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Precursor diamagnetism above the superconducting transition in YNi_2B_2C

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Abstract. High-resolution SQUID magnetization measurements in a single crystal of YNi_2B_2C around the superconducting transition are reported. The diamagnetic magnetization $-M_{fl}$ at constant field H as a function of temperature and isothermal magnetization curves $-M_{fl}$ *vs.* H are used to derive insights on precursor phenomena approaching the bulk transition temperature $T_c(H = 0) = 15.25 \pm 0.02$ K. The precursor diamagnetism is found strongly enhanced with respect to the conventional Ginzburg-Landau value for Gaussian fluctuations and the curves $-M_{fl}$ *vs.* H exhibit an upturn with the field and hysteretic effects up to $T^* = 15.4$ K. These results are interpreted in terms of a non-zero order parameter in superconducting droplets above the bulk T_c. These droplets are likely to be related to inhomogeneities resulting from small amount of boron to carbon substitutions.

PACS. 74.40.+k Fluctuations (noise, chaos, nonequilibrium superconductivity, localization, etc.) – 74.20.De Phenomenological theories (two-fluid, Ginzburg-Landau, etc.) – 74.25.Ha Magnetic properties

1 Introduction

Intermetallic borocarbide metals have attracted a great deal of interest as possible exotic superconductors, due to the high transition temperature for multielement intermetallic compounds and for the interplay with the magnetic properties related to the presence of nickel and also of magnetic rare earths. Therefore these materials appear in between traditional superconductors and high T_c cuprates. Reviews on the transport and superconducting properties of RNi_2B_2C, with R = Y and Lu, can be found in references [1,2].

One relevant aspect of superconductivity that has not yet been explored in intermetallic borocarbide metals is the one involving the superconducting fluctuations and the related diamagnetism, when fluctuating Cooper pairs are created on approaching T_c from above [3]. In cuprates and in the high temperature BCS superconductors such as MgB_2, interesting insights have recently been achieved from the study of fluctuating diamagnetism (FD) [4,5].

In this paper high-resolution magnetization measurements in a single crystal of YNi_2B_2C are reported. The diamagnetic magnetization at the zero-field transition temperature $T_c(H = 0)$ is found to be strongly enhanced with respect to the contribution expected from superconducting fluctuations of Ginzburg-Landau (GL) character. In a temperature range of about 0.2 K above the bulk T_c the isothermal magnetization curves $-M_{fl}$ *vs.* H display an upturn with the field H and history-dependent effects.

The experimental findings are discussed in the light of charge inhomogeneities and spatial fluctuations of $T_c's$ resulting from boron to carbon substitutions in the lattice positions.

2 Experimental results

The single crystal of YNi_2B_2C has been prepared and characterized as described in reference [2] and references therein.

Magnetization measurements have been carried out by means of the Quantum Design MPMS-XL7 SQUID, most with field H applied along the c-axis. A few measurements along the a and b axes confirmed the moderate anisotropy of YNi_2B_2C [1]. The temperature stabilization and resolution were better than 0.01 K for $T < 20$ K.

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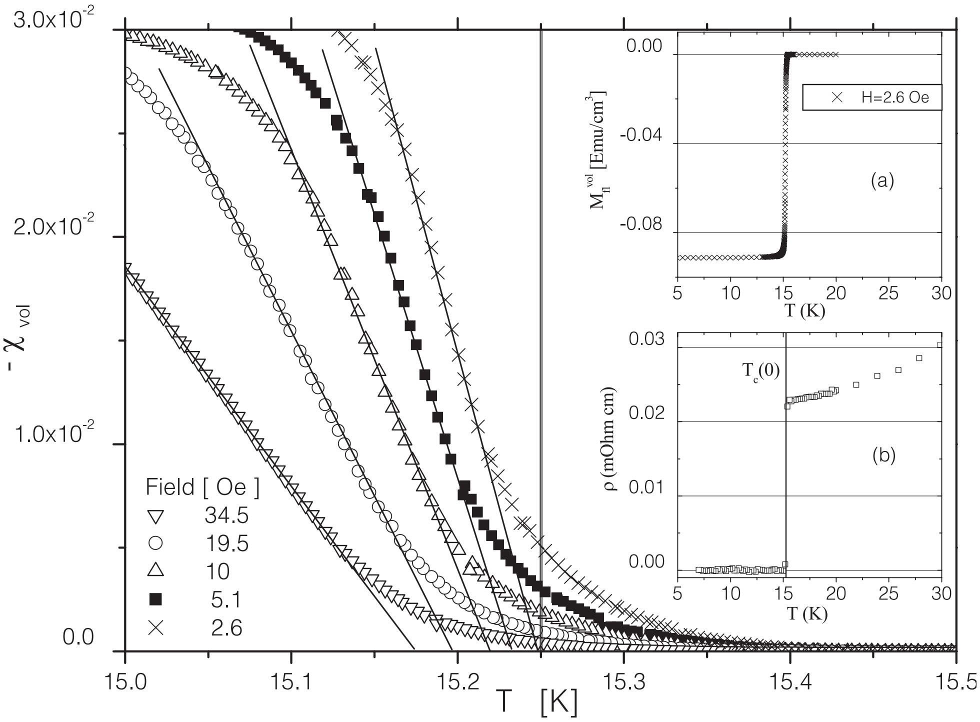

