Disorder-induced phase transition of vortex matter in MgB₂

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Measurements of single crystal MgB_2 with torque magnetometry in fields up to 90 kOe reveal a sharp peak in the irreversible torque at about $0.85 H_{c2}$. In the region between peak onset and maximum, pronounced history effects occur. Angle and temperature dependence of the characteristic peak fields track those of *Hc*2. The features observed suggest that the peak marks a disorder-induced phase transition of vortex matter between a quasiordered Bragg glass and a highly disordered phase.

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The new superconductor MgB_2 is considered to have great potential for applications, and a lot of research activity has concentrated on this compound.¹ Much light was shed on the superconducting mechanism; there is mounting evidence that MgB_2 is a two-band superconductor with a substantial difference between the superconducting gaps of the two bands. $1-4$ About the superconducting phase diagram, however, less is known. Most ''phase diagrams'' published contain only the upper critical fields H_{c2} . Here, the situation was clarified insofar as later bulk measurements all find a pronounced anisotropy γ of H_{c2} , decreasing with increasing temperature,5–9 although there are still discrepancies of the exact $\gamma(T)$ dependences reported. MgB₂ is, particularly concerning the importance of thermal fluctuations and the value of $\kappa = \lambda/\xi$, intermediate between the high- T_c cuprates and low- T_c superconductors. Studying the vortex matter phase diagram of MgB_2 may thus help in understanding the phase diagrams of various superconductors in a unified way.

From the study of cuprate superconductors is known that the *H*-*T* phase diagram contains more transition lines than the upper and lower critical fields. Identified were, for example, a melting transition between a quasiordered vortex lattice, called Bragg glass, and a disordered vortex fluid,¹⁰ as well as an order-disorder transition between the Bragg glass and a highly disordered, glassy phase. $11-13$ The latter transition¹⁴ has been observed also in low- T_c superconductors, such as $NbSe₂$, ^{15,16} and even in the elemental superconductor $Nb₁¹⁷$ but not in ultrapure Nb crystals.¹⁸ This transition is generally associated with a peak in the critical current density j_c and pronounced history effects.

In single crystals of MgB_2 , a quasiondered vortex structure has been observed in low fields by scanning tunneling spectroscopy,¹⁹ showing that at least under some conditions a Bragg glass is the stable vortex phase. Since, by tuning the amount of quenched random pointlike disorder, the stabilization of a highly disordered phase can always be favored, an order-disorder transition in fields below H_{c2} should be observable in $MgB₂$ as well, at least for certain impurity concentrations. Although a phase transition distinct from H_{c1} or H_{c2} has not been suggested yet in MgB₂, a peak effect, and accompanying history effects have been observed in transport measurements for $H||c|^9$

Here, we report the observation of a pronounced, sharp peak effect (PE) by torque magnetometry in fields close to, but clearly distinct from, H_{c2} . A minor hysteresis loop (MHL) study shows pronounced history effects in the region between the onset and the maximum of the peak. Angle and temperature dependence of the characteristic fields is reported, and we propose a phase diagram for $MgB₂$.

The measurements were performed on a high-quality single crystal of MgB_2 , sample *B* of Ref. 5. The *T* dependence of the magnetization (upper inset of Fig. 1) shows a sharp $(0.3 \text{ K with a } 10-90\%$ criterion) transition to the superconducting state at 38.2 K, indicating a high quality of the crystal. Measurements to study the PE were carried out with the torque option of a Quantum Design 9T PPMS. Measurement runs consisted in varying the field H at fixed angle θ

FIG. 1. Torque τ/H vs field *H* at 14 K and 77.5 deg. The direction of the field change is indicated by thick arrows. The irreversibility field H_{irr} and the onset and maximum fields H_{on} and H_{max} of the PE for the *H* increasing ($\hat{ }$) and decreasing ($\hat{ }$) branch are marked. Also shown are some of the MHL's (see text) measured, labeled A – D . Upper inset: $M(T)$ curve in the transition region, in a field $H\|c$ of 1 Oe, zero-field cooled (\bullet) and field cooled (\circ). Lower inset: τ/H vs *H* of the curve in the main panel, for the whole field range.

