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Published on: 27 Nov 2002 - Physical Review B (American Physical Society)

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PHYSICAL REVIEW B **66**, 180502(R) (2002)

Anisotropy of the upper critical field and critical current in single crystal MgB_2

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(Received 13 June 2002; published 27 November 2002)

We report on specific-heat, high-magnetic-field transport, and ac-susceptibility measurements on MgB_2 single crystals. The upper critical field for magnetic fields perpendicular and parallel to the basal planes is presented in the entire temperature range. A very different temperature dependence has been observed in the two directions, which yields a temperature-dependent anisotropy with $\Gamma \sim 5$ at low temperatures and ~ 2 near T_c. A peak effect is observed for $\mu_0 H \sim 2$ T parallel to the c axis, and the critical current density presents a sharp maximum for H parallel to the ab plane.

DOI: 10.1103/PhysRevB.66.180502 PACS number(s): 74.25.Bt, 74.25.Dw, 74.60.Ec

Since the discovery of superconductivity in magnesium diboride at 39 K in 2001 (Ref. 1) an enormous amount of work has been done, which helped to elucidate many of its physical properties. Among others, the two-band superconductivity[2] has been confirmed by different techniques such as specific heat[3] or Andreev reflection.[4] This picture suggests a significant anisotropy of the superconducting state but data reported on the anisotropy of the upper critical field H_{c2} scatter from 1.1 to 13, depending on the form of material (polycrystals, thin films, single crystals) and method of evaluation. The problem thus remains to be clarified on high-quality single crystals.

We have determined H_{c2} in MgB_2 single crystal for H parallel and perpendicular to the ab planes by magnetotransport (down to 5 K and up to 28 T), ac-susceptibility (up to 5 T), and specific heat (up to 7 T) measurements. $H_{c2\perp ab}$ reveals a classical linear temperature dependence near T_c [with $\mu_0 H_{c2\perp ab}(0) \simeq 3.5$ T], while $H_{c2\|ab}$ shows a positive curvature at temperatures above 20 K and saturates at $\mu_0 H_{c2\|ab}(0) \simeq 17$ T. As a consequence $\Gamma = H_{c2\|ab}/H_{c2\perp ab}$ is temperature dependent with $\Gamma(0\text{ K}) \approx 5$ and $\Gamma(\text{near } T_c) \approx 2$. On the other hand, the critical current deduced from ac-susceptibility measurements presents a sharp maximum for $H\|ab$.

Experiments have been performed on high-quality MgB_2 single crystals[5] showing clear hexagonal facets with flat and shiny surfaces (of typical dimensions $50 \times 50 \times 10\ \mu m^3$). Four-probe magnetoresistance measurements have been performed for different temperatures and angles (θ) between H and the ab planes. Gold electrodes have been evaporated as stripes overlapping the top plane of the sample with a contact resistance of $\sim 1\,\Omega$, and H was always orthogonal to the measuring current. The ac susceptibility has been deduced from the local transmittivity measured with a miniature Hall probe. The ac excitation field ($h_{ac} \sim 3$ G, $\omega \sim 23$ Hz) was perpendicular to the ab plane and to the Hall-probe plane, and superimposed over a dc field with an angle $0 < \theta < 90°$. The specific heat was measured by an ac technique.[6] Heat was supplied to the sample at a frequency ω of the order of 70 Hz by a light-emitting diode via an optical fiber. The induced temperature oscillations were measured by a 12-μm-diameter chromel-constantan thermocouple calibrated *in situ*.

Figure 1 displays the temperature dependence of the field-dependent part of the specific heat up to 7 T for $H\|ab$ and up to 2 T for $H\perp ab$. In zero field, the specific-heat discontinuity at T_c represents about 3% of the total signal. This value is five to six times smaller than that obtained in the best polycrystalline samples,[3] which can be attributed to the large contribution of the addenda, given the extremely small

