Magnetic-field-induced orientation of superconducting MgB$_2$ crystallites determined by x-ray diffraction

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X-ray diffraction studies of fine polycrystalline samples of MgB$_2$ in the superconducting state reveal that crystals orient with their c axis in a plane normal to the direction of the applied magnetic field. The MgB$_2$ samples were thoroughly ground to obtain average grain size $5-10$ μm in order to increase the population of free single crystal grains in the powder. By monitoring Bragg reflections in a plane normal to an applied magnetic field we find that the powder is textured with significantly stronger $(0,0,l)$ reflections in comparison to $(h,k,0)$, which remain essentially unchanged. The orientation of the crystals with the ab plane parallel to the magnetic field at all temperatures below T_c demonstrates that the sign of the torque under magnetic field does not alter, in disagreement with current theoretical predictions.

I. INTRODUCTION

It is well established experimentally and theoretically that the layered two-band superconductor MgB$_2$ exhibits unique and intriguing anisotropic properties. These are manifested in measurements of the ratios of the upper critical fields $\gamma_H = H_{c2}^{\perp c}/H_{c2}^{\parallel c}$ for magnetic fields along the c axis and the ab plane and of the London penetration depth $\gamma_\lambda = \lambda_c/\lambda_{ab}$.[1–14] The preponderance of experiments are in agreement with regard to the temperature dependence of γ_H, which shows increase from $\gamma_H \approx 2.0$ around T_c to $\gamma_H \approx 6$ at low temperatures.[6,7,14] However, the temperature dependence of γ_λ, in particular under applied magnetic field, is not settled yet.[13] Experimental evidence shows that $\gamma_\lambda = \gamma_H \approx 2$ around $T_c = 40$ K, and as the temperature decreases it approaches the isotropic value $\gamma_\lambda = 1$ (see Fig. 4 in Ref. 14). Some experimental studies claim that the $\gamma_H(T,H) = \gamma_\lambda(T,H)$.[12,13] For a crystal with $\gamma_\lambda = \gamma_H$ in a field along the c axis, the vortex energy is given by $\sim (\phi_0/4\pi\lambda_{ab})^2$ within a factor of order unity, where ϕ_0 is the flux quantum. The energy is approximately γ_λ times lower when the field is perpendicular to the c axis: $\sim (\phi_0/4\pi)^2/(\lambda_{ab}\lambda_c) = (\phi_0/4\pi\lambda_{ab})^2/\gamma_\lambda$. As a result, superconducting free single crystal grains placed in the magnetic field experience a torque that tends to orient their ab plane parallel to the field. Such behavior has been observed in YBCO by neutron scattering experiments[15] on powders, and in single crystal torque measurements[16] performed at temperatures close to T_c.

Recent theoretical predictions show this behavior is expected to be different when $\gamma_\lambda \neq \gamma_H$.[8] Since the coherence length ξ has the same anisotropy as H_{c2}, the vortex core has a different shape determined by the anisotropy of ξ as compared to the current distribution far from the core determined by the anisotropy of the penetration depth λ. This mismatch costs energy, which—for a sufficiently large difference between γ's—causes the energy of vortices parallel to ab to exceed their value for vortices along c. Thus, for MgB$_2$, close to T_c where $\gamma_H = \gamma_\lambda$, the theory predicts that the torque should tend to orient crystals with their ab plane parallel to

the direction of the applied magnetic field H, similar to the observation for the high T_c superconductors, whereas at low temperatures where $\gamma_H \neq \gamma_\lambda$ a single crystal will orient with its c axis along the magnetic field.