FIG. 2. $(a),(b),(c)$ Magnification of MHL's *A*, *B*, and *D* also displayed in Fig. 1. MHL's started from the field increasing branch of the full hysteresis loop are shown as dotted lines, while those started from the field decreasing branch are shown as full lines. (d) Width of the hysteresis of MHL *B* started from the field increasing/ decreasing (\bigcirc/\bullet) branch of the full hysteresis loop, as a function of cycling.

between \vec{H} and the *c* axis of the crystal, and recording the torque $\vec{\tau} = \vec{m} \times H$, where \vec{m} is the magnetic moment of the crystal.

One of the curves measured is shown in the lower inset of Fig. 1. For better comparison with magnetization curves, τ/H vs *H* is shown. The main panel shows a magnification of the PE region. The peak is well pronounced and very sharp. Various characteristic fields are indicated: The maximum of the peak for field increasing (H_{max}^{\perp}) and decreasing (H_{max}^{\perp}) branch of the hysteresis loop, and the onsets of the peak, H_{on}^{\dagger} and $H_{\text{on}}^{\downarrow}$. The separation of the two onset fields is larger, similar to the case of the cuprate superconductors (see, e.g., Ref. 20). Also indicated is the irreversibility field H_{irr} , where the two branches of the hysteresis loops meet. The peak resembles qualitatively the peaks observed in $NbSe₂$ (Refs. 15) and 21) and $Ceru_2$.²²

To investigate possible history dependences of j_c , we performed several MHL measurements in and around the peak: The field is cycled up and down by a small amount several times, ideally until the loops retrace each other, indicating that the vortex system reached a stable pinned state in the given field.15,22 MHL measured, within full loops, in four different regions of the PE are indicated in the figure (*A*–*D*).

Torque τ/H values of MHL *A* [Fig. 2(a)] vary significantly as the MHL is cycled through repeatedly. Partly, this may be explained by relatively strong normal relaxation processes. However, a pronounced difference can be seen between MHL started from the field increasing (H^{\dagger}) branch of the full hysteresis loop (FHL), and the one started from the field decreasing (H^{\downarrow}) branch. The latter has a significantly higher width initially. This effect can be explained by a dif-

FIG. 3. Angle dependence of various characteristic fields at 18 K. Shown are the upper critical field H_{c2} (\diamond , from Ref. 5), the irreversibility field H_{irr} (\bullet) (Ref. 25), the peak maximum field $H_{\text{max}}(\triangle)$, and the peak onset field $H_{\text{on}}(\blacktriangledown)$. Full lines are fits of the theoretical $H_{c2}(\theta)$ dependence (Ref. 5). Dashed lines are guides for the eye. Inset: Angle dependence of reduced [divided by $H_{c2}(\theta)$] characteristic fields.

ference in the vortex configuration between H^{\uparrow} and H^{\downarrow} in the region of MHL *A*. In the configuration on H^{\downarrow} , j_c (proportional to the width 23 of the MHL) is higher, i.e., the vortices are pinned stronger. Repeated cycling causes the width of the MHL started from H^{\downarrow} to approach the one started from H^{\uparrow} , indicating that the vortex configuration on H^{\downarrow} is only metastable. History effects are even more pronounced for MHL *B* [Fig. 2(b)]. Here, the initial H^{\uparrow} branch of the MHL started from the H^{\downarrow} branch of the FHL (full line indicated by arrows) is clearly below the H^{\uparrow} branch of the FHL (thick dashed), indicating larger hysteresis. This behavior contradicts Bean's critical state model, 24 where the hysteresis of partial hysteresis loops can never be higher than the one of the full loop. We point out that simple relaxation effects cannot account for this specific effect. It can be explained by the vortex configuration on the H^{\downarrow} branch of the FHL (where the MHL was started) having a higher j_c than the vortex configuration on the H^{\dagger} branch. The variation of the hysteresis width with cycling $[Fig. 2(d)]$ demonstrates the metastable nature of the vortex configuration on the H^{\downarrow} branch of the FHL, while the vortex configuration of the H^{\uparrow} branch of the FHL is stable, or close to stable. In contrast, no clear deviations in the cycling behavior between H^{\dagger} and H^{\dagger} branch started MHL's are visible for MHL C and MHL D [Fig. 2(c)], as well as for a MHL measured in the region around 68 kOe (not shown).

