

## Lower critical field of $\text{YNi}_2\text{B}_2\text{C}$ and the influence of granularity

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We report on the determination of the lower critical field ( $H_{c1}$ ) of the newly discovered borocarbide superconductor  $\text{YNi}_2\text{B}_2\text{C}$  (YNBC). Our  $H_{c1}(T)$  data seem to fit to the relation  $H_{c1}(0) [1 - (T/T_c)^2]$  over a wide temperature range and they give  $H_{c1}(0)$  a value (22 mT) which is consistent with a recent analysis based on Ginzberg-Landau theory. A deviation from the said relationship is observed near  $T_c(0)$  and we show that it is a consequence of the intergranularity effect in a polycrystalline superconductor.

The lower critical field ( $H_{c1}$ ) is an important physical parameter of any type-II superconductor as it is the limiting field at which a flux quantum first enters such a superconductor. The experimental determination of its precise value is difficult<sup>1,2</sup> because of flux pinning and other effects related to sample shape and morphology. Some of these effects, such as bulk pinning and surface barriers, result in overestimation of  $H_{c1}$ ; others, such as the edge penetration, granularity, etc., tend to lower it. Indeed, reliable information on  $H_{c1}$ , in many cuprate superconductors is still not available. In the case of recently discovered borocarbides,<sup>3-6</sup> a large number of magnetization studies have been made on the polycrystalline samples of the first borocarbide superconductor  $\text{YNi}_2\text{B}_2\text{C}$  (YNBC),<sup>7-13</sup> and these have projected an enormous variation in the extrapolated value of  $H_{c1}$  at zero temperature  $H_{c1}(0)$ , which ranges from 80 mT (Ref. 11) to less than 4 mT.<sup>9</sup> However, one detailed analysis,<sup>8</sup> which takes the experimental thermodynamic critical field as input and uses the Ginzburg-Landau relation, projects  $H_{c1}(0)$  to be 23 mT. The quaternary YNBC alloy crystallizes in the tetragonal  $\text{LuNi}_2\text{B}_2\text{C}$  structure<sup>6</sup> and, in principle, there is a possibility of anisotropy. However, recent magnetization data on single crystals of YNBC reveal the presence of no<sup>10</sup> or only a tiny anisotropy ( $\gamma = 1.3$ ).<sup>14</sup> The spread in the quoted values of  $H_{c1}(0)$ , therefore, cannot be reconciled in terms of the difference in the reported values of  $\gamma$ . This situation makes it pertinent to examine  $H_{c1}$  carefully. To do so, we have performed extensive magnetization measurements on a good polycrystalline sample of YNBC and have pursued a systematic method to establish reliable  $H_{c1}$  values. Through comprehensive analysis of magnetization data in our sample of YNBC, we find consistency with the estimate in Ref. 8. Furthermore, we demonstrate that the establishment of  $H_{c1}$  can become rather complicated due to intergranular effects in polycrystalline samples. More importantly, we find that near  $T_c$  a new feature appears in the  $H_{c1}(T)$  data of YNBC:  $H_{c1}(T)$  declines faster than the quadratic relation  $H_{c1}(T) \propto [1 - (T/T_c)^2]$ . We show

that the anomalous increase in  $dH_{c1}/dT$  near  $T_c$  is a manifestation of intergranular effects and such a feature could be ubiquitous in polycrystalline samples of all superconductors in which grains are weakly linked.

Our polycrystalline sample of YNBC was made using an arc furnace, in the form of a cylinder (length 7.6 mm and diameter 1.2 mm) and annealed at 900°C for 1 week. The quality of our sample was ascertained via analyses of the width of x-ray powder diffraction lines corresponding to the  $\text{LuNi}_2\text{B}_2\text{C}$  structure and the superconducting transition by ac susceptibility. The isothermal and temperature-dependent measurements at small increments of field and temperature, respectively, were made using a superconducting quantum interference device magnetometer (Quantum Design model MPMS).

