Temperature and Field Dependence of the Anisotropy of MgB_2

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The anisotropy γ of the superconducting state of high quality single crystals of MgB_2 was determined, using torque magnetometry with two different methods. The anisotropy of the upper critical field was found to be temperature dependent, decreasing from $\gamma \simeq 6$ at 15 K to 2.8 at 35 K. Reversible torque data near T_c reveal a field dependent anisotropy, increasing nearly linearly from $\gamma \simeq 2$ in zero field to 3.7 in 10 kOe. The unusual temperature dependence is a true bulk property and can be explained by nonlocal effects of anisotropic pairing and/or the $\vec{k}$ dependence of the effective mass tensor.

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The discovery of superconductivity at $T_c \approx 39$ K in MgB_2 [1] has caused a lot of interest as to its physical properties (for a review, see Ref. [2]). Measurements of the isotope effect [3] and, e.g., the ^{11}B nuclear spin-lattice relaxation rate [4] indicated a BCS type s-wave phonon-mediated superconductivity. Calculations of the band structure and the phonon spectrum predict a double energy gap [5,6], a larger gap attributed to two-dimensional p_{x-y} orbitals, and a smaller gap attributed to three-dimensional p_z bonding and antibonding orbitals. A substantial body of evidence by, among others, specific heat measurements [7], point-contact spectroscopy [8], scanning tunneling spectroscopy [9], and penetration depth measurements [10] has emerged to support this scenario. An alternative scenario with a single, but anisotropic, gap was also proposed [11], and supported by recent Raman measurements [12].

A double gap structure or an anisotropic energy gap should influence the anisotropy in the superconducting state, according to the standard anisotropic Ginzburg-Landau (GL) theory $\gamma = (m_c^*/m_{ab}^*)^{1/2} = \lambda_c/\lambda_{ab} = \xi_{ab}/\xi_c = H_{c2}^{\parallel ab}/H_{c2}^{\parallel c}$, where $\parallel ab$ ($\parallel c$) indicates the field H perpendicular (parallel) to the c axis of the sample and m^*, λ, ξ, and H_{c2} are the GL effective mass, the penetration depth, the coherence length, and the upper critical or bulk nucleation field, respectively. Most values reported for the anisotropy of polycrystalline or thin film MgB_2 span the range of values of $\gamma = 1.1-3$ [2], but there are also reports with $\gamma \approx 6-9$ [13,14]. Up to now, there are only four reports on transport measurements of the upper critical field anisotropy performed on single crystals, giving values of 2.6 [15], 2.7 [16], and 3 [17,18]. Magnetic measurements of the angular dependence of $H_{c2}(\theta)$, yielding $\gamma = 1.6$, were reported only on aligned crystallites [19].

Here, we report magnetic torque measurements on single crystals of MgB_2, performed in a wide range of temperatures from 15 to 36 K in magnetic fields of up to 90 kOe. We provide evidence that the bulk anisotropy γ is not universally constant, but is temperature dependent down to at least $0.4T_c$ and shows a pronounced field dependence near T_c. Microscopic origins of the unusual T dependence of γ in MgB_2 are discussed.

We have grown single crystals of MgB_2 with a high pressure cubic anvil technique, similar to the one described in Ref. [16]. The details of crystal growth will be published elsewhere. In brief, a mixture of Mg and B was put into a BN container and a pressure of 30–35 kbar was applied. Growth runs consisted of heating during 1 h up to the maximum temperature of 1700–1800 °C, keeping the temperature for 1–3 h and then cooling to room temperature during 1–2 h. Flat crystals were up to $0.8 \times 0.6 \times 0.05$ mm^3 in size, with sharp transitions to the superconducting state at about 38–39 K.

Measurements were performed on miniaturized piezoresistive cantilevers specifically designed for torque magnetometry [20]. The torque $\vec{\tau} = \vec{m} \times \vec{B} \simeq \vec{m} \times \vec{H}$, where $\vec{m}$ is the magnetic moment of the sample, was recorded as a function of the angle θ between the applied field $\vec{H}$ and the c axis of the crystal for various fixed temperatures and fields. For measurements close to T_c, in fields up to 14 kOe, a noncommercial magnetometer with very high sensitivity was used. For part of these measurements, a vortex-shaking process was employed to speed up the relaxation of the vortex lattice [21]. The observation of a well-resolved lock-in effect in $\tau(\theta)$ (see upper inset of Fig. 5) indicates there are no variations of crystallographic alignment throughout the samples. A crystal with a volume of about 4×10^{-4} mm^3 (sample A) was measured in this system. Another crystal with a volume of about 8×10^{-3} mm^3 (sample B) was measured in a wider range of temperatures down to 15 K in a Quantum Design PPMS (physical properties measurement system) with torque option and a maximum field of 90 kOe.