In summary, between H_{on}^{\downarrow} and H_{max}^{\parallel} , pronounced history effects occur. They can be accounted for by the coexistence of a metastable high-field vortex configuration with high pinning and a stable low-field, low pinning configuration. Above H_{max}^{\perp} and below H_{on}^{\perp} , no significant history effects are observed, indicating that there is only one vortex con-

FIG. 4. Phase diagram of $MgB₂$ single crystal at an angle of 77.5 deg between the *c* axis of the crystal and the applied field: The temperature dependence of the characteristic fields H_{c2} , H_{max} , and H_{on} is given. They mark boundaries between the normal state and the various phases of vortex matter. The irreversibility field H_{irr} is also shown. The inset shows the *T* dependence of the characteristic fields scaled by H_{c2} .

figuration, which is stable. The larger hysteresis width of MHL started from H^{\downarrow} indicates pinning in the configuration stable above $H_{\text{max}}^{\parallel}$ to be stronger than pinning in the configuration stable below $H_{\text{on}}^{\downarrow}$.

The variation of the peak onsets and maxima with angle at 18 K is shown, together with $H_{c2}(\theta)$ and $H_{irr}(\theta)$,²⁵ in Fig. 3. Since the visibility of the peaks is diminished at higher temperatures, onsets and maxima were determined from $\Delta \tau(H) = \tau(H^{\dagger}) - \tau(H^{\dagger})$ curves. The characteristic peak fields follow the angular dependence of H_{c2} , as indicated by fits to the theoretical $H_{c2}(\theta)$ dependence according to the anisotropic Ginzburg-Landau theory (see Ref. 5), while the angular scaling of the irreversibility field is less clear. This can be seen also in the inset, displaying the θ dependence of the characteristic fields, reduced by the upper critical field. The onset field is approximately constant at about $0.8H_{c2}$ and the maximum field at about $0.85H_{c2}$. H_{irr} is located at about $0.9H_{c2}$, but seems to get slightly lower as $\theta \rightarrow 0$ deg.

The characteristic fields could not be determined with enough accuracy in the whole region of angles: Since $\tilde{\tau}$ $= \vec{m} \times H$ and \vec{m} point, for $H||c$ or $H||ab$, into the same direction, the sensitivity is much lower for angles close to 0 deg and 90 deg. Due to the pronounced anisotropy of $MgB₂$ at 18 K ($\gamma \approx 5.7$) (Ref. 5) \vec{m} tends to be directed almost perpendicular to the planes, except at very high angles. Therefore, the maximum effective sensitivity of the torque magnetometer is achieved at angles in the region of 75 deg–80 deg. Superconducting quantum interference device (SQUID) measurements performed on the same crystal with $H\|c$ and $H\|ab$ showed no sign of a PE in the region around $0.8H_{c2}$. This is likely due to insufficient sensitivity of the SQUID and field inhomogeneities in the SQUID magnetometer, which, due to the movement of the sample, tend to smear such features.²⁶ However, preliminary ac susceptibility $data²⁷$ measured on the same crystal indicate the PE to be present both for $H\|c$ and $H\|ab$, confirming that the underlying mechanism is a feature for all directions of *H*.

In Fig. 4, the *T* dependence of the characteristic fields is shown, for 77.5 deg, which corresponds roughly to the angle where the PE is most visible. The peak amplitude is reduced quickly by increasing *T*, and above 22 K the PE is no longer clearly discernible in the $\tau(H)$ data. This is due to the decreased sensitivity of the magnetometer 28 and due to thermal smearing of the effective disorder potential. The inset shows that the positions of H_{on} and H_{max} relative to H_{c2} are approximately constant. It is, therefore, unlikely that they would merge with the upper critical field at some higher temperature. In contrast, H_{irr} shifts to lower fields relative to H_{c2} as *T* increases, also likely due to a smearing of the effective pinning landscape by thermal fluctuations. There is no indication that H_{irr} may correspond to a phase transition.

