# **Anisotropy in** $MgB_2$

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The first direct measurement of an anisotropic superconducting property in  $MgB_2$  was achieved for the bulk nucleation field  $H_{c2}$ , in a sample of aligned crystallites. It was found a ratio  $\gamma(T) = H_{c2}^{ab}/H_{c2}^c = 1.6 - 1.9$ , for T varying from 32 K to 26K, between H applied parallel to the ab plane, and along the c direction. The anisotropy of the induced critical current density was evaluated through the Bean model to be  $J_c^{ab}/J_c^c \approx 1.5$ . We present here a brief review of these studies in connection with current results found in the literature.

#### 1 Introduction

The strongly anisotropic crystalline structure of  $MgB_2$ , consisting of triangular layers of magnesium atoms sandwiched between hexagonal layers of boron atoms, was known[1, 2] for almost 50 years before the discovery of superconductivity in this binary compound[3]. It seemed therefore reasonable when specific heat studies done in polycrystalline samples[4], as well as band structure calculations [5], pointed to a possible anisotropic nature of the electronic and magnetic properties of  $MgB_2$ . The first direct measurement of an anisotropic superconducting property was achieved for the bulk nucleation field  $H_{c2}$ , in samples of aligned  $MgB_2$  crystallites [6, 7]. It was found a ratio  $\gamma = H_{c2}^{ab}/H_{c2}^c \approx 1.7$ , between the critical field parallel to the ab plane and parallel to the c axis direction. Since then, different groups have found values of  $\gamma$  between 1.1 and 6, using different type of samples and different techniques to characterize the normal - superconducting transition[8, 9]. In our view the large scattering of reported  $\gamma$  values could be ascribed mainly to three factors[7]: (1) the sample purity, since it affects directly the energy gap anisotropy at the microscopic level, due mainly to inter- and intra-band scattering; (2) the experimental criterion used to define a reliable superconducting bulk transition; (3) the temperature dependence of  $\gamma$ . Indeed, recent reports[10, 11] have shown that  $\gamma$  goes from  $\sim 2$  to  $\sim 6$  when T varies between  $\sim 39$  K to  $\sim 15$  K.

The critical current density anisotropy  $J_C^{ab}/J_C^c \sim 1.5$  was also evaluated[12], using the Bean's model with data taken in the same sample of aligned crystallites[6] that produced  $\gamma \approx 1.7$ . The similarity between these anisotropy values is indeed expected for clean samples with relatively low pinning interactions, since in this case  $J_C$  is proportional to[13]  $\xi^2$ , where  $\xi$  is the anisotropic coherence length. Therefore,  $J_C^{ab}/J_C^c \approx \xi_{ab}/\xi_c \approx H_{c2}^{ab}/H_{c2}^c$ . Direct determi-

nation of  $J_C^{ab}/J_C^c$ , using transport current measurements, produced values around 2.5 for c-axis-oriented thin films [14], although in this case they did not measure  $\gamma$ .

By now, a two-band Fermi surface has been clearly demonstrated [15, 16] for  $MgB_2$ : a  $\pi-$  band generated by the boron  $p_z$  orbitals, oriented perpendicularly to the layers, and a  $\sigma-$  band generated by the  $p_{xy}$  orbitals, confined in the boron layers. Several novel effects have been related to this unique case of genuine two-gap superconductivity, as for example, the temperature dependence of the  $H_{c2}$  anisotropy ratio. Intraband and interband scaterring effects has been shown to play important roles in determining some of these new effects [17]. Following, we present a brief review on the anisotropy studies of  $MgB_2$  done by our group at Unicamp, and comment on some related results reported by other people.

## 2 Aligned Crystallites

Our studies were done using samples formed by a collection of aligned crystallites. A weakly sintered sample of  $MgB_2$  was prepared, starting with a stoichiometric mixture of 99.5 at% pure boron and 99.8 at% pure magnesium. The loose mixture was sealed in a Ta tube under Ar atmosphere, which was then encapsulated in a quartz ampoule and put into the furnace. The compound formation was processed by initially holding the furnace temperature at 1200°C for 1 hour, followed by a decrease to 700°C (10°C/h), then to 600°C (2°C/h), and finally to room temperature at a rate of 100°C/h. The weakly sintered product was easily crushed and milled employing mortar and pestle. A very uniform powder was then obtained, consisting mainly of shiny crystallites with aspect ratios ranging from 2 to 5, the main surface size ranging from 5 to 40  $\mu$ m and thickness around 2  $\mu$ m. The powder was sieved into a range of particle sizes

