

Angle-resolved magnetotransport studies in anisotropic MgB₂ single crystals

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We report the angle-resolved magnetotransport measurements on MgB₂ single crystals that exhibit moderate anisotropy (γ) in upper critical fields with $\gamma=2.6\pm 0.1$. Unusual “kink” features in resistivity are observed, which appear most clearly for field parallel to the c axis. We discuss the origin of the “kink” features in relation with the vortex-lattice melting and the recently proposed model of two-gap superconductivity. The influences of anisotropy on superconducting properties including the kink features are also demonstrated.

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INTRODUCTION

The recent discovery of superconductivity in magnesium diboride (MgB₂) (Ref. 1) at 39 K presents a new possibility for significant applications²⁻⁷ at higher temperature. However, many critical issues relevant both for practical applications and fundamental research remain unresolved. One of such issues of primary importance is to what degree this superconductor is anisotropic, since MgB₂ consists of alternating B and Mg sheets. A lesson we have learned from extensive studies on high-temperature superconductors is that moderate anisotropy can suppress both upper critical and irreversibility fields by the application of magnetic field. As a matter of fact, the actual anisotropic nature of MgB₂ and its influences on various physical properties can provide a baseline for further applied research. In addition, the fundamental issues related to the recently proposed⁸ and experimentally observed⁹⁻¹² two-gap superconductivity are unclear; reason being the unavailability of high-quality single crystals until now.

There is growing evidence for the two-gap energy states from photoemission spectroscopy,⁹ specific heat,¹² and tunneling spectroscopy^{10,11} experiments in bulk samples. Although multiple gap superconductivity was predicted¹³ long ago, it has never been observed in conventional superconductors, except for very pure Nb, Ta, V (specific heat), and Nb-doped SrTiO₃ in its two-gap spectrum.¹⁴ One of the probable reasons is their large coherence length that has made the multiple spectra unrealistic, being difficult to realize the clean limit condition. The electronic structure of MgB₂ is rather complicated; particularly the Fermi surface (FS) consists of both two-dimensional (2D) cylindrical sheets perpendicular to the z direction and 3D-tubular network. Recent calculations⁸ of the phonon spectrum suggest that the manifestation of high T_c in MgB₂ is due to scattering between two pairs of sheets of the FS. This raises the possibility⁸ of having two distinct gaps. However, several issues remain unexplained, especially the behavior of two-gap-like feature in electrical transport and associated anisotropy of MgB₂ single crystal.

In this paper we show from the angle-resolved transport measurements that MgB₂ single crystals exhibit moderate

anisotropy (γ) in upper critical fields with $\gamma=2.6\pm 0.1$ and unusual “kink” features in resistivity. It is clearly demonstrated that the “kink” feature persists down to the lowest temperature at high field for field parallel to the c axis and not for field parallel to the ab plane. We discuss the origin of this feature in relation with vortex-lattice melting and two-gap superconductivity.

EXPERIMENT

Shiny golden yellow-colored single crystalline MgB₂ platelet samples of dimension $400\times 100\times 40\ \mu\text{m}^3$ were extracted from the highly dense bulk samples synthesized under high pressure using high purity starting materials by a similar technique as described earlier.⁴ The x-ray diffraction reveals that the MgB₂ bulk sample containing the used crystallites is single phase. The resistively measured superconducting transition temperature (T_c) is at ~ 38.2 K with $\Delta T_c \sim 0.2$ K, suggesting high quality and homogeneity of the crystal. In addition, the estimated normal-state resistivity ratio in zero field (RRR), $\rho(300\text{ K})/\rho(40\text{ K})$, is ≈ 5.8 [inset Fig. 1(b)] with $\rho(40\text{ K}) \approx 1.0\ \mu\Omega\text{ cm}$, suggesting the present MgB₂ crystal in the clean limit as reported by recently on similar crystals.¹⁵ The actual measurement of geometry of such a small crystal can cause inevitably large error in the determination of absolute of ρ as much as 20% of the actual value. The RRR in the present crystal is smaller than that of best polycrystalline samples,^{2,5} but similar to other reports on single crystals.^{16,17} Although the large difference in both types of samples is not understood yet, it may be related to some type of contamination that is not detected by x-ray diffraction. We have measured two crystals of similar size from the same batch, essentially showing the same behavior. As T_c in the present crystal is about 1 K lower than that for good powders, the observed RRR is moderate. The normal-state resistivity roughly obeys the $\rho(T)=\rho_0+\rho_1 T^\alpha$ power law with $\alpha\approx 3$ and is consistent with report on MgB₂ wire.⁵ Angle-resolved resistivity $\rho(\theta)$ was measured using standard four-probe ac technique with the angle resolution of less than 0.1° . The angle θ between the crystal axis and the magnetic