Before discussing the PE in terms of a disorder-induced phase transition of vortex matter, we briefly examine alternative origins of the PE. The possibility that the PE is due to inhomogeneities or extended defects is not likely. A second crystal grown with the same technique, but under slightly different conditions, also shows a PE, in similar fields. The pinning properties of the two crystals for *H* nearly aligned parallel to the *ab* planes are different in a pronounced way.²⁹ It, therefore, seems rather unlikely that the two crystals would have identical structural, nonintrinsic features leading to a similar PE. A further possibility would be a change of the elastic constants of the vortex lattice when *H* approaches H_{c2} , not associated with a phase transition.³⁰ However, the specific forms of the history effects observed in the PE region are hard to explain without a phase transition. Thermal melting can rather be excluded. Thermal fluctuations should be much less important in $MgB₂$ than in the cuprate superconductors: MgB2 has a Ginzburg number *Gi* $= \frac{1}{2} [\gamma k_B T_c / H_c^2(0) \xi_{ab}^3(0)]^2$, a measure of the importance of thermal fluctuations, of the order of 10^{-5} only, while the cuprates typically³¹ have $Gi \approx 10^{-2}$. On the other hand, thermal fluctuations should be more important than, for example, in Nb with $Gi \approx 10^{-10}$ (Ref. 18), or NbSe₂ with $Gi \approx 10^{-8}$ (Ref. 21). A calculation of the melting field H_m , using Eq. (26) of Ref. 31 leads, at 14 K, to $H_m/H_{c2} \approx 0.97$, much higher than the location of the peak and therefore hardly can account for it, 32 although it was shown that point disorder can shift H_m to slightly lower fields.³³ Also, a liquid produced by thermal melting should have weaker pinning properties than the solid lattice.

An important fact deduced from the MHL experiments is that the high-field phase has got a higher critical current density than the low-field phase. This is the case for the transition from a Bragg glass to a highly disordered glassy phase.¹¹ The fact that we indeed observed this phase transition is supported by the pronounced history dependence of j_c in the region between onset and maximum of the peak, of a form similar to observations of the PE in $NbSe₂$ and not accountable for by relaxation effects. The location relatively close to H_{c2} is expected for a superconductor with low Gi and relatively weak disorder.³¹ In NbSe₂, a superconductor with comparable, but even lower *Gi*, there is conclusive evidence that the PE is indeed due to the transition between a Bragg glass and a highly disordered phase.¹⁶ The history effects mark the region of metastability, where a macroscopic coexistence of the two phases is possible. Pinning of the phase boundary is directly responsible for the history effects. The location of the PE with respect to H_{c2} , together with the history effects studied and the observation of a higher critical current density in the high-field vortex configuration, thus indicate that the PE in $MgB₂$ marks the transition between the Bragg glass and a highly disordered phase, which may be termed ''amorphous'' or ''pinned liquid.'' If the PE observed by Welp *et al.*⁹ is of the same origin, the larger separation of the PE from H_{c2} in our case indicates that the crystal investigated by us has a higher amount of random pointlike disorder. Further investigations of the transition line with controlled tuning of the amount of disorder, as was done in the case of the cuprates, by electron irradiation 33 and chemical

- *Email address: angst@phys.ethz.ch
- ¹For a review of early research see C. Buzea and T. Yamashita, Supercond. Sci. Technol. **14**, R115 (2001).
- 2F. Bouquet, Y. Wang, R.A. Fisher, D.G. Hinks, J.D. Jorgensen, A. Junod, and N.E. Phillips, Europhys. Lett. **56**, 856 (2001).
- ³H. Schmidt, J.F. Zasadzinski, K.E. Gray, and D.G. Hinks, Phys. Rev. Lett. 88, 127002 (2002).
- ⁴M. Iavarone *et al.*, Phys. Rev. Lett. **89**, 187002 (2002).
- 5M. Angst, R. Puzniak, A. Wisniewski, J. Jun, S.M. Kazakov, J. Karpinski, J. Roos, and H. Keller, Phys. Rev. Lett. **88**, 167004 $(2002).$
- 6A.V. Sologubenko, J. Jun, S.M. Kazakov, J. Karpinski, and H.R. Ott, Phys. Rev. B 65 , 180505(R) (2002).
- 7 S.L. Bud'ko and P.C. Canfield, Phys. Rev. B 65 , 212501 (2002).
- 8M. Zehetmayer, M. Eisterer, J. Jun, S.M. Kazakov, J. Karpinski, A. Wisniewski, and H.W. Weber, Phys. Rev. B **66**, 052505 $(2002).$
- 9 U. Welp *et al.*, cond-mat/0203337, Phys. Rev. B (to be published 1 February 2003!.
- ¹⁰A. Schilling, R.A. Fisher, N.E. Phillips, U. Welp, D. Dasgupta, W.K. Kwok, and G.W. Crabtree, Nature (London) 382, 791 $(1996).$
- 11 T. Giamarchi and P. Le Doussal, Phys. Rev. Lett. **72**, 1530 (1994); Phys. Rev. B 52, 1242 (1995); T. Nattermann and S. Scheidl, Adv. Phys. 49, 607 (2000).
- ¹²B. Khaykovich *et al.*, Phys. Rev. Lett. **76**, 2555 (1996); Phys. Rev. B 56, R517 (1997).
- ¹³D. Giller *et al.*, Phys. Rev. Lett. **79**, 2542 (1997).
- 14We follow convention and treat melting and order-disorder transition separately, although there is evidence that they form one line of phase transitions, which just changes character from thermally driven to disorder driven. See N. Avraham *et al.*, Nature (London) 411, 451 (2001).
- 15G. Ravikumar, V.C. Sahni, A.K. Grover, S. Ramakrishnan, P.L. Gammel, D.J. Bishop, E. Bucher, M.J. Higgins, and S. Bhattacharya, Phys. Rev. B 63, 024505 (2001).