An example of a torque vs angle curve is given in the inset of Fig. 1. For small angles θ the torque is essentially zero. Only when H is nearly parallel to the ab plane is there an appreciable torque signal. The curve can be interpreted in a straightforward way: for H parallel to the c axis the sample is in the normal state, while for

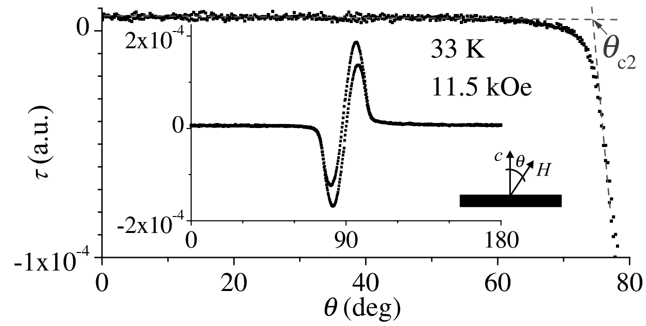

FIG. 1. Torque τ vs angle θ data, showing the definition of θ_{c2}. The inset shows the full torque curve measured.

H parallel to the ab plane it is in the superconducting state. The crossover angle θ_{c2} between the normal and the superconducting state is the angle for which the fixed applied field is the upper critical field. The inset of Fig. 1 also shows hysteretic behavior due to irreversibility. The irreversibility field $H_{\rm irr}(T, \theta)$ determined from the torque measurements is very high, close to H_{c2}. Preliminary SQUID measurements on similar crystals indicate a much lower $H_{\rm irr}$ [22]; an extended discussion of the irreversible properties of MgB_2 will be published elsewhere.

The crossing between straight lines through the background and the superconducting torque signal was used to define θ_{c2}. This definition is not unambiguous. Taking the analysis of the data more strictly we have to apply the appropriate scaling rules. The magnetization M of a 3D system in the GL theory of fluctuations in the vicinity of the transition temperature $T_c(H)$ in high magnetic fields is given by a universal function F of the distance from $T_c(H)$ [23]:

$$\frac{M}{H} = \frac{T^{2/3}}{H^{1/3}} \mathrm{F}\left(\frac{A(T - T_c)}{(TH)^{2/3}}\right), \qquad (1)$$

where A is a material constant. Combining the above dependence with the angular dependence of the torque [24] we find that the rescaled torque signal

$$\mathrm{P} = -\tau \epsilon^{1/3}(\theta)/[\sin\theta \cos\theta H^{5/3}(1 - 1/\gamma^2)T^{2/3}], \quad (2)$$

with $\epsilon(\theta) = (\cos^2\theta + \sin^2\theta/\gamma^2)^{1/2}$ is a universal function of the distance from T_c with a fixed value F(0) at $T = T_c(H)$. Taking into account the F(0) value for the theoretical dependence of the universal function for a 3D system [25] we can estimate that for a volume of the sample of 8×10^{-3} mm^3 P reaches at $T = T_c(H)$ a value of about 2×10^{-10} dyn cm Oe$^{-5/3}$ K$^{-2/3}$. The inset in Fig. 2 presents the angular dependence of the rescaled torque P in different magnetic fields at 22 K. The crossing of the P(θ) dependence for each field with the line of the constant value of 2×10^{-10} dyn cm Oe$^{-5/3}$ K$^{-2/3}$ determines the $H_{c2}(\theta)$ dependence as it is shown in the main panel of Fig. 2. It is important to stress that the results obtained depend not very sensitively on the criterion chosen and it will be shown later (see Fig. 4) that even with a 3 times higher criterion we get very similar temperature dependences of H_{c2} and γ. Additional $\tau(H)$ measurements at fixed angle give $H_{c2}(\theta)$ values corresponding well to those from $\tau(\theta)$ measurements.

