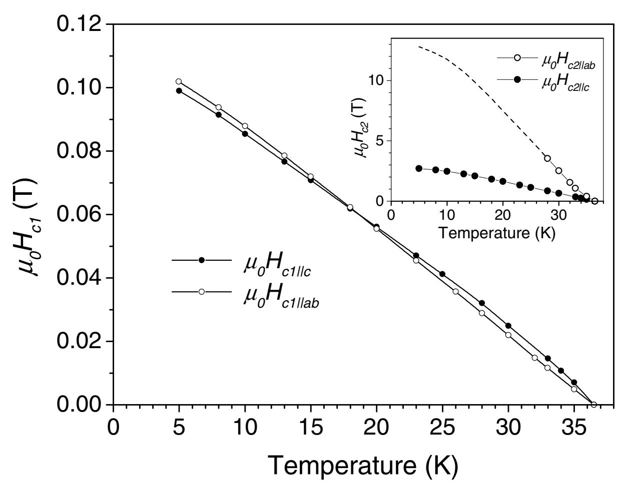

FIG. 3. Temperature dependence of the lower critical field for $H \parallel c$ and $H \parallel ab$ determined from the minimum in the $M_{rev}(H)$ curves (see Fig. 2). The error bars using this determination criterion are on the order of the dimension of the symbols. In the inset: H_{c2} vs T for the same directions. The dotted line is an extrapolation below 26 K deduced from our previous magnetotransport measurements [5].

26 K (open circles) have been extrapolated down to 5 K (dotted line) using our previous high field magnetotransport data performed on another sample coming from the same batch [5] (those previous H_{c2} values were about 20% larger in both directions and have been rescaled to match our data in the common temperature range). As previously observed [5,6,8], close to T_c, the temperature dependence of H_{c2} is almost linear for $H \parallel c$ and presents a positive curvature for $H \parallel ab$ leading to a strong decrease of $\Gamma_{H_{c2}}$ for $T \rightarrow T_c$ (see Fig. 4, open circles).

The situation is very different for the lower critical field which is almost linear for $H \parallel ab$ and reveals a *negative* curvature for $H \parallel c$ at high temperature. This unusual negative curvature close to T_c has been previously observed in thin films and polycrystalline samples [15] and can be explained by the two-band Ginzburg-Landau theory [16]. As shown in Fig. 3, $H_{c1\parallel ab} \sim H_{c1\parallel c}$ at low temperature (i.e., $\Gamma_{H_{c1}} \sim 1 \ll \Gamma_{H_{c2}}$) and the *negative* curvature for $H_{c1\parallel c}$ leads to an *increase* of $\Gamma_{H_{c1}}$ with temperature (see Fig. 4, solid circles). This behavior is in contrast with that previously obtained by Caplin *et al.* [7], who suggested that $\Gamma_{H_{c1}} \sim 2$ down to low temperature and that $\Gamma_{H_{c1}} \sim \Gamma_{H_{c2}}$ for $T > 25$ K. Our result is also in *apparent* contrast with the Zehetmayer *et al.* [8] measurements ($\Gamma_{H_{c1}}(0) \sim 3$). This discrepancy will be discussed below.

These temperature dependencies can be compared to those predicted for the anisotropy of the coherence length $\Gamma_\xi = \xi_{ab}/\xi_c$ and penetration depth $\Gamma_\lambda = \lambda_c/\lambda_{ab}$. "Classical" superconductors can be characterized by one field independent anisotropy parameter: $\gamma = \xi_{ab}/\xi_c (= \Gamma_{H_{c2}}) = \lambda_c/\lambda_{ab}$, whatever the anisotropy of the superconducting gap. However, it has been suggested

