PHYSICAL REVIEW B, VOLUME 65, 180505(R)

Temperature dependence and anisotropy of the bulk upper critical field H_{c2} of MgB_2

A. V. Sologubenko, J. Jun, S. M. Kazakov, J. Karpinski, and H. R. Ott
Laboratorium für Festkörperphysik, ETH Hönggerberg, CH-8093 Zürich, Switzerland
(Received 8 March 2002; published 26 April 2002)

The bulk upper critical field $H_{c2}(T)$ of superconducting MgB_2 and its anisotropy are established by analyzing experimental data on the temperature and magnetic-field dependences of the ab-plane thermal conductivity of a single-crystalline sample in external magnetic fields oriented both parallel (H_{c2}^{c}) and perpendicular (H_{c2}^{ab}) to the c axis of the hexagonal lattice. From numerical fits we deduce the anisotropy ratio $\gamma_0 = H_{c2}^{ab}(0)/H_{c2}^{c}(0) = 4.2$ at $T=0$ K. Both the values and the temperature dependences of H_{c2}^{c} and H_{c2}^{ab} are distinctly different from previous claims based on measurements of the electrical resistivity.

DOI: 10.1103/PhysRevB.65.180505 PACS number(s): 74.70.−b, 74.25.Fy, 74.60.Ec

Since the recent discovery of superconductivity in MgB_2 at a critical temperature $T_c \simeq 40$ K,[1] a large number of experimental results on different properties of this compound have been reported in the literature. Most experiments were made using powder or polycrystalline samples. The hexagonal crystal structure of MgB_2, however, is expected to cause pronounced anisotropies in the electrical and magnetic properties, which can unambiguously be probed only by experiments using single crystals. In particular, the upper critical field $H_{c2}(T)$ is an important parameter for characterizing the superconducting state of type-II superconductors.[2–4] For anisotropic materials, such as hexagonal MgB_2, the values of H_{c2} may vary considerably for different orientations of the external magnetic field H. Choosing the field directions either perpendicular or parallel to the c axis, the anisotropy may be expressed by a parameter $\gamma = H_{c2}^{ab}/H_{c2}^{c}$, which, in the most general case, may be temperature dependent. Earlier experimental results, mainly based on measurements of the electrical resistivity $\rho(T,H)$, have resulted in a broad range of values of γ and of extrapolated zero-temperature values of $H_{c2}^{c}(0)$ and $H_{c2}^{ab}(0)$ (for a review, see Ref. 5). Most of these experiments, also on single crystals,[6–8] indicate a positive curvature of $H_{c2}(T)$ in a wide range of temperature below T_c and correspondingly, rather high critical fields at $T=0$. Attempts to explain these features have led to theoretical work suggesting the existence of some soft bosonic modes[2] and even unconventional mechanisms of superconductivity have been considered.[9]

In this paper we present an evaluation of $H_{c2}^{c}(T)$ and $H_{c2}^{ab}(T)$ of single-crystalline MgB_2, based on measurements of the thermal conductivity $\kappa(H,T)$. Complementary results of $\rho(T,H)$, obtained on the same single-crystalline sample, indicate that electrical transport measurements are not well suited to probe the bulk upper critical field $H_{c2}(T)$ of MgB_2. Inspecting the temperature dependences of both $H_{c2}^{c}(T)$ and $H_{c2}^{ab}(T)$ close to T_c, our results indicate that even the zero-field critical temperature $T_c(0)$ of the bulk may be lower than commonly believed up to now. This indicates that, in relation to superconductivity of MgB_2, surface effects must be considered.