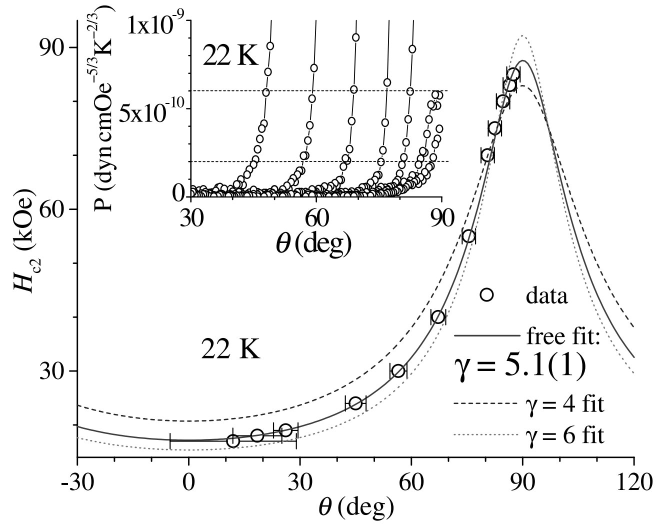

FIG. 2. Upper critical field H_{c2} vs θ at 22 K. The full line is a free fit of Eq. (3) to the data. Alternative fits with fixed values of the anisotropy γ are also shown. The inset shows selected rescaled torque P [see Eq. (2)] vs angle θ. From left to right, curves shown were measured in H = 24, 30, 40, 55, 70, 80, and 85 kOe. The two criteria used for the determination of θ_{c2} are indicated by dashed lines. The data in the main panel were obtained employing the lower criterion.

Within the applicability of the anisotropic GL theory the angle dependence of the upper critical field is predicted to be [26]

$$H_{c2}(\theta) = H_{c2}^{\|c}(\cos^2\theta + \sin^2\theta/\gamma^2)^{-1/2}. \qquad (3)$$

A fit of Eq. (3) to the data at 22 K yields $\gamma = 5.1(1)$ and $H_{c2}^{\|c} = 17.2(1)$ kOe. This fit ($\gamma = 5.1$) describes the data well, while alternative fits with γ fixed to 4 and 6 are clearly incompatible with the data, as shown in Fig. 2.

Figure 3 shows the angular dependence of H_{c2} scaled by $H_{c2}^{\|c}$ to directly compare the anisotropy at different temperatures. The 15 K data are well described by the line corresponding to $\gamma = 6$, while the 34 K data lie below the line for $\gamma = 3.5$. The data indicate an anisotropy systematically decreasing with increasing temperature. To show that this is not an artifact related to fitting, we present the angular dependence of the rescaled torque P for fixed $H/H_{c2}^{\|c}$ in the inset. The curves clearly shift to higher angles with increasing temperature. Furthermore, we directly checked $\tau(\theta)$ raw data in fields above and below $H_{c2}^{\|c}$ and $H_{c2}^{\|ab}$ to give absolute limitations of γ. We find that at 22 K the anisotropy *must* be higher than 4.4, while at 34 K it *must* be lower than 3.5.

All data are summarized in Fig. 4. The $H_{c2}^{\|c}$ data obtained from fits to Eq. (3) do not vary much with the criterion used for the determination of θ_{c2}, and agree well with $H_{c2}^{\|c}$ calculated from thermal conductivity data [28] measured on a single crystal grown with the same technique. The T dependence of $H_{c2}^{\|c}$ is in agreement with

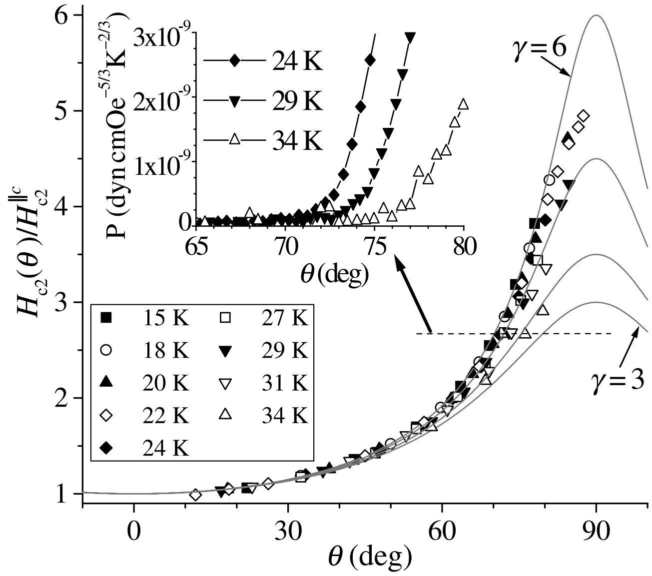

FIG. 3. Angular dependence of $H_{c2}(\theta)/H_{c2}^{\|c}$, employing the lower criterion, at various temperatures. Curves correspond to $H_{c2}(\theta)$ according to Eq. (3) for $\gamma = 3$, 3.5, 4.5, and 6. Inset: Some rescaled torque P [see Eq. (2)] vs angle θ curves, corresponding to $H/H_{c2}^{\|c} \simeq 2.67$.

