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Coexistence of superconductivity and magnetism in $Y_{1-x}Dy_xNi_2B_2C$ single crystals

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Abstract

Single crystals of $Y_{1-x}Dy_xNi_2B_2C$ were obtained in the wide concentration range ($0 < x < 0.95$). Quality of plate-like crystals was examined by X-ray diffraction and XPS measurements. The superconducting and magnetic properties of the samples were determined by means of magnetisation measurements as a function of temperature and magnetic field. The intrinsic properties of $Y_{1-x}Dy_xNi_2B_2C$ superconductors are strongly influenced by the magnetic subsystem. The effect of the 4f moment ordering manifests in the suppression of the diamagnetic signal and the shift of the superconducting transition towards the lower temperatures. In the FC plots paramagnetic magnetisation is observed in the Dy substituted superconductors. The applied magnetic field can induce metamagnetic transition in the Y–Dy magnetic sublattice. A high anisotropy in superconducting as well as magnetic properties of pseudoquaternary $Y_{1-x}Dy_xNi_2B_2C$ single crystals is found. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: High- T_c superconductors; Crystal growth; X-ray diffraction; Magnetisation; Anisotropy; Magnetic measurements

1. Introduction

Since their discovery in 1994, the quaternary borocarbides $RENi_2B_2C$ ($RE=Y, La$ or the other rare earth) [1] have been intensively investigated because of their interesting superconducting and magnetic properties. Relatively few studies have been done on single crystals [2,3]. YNi_2B_2C is a superconductor with a relatively high superconducting transition temperature $T_c = 16$ K. For the magnetic rare earth substitutions superconductivity is also detected. $DyNi_2B_2C$ shows superconductivity below anti-ferromagnetic Neel temperature ($T_c = 6$ K, $T_N = 10.5$ K [3]). The $Y_{1-x}Dy_xNi_2B_2C$ system allows investigation of the coexistence of superconductivity and magnetism which is one of the fundamental problems in condensed matter physics.

2. Experimental

$Y_{1-x}Dy_xNi_2B_2C$ compounds were prepared by arc melting of high purity constituents and followed by long annealings at 920°C for 1 week. It was possible to find small, well-formed single crystals which have been ex-

tracted. Quality of the crystals was examined by X-ray diffraction and stoichiometry was tested by XPS measurements. The XPS measurements were carried out with a Multitechnique Electron Spectrometer PHI 5700/660 from Physical Electronics using a monochromatized Al $K\alpha$ radiation of 1486.6 eV. The samples were broken or scraped with a diamond file under ultra high vacuum conditions. Some samples were sputtered using Ar or Xe ions. The magnetic measurements were carried out as a function of external magnetic field in the temperature range 1.7–300 K using a SQUID magnetometer. Measurements of dc magnetisation were performed in a magnetic field applied parallel and perpendicular to the c -axis of the single crystals under zero-field and field cooling conditions.

3. Results and discussion

3.1. Physical characterisation of the samples

The samples consisted of large grains. Small plate-like crystals were found at the top of the samples (see Fig. 1). These plates with dimensions of about $1 \times 1 \times 0.1$ mm³ (Fig. 1a) were extracted from the samples for further investigations. The crystal orientation and quality were