between 5 - 20  $\mu$ m, in which the crystallites fraction was almost 100%. Small amounts of this powder were then spread carefully on both sides of a piece of paper, producing an almost perfect alignment of the crystallites, as revealed[6] by SEM pictures and X-ray analysis. Finally, several samples were mounted consisting of a pile of five squares of  $3\times3$  mm², cut from the *crystallite-painted paper* and glued with Araldite resin. These samples produced enough signal for the measurement of magnetization and AC susceptibility, using, respectively, a SQUID and PPMS machines (made by Quantum Design).

## 3 Upper Critical Field Anisotropy

Figure 1 shows our data (crossed circles) for the temperature dependent ratio  $\gamma = H_{c2}^{ab}/H_{c2}^c$ , with other representative results taken from the literature, for  $MgB_2$  single crystals[11, 10] and thin films[18]. Our data shows consistently smaller values for  $\gamma \sim 1.6$  - 1.9 in the probed temperature interval of 32 K - 26 K, while the other results are scattered between  $\sim 2$  and  $\sim 4$ . We ascribe these differences mainly to the different sample purities as well as to the different criteria employed to define the relevant transition between the normal and superconducting states. Our results may also contain a small contribution to the depressed values of  $\gamma$  in consequence of the average misalignment of 4.6 degrees between the crystallites **c**-axis[7].

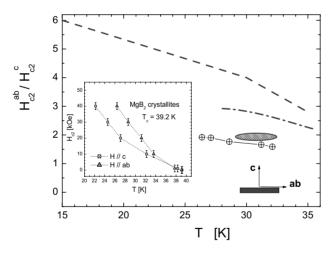


Figure 1. Anisotropy ratio of the upper critical field in  $MgB_2$  as a function of temperature for aligned crystallites (crossed circles), textured thin films of different purities[18] (hatched ellipse), single crystal using resistivity measurements[11] (dash-dotted line), single crystal using torque magnetometry[10] (dashed line). The inset shows the original  $H_{c2}$  data from Ref.[6]. Note: The data quoted from other authors are only approximately represented in this figure.

Using the Ginzburg-Landau mean field expression[19] (in CGS units)  $\xi(T)=\xi_o~(1-T/T_c)^{-1/2}$  and the results for anisotropic situations[20, 13]  $H_{c2}^c(T)=\phi_o/(2\pi~\xi_{ab}^2)$  and  $H_{c2}^{ab}/H_{c2}^c=1/\varepsilon$ , where  $\phi_o=2.07\times 10^{-7}~{\rm G~cm}^2$  is the quantum of flux and  $\varepsilon^2=m_{ab}/m_c$  is the mass anisotropy

ratio, we find that  $\xi_{o,ab}/\xi_{o,c}=\xi_{ab}(T)/\xi_c(T)=\gamma\simeq 1.73$  and  $\varepsilon^2\simeq 0.3$ , for  $T\approx 27$  K. Since at that temperature we have  $H_{c2}^c\approx 20$  kOe, this would imply  $\xi_{o,ab}\simeq 70$  Å and  $\xi_{o,c}\simeq 40$  Å. However, these values may be underestimated, in view of recent experiments[21] and theoretical calculations[22] based on the almost independent two-gap structure, originated by the two separate  $\pi$  and  $\sigma$  bands.

## 4 Critical Current Density Anisotropy

Measurements of the magnetic moment as a function of the applied magnetic field, for several temperatures below  $T_c =$ 39 K, were performed with a SQUID magnetometer. The average crystallites dimensions were  $10 \times 10 \times 2 \mu m^3$ , determined by visual inspection, using an optical microscope with a micrometer scale. An estimate of the total volume of crystallites in the sample gives 0.06 mm<sup>3</sup>, in total agreement with a value that produces a slope  $\Delta M/\Delta H = -1/4\pi$  for the region of diamagnetic shielding at  $H \approx 0$ . In order to subtract the magnetic background present in all curves[6], the same type of measurements were repeated at temperatures above  $T_c$  (not shown here). No significant temperature dependence was observed for the hysteresis loops measured at several temperatures from 45 K to 80 K, in the low field region. Thus, the loop obtained at T = 45 K was considered to be a good approximation for the magnetic background, in the entire temperature range going from 5 K to 35 K. The fully corrected magnetization curves are presented in Fig. 2, for (a) H // c and (b) H // ab. The remanent magnetization values, in both field directions, are shown in the enlarged plot of Fig. 3. To avoid complications associated with demagnetizing effects we treat only the case of H = 0.