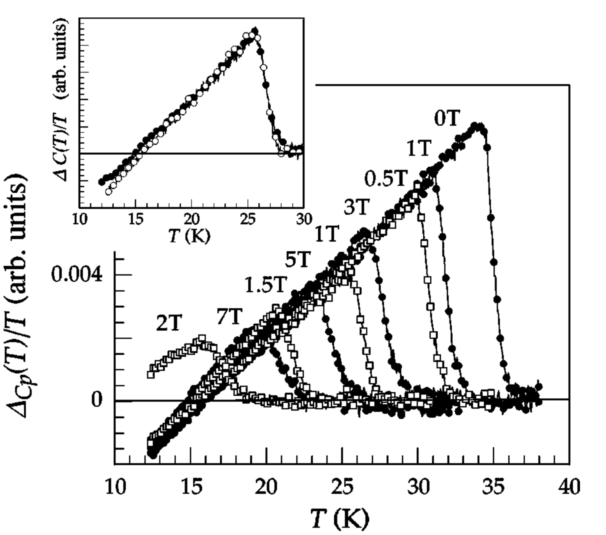

FIG. 1. Temperature dependence of the specific heat at different magnetic fields for $H\|ab$ (closed circles) and $H_{\perp ab}$ (open squares). Inset: specific-heat transition at $\mu_0 H_{\perp ab} = 1$ T and $\mu_0 H_{\|ab} = \Gamma \mu_0 H \perp ab \simeq 3.5$ T.

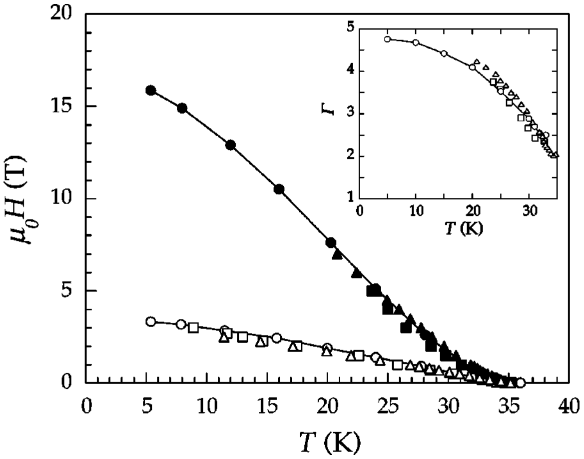

FIG. 2. Temperature dependence of the upper critical field obtained by transport (circles), ac-susceptibility (squares), and specific-heat (triangles) measurements. Inset: temperature dependence of the anisotropy.

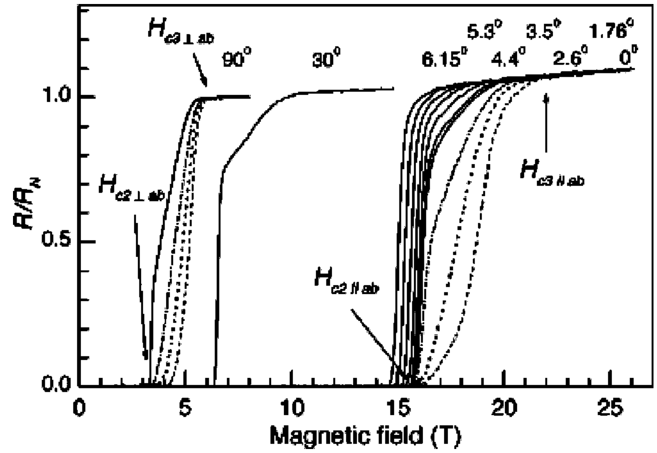

FIG. 3. Magnetic-field dependence of the resistance at $T = 5.4$ K for various angles between H and the ab plane. H_{c2} and H_{c3} are marked by arrows for the two principal field orientations. The influence of the current density is presented for these two orientations (dashed lines from right to left, $j = 17,50,170$ A/cm^2, solid lines $j = 500$ A/cm^2).