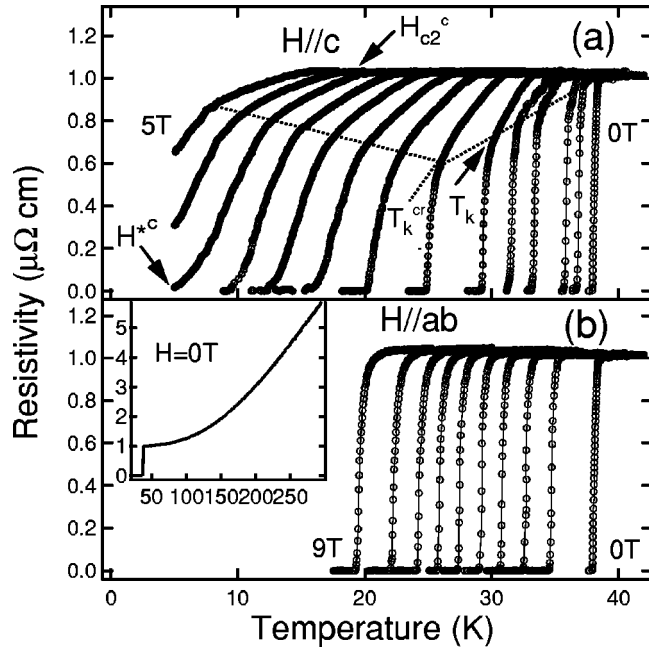


FIG. 1. (a) Temperature dependence of the in-plane resistivity of MgB₂ single crystal for the $H//c$ axis at $H=0, 0.1, 0.2, 0.5, 0.7, 1$ T and with a 0.5 T interval up to 5 T. The dashed lines show the decreasing tendency of the “kink” height T_k from a critical temperature T_k^{cr} . The arrows indicate H_{c2}^c and H^{*c} . (b) T vs ρ for $H//ab$ at $H=0$ up to 9 T with 1 T interval. The current density was 10 A/cm². The inset in (b) shows the normal state ρ for $H=0$ T.

field (H) is varied from 130° to -20° , where 90° correspond to $H//ab$ plane and 0° to $H//c$ axis of the crystal. The contact resistance was found to be typically less than 1 Ω .

RESULTS AND DISCUSSION

In anisotropic type-II superconductors, the magnetic field destroys superconductivity at the upper critical fields H_{c2}^{ab} and H_{c2}^c for field applied $H//ab$ planes and $H//c$ axis of the crystal, respectively. The T_c varies between two field orientations, depending on the anisotropy $\gamma = H_{c2}^{ab}/H_{c2}^c$. Figures 1(a) and 1(b) show the temperature dependent resistivity $\rho(T)$ curves of MgB₂ crystal at several fields applied both for $H//c$ and $H//ab$ planes. The experiments show three primary results. First, the resistive transition under magnetic field for $H//c$ is significantly suppressed compared to $H//ab$, indicating a moderate anisotropy in upper critical fields H_{c2}^c and H_{c2}^{ab} for respective orientations. Second, for $H//c$ the significant broadening of the transition causes large suppression of the irreversibility field H^{*c} where bulk supercurrent disappears. Third, at all fixed field values $\rho(T)$ exhibits a “kink” at a temperature T_k only for $H//c$ where resistivity falls very sharply up to $H \leq 1.5$ T and changes slope at higher fields.