Fig. 1. $-\chi = -M/H$ for $H = 2.6$ Oe, as a function of temperature and evaluation of the transition temperature $T_c(H)$. One derives $T_c(0) = 15.25 \pm 0.02$ K. In the insets, the behaviors of the total magnetization (a) and of the resistivity (b) measured in a rather wide temperature range, are reported.

From magnetization curve M *vs.* H up to the field $H = 6$ tesla, at $T \simeq 300$ K, the volume Pauli susceptibility $\chi_P = 8 \times 10^{-6}$ was extracted. For $H \leq 0.5\,\mathrm{T}$ a steep increase of M with the field, fastly saturating, was detected and attributed to ferromagnetic impurities. Assuming a magnetic moment of 1 Bohr magneton, the amount of impurities turns out to be around 70 ppm. In the magnetization measurements from $T \simeq 25$ K down to $T \simeq 15.8$ K, the Pauli and impurities contributions M_P and M_{imp} were found practically temperature independent. Then from the raw data of the magnetization around T_c, the contribution $(M_P + M_{imp})$ was eliminated by subtracting the value of M measured at $T \simeq 15.8$ K, where the fluctuating negative magnetization $-M_{fl}$ is practically zero.

In Figure 1 we show how the transition temperature $T_c\,(H\,)$ was obtained from the extrapolation at zero magnetization of the behavior in the region where $|-M|$ decreases linearly with T. The Meissner fraction was estimated to be 45% (inset a). The sharpness of the transition for the bulk sample is also indicated by the resistivity data (inset b). The bulk transition temperature at zero field is evaluated $T_c(H \to 0) = 15.25 \pm 0.02$ K.

The field dependence of T_c is in agreement with the data of Figure 7 in reference [1]. A detailed temperature behavior in the range 1 Oe $\leq H \leq$ 1 KOe shows a crossover region, with positive curvature H_{c2} *vs.* T. For $H \leq 100$ Oe one has a linear behavior of H_{c2} *vs.* T, with $(dH_{c2}/dT)_{T\longrightarrow T_c} = -0.4$ kOe/ K, (field along the c-axis).

According to the GL theory for FD in finite field [6,7] and to scaling arguments in isotropic or moderately anisotropic systems [8,9] the scaled magnetization $m = -M_{fl}/\sqrt{H}T_c$ should be $m = ck_B/\phi_0^{\frac{3}{2}}$, with $c = 0.32$ for 3D system and $c = 0.32\,\gamma$, with γ anisotropy parameter, in 3D anisotropic crystals. As it will be discussed in the following the reduced magnetization at T_c exceeds by a large factor the GL value, being about a factor 6 larger than in optimally doped YBCO (Fig. 2).

3 Analysis of the data and summarizing conclusions

The analysis of the experimental findings should first be attempted on the basis of the Ginzburg-Landau theory in finite fields, for Gaussian fluctuations in anisotropic systems [6,7,10,11]. According to the GL scenario the fluctuating diamagnetic magnetization should be

$$-M_{fl}(\epsilon, H) = \frac{3\sqrt{\pi}}{\phi_o^{\frac{3}{2}}}\,k_B\,T\,\sqrt{H}\,\zeta\left(-\frac{1}{2}, \frac{1}{2}, +\frac{\phi_0\epsilon}{4\,\pi\,\xi^2(0)\,H}\right) - \frac{k_B\,T\,\epsilon}{4\,\xi^2(0)\,\sqrt{\pi}\,\phi_0\,H}\,\zeta\left(+\frac{1}{2}, \frac{1}{2}, +\frac{\phi_0\,\epsilon}{4\,\pi\,\xi^2(0)\,H}\right) \quad (1)$$