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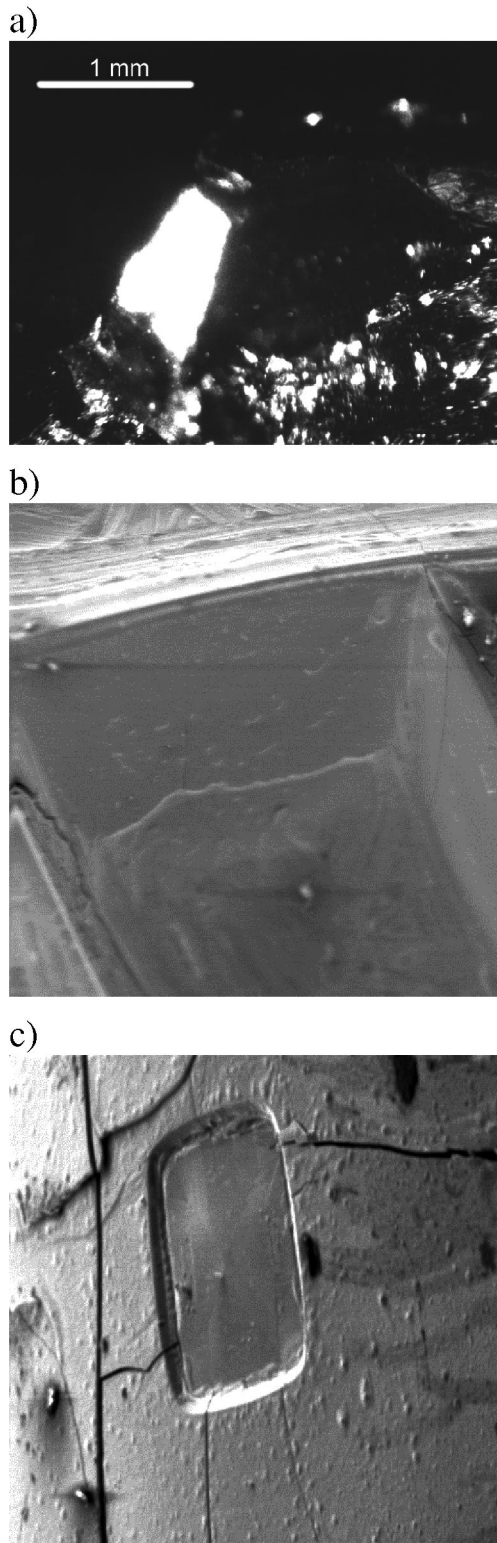


Fig. 1. (a) 'As-grown' plate-like $\text{DyNi}_2\text{B}_2\text{C}$ crystal on the surface of an arc-melted sample; (b) S.E.M. pictures of plate-like crystal $\text{Y}_{0.5}\text{Dy}_{0.5}\text{Ni}_2\text{B}_2\text{C}$; (c) $\text{Y}_{0.6}\text{Dy}_{0.4}\text{Ni}_2\text{B}_2\text{C}$ — S.E.M. picture of a small rectangular plate on the surface of a greater plate.

examined using the Laue method. The plate-like crystals were single crystals, where the c -axis was perpendicular to the plate surface. The plates were (001) oriented and the common a long edge in plane was parallel to the crystallographic direction [110]. Optical microscopy and scanning electron microscopy observation on these crystals does not show the presence of any second phase (Fig. 1b). Sometimes there were found small rectangular plates which grew on the surface of a greater plate of the same compound (Fig. 1c).

These compounds crystallise in the layered tetragonal body-centred $\text{LuNi}_2\text{B}_2\text{C}$ -type structure. X-ray powder diffraction measurements on ground samples confirmed generally the formation of single-phase $\text{Y}_{1-x}\text{Dy}_x\text{Ni}_2\text{B}_2\text{C}$ solid solutions. Some weak impurity lines identified as $\text{Y}_{1-x}\text{Dy}_x\text{NiBC}$ were found with approximately 4% of the total intensity. Lattice parameters of the powdered samples with various concentrations of Dy in the $\text{Y}_{1-x}\text{Dy}_x\text{NiBC}$ system (see Fig. 2) nearly obey Vegard's law (including the Dy-rich samples). A marked change in the c -parameter and a negligible change in a -parameter were obtained when the Y atoms substituted by the Dy ones. It is worth to note that the ratio of c/a parameters increases monotonically with decreasing the Dy concentration in the $\text{Y}_{1-x}\text{Dy}_x\text{NiBC}$ system. This effect is caused by the different atomic radii of Dy and Y as well as by a change in numbers of