The thermal conductivity was measured in the basal plane of hexagonal MgB_2 exposed to varying magnetic fields H, oriented parallel and perpendicular to the basal ab plane with small misalignments of $3.5 \pm 0.5°$ between the field directions and the orientation of the plane. A standard uniaxial heat-flow method, as described in Ref. 10, was used for the $\kappa(H,T)$ measurements. The temperature difference between the two thermometers was about 1% of the absolute average temperature. The measurements of the electrical resistivity $\rho(H,T)$ were made using a four-contact scheme and a dc current of density 50 A/cm^2 in the ab plane with H along the c direction. The investigated single crystal has lateral dimensions of $0.5 \times 0.17 \times 0.035$ mm^3 and was grown employing a high-pressure cubic anvil technique as described elsewhere.[11]

Low-temperature $\rho(T)$ curves measured in constant external magnetic fields H are presented in Fig. 1 for $T<50$ K. The zero-field resistive superconducting transition at $T_c = 38.1$ K is rather narrow ($\Delta T_c \sim 0.15$ K), but the application of magnetic fields broadens the transition considerably. If the field dependence of the onset of the resistive transition, as illustrated in Fig. 1 for $H=50$ kOe, is plotted in an $[H,T]$ diagram, the curve denoted as H_{ρ}^{c} in Fig. 4 is obtained. These H_{ρ}^{c} data are qualitatively and quantitatively very similar to results previously obtained on single crystals,[6–8] in particular, with respect to the positive curvature of $H_{\rho}^{c}(T)$. In these earlier works $H_{\rho}^{c}(T)$ was associated with $H_{c2}^{c}(T)$.

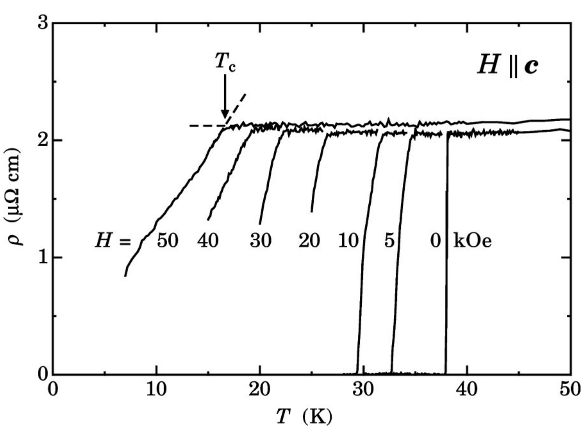

FIG. 1. The low-temperature electrical resistivity of MgB_2 and its magnetic-field dependence for a current in the ab plane. The broken lines for $H=50$ kOe indicate how the onset of the resistive transition defining H_{ρ}^{c} has been established.

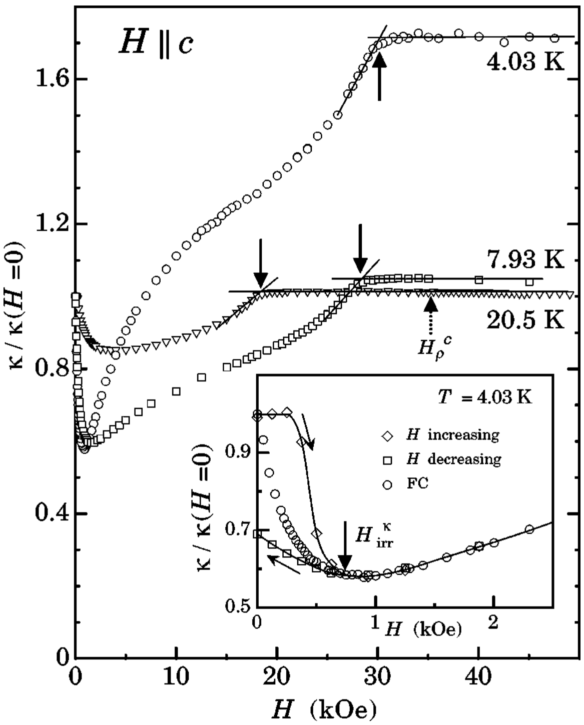

FIG. 2. The magnetic-field dependence of the thermal conductivity $\kappa(H)$ for $H\|c$ at $T=4.03$, 7.93, and 20.5 K. The solid vertical arrows mark H_κ^c, set equal to the upper critical field H_{c2}^c. The dotted vertical arrow denotes H_ρ^c (see Fig. 1). The inset demonstrates the irreversible behavior of $\kappa(H)$ below $H_{\rm irr}^\kappa$ for $T=4.03$ K.