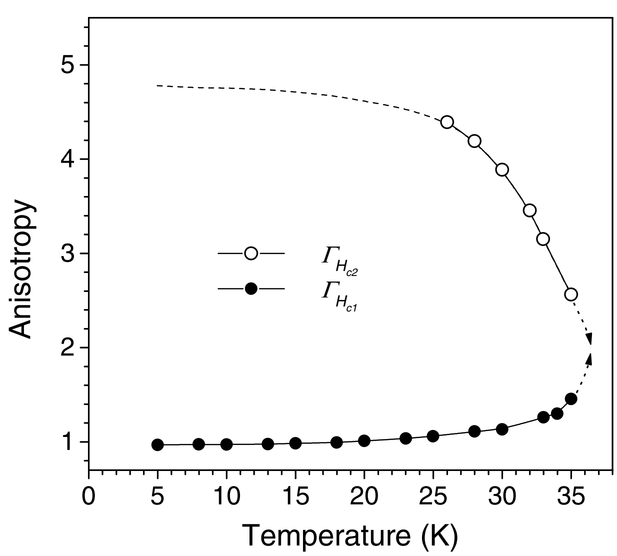

FIG. 4. Temperature dependence of the anisotropy parameters $\Gamma_{H_{c1}}$ and $\Gamma_{H_{c2}}$. The dotted line is an extrapolation of $\Gamma_{H_{c2}}$ at low temperature deduced from our previous magnetotransport measurements [5].

that $\Gamma_{H_{c2}}$ could differ considerably from Γ_λ at low temperature in MgB_2 due to the presence of two superconducting energy gaps with different anisotropies [3]. Indeed, at low temperature the anisotropy of the upper critical field is mainly related to the anisotropy of the Fermi velocities *over the quasi-2D σ sheet*: $\Gamma_{H_{c2}} \approx \sqrt{\langle v_{ab}^2\rangle^\sigma/\langle v_c^2\rangle^\sigma} \sim 6$. $\Gamma_{H_{c2}}$ then decreases with temperature as the influence of the small, nearly isotropic gap increases due to thermal mixing of the two gaps [3,4]. On the other hand, the anisotropy of λ (when deduced from low field measurements) is related to the anisotropy of the Fermi velocities *over the whole Fermi surface* which is expected to be of the order of 1.1 in good agreement with recent neutron scattering data [17] and scanning tunneling imaging [18]. The two anisotropies finally merge at T_c as they are then determined by the same "mass tensor" [3].

In the London model, the penetration depth for $H \parallel c$ can in principle be deduced from H_{c1} through [11]: $\mu_0 H_{c1} = (\Phi_0/4\pi\lambda_{ab}^2)[\ln(\lambda_{ab}/\xi_{ab}) + c]$, where $c \sim 0.5$ in isotropic superconductors. Apart from the uncertainty in c, another difficulty arises in MgB_2 as the superconducting parameters here depend on the value of the magnetic field. Indeed, in a classical system ξ can be obtained from the upper critical field ($\mu_0 H_{c2\parallel c} = \Phi_0/2\pi\xi_{ab}^2$), which here leads to $\xi_{ab}(0) \sim 10$ nm and then to $\lambda_{ab}(0) \sim 60$ nm (taking $c = 0.5$). However, the radius of the vortex core deduced from tunneling measurements at low field (i.e., below the "upper critical field" H_{c2}^π of the π band) [19] is actually much larger (i.e., $\xi \sim 50$ nm) than the one obtained from H_{c2}. The condition $\lambda/\xi \gg 1$ would not be valid any longer, and the use of this equation in MgB_2 should thus be considered carefully.