size of our single crystals (~ 100 ng). The data are therefore given in arbitrary units and the temperature range is limited to $T > 12$ K. No significant increase of the transition width, $\Delta T \sim 2$ K, is observed for $\mu_0 H || c < 2$ T and $\mu_0 H || ab < 5$ T. Despite a clear broadening at higher fields, the specific-heat anomaly remains well defined in our explored temperature range. This implies that the broadening observed in polycrystalline samples[3,7] is due to randomly oriented anisotropic grains rather than thermodynamic fluctuations.[7] The curves are remarkably identical for both directions once the field is renormalized by the temperature-dependent anisotropy coefficient (see inset of Fig. 1). It is also worth noticing that the amplitude of the specific-heat jump decreases very rapidly with H, implying a rapid increase of the Sommerfeld coefficient γ and therefore of the density of states. An entropy conservation construction shows that $\gamma \sim 0.9 \gamma_N$ (where γ_N is related to the normal-state density of states) for $H \sim 0.5\ H_{c2}$, in good agreement with recent low-temperature measurements by Bouquet *et al.*[8]

The $T_c(H)$ values deduced from the classical entropy conservation construction, as well as those deduced from susceptibility (onset of the diamagnetic response) and transport measurements have been reported in Fig. 2. In the latter case, $T_c(H)$ has been defined at the *onset of finite resistivity* (see Fig. 3). All three methods reveal the same results in the common field range, and the entire H-T phase diagram could be determined from magnetotransport data for both field directions. Note that the resistivity reaches the normal-state value at a field $H_{R_N} > H_{c2}$, as previolusly shown by Welp *et al.*[9] for $H \perp ab$ (see discussion below). As expected for a type-II superconductor, $H_{c2\perp ab}$ varies linearly close to T_c and saturates at low temperature ($\mu_0 H_{c2\perp ab} \simeq 3.5$ T). On the other hand, $H_{c2||ab}$ reveals a positive curvature close to T_c, which changes to negative below 20 K and saturates at a much higher value ~ 17 T. A direct consequence of these two different shapes is that Γ is temperature dependent, decreasing from ~ 5 at low T down to ~ 2 near T_c (see inset of Fig. 2). Despite this T-dependent anisotropy, the $H_{c2}(\theta)$ deduced from the magnetotransport measurements (see Fig. 3) can be well fitted by the ellipsoidal Lawrence-Doniach form for anisotropic three-dimensional superconductors:[10] $[H_{c2}(\theta) \sin\theta / H_{c2\perp ab}]^2 + [H_{c2}(\theta) \cos\theta / H_{c2||ab}]^2 = 1$ with $\Gamma = 4.8$ at $T = 5.4$ K (Fig. 4) and $\Gamma = 3.4$ at $T = 26$ K (not shown).

It has been suggested that the positive curvature observed for $H||ab$ is a consequence of the two-gap structure.[11] However, it is worth mentioning that a very similar behavior has also been observed in single-gap superconductors such as $NbSe_2$ (Ref. 12) or borocarbides.[13] This behavior was believed to be a characteristic of layered compounds,[12] but it is even more "general" as it has also been observed in the isotropic $(K,Ba)BiO_3$ system.[14] The origin of this effect thus still has to be clarified.

Our specific-heat measurements clearly show that H_{c2} is located at the onset of finite resistivity and the resistance R

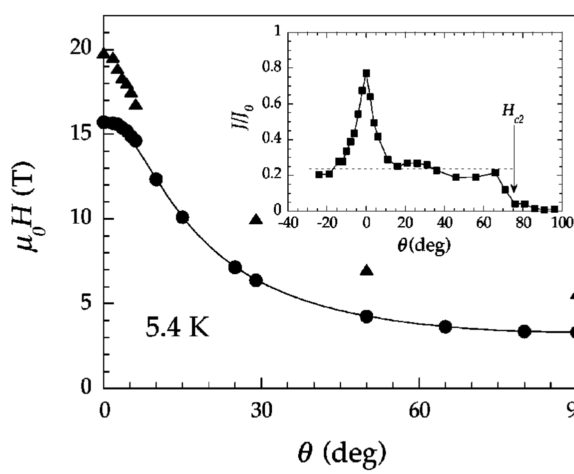

FIG. 4. Angular dependence of the upper critical field (open circles) at 5.4 K. The solid line is a fit to the data by the Lawrence-Doniach formula. Triangles are critical fields taken at the end of the resistive transition H_{R_N}. Inset: angular dependence of the critical current at $\mu_0 H = 2$ T and $T = 18$ K ($J_0 \sim 5 \times 10^3$ A/cm^{-3}).