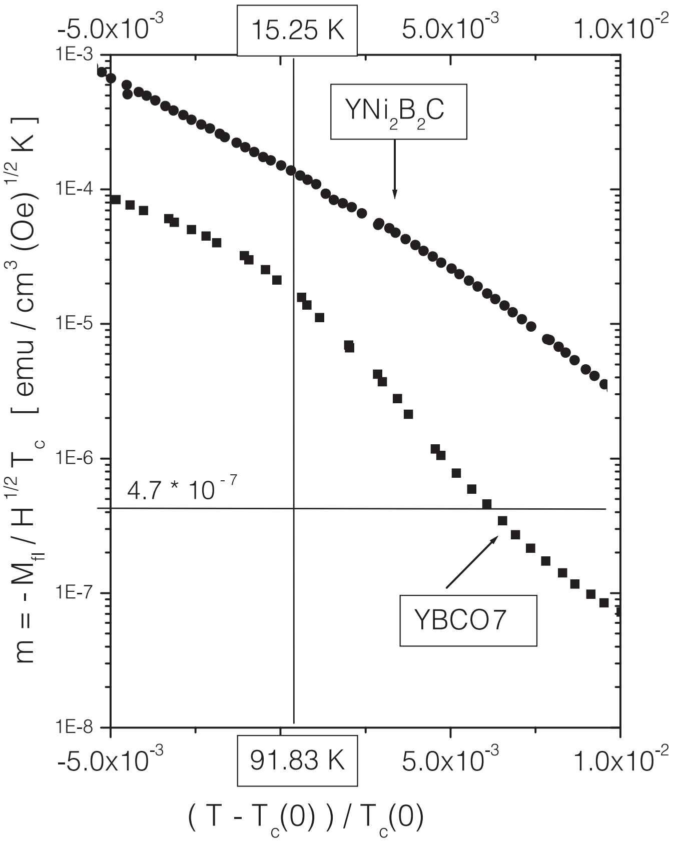

Fig. 2. Comparison of the reduced magnetization $m = -M_{fl}/\sqrt{H}T_c$, measured in a field of 34.5 Oe along the c-axis in oriented powder of optimally doped YBCO and in the single crystal of YNi_2B_2C, as a function of the reduced temperature $\epsilon = \frac{T-T_c(0)}{T_c(0)}$. The theoretical value (4.7×10^{-7}) for isotropic 3D conventional superconductors (see Ref. [3]) is indicated.

where $\epsilon = (T - T_c(0))/T_c(0)$, $\xi(0)$ is the coherence length and ζ the Hurwitz function [10].

From the comparison of the experimental data with the magnetization expected according to equation (1) (Fig. 3), one can see that the experimental results are well beyond any possible prediction based on Gaussian GL model. The shift of the theoretical behavior, by $\Delta = +0.2$ K (solid line in the inset) indicates that the range of validity of the Gaussian approximation could possibly occur only in a temperature range where $-M_{fl}$ is larger than the SQUID noise only by a factor around 2÷3.

A significant way to analyze the detail of the FD is to look at the isothermal magnetization curves $-M_{fl}(T = \text{const.})$ *vs.* H (Fig. 4). A remarkable difference with respect to the predictions based on equation (1) is noted. Not only the magnetization values for $H \leq 10$ K are quantitatively well outside the theoretical estimates, but also the field dependence is very different. In fact $-M_{fl}$ show an upturn at a field $H_{up} \simeq 10$ Oe and it is practically quenched for H around $1 \div 2$ kOe.