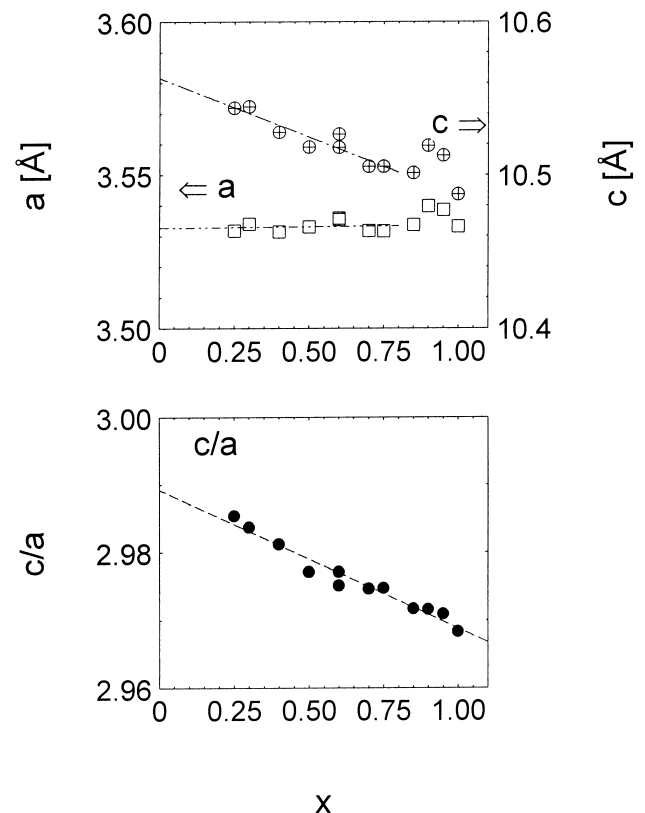


Fig. 2. Lattice parameters a , c , and ratio c/a for $\text{Y}_{1-x}\text{Dy}_x\text{Ni}_2\text{B}_2\text{C}$ versus Dy concentration.

electrons in the conductivity bands of the samples with various Dy concentrations.

3.2. Superconducting and magnetic properties of $Y_{1-x}Dy_xNi_2B_2C$

The representative results of magnetic measurements as a function of temperature and applied field for the samples of $Y_{1-x}Dy_xNi_2B_2C$ with $x=0.25$, 0.4 and 0.6 are presented in Figs. 3 and 4.

Fig. 3 shows the temperature dependence of the dc magnetic susceptibility measured in a magnetic field of 50 Oe, applied perpendicular and parallel to the c -axis of the crystal under zero-field cooling (ZFC) and field cooling (FC) conditions.

Superconductivity was detected for all zero-field cooled samples. As one can see the diamagnetic signal decreased with increasing Dy content in pseudoquaternary $Y_{1-x}Dy_xNi_2B_2C$ compounds. Temperature in which the diamagnetic contribution vanished in ZFC plots could be taken as the superconducting transition temperature T_c . The small transition widths $T_{c,90\%} - T_{c,10\%}$ (0.5 and 1 K for $x=0.25$ and 0.6, respectively) indicate the good quality of crystals. The superconducting transition shifts monotonically to lower temperatures with increasing Dy content for $x < 0.7$ according to Abrikosov-Gorkov theory. State the anomalies in the 'Tc' for high Dy contents is observed [4]. The superconducting transition temperature is not only a function of x , but also depends on the direction of the applied magnetic field. The observed anisotropy is due to the characteristic layered structure of the $RENi_2B_2C$ system, which may be viewed as a stacking of Ni_2B_2 and REC layers [1]. The high magnetic anisotropy is a noticeable feature of both ZFC and FC plots. The FC plots obtained in the same field (50 Oe) show anomalous behaviour, with positive values below T_c . There appears to be a strong paramagnetic contribution in the superconducting state due to the ordering the Dy moments in $Y_{1-x}Dy_xNi_2B_2C$ system. Probably a paramagnetic magnetisation is originated by the superposition of the paramagnetic signal of the Dy ions in the mixed state of the superconductor. It should be noticed that positive contribution is more pronounced when a magnetic field is applied in base plane while the stronger diamagnetic signal occurs for a magnetic field applied parallel to the c -axis. Similar anisotropy behaviour was reported earlier for $Bi_2Sr_2CaCu_2O_x$ superconductor [5]

Fig. 4a presents magnetisation curves measured at 1.8 K versus applied field. The plotted curves are composed of two components: one typical of superconducting system and second, due to the antiferromagnetically ordered system. One can see that the introduction of the magnetic atoms of Dy decreases markedly the superconductivity (Fig. 4b). Then the magnetic field induces some magnetic ordering of Dy moments. For $Y_{0.75}Dy_{0.25}Ni_2B_2C$ applica-