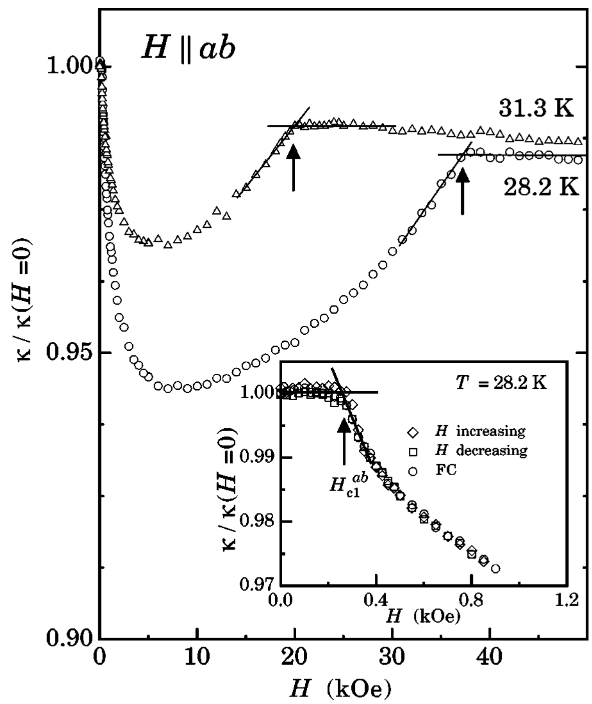

FIG. 3. The magnetic-field dependence of the thermal conductivity $\kappa(H)$ for $H\perp c$ at $T=28.2$ and 31.3 K. The solid vertical arrows mark H_κ^{ab}, set equal to the upper critical field H_{c2}^{ab}. The inset demonstrates the small hysteresis of $\kappa(H)$ at low fields for $T=28.2$ K.

The H dependence of the thermal conductivity was measured at selected constant temperatures in the range between 2 and 50 K and in fields up to 60 kOe. Representative $\kappa(H)$ curves at selected temperatures are displayed in Figs. 2 and 3 for $H\|c$ and $H\|ab$, respectively. As demonstrated in the inset of Fig. 2, a hysteretic behavior of $\kappa(H)$, caused by vortex pinning, is observed in the low-field regime for $H\|c$. In order to avoid ambiguities, each new field setting at a constant temperature was achieved by heating the sample to the normal state above 50 K, and subsequently cooling it to the set temperature in the chosen field. In this way, a smooth variation of $\kappa(H)$, as demonstrated by the open circles in the inset of Fig. 2, was obtained. The field values $H_{\rm irr}^\kappa$, below which the irreversibility is discernible, are rather low. For $T=4.03$ K, e.g., $H_{\rm irr}^\kappa\sim0.7$ kOe. At elevated temperatures and for $H\|ab$ the irreversibilities are reduced, as demonstrated in the inset of Fig. 3.