only reaches its normal-state value (R_N) for a higher field H_{R_N}. For $H_{R_N}>H>H_{c2}$, the highly non-Ohmic current dependence of the transition has been attributed to surface conductivity by some of us.[9] As shown in Fig. 3, for low current densities ($j=17$, 50, and 170 A/cm^2) the transitions are smooth for both field orientations and, for $H\perp ab$, the onset of finite resistivity even shifts towards higher fields as the current density is decreased. This onset becomes sharp and current independent above 500 A/cm^2 for $H\perp ab$ and 170 A/cm^2 for $H||ab$. The ratio between the fields $H_{R=0}=H_{c2}$ (for $j=500$ A/cm^2) and H_{R_N} (measured at the lowest current) is about 1.8 for $H\perp ab$ and 1.4 for $H||ab$. This ratio remains constant for all temperatures apart from those very close to T_c, where it is affected by the width of the transition ΔT_c.

For $H||ab$, our data are strikingly similar to those obtained by Hempstead and Kim[15] in their pioneering work on surface surperconductivity in $Nb_{0.5}Ta_{0.5}$ and $Pb_{0.83}In_{0.17}$ sheets. The resistive transition can be analyzed as follows: up to some characteristic j value, the onset of finite resistance is sensitive to j due to the existence of a surface sheath which can bear a current keeping the zero resistance above H_{c2}. For higher j values, this sheath is destroyed and the resistance increases sharply at $H=H_{c2}$ to a fraction of R_N which increases with j. For the lowest j values, $H_{R_N}=H_{c3}\approx 1.7H_{c2}$, but the experimental H_{c3}/H_{c2} ratio in Ref. 15 was scattered between 1.6 and 1.96 and even droped down to 1.14 for copper coated Pb-In sheets. Surface superconductivity effects should be negligible for $H\perp ab$, but in our experimental setup, the current and voltage electrodes overlapping the sample go over the vertical side planes, which thus inevitably contribute to surface superconductivity as well.[16] With triangles, we have plotted in Fig. 4 the angular dependence of H_{R_N} at $T=5.4$ K. If surface conductivity is important for $H\perp ab$ and $H||ab$, this effect is diminished for other field orientations and H_{R_N}/H_{c2} drops down to ≈ 1.1. This increase of surface conductivity close to the ab planes can thus account for the cusplike behavior of H_{R_N} at small angles which has been first attributed to H_{c2} by other authors.[17,18]

The anisotropy of H_{c2} in MgB_2 crystals has been measured recently by several authors in a limited temperature range by either magnetometric[19,20] or thermal-conductivity[21] measurements. Those measurements yield results very similar to ours, but it was of fundamental importance to show that the "critical fields" obtained from all those measurements coincide with the thermodynamic transition field deduced from specific-heat measurements. Indeed, in many novel superconductors, fluctuations broaden the transitions from the superconducting to the normal state. One of the most striking phenomenon that has been observed in these systems is the existence of a melting line $T_m(H)$. The presence of this vortex liquid phase above T_m complicates the determination of the upper critical field H_{c2}. For instance, the onset of a finite R in these systems indicates the melting transition, and H_{c2} is shifted to a much higher resistance (close to R_N; see, for instance, Ref. 22). However, fluctuations are not expected to play any significant role in MgB_2 as confirmed by specific-heat measurements. A incorrect criterion for the H_{c2} determination partly explains the discrepancy in the H_{c2} values deduced from transport measurements.[23,17]