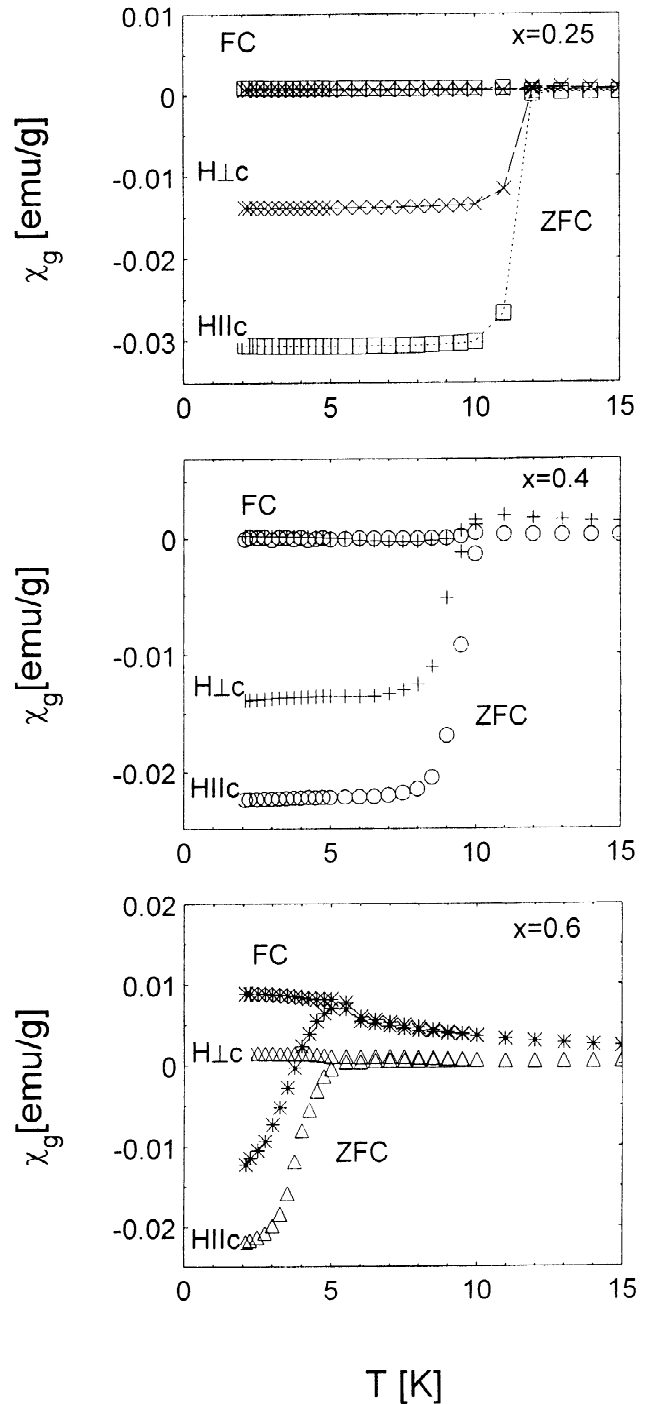


Fig. 3. Temperature dependent magnetic susceptibility of $Y_{1-x}Dy_xNi_2B_2C$ measured at 50 Gs.

tion of sufficiently strong a magnetic field parallel to the c -axis causes a metamagnetic transition probably due to ordering of the Dy moments. Antiferromagnetic Neel temperature can be measured for the Dy-rich compositions [4]. For the presented compositions, superconductivity coexists with antiferromagnetic order. Magnetisation