The magnetization values in Fig. 1 correspond to a

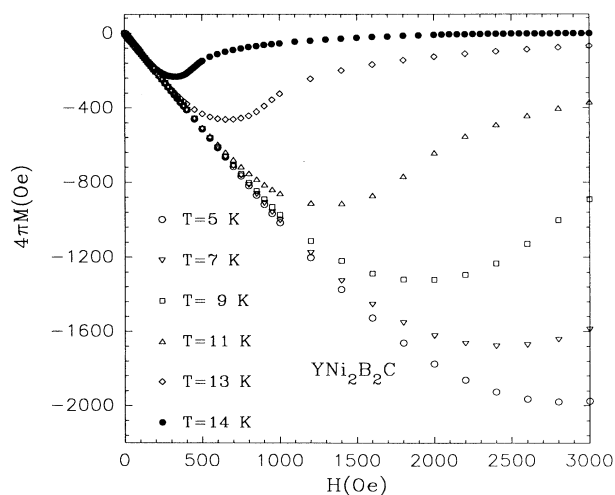


FIG. 1. Isothermal magnetization ( $4\pi M$  vs  $H$ ) at different temperatures in cylindrical sample ( $l=7.6$  mm,  $\phi=1.2$  mm) of  $\text{YNi}_2\text{B}_2\text{C}$  for  $H \parallel$  cylindrical axis.

density of  $6.03 \times 10^3 \text{ kg/m}^3$ , a value consistent with lattice constant data and perfect stoichiometry. The  $4\pi M$  vs  $H$  data in Fig. 1 confirm that the initial slope value ( $\Delta M/\Delta H$ ) does not change between 5 K and 14 K. Assuming perfect diamagnetism,  $(\Delta M/\Delta H) = 1/(1-N_{\parallel})$ , where  $N_{\parallel}$  is the demagnetization factor for  $H$  parallel to the cylinder axis, and so  $N_{\parallel} = 0.04$ . In Fig. 1, the  $4\pi M$  vs  $H$  curve for any temperature appears nominally linear up to high field values and this can be a source of higher estimates for  $H_{c1}$ . To correct for this error, we adopt the following recipe: We choose some field value  $H_0$  at each temperature and fit the initial magnetization data up to  $H_0$  using the expression  $M = a + bH$ . We then compute the difference between the observed ( $M_{\text{obs}}$ ) and the fit ( $M_{\text{fit}}$ ) values at each field and plot  $(M_{\text{obs}} - M_{\text{fit}})$  vs  $H$ . The field at which  $(M_{\text{obs}} - M_{\text{fit}})$  deviates from zero gives  $H_{c1}/(1-N_{\parallel})$ . This field has to be higher than the chosen  $H_0$  value. To cross check the  $H_{c1}$  value so determined, we have repeated this exercise for different choices of  $H_0$ . Figure 2 shows the results at 5 and 14.4 K, where we plot  $4\pi(M_{\text{obs}} - M_{\text{fit}})$  vs  $H$ . The scatter in ordinate values is only of the order of  $\pm 0.01$  mT. In Fig. 2(a), the linear fits are made for  $H_0 = 5.5, 10,$  and  $15$  mT and  $H_{c1}/(1-N_{\parallel})$  is