calculations by Helfand and Werthamer [29]. The corresponding $H_{c2}^{\|c}(0) \simeq 31$ kOe is relatively small compared to literature values, which may indicate that the crystals investigated are relatively free of defects. The $H_{c2}^{\|ab}$ values were obtained from the two fit parameters $H_{c2}^{\|c}$ and γ. There is a slight positive curvature of $H_{c2}^{\|ab}(T)$, which can be attributed to the T dependence of γ. The $\gamma(T)$ dependence may also be the origin of the positive curvature of H_{c2} observed in other measurements of bulk, thin film and single crystal measurements [2]. Because of the lack of low temperature data and the variation of γ, only a rough estimation $H_{c2}^{\|ab}(0) \approx 230$ kOe can be given. The anisotropy data show that γ *systematically decreases* with increasing temperature. A change of the criterion used for the determination of θ_{c2} leads to small shifts of the magnitude of γ, but the temperature dependence is always the same. The highest upper critical field anisotropy $\gamma \simeq 6$ was obtained at 15 K, the lowest anisotropy $\gamma \simeq 2.8$ at 35 K. From Fig. 4 we estimate $\gamma(0) = 7$–8 and $\gamma(T_c) = 2.3$–2.7.

Small systematic deviations from Eq. (3), observed near T_c, indicate that the field influence on γ may be important as well. To clarify this point, we have measured the reversible torque τ as a function of angle θ for various fields and temperatures near T_c. The data were analyzed with an equation derived by Kogan *et al.* [27], based on the anisotropic London model, which contains the GL anisotropy γ as a parameter.

Without shaking, the irreversibility of the torque was relatively large and a clear lock-in effect was observed (see upper inset of Fig. 5). The lower inset of Fig. 5 shows the corresponding shaked torque data, together with the fitted line. An evaluation of the data measured for various T and H up to 10 kOe with the Kogan formula [27] reveals that γ is field dependent with larger γ in larger fields, while temperature variations do not affect γ appreciably

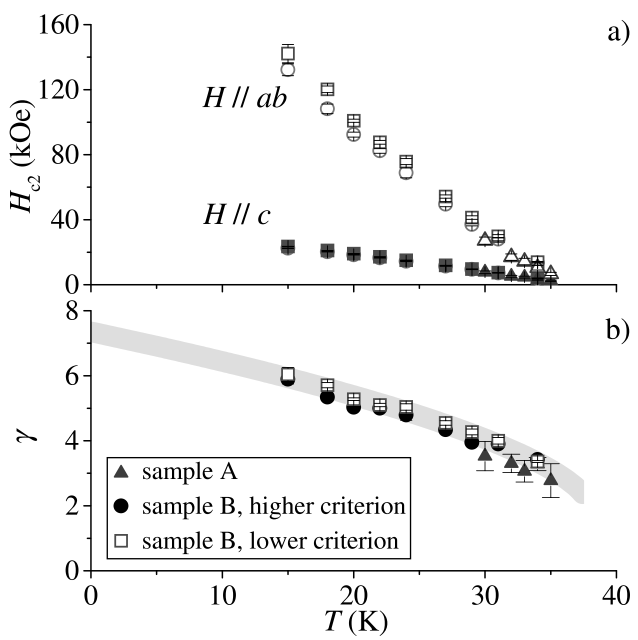

FIG. 4. (a) Upper critical field H_{c2} vs temperature T. Open symbols correspond to $H \parallel ab$, full symbols to $H \parallel c$, from fits of $H_{c2}(\theta)$ data to Eq. (3). Up triangles are from measurements on sample A (with θ_{c2} determined as shown in Fig. 1) and squares (circles) are from measurements on sample B, using the lower (higher) criterion of constant rescaled torque P (see inset of Fig. 2). (b) Temperature dependence of the upper critical field anisotropy $H_{c2}^{\|ab}/H_{c2}^{\|c}$, determined from fits of $H_{c2}(\theta)$ to Eq. (3).

