## Anisotropy of the Sommerfeld Coefficient in Magnesium Diboride Single Crystals

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The anisotropic field dependence of the Sommerfeld coefficient  $\gamma$  has been measured down to  $B \to 0$ by combining specific heat and Hall probe magnetization measurements in MgB<sub>2</sub> single crystals. We find that  $\gamma(B, \theta)$  is the sum of two contributions arising from the  $\sigma$  and  $\pi$  band, respectively. We show that  $\gamma_{\sigma}(B, \theta) = B/B_{c2}(\theta)$  where  $B_{c2}(\theta) = B_{c2}^{ab}/\sqrt{\sin^2\theta + \Gamma^2\cos^2\theta}$  with  $\Gamma \sim 5.4$  ( $\theta$  being the angle between the applied field and the *c* axis) and  $\gamma_{\pi}(B, \theta) = \gamma_{\pi}(B) = B/B_{\pi}(B)$ . The "critical field" of the  $\pi$  band  $B_{\pi}$ is fully isotropic but field dependent increasing from  $\sim 0.25$  T for  $B \leq 0.1$  T up to 3 T  $\sim B_{c2}^c$  for  $B \to 3$  T. Because of the coupling of the two bands, superconductivity survives in the  $\pi$  band up to 3 T but is totally destroyed above for any orientation of the field.

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It is now well established that the anisotropy parameter ( $\Gamma$ ) of magnesium diboride (MgB<sub>2</sub>) is strongly field and temperature dependent [1,2]. This is a direct consequence of the coexistence of two weakly coupled superconducting bands. As suggested by point contact spectroscopy [3] or small angle neutron scattering [4] experiments, the so-called  $\pi$  band is very sensitive to magnetic field and, above some "crossover" field on the order of 0.5–1 T, the anisotropy is then mainly given by the parameters of the quasi-2D  $\sigma$  band leading to  $\Gamma = \Gamma_{H_{c2}} = H_{c2}^{ab}/H_{c2}^c$  (~5–6 at low temperature,  $H_{c2}^{ab}$  and  $H_{c2}^c$  being the upper critical fields parallel to the *ab* planes and *c* axis, respectively). On the other hand, at low field, the anisotropy has to be averaged over the entire Fermi surface [5] leading to  $\Gamma \sim \Gamma_{H_{c1}} \sim 1$  in good agreement with  $H_{c1}$  measurements [2].

Similarly, as a consequence of this suppression of the  $\pi$  band, preliminary measurements of the Sommerfeld coefficient  $\gamma = \lim_{\sigma \in I} T|_{T \to 0}$  ( $C_{el}$  being the electronic contribution, to the specific heat) by Bouquet *et al.* [6] clearly showed that its field dependence is highly non-linear. However, the details of the nature of the super-conducting state remained unknown. We will show here that, above ~0.3 T, superconductivity is induced in the  $\pi$  band by coupling with the  $\sigma$  band leading to a shrinking of the vortex core from  $\xi_{\pi}(0) \sim 350$  Å down to  $\xi_{\pi} = \xi_{\sigma}^c \sim 100$  Å for  $B \sim 3$  T. Superconductivity is completely destroyed in this band above 3 T for any orientation of the magnetic field.

We present the first detailed analysis of the angular and field dependence of  $\gamma$  by combining specific heat and Hall probe magnetization measurements. In classical superconductors, the angular dependence of  $\gamma$  is determined by the  $B/B_{c2}$  ratio with  $B_{c2} = B_{c2}^{ab}/\sqrt{\sin^2\theta + \Gamma^2\cos^2\theta}$  ( $\theta$  being the angle between the *c* axis and the field). We show that, in

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MgB<sub>2</sub>, the contribution of the  $\sigma$  band to  $\gamma$  is directly proportional to  $B/B_{c2}(\theta)$ , whereas the contribution of the  $\pi$  band is isotropic but highly nonlinear in field. For  $T \ge$ 10 K, the influence of the small gap is smeared out by the temperature and  $\Delta C_p/T = f(B/B_{c2}(T, \theta))$ . Specific heat measurements have been performed on single crystals of  $MgB_2$  grown under high pressure [7,8] (with typical dimensions of few hundred microns) using an ac technique. This high sensitivity technique is very well adapted to measure  $C_p$  of very small samples and to carry continuous measurements during field sweeps at a given temperature. We were thus able to obtain the field dependence of the Sommerfeld coefficient continuously on the entire field range for different  $\theta$  values. A precise *in situ* calibration of the thermocouple used to record the temperature oscillations was obtained from measurements on ultrapure silicon.