We have undertaken synchrotron x-ray diffraction studies to determine the orientation of free-grain powdered samples under applied magnetic field. The preferential orientation can be detected in the x-ray scattering experiment on fine-grain MgB$_2$ powders provided a sufficient number of grains are single crystals and the difference in magnetic energy between the two principal orientations is large enough to overcome the average intergrain energy barrier for reorientation. In YBCO similar experiments found that the crystallites tend to orient themselves with the c axis perpendicular to the applied field.[15]

II. EXPERIMENTAL DETAILS

The experimental setup used in this study and expected diffraction patterns for different scenarios are shown schematically in Fig. 1. The magnetic field is normal to the scattering plane spanned by the incident and scattered beams, $\mathbf{H} \perp \mathbf{Q}$; $\mathbf{Q} \equiv \mathbf{k_i} - \mathbf{k_f}$ is the scattering vector. Figure 1 shows the consequences of applying a magnetic field on $(0,0,l)$ and $(h,0,0)$ reflections as a function of the scattering angle 2θ represented with the same peak shape and intensity at zero field (all grains are randomly oriented). For simplicity, we assume the Bragg reflections are normalized to their structure factor, multiplicity, and Lorentz factor. If the grains orient with their c axis parallel to the applied magnetic field, we expect a near-complete suppression of the intensity of $(0,0,l)$ reflections and a significant increase in the $(h,0,0)$. In the extreme case of perfect orientation ($\mathbf{H} \parallel \mathbf{c}$) and perfect instrumental resolution function (δ-function-like) the $(0,0,l)$ reflections vanish. In the case $\mathbf{H} \perp \mathbf{c}$, due to the axial symmetry, we expect a strong increase in intensity of the $(0,0,l)$ reflections and almost no change in the $(h,0,0)$, as depicted in Fig. 1.
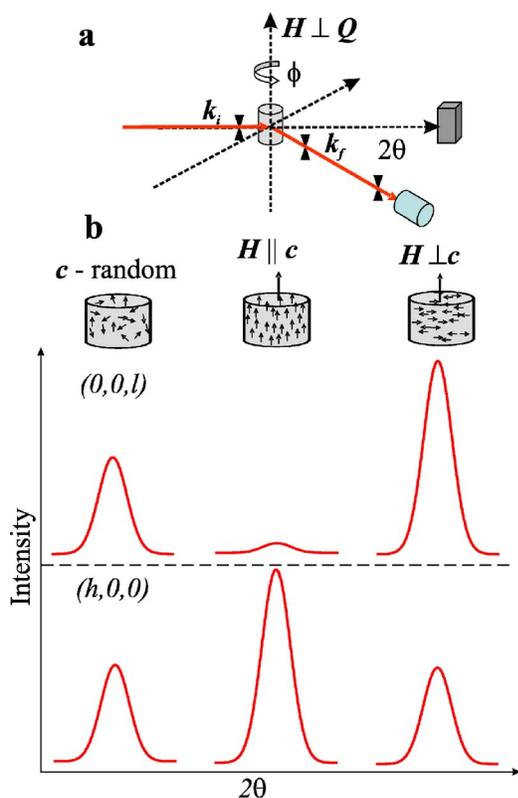
FIG. 1. (Color online) (a) Sketch of the diffractometer-magnet configuration. The scattering vector is in the plane normal to the direction of the applied magnetic field. (b) Illustration of the effect of single-crystal grains reorientations under magnetic field on the $(0,0,l)$ and $(h,k,0)$ reflections as a function of the scattering angle 2θ. The two sets of reflections at zero field (random orientation) are represented by same shape and intensity peaks for simplicity. Preferred alignment of the c axis along the magnetic field, reduces the intensity of the $(0,0,l)$ increasing that of the $(h,0,0)$. If the grains orient with the ab plane parallel to the field $\mathbf{H} \perp \mathbf{c}$, the $(0,0,l)$ will become more intense and there will be only a slight effect on the intensity of the $(h,0,0)$.