We now provide a direct experimental manifestation of anisotropy in Figs. 2(a)–2(d) from our angle-resolved transport measurements. Two excitation currents (10 and 50 A/cm²) were used mainly to obtain sharper transition at lower current and to observe the influences of higher excita-

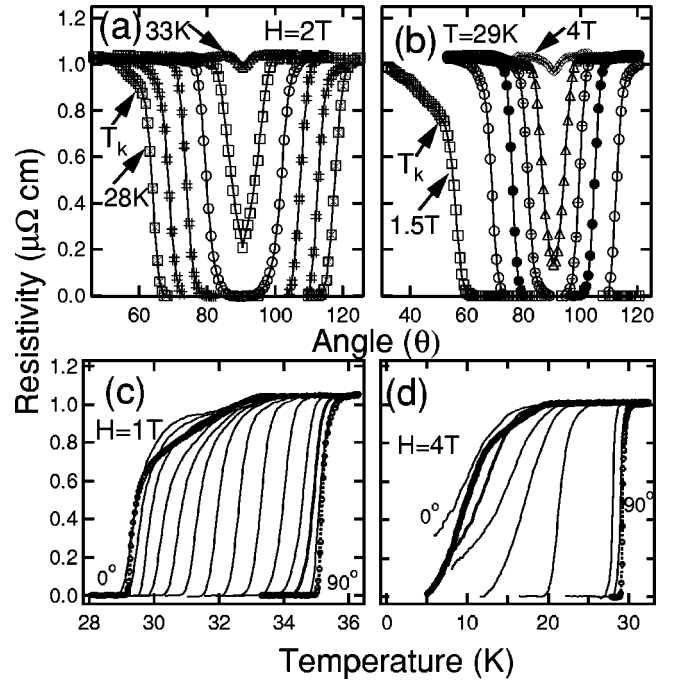


FIG. 2. Upper panel: Isothermal resistivity versus angle θ (a) at $H=2$ T for $T=28$ to 33 K with a T interval of 1 K, (b) for $T=29$ K at a fixed field of 1.5 to 4 T at a field interval of 0.5 T. Lower panel: T vs ρ (c) at $H=1$ T, varying θ from 90° to 0° at an interval of 7.5° and (d) at 4 T at an interval of 15°. The current density was 50 A/cm². The dashed lines with open circles shown in (c) and d for $\theta=0^\circ$ and 90° are for lower current of 10 A/cm². No heating effects were noticed due to low contact resistance ($<1 \Omega$).

tion current on the resistive transitions. The angle-dependent resistivity $\rho(\theta)$ shows a remarkably sharper transition on decreasing both temperature and field when θ is away from 0° [see Figs. 2(a) and 2(b)], indicating a modest anisotropy. Note that the cup-shaped feature in $\rho(\theta)$ centered at $\theta=90^\circ$ is the first visual evidence for the anisotropic nature of MgB₂ single crystal. Using the scaling equation for the field-dependent upper critical field $H_{c2}(\theta) = H_{c2}^{ab}/\sqrt{\varepsilon(\theta)}$ with $\varepsilon(\theta) = 1 + (\gamma^2 - 1)\cos^2\theta$, the estimated γ value becomes 2.6 ± 0.1 for $H \leq 3.5$ T. This suggests that the broadening of T_c at higher magnetic field for $\theta < 90^\circ$ is due to weak flux pinning and paucity of pinning centers in MgB₂. Further evidence can be seen from the large broadening of $\rho(\theta=0^\circ)$ for higher excitation current [see Figs. 2(c)–2(d)] due to the influence of Lorentz force, especially at higher fields. The Lorentz-force driven dissipation is considerably high at high fields and consequently the field-induced transition broadens, and is pushed down to the low-temperature regime.

The most unusual and remarkable feature in Figs. 1(a) and 2 is the presence of “kink” at T_k . At a first glance, it is tempting to suspect that T_k is probably associated with the melting of the Abrikosov vortex lattice. A sharp drop in resistivity below T_k [shown in Fig. 1(a) for $H//c$] suggests the possibility of vortex-lattice melting scenario. The flux-flow type behavior in the current-voltage curves accompanied by Ohmic behavior above and non-Ohmic below the transition