Figure 1 displays this field dependence for  $H \parallel c$  and  $H \parallel$  ab (at T = 2 K). As previously observed by Bouquet et al., the  $\gamma$  vs  $H_a$  curve is nonlinear. Those measurements suggested that  $\gamma$  is isotropic below 0.2 T, becoming anisotropic for  $H \ge 0.5$  T. However, for fields up to a few  $H_p$ , the first penetration field, the measurements are strongly hysteretic reflecting different vortex distributions in the sample [see [9] and inset of Fig. 1(a)]. The proximity of  $H_p$  may thus cast some doubt on the field dependence of  $\gamma$ observed in [6] at low field. As shown in the inset of Fig. 1(a), the first penetration field  $H_p$  can be clearly identified on the ascending branch of zero field cooled cycles as  $\gamma = 0$  up to  $H_a = H_p$  and rises sharply above this field due to the fast proliferation of vortices in the sample. Vortices remain pinned in the samples for decreasing fields and  $\gamma \neq 0$  down to  $H_a = 0$ . To clearly identify the field dependence of  $\gamma$  in this region it was important to

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FIG. 1. Magnetic field dependence of the Sommerfeld coefficient  $\gamma$  at T = 2.5 K for  $H \parallel c$  and  $H \parallel$  ab. In the inset: low field dependence showing the hysteretic behavior related to flux penetration and pinning. (b) Sommerfeld coefficient  $\gamma$  for  $H \parallel c$  and  $H \parallel$  ab as a function of the average field *B*. In the inset: average field *B* deduced by Hall probe magnetometry (see sketches) as a function of the applied field  $H_a$ .

determine the *true* induction *B* in the sample. We thus performed Hall probe magnetization measurements on the same sample for both field directions using a miniature Hall probe array [see sketches in Fig. 1(b)]. The induction *B* at the surface of the sample has been averaged over ~25 points for  $H \parallel c$  and ~10 points for  $H \parallel$  ab (the average field is hereafter noted *B*). The *B* vs  $H_a$  curves are displayed in the inset of Fig. 1(b) and the corresponding  $\gamma$  vs *B* curves in Fig. 1(b). As shown,  $\gamma$  is perfectly linear and isotropic at low field (note that, as expected  $\gamma$  vs *B* is completely reversible). However, the linear regime is only visible up to ~0.1 T and  $\gamma$  remains isotropic up to  $B \sim 0.3$  T. As discussed in [6], in MgB<sub>2</sub> the nonlinear behavior can be qualitatively understood by writing  $\gamma = \omega \gamma_{\pi} + (1 - \omega) \gamma_{\sigma}$ , where  $\omega$  is the relative weight of the  $\pi$  band (on the order of  $\frac{1}{2}$  [10]). Assuming that all excitations are localized in the vortex cores, i.e., that  $\gamma_i \propto \gamma_N (\xi_i/a_0)^2$  for  $B < B_i = \Phi_0/2\pi\xi_i^2$  and  $\gamma_i = \gamma_N$  for  $B > B_i$  (with  $i = \pi$  or  $\sigma$ ), one gets two linear behaviors corresponding to  $B < B_{\pi}$  and  $B > B_{\pi}$ , respectively [introducing  $a_0 \sim \sqrt{(\Phi_0/B)}$ ]. The low field linear behavior is hence clearly visible but limited on a very restricted field range and, as discussed below, the high field linear behavior is only observed for  $\theta \neq 0$  (for B > 3 T).

To get a better description of  $\gamma(B, \theta)$ , we measured the angular dependence of the Sommerfeld coefficient for various applied fields [see inset of Fig. 2(a), at T = 2 K]. In a classical single gap superconductor  $\gamma$  is fully determined by the  $B/B_{c2}$  ratio. In clean systems, deviations from the above mentioned linear behavior may be expected [11], but  $\gamma$  still remains a function of  $B/B_{c2}$ . Obviously such a simple behavior does not hold in MgB<sub>2</sub> since  $\gamma$  is isotropic up to  $\approx 0.3$  T and its anisotropy then rises up to 5-6 for  $B \rightarrow B_{c2}$ . However, as shown in Fig. 2(a), substracting from  $\gamma(\theta)$  a constant for each  $H_a$  value, i.e., taking  $[\gamma/\gamma_N - \omega a(B)]/(1 - \omega)$  instead of  $\gamma/\gamma_N$ , leads to a collapse of all the data on a single curve when plotted as a function of  $B/B_{c2}(\theta) \propto B\sqrt{\sin^2\theta} + \Gamma^2 \cos^2\theta$  [12] with  $\Gamma = \Gamma_{H_{c2}} = 5.4$ . We here assumed that  $\gamma_{\pi}(B, \theta)$  is isotropic and hence depends only on  $B [\gamma_{\pi}/\gamma_N = a(B)]$ and that  $\gamma_{\sigma}(B,\theta)/\gamma_{N} = [\gamma/\gamma_{N} - \omega a(B)]/(1-\omega) =$  $f(B/B_{c2}(\theta))$ . It is important to note that we made no assumption on the form of the function f and directly got  $\gamma_{\sigma}(B, \theta) = [B/B_{c2}(\theta)]\gamma_N$  as expected in classical (dirty) superconductors. Note also that  $\gamma$  appears to be fully isotropic below 0.3 T but, in this field range, the contribution of the  $\sigma$  band is less than a few percent and the corresponding variation is at the limit of our experimental resolution [13].