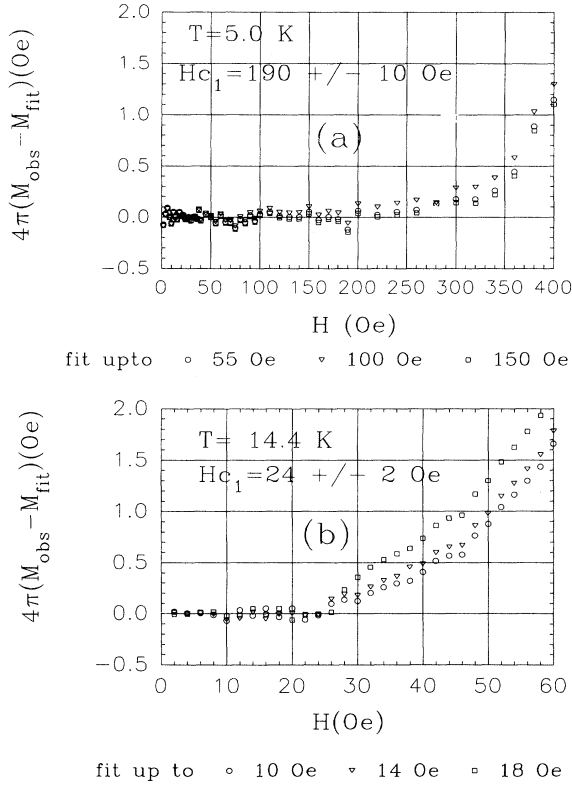


FIG. 2. Plots of  $4\pi(M_{\text{obs}} - M_{\text{fit}})$  vs  $H$  at (a) 5 K and (b) 14.4 K for different choices of  $H_0$  (as indicated), up to which linear fits are made. The lifted  $H_{c1}$  values at 5 K and 14.4 K correspond to the threshold fields above which data points systematically lie above the x axis.

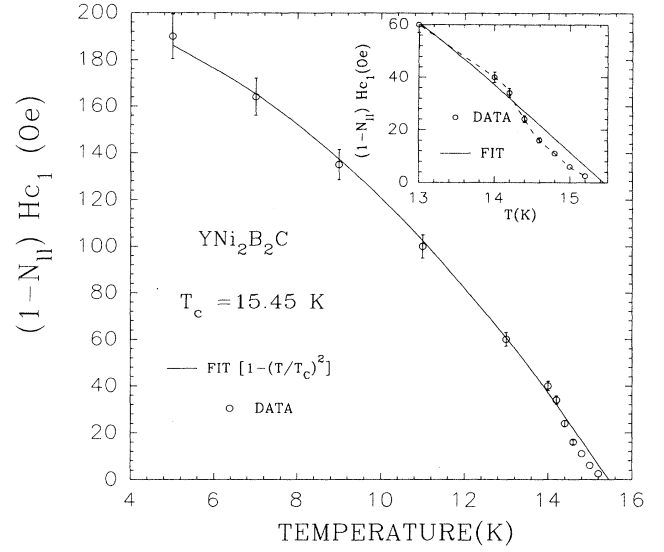


FIG. 3. Plot of  $(1-N_{\parallel})H_{c1}$  vs  $T$  in  $\text{YNi}_2\text{B}_2\text{C}$ . The solid line corresponds to the fit  $21[1-(T/T_c)^2]$  mT. The inset shows  $H_{c1}$  vs  $T$  data near  $T_c$  on an expanded scale.

$19 \pm 1$  mT at 5 K. In Fig. 2(b), the fits are made for  $H_0 = 1.0, 1.4,$  and  $1.8$  mT and  $H_{c1}/(1-N_{\parallel})$  at 14.4 K is  $2.4 \pm 0.2$  mT.

Figure 3 shows  $(1-N_{\parallel})H_{c1}(T)$  from 5 K to 15.2 K. It is clear that the data from 5 K to 14 K fit the relation  $(1-N_{\parallel})H_{c1}(T) = 21\{1-[T/T_c(0)]^2\}$  mT, which gives  $H_{c1}(0)$  to be about 22 mT in agreement with an estimate