Two types of MgB_2 samples distinguished by the growth method and morphology, loosely sintered pellet,[17] and thin-wire segment (or fiber),[18] were used in this study. A portion of each sample was coarsely crushed to grains of $50-100$ μm in size and another portion was thoroughly ground to $5-10$ μm. The four polycrystalline samples were loaded into thin walled (250 μm) cylindrical aluminum cans, and indium-sealed with copper caps in a helium environment to enhance thermal contact. The x-ray scattering studies were conducted on the x-ray operations and research (XOR) beamline 4IDD, at the Advanced Photon Source, Argonne National Laboratory. A Si(111) double crystal monochromator was used to select the 36 keV ($\lambda=0.344\,40$ Å) incident x-ray beam from an undulator fifth harmonic. A cryogen-free superconducting magnet with maximum field of 4 Tesla mounted on a Huber diffractometer was used to produce a magnetic field normal to the scattering plane as schematically shown in Fig. 1. The magnet is equipped with three optical beryllium windows providing access to a wide range of scattering angles. A magnetically shielded scintilla-
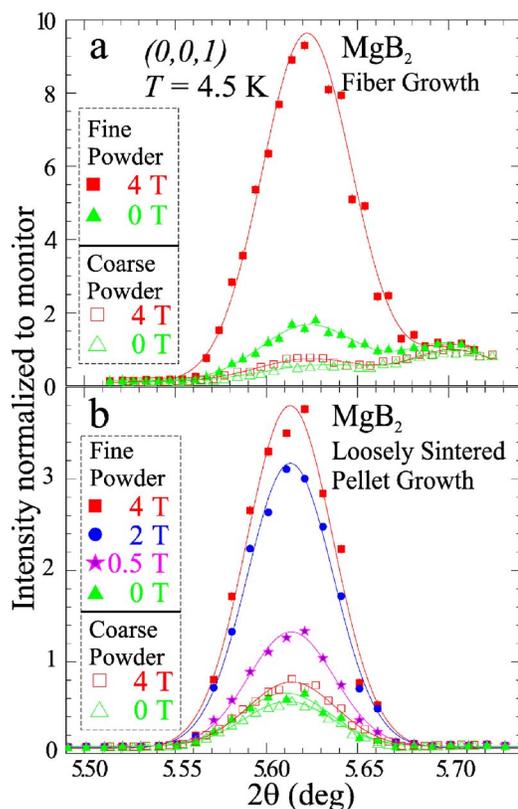

FIG. 2. (Color online) Intensity versus scattering angle 2θ at $T=4.5$ K of the $(0,0,1)$ for types of MgB_2 samples (a) fiber grown (b) loosely sintered pellet grown at various magnetic fields as indicated. The intensity of each point is integrated over 60 degrees rotation of the sample angle ϕ. Whereas a significant increase in intensity is observed for finely ground powder (filled symbols), there is almost no change in intensity for samples consisting of coarse grains (open symbols). After lowering the magnetic field the powder was randomized spontaneously by mechanical vibrations of the compressor-cooled magnet with a typical time constant of a few minutes with small remnant alignment (Ref. 20).

tor point detector was used for measuring photon intensity. The intensity of each point in 2θ scans was integrated over 60 degrees of sample rotation ϕ to improve powder averaging, and normalized to a monitor count of the incident beam intensity. Samples were shaken thoroughly prior to being loaded in the cryomagnet. The superconducting magnet is cooled by a closed-cycle helium refrigerator that produces ~ 1 Hz of continuous mechanical vibration of the sample stage.