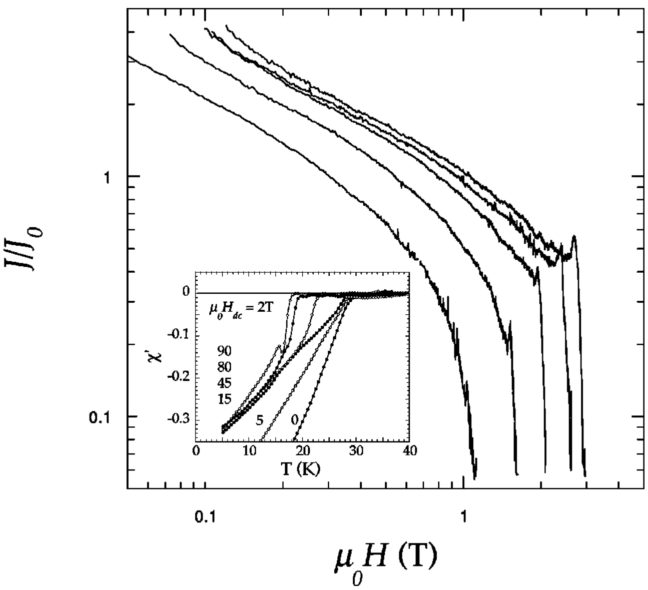

FIG. 5. Magnetic-field dependence of the critical current for $H\perp ab$ at $T=8.5$, 13, 17, 21.5, and 26 K (from top to bottom), showing a peak effect at low temperatures ($J_0\approx 5\times 10^3$ A/cm^3). Inset: temperature dependence of the ac suscetibility at $\mu_0 H=2$ T for different angles between H and the ab plane.

Another interesting phenomenon is the observation of a peak effect in the critical current (J), which shows up in our transport data in the same way as in Ref. 9. Similarly, this increase in J appears as a dip in our ac-susceptibility measurements, which could be observed for $H\perp ab$ in a narrow magnetic-field range $\sim 2-3$ T (see inset of Fig. 5). As recently reported by Pissas *et al.*,[24] this dip is replaced by a sharp drop for lower magnetic fields and has been attributed to a possible melting of the vortex solid,[24] in contradiction with our specific-heat data, which show that the onset of diamagnetism actualy coincides with the H_{c2} line. As shown in Fig. 5, the J value at the peak position—deduced from the susceptibility, following the procedure introduced by Pasquini *et al.*[25]—rapidly decreases for increasing temperature (i.e., for decreasing magnetic field). Similarly, as shown in the inset of Fig. 5, the peak effect also "disappears" in the susceptibility measurements when the magnetic field is turned away from the c axis. However, this peak is still visible in our transport measurements for $H||ab$, suggesting that it could exist for all H and θ values. In MgB_2 this effect most probably reflects the fact that the shear energy of the flux lattice drops towards zero more rapidly than the pinning energy in the vicinity of the normal state, allowing the vortex matter to accomodate the disorder close to the transition more efficiently.[26]

Our J values for $H\perp ab$ are much smaller than those previously obtained in polycrystals.[27] However, as shown in the inset of Fig. 5, if the susceptibility only weakly depends on θ down to $\sim 10°$, it decreases rapidly close to the ab plane, showing that the pinning of the vortices is much more efficient for $H||ab$. The corresponding J values at 18 K are displayed in the inset of Fig. 4, showing that J increases by a factor of about 4 for $H||ab$. A similar behavior has been observed in the critical current deduced from transport measurements in c axis oriented thin films[28] and Eltsev *et al.* recently reported on a rapid drop of the dissipation when the magnetic field is applied parallel to the ab planes in single crystals.[17] A similar cusp in the critical current has been observed in high-T_c cuprates in which vortices are strongly pinning by the weakly superconducting layers between the CuO_2 planes when the field is applied parallel to those planes (so-called intrinsic pinning[29]). However, the origin of the strong pinning of the ab planes in diborides still has to be clarified, given the rather small anisotropy of this material.

In summary, the upper critical field has been deduced from specific-heat, high-magnetic-field transport, and ac-susceptibility measurements for H parallel and perpendicular to the ab planes. $H_{c2}\perp ab$ reveals a conventional temperature dependence with $\mu_0 H_{c2\perp ab}(0) \simeq 3.5$ T, but the parallel critical field has a positive curvature above 20 K and saturates at $\mu_0 H_{c2||ab}(0) \simeq 17$ T. Consequently, the anisotropy factor Γ is temperature dependent, decreasing from ~ 5 at low temperature to ~ 2 close to T_c. The critical current deduced from the susceptibility measurements presents a peak effect for $\mu_0 H \perp ab \sim 2-3$ T and rapidly increases for $H||ab$.

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