The critical current density can be estimated from the corrected magnetization curves of Figure 2, if one assumes the occurrence of uniform gradients in the flux density distribution inside the crystallites. According to the Bean's critical state model[23],  $J_C$  is proportional to the width of the hysteresis loop and for a slab geometry it is

$$J_C = \frac{40 |\Delta M|}{t (1 - t/3w)} \tag{1}$$

where t < w are the sample dimensions perpendicular to the applied field and  $|\Delta M|$  is the magnetization loop width. Notice that the sample used in this work is formed by a collection of isolated crystallites, so t and w appearing in Equation (1) refer to the crystallites average dimensions.

The ratio  $J_C^{ab}/J_C^c$  was determined by evaluating  $|\Delta M|$  at H=0, followed by the use of Equation (1) to both field orientations. Fig. 4 shows that  $J_C^{ab}/J_C^c=1.5\pm0.1$ , between 5 K - 30 K and drops suddenly at T=35 K. The large error bars shown in Fig. 4 reflect essentially the uncertainty in the crystallites sizes, taken to be  $a=b=10\pm4$   $\mu m$  and  $c=2\pm1$   $\mu m$ . Based on this evaluation it is safe to conclude that we found an anisotropy ratio of the same order of the  $H_{c2}$  anisotropy, measured in the same sample[6]. A confirmation of this result through measurements taken

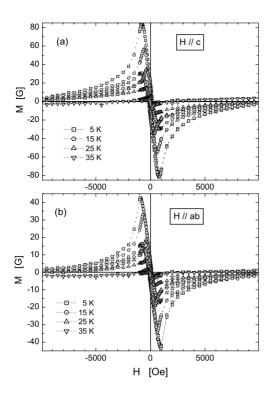


Figure 2. Magnetization loops for applied fields H (a) perpendicular and (b) parallel to the crystallites planes, after subtracting the magnetic background measured at  $T=45~\mathrm{K}$  (see text).

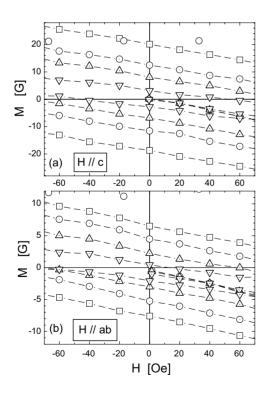


Figure 3. Enlarged view of the magnetization loops shown in Fig. 2, for fields near H=0. This plot allowed us to extract the remanent magnetization and  $\Delta M$  values for each temperature.

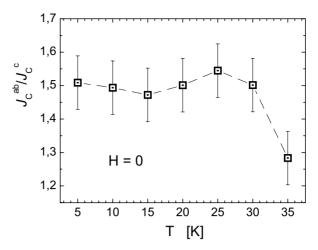


Figure 4. Critical current density anisotropy, given by the ratio between the in-plane and out of plane values, calculated from the remanent magnetization at H=0 and using the Bean model.

on larger single crystals, or well textured samples, would be highly desirable. In this case, we would expect also a large scattering of values from different works. Perhaps, these differences will be even larger than the ones reported[8, 9] for  $H_{c2}^{ab}/H_{c2}^{c}$ , since the critical current density is a very complex variable that depends also on pinning of vortices and, hence, should depend more strongly on sample quality.

Transport current measurements of the critical current density anisotropy has also been done in thin films[14], where values ranging between 2.17 and 2.88 were obtained. However, in this work they have not measured  $\gamma$  and  $J_C^{ab}/J_C^c$  in the same sample. We feel that more measurements involving these two anisotropy ratios in the same sample would be desirable.

#### 5 Conclusion

We presented here a brief review of the studies done by our group at Unicamp, on the anisotropic properties of  $MgB_2$ . We have employed samples of well aligned crystallites that fournished  $\gamma = H_{c2}^{ab}/H_{c2}^c = 1.6$  - 1.9, for temperatures varying between 32 K - 26 K. The critical current density anisotropy was found to be  $J_C^{ab}/J_C^c = 1.5 \pm 0.1$ , between 5 K - 30 K. The similarity between these values of  $\gamma$  and  $J_C^{ab}/J_C^c$  is an indication of the crystallites high purity level and low density of pinning centers. Although these anisotropy values are relatively small, compared with the values found in the cuprate superconductors[13], it is clear that some texturization technique will be required in order to produce wires and cables of  $MgB_2$  optimized for applications.