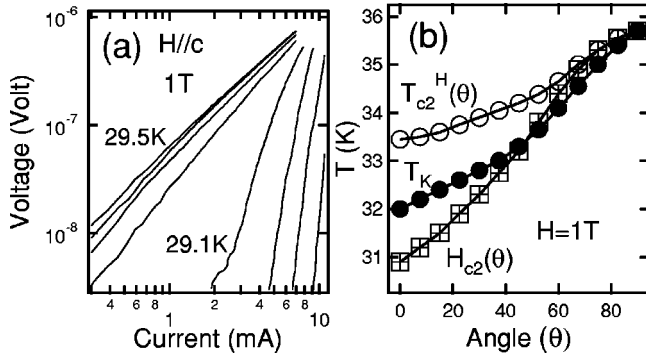


FIG. 3. (a) Logarithmic plot of current-voltage curves at $H = 1$ T for $H//c$ axis of the MgB_2 crystal in the T range 29.5 to 28.8 K (0.1 K step). (b) The angular dependence of $T_{c2}^H(\theta)$, T_k , and $H_{c2}(\theta)$ for $H = 1$ T is shown. The difference in $T_{c2}^H(\theta)$ and $H_{c2}(\theta)$ is basically due to the criterion (extrapolation of the resistivity curves of the second slope below T_k) fixed for the determination of $H_{c2}(\theta)$.

as shown in Fig. 3(a) also supports the melting of flux lattice¹⁸ as observed in $\text{YBa}_2\text{Cu}_3\text{O}_7$. The resistivity height at T_k at 1 T is about, $\rho(T_m) \approx 0.6 \rho(\text{normal})$, which is much larger than that of ≈ 0.15 of $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals undergoing melting,¹⁸ can be understood by considering smaller anisotropy in MgB_2 . However, the absence of the “kink” for $H//ab$ certainly raises question regarding the origin of “kink,” which cannot be simply attributed to the sample inhomogeneity considering the systematic angular dependence of resistivity as shown in the following.

As electrical transport is related with the density of states (DOS) at the FS, the unusual behavior may be related to the change of DOS at the FS. One convincing way to explain the unusual resistivity feature may be the existence of proposed two kinds of gaps in MgB_2 .⁸ This model is based on the coexistence of a two-dimensional (2D) FS (p_{x-y} orbitals) perpendicular to the z direction and 3D one (p_z and antibonding bands), predicting a larger and a smaller energy gaps, respectively. Our results show that T_k is smeared by temperature for $T > 25$ K and $H \leq 1.5$ T until $T \approx T_c$ [see Fig. 1(a)], suggesting that both small and large gaps close near the zero-field T_c with the small gap closing more rapidly near T_c . The combination of two gaps is better resolved in moderate magnetic fields (1 to 2 T). The gap at high temperature reduces gradually with increasing magnetic field up to 5 T, while the gap at low temperature broadens due to pair-breaking effects, confirming that two gaps coexist from T_c in zero-field down to 5 K in a field of 5 T with a single T_c . For a two-band, two-gap superconductor, interband coupling ensures that the two gaps open at the same T_c .⁸ The resistivity reveals the transition to the superconducting state in two stages, associated with two energy gaps: the first, a partial transition until $T = T_k$, the second, which completes the transition, is probably associated with the second smaller energy gap. The absence of any second or impurity phases in x-ray diffraction suggests that sample inhomogeneity may not be the reason for the observation of the above nontrivial features in resistivity over the whole field range. In addition, the energy gap distribution cannot explain our results be-

cause of absence of any anomaly due to effects of impurity scattering, thus ruling out the argument based on inhomogeneity. Hence, two-gap scenario remains one of the attractive and plausible models for the explanation. On the other hand, we have not observed any such anomaly in resistance on similar MgB_2 crystal with slightly broader transition in our previous report.¹⁵ However, Lee *et al.* have reported very similar resistive transition for $H//c$ in their MgB_2 single crystal.