The curves presented in Figs. 2 and 3 reveal the general features observed at all temperatures below 38.1 K. Starting at $H=0$, κ drops with a steep slope and, after passing through a minimum, increases again until a region of very weak field dependence above some critical field, denoted as H_κ is reached. It is remarkable that, for each temperature, H_κ^c is distinctly lower than H_ρ^c and that no distinct feature of $\kappa(H)$ is observed in the region of H_ρ^c. This is explicitly demonstrated in Fig. 2. With increasing temperature, H_κ decreases towards zero as T approaches T_c. This general $\kappa(H)$ feature is typical of type-II superconductors and can be explained as follows.[12] The thermal conductivity of a superconductor is due to itinerant electrons (κ_e) and phonons ($\kappa_{\rm ph}$). Enhancing H from zero eventually causes the formation of vortices in the bulk of a type-II superconductor. After zero-field cooling, the first vortices form at the lower critical field H_{c1}. Consequentially, some additional scattering of phonons by normal electrons in the cores of the vortices will reduce $\kappa_{\rm ph}$. With further increase in the field the decrease of $\kappa_{\rm ph}$ is compensated by an enhancement of κ_e. Above H_{c2}, in the normal state, the field dependence of both $\kappa_{\rm ph}$ and κ_e is expected to be weak. The overall behavior of the $\kappa(H)$ curves shown in Figs. 2 and 3 reflects these expectations, and as may be seen, $\kappa(H)$ is virtually field independent for $H>H_\kappa$.

A more complete analysis of the $\kappa(H)$ data will be presented in a forthcoming paper.[13] Here, we concentrate on the opportunity that these data allow for a reliable evaluation of the bulk upper critical field $H_{c2}(T)$, which obviously coincides with $H_\kappa(T)$ as derived from our $\kappa(H)$ curves for both field orientations. It may be seen that $H_{c2}^c(T)\equiv H_\kappa^c(T)$ is distinctly different from $H_\rho^c(T)$. The solid line in Fig. 4, representing a general prediction for $H_{c2}(T)$ of a conventional type-II superconductor in the case where the coherence length ξ and the electron mean free path l are of similar magnitude,[14] is in fair agreement with the measured $H_{c2}^c(T)$. It is obvious that $H_\rho^c(T)$ does not follow the same general T dependence. Since thermal conductivity experiments probe the bulk of the sample, it is $H_\kappa^c(T)$ rather than $H_\rho^c(T)$ that ought to be identified as the upper critical field $H_{c2}^c(T)$. Recent magnetization measurements[15] on single crystals of MgB_2 using a torque magnetometer result in values and a temperature dependence of $H_{c2}^c(T)$ consistent with our $H_\kappa^c(T)$ and thus support our conclusion. Employing the equation given by anisotropic Ginzburg-Landau theory $H_{c2}(\theta)=[(\sin\theta/H_{c2}^c)^2+(\cos\theta/H_{c2}^{ab})^2]^{-1/2}$, where θ is the angle between the magnetic field and the ab plane,[16] we estimate the errors in calculating H_{c2} caused by the above-mentioned misalignment of $3.5\pm0.5°$ to be about $3\pm1\%$ for H_{c2}^{ab} and below 0.2% for H_{c2}^c.

As displayed in Fig. 4, at temperatures above about 27 K, H_{c2}^c varies linearly with temperature with a slope $dH_{c2}^c/dT=-1.17$ kOe/K. This behavior leads to an extrapolated zero-field $T_c'=36.6$ K, 1.5 K below T_c obtained from $\rho(T,0)$. Using the equations that are given by the Ginzburg-Landau theory considering anisotropies,[16] $\xi_{ab}(T)=[\Phi_0/2\pi H_{c2}^c(T)]^{1/2}$ and $\xi_{ab}(T)=0.74(1-T/T_c)^{-1/2}\xi_{ab,0}$, where ξ_{ab} is the coherence length in the basal ab plane, we obtain the zero-temperature value of $\xi_{ab,0}=11.8$ nm.