The  $a(B) = \gamma_{\pi}/\gamma_{N}$  values are displayed in the inset of Fig. 2(b) (solid symbols) together with direct determinations from magnetic field sweeps for the indicated  $\theta$  val- $\gamma_{\pi}/\gamma_{N} = [\gamma/\gamma_{N} - (1-\omega)B/B_{c2}(\theta)]/\omega$ ues: (open symbols). For  $B \ge 3$  T,  $\gamma_{\pi} = \gamma_N$  showing that superconductivity is completely destroyed in this band at high field. This is further emphasized in the field dependence of  $\gamma$  at fixed  $\theta \neq 0$  values [Fig. 2(b)] which clearly shows that  $\gamma$ becomes perfectly linear for  $B \ge 3$  T in all directions. Note that the linear fits intercept the B = 0 axis at  $\omega \sim$ 0.5 in good agreement with numerical calculations [10]. With this  $\omega$  value, we did *not* observe any linear high field behavior for  $H \parallel c$  but  $\gamma$  can be rather described by a  $(B/B_{c2}^c)^{\alpha}$  law (with  $\alpha \sim 0.4$ –0.5). As discussed in [14], it is then possible to introduce an effective field dependent  $\xi_{\text{eff}}$  value:  $\xi_{\text{eff}}(B) = \sqrt{\omega \xi_{\pi}(B)^2 + (1-\omega)\xi_{\sigma}^2}$  with  $a(B) = [\xi_{\pi}(B)/a_0]^2$  (for  $B \le 3$  T). This effective value, with combined with a field dependent penetration depth can then be used to described all physical properties (see also



FIG. 2. (a)  $[\gamma/\gamma_N - \omega a(B)]/(1 - \omega)$  as a function of  $B/B_{c2}$ with  $B_{c2} = B_{c2}^{ab}/\sqrt{\sin^2\theta + \Gamma^2\cos^2\theta}$  (with  $\omega \sim \frac{1}{2}$  and  $\Gamma \sim 5.4$ ), in the inset  $\gamma/\gamma_N$  as a function of  $\theta$  for the indicated values of the applied field. (b) Field dependence of the Sommerfeld coefficient for the indicated field directions showing that  $\gamma$  becomes linear for  $B \ge 3$  T. In the inset: field dependence of the contribution of the  $\pi$  band:  $\gamma_{\pi}/\gamma_N = a(B)$  deduced from the angular measurements (solid squares) and magnetic sweeps at the indicated angles (open symbols, see text for details).

[15]). The  $\xi_{\pi}(0)$  value would correspond to a critical field for the  $\pi$  band on the order of  $B_{\pi}(0) = 0.25$  T but superconductivity still survives in this band due to the coupling with the  $\sigma$  band leading to a shrinking of the vortex core from  $\xi_{\pi} \approx 350$  Å below 0.1 T down to  $\xi_{\pi} = \xi_{\sigma}^{ab}$  for  $B \sim$ 3 T =  $B_{c2}^c$ . However, it is important to note that, since the  $\pi$  band is isotropic, it can have only one  $\theta$  independent  $B_{c2}$ value and superconductivity is hence destroyed in this band in all direction for  $B \ge 3$  T.

As expected the  $\xi_{\sigma}^{ab} \sim 100$  Å and  $\xi_{\sigma}^{c} \sim 20$  Å values are very close to the BCS single band estimate:  $\hbar v_{F,\sigma}/\pi \Delta_{\sigma} \sim$ 130 Å and ~20 Å in the *ab* planes and along the *c* direction, respectively  $(v_{F,\sigma})$  being the Fermi velocity of the  $\sigma$  band ~4.6 × 10<sup>7</sup> cm/s and ~0.7 × 10<sup>7</sup> cm/s for the two crystallographic directions and  $\Delta_{\sigma}$  the large gap value ~7.0 meV). More surprisingly, the as-deduced  $\xi_{\pi}$  value (~350 Å) is also quite close to  $\hbar v_{F,\pi}/\pi \Delta_{\pi} \sim 400-500$  Å [taking an average  $v_{F,\pi}$  value on the order of (5–6) × 10<sup>7</sup> m/s [5,16] and  $\Delta_{\pi} \sim 2.4$  meV]. Indeed, it has been suggested by Zhitomirsky *et al.* [16] that this single band estimate should not be applicable in MgB<sub>2</sub> and that in the clean limit  $\xi_{\pi}^c/\xi_{\sigma}^c \leq \sqrt{\omega \langle v_{F,\pi}^2 \rangle/(1-\omega) \langle v_{F,\sigma}^2 \rangle} \sim 1-2$  for reasonable  $\langle v_{F,\pi}^2 \rangle/\langle v_{F,\sigma}^2 \rangle$  values [5,16]. Similarly, it has been shown that, in the dirty limit,  $\xi_{\pi}^c/\xi_{\sigma}^c \sim 3$  for  $D_{\pi}/D_{\sigma} \sim 4$  [17] (where  $D_i$  is the diffusivity of the *i* band), i.e., for a  $\sigma$  band which would be much dirtier than the  $\pi$  band. This numerical  $\xi_{\pi}$  value obviously deserves further theoretical investigation.