The coexistence of two-gap superconductivity may originate from the interband anisotropic coupling, especially due to layered structure. The angle-dependent resistivity results shown in Fig. 2 clearly reveal a systematic evolution of T_k when field is tilted away from $H//ab$ plane, elucidating the dominating influence of anisotropy. The gradual emergence of T_k when θ is tilted away from 90° and exhibiting a pronounced “kink” at $\theta = 0^\circ$ are distinctly seen in Figs. 2(c) and 2(d). The anisotropic nature of both onset of first transition $T_{c2}^H(\theta)$, T_k and $H_{c2}(\theta)$ is highlighted for $H = 1$ T in Fig. 3(b), confirming that the two-gap feature is mainly associated with the B - B plane for the $H//c$ axis. The difference in $T_{c2}^H(\theta)$ and $H_{c2}(\theta)$ is obvious, mainly due to the criterion (extrapolation of the resistivity curves of the second slope below T_k) fixed for the determination of $H_{c2}(\theta)$. Furthermore, the results of Fig. 1 for $H//c$ seem to indicate qualitatively that magnetic field separates the two-gap feature distinctly with different field dependencies. This is consistent with the unusual, however broad, field-induced temperature dependence of the electronic contribution to specific heat in bulk samples.¹² Two-gap behavior was better demonstrated close to the bulk T_c by point-contact spectroscopy when a magnetic field is applied.¹¹ In the two-gap scenario, resistive onset of superconductivity and the “kink” corresponds to the opening of large and small gaps, respectively. However, the temperature dependence of magnetization for the same single crystal under high magnetic field shown in Fig. 4 is at odd in this respect. For each field orientation, only one onset of diamagnetism is observed, which roughly corresponds to the H^* for each direction. This behavior of magnetization is also not consistent with the vortex-lattice melting scenario. In high-temperature superconductors, the sudden decrease in resistivity accompanies the step in equilibrium magnetization. However, the step occurs below the onset of diamagnetism where appreciable diamagnetic signal is established. Obviously more extensive studies are necessary to resolve the mystery related to the origin of the “kink” features in resistivity when a magnetic field is applied. Furthermore, no theoretical models exist to the best of our knowledge to explain the two-gap superconductivity from the magnetoresistance.

Next we address the magnetic field-temperature phase diagram summarized in Fig. 5. The upper critical fields exhibit pronounced anisotropy with anisotropy ratio $\gamma = H_{c2}^{ab}/H_{c2}^c \approx 2.6 \pm 0.1$, which is similar to the reported values.^{16,17} The anisotropy ratio can be over estimated depending on the shape of the resistivity curves¹⁵ and criteria for the determination of H_{c2} . Our measured γ value is larger than that of other samples of different forms^{19,20} ranging from 1.1 to 2, but smaller than the estimation by CESR,²¹

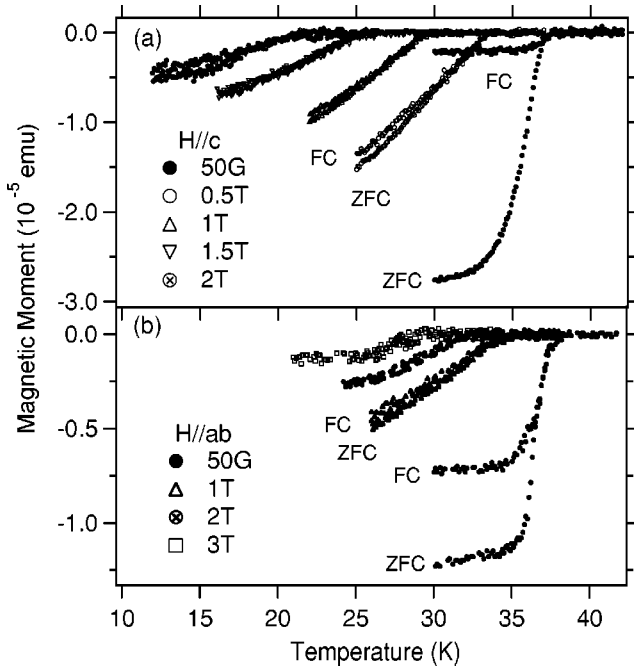


FIG. 4. Temperature dependent zero-field-cooled and field-cooled magnetization curves at various applied fields for the (a) $H//c$ axis and (b) $H//ab$ plane of the MgB_2 crystal.