Turning to the temperature dependence of the critical field $H_{c2}^{ab}(T)$ for $H\perp c$, we again note a sizable temperature interval where $H_{c2}^{ab}(T)$ varies linearly with T. This is emphasized by the dotted line in Fig. 4. The slope $dH_{c2}^{ab}/dT=-5.15$ kOe/K gives $\sqrt{\xi_{ab,0}\xi_{c,0}}=5.75$ nm and therefore, the zero-temperature value of the c-axis correlation length $\xi_{c,0}=2.8$ nm. Another important parameter that can be estimated from our $\kappa(H)$ data is the lower critical field H_{c1}^{ab}, as demonstrated in the inset of Fig. 3. In the temperature region between 28 and 35 K, where an evaluation of H_{c1}^{ab} with reasonable accuracy of about $\pm10\%$ was possible, $H_{c2}^{ab}/H_{c1}^{ab}\approx130$. From this ratio, using the equation $H_{c2}/H_{c1}=2\kappa_{\rm GL}^2/\ln\kappa_{\rm GL}$,[16] the parameter $\kappa_{\rm GL}^{ab}$ of the Ginzburg-Landau theory is estimated to be about 13.

As may be seen in Fig. 4, above approximately 33 K, $H_{c2}^{ab}(T)$ deviates from the linear in T variation and, with increasing temperature, approaches zero also at T_c' defined above. This is reflected in the temperature dependence of the anisotropy ratio γ, which seems to decrease with T approaching T_c'. The positive curvature of $H_{c2}(T)$ is typical of strongly anisotropic, layered superconductors[17] and has often been explained in terms of the Lawrence-Doniach model,[18] which treats a layered superconductor as a stacked array of weakly coupled two-dimensional superconducting sheets. Various other theoretical models have been proposed to explain this feature (for a critical review see, e.g., Ref. 19). At this point we cannot commit ourselves to any of these models. It is important, however, that the anomaly is absent for $H\|c$ and small and restricted to a rather narrow temperature region for $H\perp c$. At lower temperatures, with decreasing temperature the anisotropy ratio $\gamma(T)$ tends to a constant value and is approaching $\gamma_0=H_{c2}^{ab}(0)/H_{c2}^c(0)=\xi_{ab,0}/\xi_{c,0}=4.2$.

The extrapolation to zero temperature gives $H_{c2}^c(0)\approx31$ kOe, a considerably lower value than is typically claimed for MgB_2.[5] Exceptions are the reports of Refs. 20 and 21. The rather low value of $H_{c2}^c(0)$ and the observation of a Helfand-Werthamer-type[14] temperature dependence of

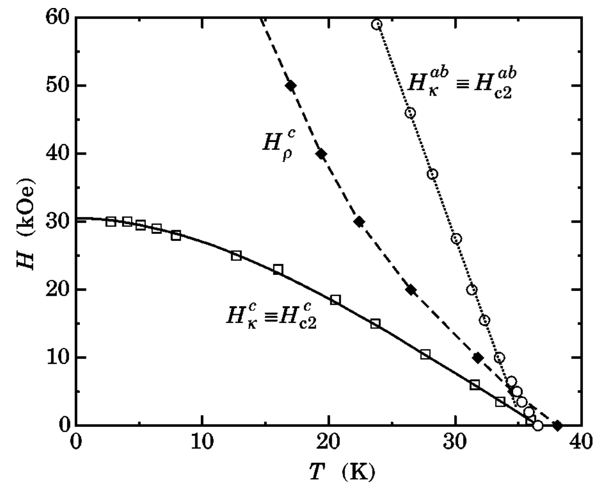

FIG. 4. Critical fields H_ρ and $H_\kappa\equiv H_{c2}$, as determined from electrical-resistivity and thermal-conductivity measurements, respectively. The solid line is compatible with calculations due to Helfand and Werthamer (Ref. 14). The dashed and dotted lines are to guide the eye.

$H_{c2}^c(T)$ have important consequences for possible models of the superconducting state of MgB_2.

Our result obviously questions the intrinsic nature of $H_{c2}(T)$ derived from measurements of $\rho(T)$. Since the resistive transition is not manifest in $\kappa(T)$, which may be considered as a bulk property, $H_\rho(T)$ must correspond to a minor fraction of an additional phase (or phases) with enhanced H_{c2} and T_c. The spatial extension of this phase is, however, large enough to short circuit the electrical current path and produce a narrow superconducting transition at a temperature T_c higher than the bulk transition temperature T_c'. Based on an analysis of their magnetization and ac susceptibility data on polycrystalline samples, the authors of Ref. 22 came to a similar conclusion. The most likely origin of the second phase with enhanced superconducting parameters seems to be related to surface effects. A considerable enhancement of the electron density of states near the Fermi level and, therefore, an enhanced trend to superconductivity at the surface of MgB_2 have been predicted,[23–25] in agreement with our observations.