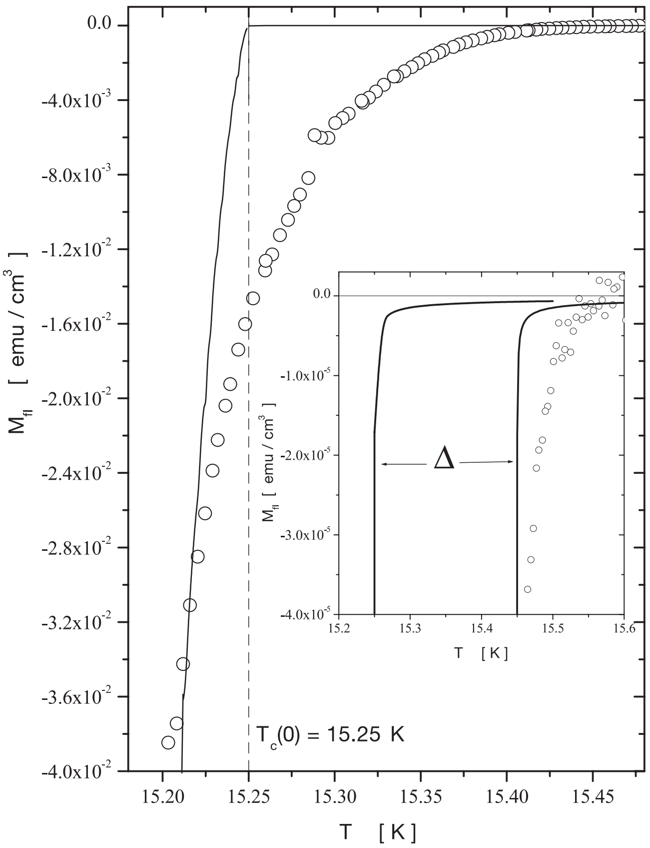

Fig. 3. Comparison of the experimental data for the fluctuating magnetization measured in YNi_2B_2C, in a field $H = 5$ Oe along the c-axis, around the transition temperature with the theoretical behavior (solid line) expected from equation (1) in the text. The prediction for Gaussian GL fluctuations is more than an order of magnitude smaller than the experimental data (blow up in the inset). In the inset the theoretical line resulting from equation (1) in the text has been moved by $\Delta = +0.2$ K and again compared with the experimental results.

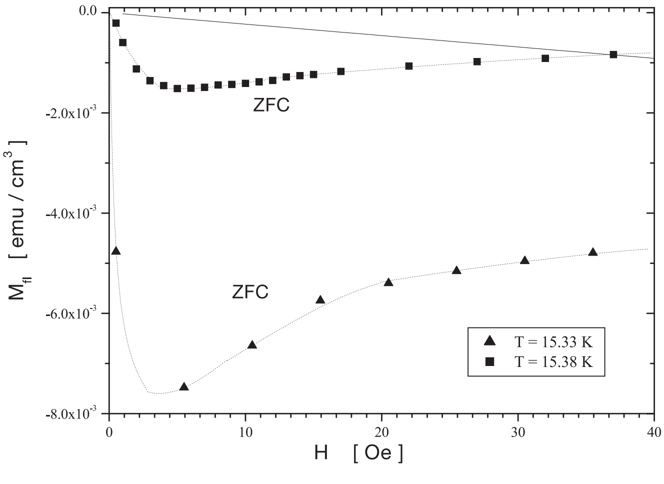

Fig. 4. Zero-field cooled isothermal magnetization curves $-M_{fl}$ *vs.* H (along the c-axis) in YNi_2B_2C, for temperature above the bulk $T_c(0) = 15.25 \pm 0.02$ K. The upward solid line is the theoretical behavior according to equation (1) in the text (increased by a factor 100), while the other lines are guides for the eye.