III. RESULTS AND DISCUSSION

Figure 2 shows intensity versus 2θ scans of the $(0,0,1)$ Bragg reflection at $T=4.5$ K demonstrating the effect of applied magnetic field on the coarse and fine powders of both the sintered pellet and the fiber grown.[19] Similar behavior with increase in intensity under applied magnetic field was also observed for the $(0,0,2)$ Bragg reflection. The coarse powders exhibit a 10%–20% increase between zero and the highest magnetic field (open symbols), whereas the increase
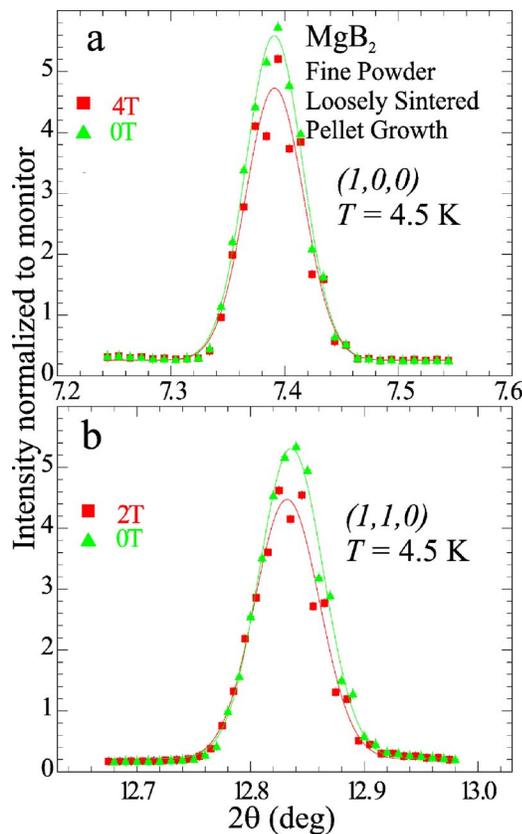
FIG. 3. (Color online) Intensity versus scattering angle 2θ at $T=4.5$ K of $(1,0,0)$ and $(1,1,0)$ in-plane Bragg reflections in magnetic field.
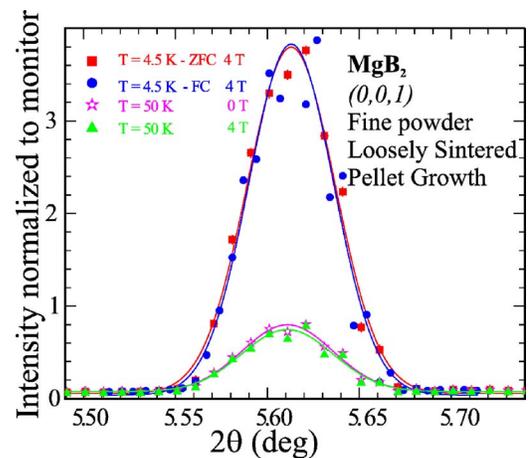

FIG. 4. (Color online) Intensity versus scattering angle 2θ at $T=50$ K at 0 and 4 Tesla of the $(0,0,1)$ of a finely ground sample. The sample was then cooled in field to reproduce the results obtained for the zero-field cooled sample shown in Fig. 2(b).

in intensities for the fine powders is sevenfold or eightfold stronger at 4 Tesla compared to zero field (solid symbols). The field independent peak at $2\theta \approx 5.7°$ is due to unidentified impurity phase present in the fiber grown sample.[18] The difference in behavior of finely and course ground powders is in fact expected, as it is known that as grown MgB_2 even moderately ground consists of bound crystalline clusters and only thorough grinding yields isolated single crystals (5–10 μm in size). By contrast to the $(0,0,l)$ reflections, the intensities of $(h,k,0)$ reflections yielded significantly smaller changes in intensity. A very small decrease in the intensities of the inplane $(1,0,0)$ and $(1,1,0)$ reflections were detected, as shown in Fig. 3. These observations clearly prove that the crystallites tend to orient with their c axis normal to the magnetic field in the superconducting state. After lowering the magnetic field to zero, the peaks intensities nearly recover their values in the randomized state within minutes after the field is removed.[20]

The difference in the reorientation of field cooled (FC) or zero field cooling (ZFC) samples was monitored by the intensity of the $(0,0,1)$ reflection. Figure 4 shows scans of the $(0,0,1)$ reflection for a finely ground sample that was cooled under 4 Tesla from $T=50$ K to base temperature (4.5 K, circles) in comparison with a similar scan of a ZFC sample (square symbols). There is no observable difference of FC or ZFC protocols on grain orientation. Figure 4 also includes two scans of the $(0,0,1)$ reflection at $T=50$ K, at zero and

4 Tesla with practically no change in either intensity or shape. Hence, the orientation occurs only in the superconducting state.