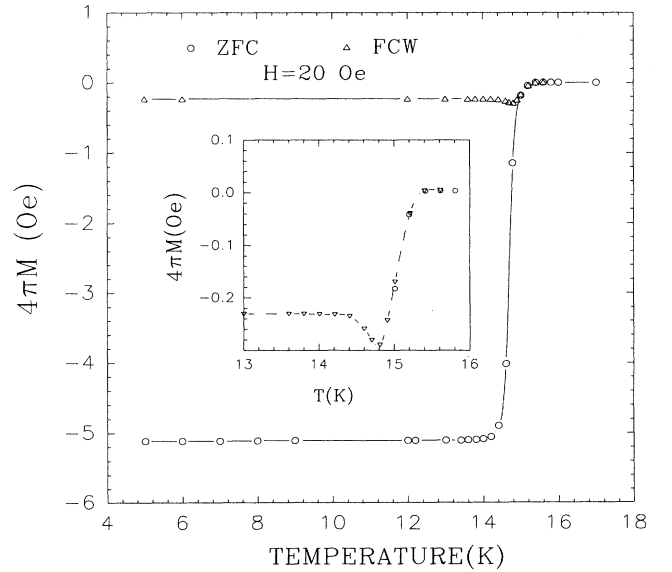


FIG. 4. Temperature variation of zero-field-cooled (ZFC) and field-cooled warmup (FCW) magnetization values in  $\text{YNi}_2\text{B}_2\text{C}$  in 20 Oe. The inset shows the negative peak in FCW data on an expanded scale.

made<sup>8</sup> using thermodynamic critical field data. We think the good fit of our  $H_{c1}(T)$  data to the relationship  $H_{c1}(0)\{1 - [T/T_c(0)]^2\}$  and the consistency of these data with the thermodynamical critical field data<sup>8</sup> represent that our  $H_{c1}(T)$  values are intrinsic to YNBC. The inset in Fig. 3 shows that the fit curve lies above the measured  $H_{c1}(T)$  values for  $14.4 \text{ K} < T < T_c(0)$ . The observation of a sharper drop in  $H_{c1}(T)$  above 14.4 K is significant, as similar behavior in  $H_{c1}(T)$  data (for  $H \parallel c$ ) was seen by Brawner *et al.*<sup>15</sup> for a single crystal of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212). The Bi2212 is a two-dimensional (2D) superconductor, with the  $\text{CuO}_2$  planes Josephson coupled to each other. The faster decline of  $H_{c1}(T)$  values in Bi2212 just below  $T_c$  is thought to be a result of either fluctuations in the  $\text{CuO}_2$  planes<sup>16</sup> or the appearance of weak links.<sup>17,18</sup> In contrast, the YNBC system, though layered, is nearly isotropic, and has a coherence length much larger than the interlayer spacing,<sup>8</sup> hence, the presence of fluctuations can be discounted. The presence of weak links appears possible in YNBC as it is a brittle intermetallic compound. Such weak links should leave their imprint in the magnetization data recorded under different thermomagnetic histories. We, therefore, purposefully examined such data in our YNBC sample.

Figure 4 shows zero-field-cooled (ZFC), measured upon warming, and field-cooled warmup (FCW), also measured upon warming, magnetization data in a field of 2 mT.

The FCW curve reveals the presence of an anomalous negative peak effect (see inset of Fig. 4), i.e., an increase in diamagnetic response upon increasing the temperature above 14.4 K. Such an effect, noted in polycrystalline samples of a conventional superconductor  $\text{Nb}_3\text{Sn}$  (Ref. 19) and ceramic samples of oxide superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_7$ ,<sup>20</sup> has been interpreted in terms of expulsion of additional magnetic flux as the sample warms up due to interplay between the magnetic responses from intragrain and intergrain regions.<sup>18-20</sup> The onset temperature of the negative peak in YNBC is seen to coincide with the temperature at which  $H_{c1}$  starts to deviate from quadratic behavior [cf. inset of Fig. 3 and inset of Fig. 4]. We conclude that just as the negative peak effect, the drop in  $H_{c1}(T)$  is also caused by weak links. The values of  $H_{c1}$  above 14.4 K thus correspond to preferential penetration of the field in the intergranular regions. We believe that the fit line represents the field values at which the field would penetrate the intragrain regions. In this sense,  $H_{c1}(T)$  data above 14.4 K are not representative of the bulk response of YNBC.

To conclude, we have measured  $H_{c1}(T)$  in the borocarbide compound  $\text{YNi}_2\text{B}_2\text{C}$ . These data fit the relationship  $H_{c1}(0) [1 - (T/T_c)^2]$  over a wide temperature range, and give an  $H_{c1}(0)$  value consistent with an earlier estimate.<sup>8</sup> A departure from quadratic relationship is observed near  $T_c$ , which is ascribed to weak links.

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