$\gamma=6$ to 9. We mention that our analysis of angle dependence of H_{c2} (Fig. 2) show a moderately good fitting up to $\gamma = 3.5$ using anisotropic Ginzburg-Landau (GL) model. Both irreversibility field H^* and H_{c2} measured using a SQUID magnetometer (Fig. 4) for both directions show similar anisotropy (see Fig. 5). H^* values determined from magnetization are remarkably consistent with that of the resistivity measurements. However, H_{c2} values measured magnetically show a slight discrepancy (slightly lower value compared to the resistivity). This is mainly due to the very small value of the diamagnetic moment of such a small sample which poses a problem for the accurate determination of H_{c2} values. A significant suppression of the irreversibility field H^* occurs for $H//c$, giving $H^{*c}(T) \approx 0.55H_{c2}^c(T)$. By contrast, for $H//ab$ we found $H^{*ab}(T) \approx 0.93H_{c2}^{ab}(T)$. We estimate $H_{c2}^c(0) \approx 6.1$ T and $H_{c2}^{ab}(0) \approx 15.6$ T from the slopes $dH_{c2}^c(T)/dT \approx -0.22$ T/K and $dH_{c2}^{ab}(T)/dT \approx -0.56$ T/K, assuming the extrapolation formula for an isotropic s -wave superconductor²² $H_{c2}(0) = 0.73T_c[-dH_{c2}(T)/dT]$. Using the anisotropic GL model, $H_{c2}^c(0) = \phi_0/(2\pi\xi_{ab}^2\xi_c)$, and $H_{c2}^{ab}(0) = \phi_0/(2\pi\xi_{ab}\xi_c)$, ϕ_0 is the flux quantum, the GL coherence lengths $\xi_c(0)$ and $\xi_{ab}(0)$ are estimated to be ~ 2.8 and ~ 7.2 nm, respectively. Such short and anisotropic coherence lengths and multiple FS can be responsible for the appearance of two-gap energy feature in MgB_2 in the clean limit. The first remarkable observation of anisotropic “kink” features in electrical transport, that is consistent with that of other techniques,^{9–12} especially from the thermodynamic evidence of the specific heat¹² and Raman spectroscopy,²³ argue

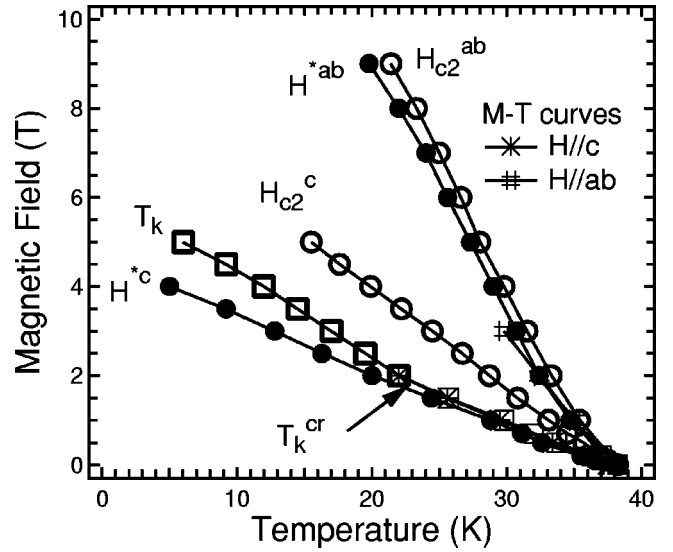


FIG. 5. Temperature-magnetic field phase diagram of a MgB_2 single crystal, displaying upper critical (H_{c2}^c, H_{c2}^{ab}), irreversibility field (H^{*c}, H^{*ab}) anisotropy, and irreversibility from magnetization measurements for the $H//c$ axis and $H//ab$ plane. The field dependence of T_k for the $H//c$ axis exhibiting a crossover point is shown by squares. The onset of diamagnetism for two field orientations measured by SQUID magnetometer is also plotted.

persuasively for the existence of two-gap-like superconductivity and a high degree of crystalline perfection in our MgB_2 single crystal. It is needless to say from the early history of cuprate high-temperature superconductors, that we cannot stress the importance of reliable single-crystal data too much, even after unusual feature has been reported on polycrystalline samples. However, the difference in anisotropy and resistive transition in different MgB_2 single crystals and polycrystals remains unclear, and calls for further investigation.

CONCLUSION

In conclusion, we have demonstrated from the angle-resolved magnetotransport measurements that MgB_2 single crystals exhibit moderate anisotropy in upper critical fields with $\gamma = 2.6 \pm 0.1$. A remarkable appearance of unusual “kink” features was observed in resistivity and the influence of anisotropy on the “kink” features and resistive transitions including the large suppression of the irreversibility fields are demonstrated. We interpret the “kink” features in relation with vortex-lattice melting and the recently proposed model of two-gap superconductivity. Our results thus lead to the conclusion that MgB_2 provides immense interest for basic research as well as applied research.

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