As discussed in [6], an effective anisotropy ratio can be defined as the ratio of the magnetic fields in the *ab* plane and *c* axis which correspond to the same  $\gamma$  value but, as pointed out in [6], the choice of the corresponding field is then arbitrary and an almost linear increase of  $\Gamma$  with field was proposed. However, we have seen that superconductivity is completely destroyed in the  $\pi$  band for  $B \ge 3$  T and  $\Gamma$  is hence expected to be equal to 5.4 above 3 T. We have calculated this field dependent anisotropy  $\Gamma(B)$ , writing  $\gamma(B, \theta) = \gamma(B\sqrt{\sin^2\theta + \Gamma(B)^2\cos^2\theta}, \theta = 90^\circ)$ , i.e., introducing an effective field dependent  $B_{c2}^* = \frac{B_{c2}^{ab}}{\sqrt{\sin^2\theta + \Gamma(B)^2\cos^2\theta}}$  and writing that  $\gamma = f(B/B_{c2}^*)$ . The corresponding  $\Gamma(B)$  values have been reported on Fig. 3 (open symbols and lines) for  $\theta = 0^\circ$ ,  $30^\circ$ , and  $60^\circ$  together with the  $\Gamma_{\lambda} = \frac{\lambda_c}{\lambda_{ab}}$  values deduced from



FIG. 3. Field dependence of the anisotropy ratio (see text for details) together with  $\Gamma_{\lambda} = \frac{\lambda_{c}}{\lambda_{ab}}$  values deduced from small angle neutron scattering data (closed squares: from [4], closed triangle: from [16].



FIG. 4. (a) Magnetic field dependence of the specific heat at T = 9 K showing that, at high temperature,  $C_p/T$  scales as  $B/B_{c2}(\theta)$ . In the inset: same dependence plotted as a function of the applied field  $H_a$  for  $\theta = 0^\circ$ ,  $30^\circ$ ,  $60^\circ$ , and  $90^\circ$ .

small angle neutron scattering data (closed squares: from [4], closed triangle: from [18]). As shown the same effective anisotropy (saturating at 5.4 for  $B \ge 3$  T) is obtained for all angles and a very reasonable agreement is obtained between specific heat and SANS data confirming that  $\Gamma_{\lambda} = \Gamma = \Gamma_{\xi}$ . Note that  $\Gamma$  stays on the order of 1 up to  $B_{\pi}$  and then sharply increases reaching  $\Gamma_{H_{c2}}$  for  $B \ge B_{c2}^c$ .

Finally, we have investigated the influence of the temperature on the field and angular dependence of  $C_p$ . The inset of Fig. 4 displays the field dependence of  $C_p/T$  at T = 9 K for the indicated  $\theta$  values. In this temperature range superconductivity in the  $\pi$  band is reduced due to thermal activation over the small gap and, as shown in the main panel of Fig. 4, all curves can then be rescaled directly as a function of  $B/B_{c2}(\theta)$  clearly showing that a *classical* behavior is recovered at high temperature (except that  $\Gamma$  is temperature dependent).

We have shown that, the contribution of the  $\sigma$  band to the specific heat  $\gamma_{\sigma}(B, \theta) = [B/B_{c2}(\theta)]\gamma_N$  whereas the contribution of the  $\pi$  band is isotropic but highly nonlinear in field:  $\gamma_{\pi}(B, \theta) = \gamma_{\pi}(B) = [B/B_{\pi}(B)]\gamma_N$  for  $B \leq$ 3 T ~  $B_{c2}^c$  and  $\gamma_{\pi} = \gamma_N$  for  $B \geq$  3 T. We hence show that superconductivity can be induced in the  $\pi$  band by coupling with the  $\sigma$  band but only up to  $B_{c2}^c$  and no superconductivity is observed in this band for  $B \geq$  3 T.

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