On the other hand, still assuming that the London model for type II superconductors is valid, λ can be estimated from the slope of the magnetization curve: $1/\lambda^2 = [8\pi/\Phi_0]d(\mu_0 M_{\rm rev})/d\ln(H)$ [11]. As shown in the upper inset of Fig. 2, at low field, this slope rapidly decreases for increasing fields and saturates above some characteristic field $H^*(T)$. Such a decrease of $d(\mu_0 M_{\rm rev})/d\ln(H)$ is expected in type II superconductors as the slope of the $M_{\rm rev}(H)$ curve is expected to be vertical for $H = H_{c1}$ [11]. However, it is important to note that here it occurs for the same H^* value in both directions despite very different H_{c2} values. H^* can thus probably be associated with the rapid filling of the small gap (i.e., of the π band) observed in point-contact spectroscopy experiments [20] in good agreement with specific heat [21] and tunneling [19] measurements ($H^* \sim H_{c2}^\pi$). λ thus rapidly increases with increasing fields [22] reaching $\lambda_{ab} \approx 70$ nm at 5 K for $H > H^* \sim 0.5$ T. This increase is related to the lowering of the superfluid density ($\propto 1/\lambda^2$) as the superconductivity in the π band is destroyed [23]. Our "high field" $\lambda_{ab}(5\ {\rm K})$ estimation is larger than the λ value previously estimated in single crystals by Caplin *et al.* [7] ($\sim$43 nm) but lower than estimates obtained for thin films and polycrystalline samples from rf [24] techniques (~100 nm; for a review, see [15]). However, this value is comparable to the one deduced from μSR measurements ~85 nm [25] (which have been performed above H_{c2}^{π}) as well as the one obtained by Zehetmayer *et al.* [8] (~82 nm) from high field magnetization measurements. On the other hand, the $\xi_{ab}(0)$ values obtained from tunneling measurements at low field (~50 nm [19]) are much *larger* than those deduced from H_{c2} (~10 nm) suggesting that ξ_{ab} *decreases* with field. This would imply that κ_{ab} rapidly increases, ranging from ~1 at low field to ~7 above H^*.

The estimation of λ_c from H_{c1} for $H \parallel ab$ is even more difficult as it is not obvious how the logarithmic term is affected by the anisotropy of the system. However, it has been suggested by Balatskii *et al.* [26] that for uniaxial superconductors this term does not depend on the orientation between the field and the c axis which would directly lead to $\Gamma_{H_{c1}} = \Gamma_\lambda \sim 1$ at low temperature in good agreement with previous experimental data [23,18]. Assuming that $\Gamma_{H_{c1}} \approx \Gamma_\lambda$, the increase of $\Gamma_{H_{c1}}$ with temperature is then in good agreement with theoretical predictions for the weakly coupled two bands superconductor [3]. Note that we did not observe any significant change in the temperature dependence of Γ_λ, whatever the choice of the logarithmic correction. The choice of this correction and the uncertainties on the absolute value of $H_{c1}(0)$ indeed lead to some uncertainty on $\Gamma_\lambda(0)$. However, using the fact that $\Gamma_\lambda \rightarrow \Gamma_{H_{c2}}$ close to T_c, we can estimate that $\Gamma_\lambda(0) < 1.5$.

Γ_λ can also be estimated through $\Gamma_\lambda = [dM_{\rm rev}/d\ln(H \parallel c)]/[dM_{\rm rev}/d\ln(H \parallel ab)]$. As shown in the upper inset of Fig. 2, these derivatives are very similar in both directions for low magnetic fields confirming that the "low field" Γ_λ value remains of the order of 1 up to $T \approx 28$ K (below 25 K, the H_{c2} value for $H \parallel ab$ was larger than our maximum field and we could thus not deduce $M_{\rm rev}$ very accurately at high field). However, it is important to note that this ratio rapidly increases with a field approaching ~3 above H_{c2}^{π}. This high field Γ_λ value is then on the order of $\Gamma_{H_{c2}} \approx 3.7$ at 28 K. Indeed, above H_{c2}^{π} the anisotropy of λ is no more given by that of the whole Fermi surface and the two anisotropy parameters are thus expected to merge. A similar increase of Γ_λ with field has also been observed in [23] at low temperature, and Γ_λ is thus very different from $\Gamma_{H_{c1}}$ above H_{c2}^{π}. This high field ratio is then consistent with the one obtained by [8] from high field torque measurements. More details on the field dependence of the anisotropy parameters will be given elsewhere.

In conclusion, we have shown that $\Gamma_{H_{c1}}$ and $\Gamma_{H_{c2}}$ not only differ in absolute value at low temperature but also have opposite temperature dependencies in MgB_2. Γ_λ is $\approx \Gamma_{H_{c1}}$ at low field but increases for $H > H_{c2}^{\pi}$. This suggests that MgB_2 could be described by an unique field dependent anisotropy parameter $\gamma(H)$ ($= \Gamma_\lambda = \Gamma_\xi$) rising from $\Gamma_{H_{c1}}$ at low field to $\Gamma_{H_{c2}}$ at high field.