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### References

- V. Russel, R. Hirst, F. A. Kanda, and A. J. King, Acta Crystallogr. 6, 870 (1953).
- [2] M. E. Jones and R. E. Marsh, J. Am. Chem. Soc. 76, 1434 (1954).
- [3] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, Nature 410, 63 (2001).
- [4] Y. Wang, T. Plackowski, and A. Junod, Physica C 355, 179 (2001).
- [5] J. Kortus, I. I. Mazin, K. D. Belashchenko, V. P. Antropov, and L. L. Boyer, Phys. Rev. Lett. 86, 4656 (2001).
- [6] O. F. de Lima, R. A. Ribeiro, M. A. Avila, C. A. Cardoso, and A. A. Coelho, Phys. Rev. Lett. 86, 5974 (2001).
- [7] O. F. de Lima, C. A. Cardoso, R. A. Ribeiro, M. A. Avila, and A. A. Coelho, Phys. Rev. B 64, 144502 (2001)
- [8] C. Buzea, and T. Yamashita, Supercond. Sci. Technol. 14, R115 (2001).
- [9] M. Angst, and R. Puzniak, cond-mat/0305048v2 at <a href="http://arxiv.org/>(2003).">http://arxiv.org/>(2003).</a>
- [10] M. Angst, R. Puzniak, A. Wisniewski, J. Jun, S. M. Kazakov, J. Karpinski, J. Roos, and H. Keller, Phys. Rev. Lett. 88, 167004 (2002).
- [11] Yu. Eltsev, S. Lee, K. Nakao, N. Chikumoto, S. Tajima, N. Koshizuka, and M. Murakami, Phys. Rev. B 65, 140501 (2002)
- [12] O. F. de Lima, and C. A. Cardoso, Physica C 386, 575 (2003).
- [13] G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. 66, 1125 (1994).

- [14] S. Sen, A. Singh, D. K. Aswal, S. K. Gupta, J. V. Yakhmi, V. C. Sanhi, E.-Mi Choi, H.-Jin Kim, K. H. P. Kim, H. -Sook Lee, W. N. Kang, and S. -Ik Lee, Phys. Rev. B 65, 214521 (2002).
- [15] H. J. Choi, D. Roundy, H. Sun, M. L. Cohen, and S. G. Louie, Nature 418, 758 (2002).
- [16] S. Tsuda, T. Yokoya, Y. Takano, H. Kito, A. Matsushita, F. Yin, J. Itoh, H. Harima, and S. Shin, Phys. Rev. Lett. 91, 127001 (2003).
- [17] A. Gurevich, Phys. Rev. B **67**, 184515 (2003). See also a paper by A. Gurevich in this Special Volume of the Braz. J. Phys (*U-Super Proceedings*).
- [18] S. Patnaik, L. D. Cooley, A. Gurevich, A. A. Polyanskii, J. Jiang, X. Y. Cai, A. A. Squitieri, M. T. Naus, M. K. Lee, J. H. Choi, L. Belenky, S. D. Bu, J. Letteri, X. Song, D. G. Schlom, S. E. Babcock, C. B. Eom, E. E. Hellstrom, and D. C. Larbalestier, Supercond. Sci. Technol. 14, 315 (2001).
- [19] M. Tinkham, *Introduction to Superconductivity*, 2<sup>nd</sup> ed. (McGraw-Hill, New York, 1996).
- [20] L. N. Bulaevskii, V. L. Ginzburg, and A. A. Sobyanin, Zh. Eksp. Teor. Fiz. 94, 355 (1988) [Sov. Phys. JETP 68, 1499 (1988)].
- [21] M. R. Eskildsen, M. Kugler, S. Tanaka, J. Jun, S. M. Kazakov, J. Karpinski, and F. Fischer, Phys. Rev. Lett. 89, 187003 (2002).
- [22] A. E. Koshelev, and A. A. Golubov, Phys. Rev. Lett. 90, 177002 (2003).
- [23] C. P. Bean, Rev. Mod. Phys. 36, 31 (1964).