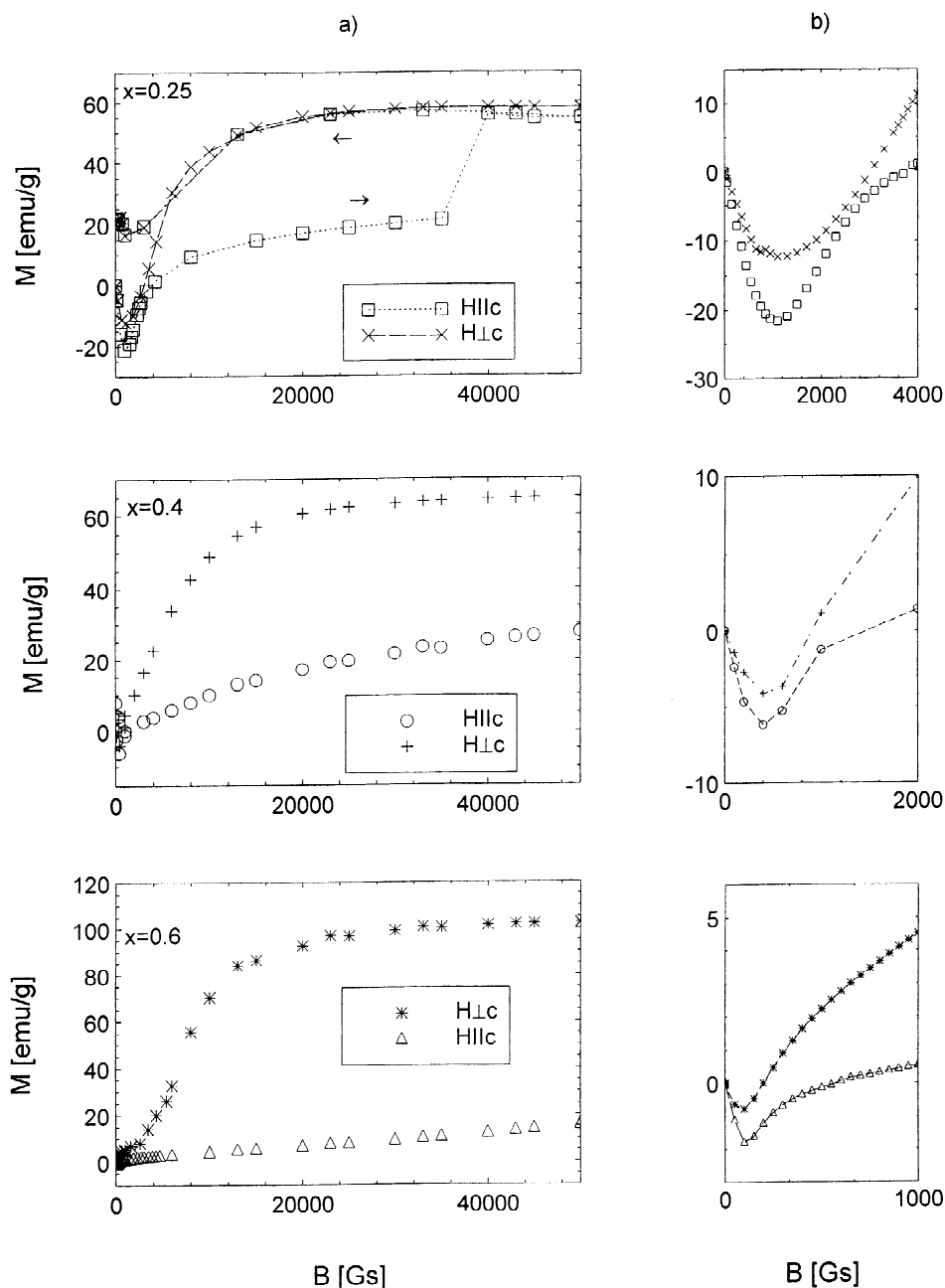


Fig. 4. (a) Magnetisation isotherms of $Y_{1-x}Dy_xNi_2B_2C$ at $T=1.8$ K. (b) Superconducting component of magnetisation curve for $Y_{1-x}Dy_xNi_2B_2C$.

curves reveal the high magnetocrystalline anisotropy that is enhanced with increasing Dy content in the compound.

4. Conclusion

The intrinsic properties of $Y_{1-x}Dy_xNi_2B_2C$ superconductors are strongly influenced by the magnetic subsystem. The effect of the 4f moment ordering manifests itself as a suppression of the shielding signal and as a decrease of the superconducting temperature. The paramagnetic magneti-

sation is observed in all the Dy substituted superconductors. In some cases a metamagnetic transition occurs due to the spin flip. The pseudoquaternary $Y_{1-x}Dy_xNi_2B_2C$ system studied exhibits a high anisotropy in both its superconducting and magnetic behaviour.

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