The degree of crystalline orientation is both temperature and field dependent. Figure 5 shows the integrated intensity of the $(0,0,1)$ peak at various temperatures versus magnetic field. At all temperatures the intensity of the $(0,0,1)$ initially increases with field providing evidence that the ground state orientation does not vary with temperature. The more detailed field-dependent measurement, conducted at $T=32.5$ K (open squares), shows features that can be correlated with $H_{c2}^{\|c}$ and $H_{c2}^{\perp c}$, indicated by arrows. We suggest to identify $H_{c2}^{\|c} \approx 0.55$ T as the point of deviation from the trend of the low field data, and with the development of the first minimum, and to identify $H_{c2}^{\perp c} \approx 3$ T with the point of devia-
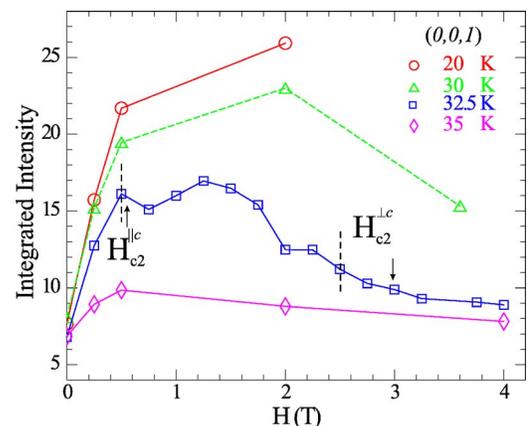

FIG. 5. (Color online) Integrated intensity of $(0,0,1)$ versus magnetic field at various temperatures, for the finely ground loosely sintered pellet grown sample. We suggest to identify $H_{c2}^{\|c} \approx 0.5$ T as the point of deviation from the trend of the low field data, i.e., with development of the first minimum and to assign $H_{c2}^{\perp c} \approx 3$ T with the point of deviation from the asymptotic behavior at high fields. Magnetization measurements of $H_{c2}^{\perp c}$ and $H_{c2}^{\|c}$ at 32.5 K on similar powders from Ref. 3 are shown with dashed lines.

tion from the asymptotic behavior at high fields (the curves shown in Fig. 5 for all temperatures other than 32.5 K are not sufficiently detailed to extract either $H_{c2}^{\parallel c}$ or $H_{c2}^{\perp c}$). It should be noted that above $H_{c2}^{\parallel c}$ the particles with the c axis parallel to the field are in the normal state, and only by virtue of mechanical vibrations may tilt to become superconducting and orient with the c axis perpendicular to the field. This explains the decrease in orientation at $H_{c2}^{\parallel c}$. The dashed lines in Fig. 5 are the results obtained from magnetization measurements.[3] Although the x-ray diffraction technique described here appears promising as a method to determine upper critical fields, more systematic studies are obviously required before it can be utilized as a reliable tool for this purpose.

In conclusion, our x-ray diffraction data show that the preferred orientation of an MgB_2 crystallite at low temperatures is to align with $c \perp H$ whereas theory predicts[8] the $c \parallel H$ orientation. Furthermore, the orientation $c \perp H$ persists at all temperatures showing no evidence of orientation change. One can trace the disagreement with theory to the assumption that the anisotropy parameter γ_λ and its temperature dependence used to interpret the high-field properties of the mixed state in Ref. 8 is the same as the one calculated[21] and measured[14] for small fields. The theoretical calculation[21] was done for a fixed ratio of gaps on two major sheets of the Fermi surface of MgB_2 (π and σ bands), whereas by now it

is accepted that the small π gap is suppressed even in moderate fields of a few kOe.[22] Thus, in the fields used in this experiment, the material behaves as having only one gap at the σ band and therefore a single anisotropy parameter for both H_{c2} and λ (i.e., γ_λ is nearly the same as γ_H). In other words, calculations of Ref. 8 do not apply. To construct a model applicable for all fields, one would need information on the angular dependence of the π-band suppression field. In principle, this can be acquired by detailed measurements of the field and angular dependencies of the magnetization and torque. The field and temperature dependence of the alignment show features that can be associated with the H_{c2} transitions yielding yet another way to measure the superconducting anisotropy in powder samples.

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[19] The intensities of the fine powders, fiber and the loosely sintered pellet, at zero field are different (Fig. 2) possibly due to differences in average grain size and scattering-volume filling ratios, however, the degree of alignment in the highest magnetic field (4 T) for both samples is similar, we therefore report on the loosely sintered pellet fine powder henceforth.

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