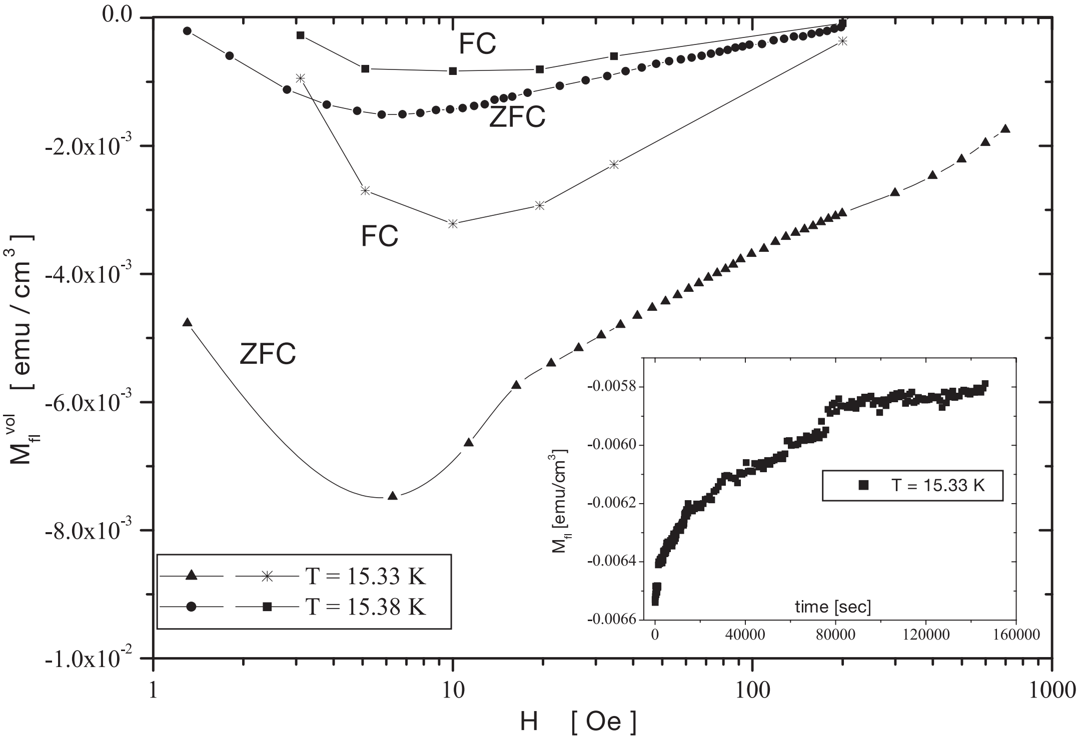

Fig. 5. Comparison of the FC and ZFC magnetization curves at $T = 15.33$ K and $T = 15.38$ K. In the inset the evolution with time of the magnetization measured after cooling in zero field and then applying a field $H = 10$ Oe is reported, for $T = 15.33$ K. The solid lines are guides for the eye.

In the GL weak fluctuation regime a saturation of the magnetization at high field has to be expected [10,12], the superconducting coherence being broken for fields larger than $H_{c2}(0)\sqrt{\epsilon}$. However, the decrease of $-M_{fl}$ on increasing field can be justified only by taking into account the short-wavelength fluctuations or within the framework of exactly solvable, for any magnetic field, zero-dimensional ($D = 0$) models [3,7]. An estimate of the order of magnitude of the upturn field H_{up} can be obtained by the assumption of fluctuation-induced superconducting droplets of radius d smaller than the coherence length $\xi(T)$, so that the order parameter is homogeneous. Then the exact solution of the GL model for any field not close to H_{c2}, yields [7]

$$M_{fl} = -\frac{k_B\, T\, \frac{2}{5}\, \frac{\pi^2\, \xi_0^2}{\phi_0^2}\, d^2\, H}{\epsilon + \frac{\pi^2\, \xi_0^2}{5\, \phi_0^2}\, H^2\, d^2}. \qquad (2)$$

By considering that the most sizeable contribution to FD comes from droplets of radius $d \sim \xi(T)$, extending the $D = 0$ condition to these droplets, then from equation (2) one would derive an upturn field $H_{up} \approx \epsilon\, \phi_0/\xi^2(0)$. For ϵ in the range $10^{-2} \div 10^{-3}$ and $\xi(0) \approx 70$ Å [1] one should have $H_{up} \approx (0.5 \div 5)$ KOe, a value much larger than the ones in Figure 4, where H_{up} is almost T-independent. Although the zero-dimensional assumption is obviously rather crude, the considerations given above prove that the upturn field observed in YNi_2B_2C is not related to the effect of the field on the life time of the Cooper pairs.

An experimental observation which helps in clarifying the character of the SC droplets is the one involving history-dependent effects. As shown in Figure 5, the zero field cooled (ZFC) magnetization curves are different from the one obtained by cooling at the same temperature in the presence of a given value of H (FC). Furthermore relaxation effects are observed (inset in Fig. 5). The negative magnetization detected at a given temperature, after zero-field cooling, is time-dependent and displays a progressive decrease from the ZFC value toward the one measured in FC condition. The magnetic-history dependent effects have been found to occur up to an irreversibility temperature $T_{irr} \simeq 15.4$ K. In the narrow temperature range above T_c that could be explored, the upturn field H_{up} was found practically temperature independent. We remark that in case of diamagnetism related to phase fluctuations, at variance H_{up} increases with T [13].

From these observations one is led to conclude that a large part of the diamagnetism detected in the temperature range of about 0.2 K above the bulk transition temperature $T_c(0)$ must be attributed to non-percolating superconducting regions having a local T_c higher than the bulk transition temperature. These SC "droplets" represent a kind of precursor diamagnetism, similar to one occurring when the order parameter is locally different from zero as for instance in Al-doped MgB_2 [14]. For YNi_2B_2C

is feasible that we are observing some “frozen” superconducting fluctuations related to spatial dependence of the transition temperature due to microscopic defects. Small amounts of boron to carbon substitutions in the lattice positions are the most probable source of such small random distribution of T_c’s.

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