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[1] A. Y. Liu *et al.*, Phys. Rev. Lett. **87**, 087005 (2001).
[2] P. Szabó *et al.*, Phys. Rev. Lett. **87**, 137005 (2001); F. Giubileo *et al.*, Phys. Rev. Lett. **87**, 177008 (2001); F. Bouquet *et al.*, Europhys. Lett. **56**, 856 (2001).
[3] V. G. Kogan, Phys. Rev. B **66**, 020509(R) (2002); P. Miranovic *et al.*, J. Phys. Soc. Jpn. **72**, 221 (2003).
[4] T. Dahm and N. Schopol, Phys. Rev. Lett. **91**, 017001 (2003).
[5] L. Lyard *et al.*, Phys. Rev. B **66**, 180502(R) (2002).
[6] S. L. Bud'ko *et al.*, Phys. Rev. B **64**, 180506 (2001); M. Angst *et al.*, Phys. Rev. Lett. **88**, 167004 (2002); U. Welp *et al.*, Phys. Rev. B **67**, 012505 (2003); A. Rydh *et al.*, cond-mat/0308319.
[7] A. D. Caplin *et al.*, Supercond. Sci. Technol. **16**, 176 (2003).
[8] M. Zehetmayer *et al.*, Phys. Rev. B **66**, 052505 (2002).
[9] K. H. P. Kim *et al.*, Phys. Rev. B **65**, 100510(R) (2002).
[10] H. H. Wen *et al.*, Supercond. Sci. Technol. **15**, 31 (2002).
[11] Z. Hao and J. R. Clem, Phys. Rev. Lett. **67**, 2371 (1991); E. H. Brandt, Phys. Rev. B **68**, 054506 (2003).
[12] E. Zeldov *et al.* Phys. Rev. Lett. **73**, 1428 (1994).
[13] C. P. Bean and J. D. Livingston, Phys. Rev. Lett. **12**, 14 (1964).
[14] M. Konczykowski *et al.*, Phys. Rev. B **43** , 13707 (1991).
[15] C. Buzea *et al.*, Supercond. Sci. Technol. **14**, R115 (2001).
[16] I. N. Askerzade *et al.*, Supercond. Sci. Technol. **15**, L17 (2002). The isotropic Ginzburg-Landau theory allows only for a linear temperature dependence of H_{c1} close to T_c [A. A. Abrikosov, in *Fundamentals of the Theory of Metals* (North-Holland, Amsterdam, 1988)].
[17] R. Cubitt *et al.*, Phys. Rev. Lett. **90**, 157002 (2003).
[18] M. R. Eskildsen *et al.*, Phys. Rev. B **68**, 100508(R) (2003).
[19] M. R. Eskildsen *et al.*, Phys. Rev. Lett. **89**, 187003 (2002).
[20] P. Samuely *et al.*, Physica (Amsterdam) **385C**, 244 (2003).
[21] F. Bouquet *et al.*, Phys. Rev. Lett. **89**, 257001 (2002).
[22] The "low field" λ value is expected to be very sensitive to interband scattering ranging from $\lambda_{ab}^{\rm dirty}(0) \approx 100$ nm in dirty systems to $\lambda_{ab}^{\rm clean}(0) \approx 40$ nm for clean samples [A. A. Golubov *et al.*, Phys. Rev. B **66**, 054524 (2002)].
[23] R. Cubitt *et al.*, Phys. Rev. Lett. **91**, 047002 (2003).
[24] F. Manzano *et al.*, Phys. Rev. Lett. **88**, 047002 (2002); A. Carrington and F. Manzano, Physica (Amsterdam) **385C**, 205 (2003).
[25] C. Niedermayer *et al.*, Phys. Rev. B **65**, 094512 (2002); C. Panagopoulos *et al.*, Phys. Rev. B **64**, 094514 (2001).
[26] A. B. Balatskii *et al.*, Sov. Phys. JETP **63**, 866 (1986).