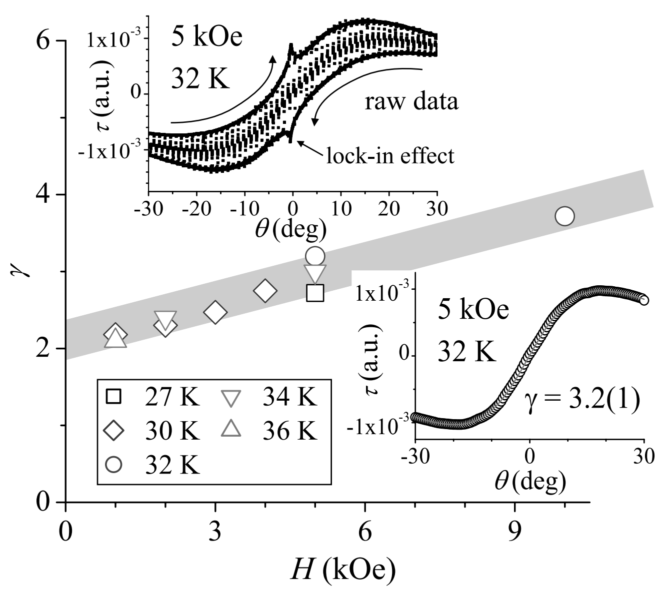

FIG. 5. Field dependence of the anisotropy γ as determined from the angular dependence of the reversible torque $\tau(\theta)$. Upper inset: raw data, demonstrating the effect of shaking and the lock-in effect. Lower inset: fitting of the shaked data to a formula derived by Kogan *et al.* [27], yielding $\gamma \simeq 3.2$.

(see Fig. 5). Indications of a field dependence of the coherence length ξ (a precondition of a field dependence of its anisotropy) have been observed by specific heat [30] and muon spin rotation [31] measurements on $NbSe_2$. We should stress that Kogan's formula assumes equivalence of the anisotropies of the coherence length ξ and the penetration depth λ. Since the anisotropy of λ might be different from the one of ξ in our case, the two different methods used in this work can lead to different effective values of γ. The field dependence of γ may be related to the peculiar double gap structure of MgB_2 with a large gap of two-dimensional nature and a small three-dimensional gap, which is very rapidly suppressed in a magnetic field [8].

A temperature dependent H_{c2} anisotropy was previously observed, e.g., in $NbSe_2$ [32] and $LuNi_2B_2C$ [33]. However, in MgB_2, the effect is much more pronounced. It was shown that any theory capable of explaining a temperature dependence of γ needs to take into account nonlocal effects [34,35], which can be pronounced in samples of high purity. In the vicinity of T_c, nonlocality is not important [33]. Therefore, $m_c^*/m_{ab}^* = \gamma^2(T_c) \simeq 5-7$ corresponds to the standard GL effective mass anisotropy. We note that this is significantly higher than the calculated [5,36] anisotropy of the band effective mass (BEM) averaged over the Fermi surface (1.0–1.2). The microscopic theories [34,35] show that the T dependence of γ cannot be attributed to an anisotropy of the BEM tensor, unless it is also wave vector dependent. An anisotropic energy gap, caused, e.g., by an anisotropy in the electron-phonon coupling (EPC), can also lead to variations of γ with temperature.

In MgB_2, first principles calculations suggest both a pronounced wave-vector dependence of the BEM tensor [36] and a strong anisotropy of the EPC (see, e.g., [6,37]). The latter leads naturally [6] to the observed double energy gap and was suggested [38] to be responsible for the unusually high T_c of MgB_2. To our knowledge, there is only one theory calculating a temperature dependent H_{c2} anisotropy of MgB_2 [11], which predicts, however, $H_{c2}^{\|ab}/H_{c2}^{\|c}$ to increase with increasing T. A quantitative explanation of the measured $\gamma(T)$ apparently needs to take into account both the wave-vector dependence of the BEM tensor and the anisotropic EPC, and is beyond the scope of this work.

In conclusion, the upper critical field anisotropy γ of MgB_2, determined by torque magnetometry, decreases with increasing temperature. Measurements of the reversible torque near T_c reveal an almost linear field dependence of the anisotropy of the coherence length and/or the penetration depth as well. Our results imply a breakdown of standard anisotropic GL theory with a (temperature and field independent) effective mass anisotropy. The temperature dependence of γ can be approximated tentatively by $\gamma(T) = \gamma^* + \tilde{\gamma}(1 - T/T_c)^n$ with n close to 1. Here, $\gamma^* \simeq 2.3-2.7$ is the band effective mass anisotropy and $\tilde{\gamma} \simeq 4.5-5.5$ arises from the anisotropy of the attractive electron-electron interaction and/or the wave-vector dependence of the effective mass tensor.

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