In conclusion, we observe a striking disagreement in the values and the temperature dependences of the upper critical field H_{c2}^c of MgB_2 evaluated from results of electrical- and thermal-conductivity measurements on the same sample. The shape of $H_{c2}^c(T)$ as established by $\kappa(H)$ with $H\|c$ does not reveal an anomalous positive curvature near T_c and therefore no exotic mechanism needs to be involved to explain the upper critical field, at least not for the bulk. Our data also indicate that the bulk transition temperature T_c' is lower than T_c obtained from results of $\rho(T)$ measurements.

We acknowledge useful discussions with I.L. Landau, M. Angst, and R. Monnier. This work was financially supported in part by the Schweizerische Nationalfonds zur Förderung der Wissenschaftlichen Forschung.

[1] J. Nagamatsu *et al.*, Nature (London) **410**, 63 (2001).
[2] S.V. Shulga *et al.*, cond-mat/0103154 (unpublished).
[3] C. Joas, I. Eremin, D. Manske, and K. H. Bennemann, Phys. Rev. B **65**, 132518 (2002).
[4] S. Haas and K. Maki, Phys. Rev. B **65**, 020502(R) (2002).
[5] C. Buzea and T. Yamashita, Semicond. Sci. Technol. **14**, R115 (2001).
[6] S. Lee *et al.*, J. Phys. Soc. Jpn. **70**, 2255 (2001).
[7] M. Xu *et al.*, Appl. Phys. Lett. **79**, 2779 (2001).
[8] K.H.P. Kim *et al.*, cond-mat/0105330 (unpublished).
[9] G.M. Zhao, cond-mat/0111260 (unpublished).
[10] K. Giannò *et al.*, Phys. Rev. B **62**, 292 (2000).
[11] J. Karpinski *et al.* (unpublished).
[12] W.F. Vinen, E.M. Forgan, C.E. Gough, and M.J. Hood, Physica A **55**, 94 (1971).
[13] A. V. Sologubenko *et al.* (unpublished).
[14] E. Helfand and N.R. Werthamer, Phys. Rev. **147**, 288 (1966).
[15] M. Angst *et al.*, cond-mat/0112166 (unpublished).
[16] M. Tinkham, *Introduction to Superconductivity*, 2nd ed. (McGraw-Hill, New York, 1996).
[17] J.A. Woollam, R.B. Somoano, and P. O'Connor, Phys. Rev. Lett. **32**, 712 (1974).
[18] W.E. Lawrence and S. Doniach, in *Proceedings of the 12th International Conference on Low Temperature Physics, Kyoto, 1970*, edited by E. Kanda (Keigaku, Tokyo, 1971), p. 361.
[19] B. Brandow, Phys. Rep. **296**, 1 (1998).
[20] F. Simon *et al.*, Phys. Rev. Lett. **87**, 047002 (2001).
[21] S.L. Bud'ko, V.G. Kogan, and P.C. Canfield, Phys. Rev. B **64**, 180506(R) (2001).
[22] G. Fuchs *et al.*, Solid State Commun. **118**, 497 (2001).
[23] I.G. Kim, J.I. Lee, B.I. Min, and A.J. Freeman, Phys. Rev. B **64**, 020508(R) (2001).
[24] E. Bascones and F. Guinea, Phys. Rev. B **64**, 214508 (2001).
[25] V.M. Silkin, E.V. Chulkov, and P.M. Echenique, Phys. Rev. B **64**, 172512 (2001).