substitution, 2^{0} may help finding a unified description of the phase diagrams of different superconductors.

In summary, using torque magnetometry, we observed a pronounced, sharp peak effect in single crystal MgB_2 . Onset and maximum of the peak are located at about $0.8H_{c2}$ and $0.85H_{c2}$, with little dependence on the temperature or the direction of the applied field. Peak form, history effects between onset and maximum, as well as the location of the peak are consistent with the peak effect marking a phase transition between the Bragg glass and a highly disordered phase of vortex matter.

Note added. After submission of this manuscript, we became aware of a report³⁴ on ac susceptibility measurements for $H||c$, coming to similar conclusions.

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- 16 M. Marchevsky, M.J. Higgins, and S. Bhattacharya, Nature (London) 409, 591 (2001).
- 17X.S. Ling, S.R. Park, B.A. McClain, S.M. Choi, D.C. Dender, and J.W. Lynn, Phys. Rev. Lett. **86**, 712 (2001).
- 18E.M. Forgan, S.J. Levett, P.G. Kealey, R. Cubitt, C.D. Dewhurst, and D. Fort, Phys. Rev. Lett. 88, 167003 (2002).
- 19 M.R. Eskildsen (private communication).
- 20M. Angst, S.M. Kazakov, J. Karpinski, A. Wisniewski, R. Puzniak, and M. Baran, Phys. Rev. B 65, 094518 (2002).
- 21V.V. Eremenko, V.A. Sirenko, Y.A. Shabakayeva, R. Schleser, and P.L. Gammel, Low Temp. Phys. **28**, 6 (2002).
- 22S.B. Roy, P. Chaddah, and S. Chaudhary, Phys. Rev. B **62**, 9191 $(2000).$
- ²³The form of the MHL clearly indicates that a full reversal of the critical state (Ref. 24) is achieved.
- ²⁴ C.P. Bean, Phys. Rev. Lett. **8**, 250 (1962).
- ²⁵A criterion τ_{irr} /*H* sin(2 θ)=10⁻⁶ dyn cm Oe⁻¹ was used for de-
fining H_{irr} .
- ²⁶G. Ravikumar *et al.*, Physica C **276**, 9 (1997).
- 27 R. Puzniak *et al.* (unpublished).
- ²⁸The signal-to-noise ratio is decreased in lower fields, since τ \propto *H*.
- 29M. Angst, R. Puzniak, A. Wisniewski, J. Roos, H. Keller, and J. Karpinski, cond-mat/0206407 (unpublished).
- ³⁰ A.B. Pippard, Philos. Mag. **19**, 217 (1969).
- 31 G.P. Mikitik and E.H. Brandt, Phys. Rev. B 64 , 184514 (2001).
- ³² Combined ac susceptibility and dc magnetization measurements found the PE maximum to be located in fields equal to or even higher than the magnetization step signifying melting. See, e.g., J. Shi *et al.*, Phys. Rev. B **60**, R12 593 ~1999!; T. Ishida *et al.*, *ibid.* **56**, 5128 (1997).
- 33T. Nishizaki, T. Naito, S. Okayasu, A. Iwase, and N. Kobayashi, Phys. Rev. B 61, 3649 (2000).
- 34M. Pissas, S. Lee, A. Yamamoto, and S. Tajima, Phys. Rev. Lett. **89**, 097002 (2002).