Preparation of c-axis fibre textured YNi_2B_2C thin films by pulsed laser deposition: film structure and superconducting properties

K. Häse, S. C. Wimbush, S. Paschen and B. Holzapfel

***Abstract*—Thin films of YNi_2B_2C have been prepared in situ by pulsed laser deposition onto MgO(100) substrates. The films exhibit c-axis fibre textured but without in-plane texture. The superconducting transition temperature T_c increases with increasing deposition temperature up to 14.7K at a deposition temperature of 735°C. We observed high values of the upper critical field H_{c2} in thin films of about 7T at 3K and the expected temperature dependence of H_{c2}. An anisotropic behavior of H_{c2} was measured for the different temperatures 2K, 6K and 10K. The critical current density j_c shows different dependencies in applied magnetic fields H||c and H⊥c at 3K.**

***Index Terms*—borocarbides, thin films, upper critical field**

I. INTRODUCTION

THE class of quaternary intermetallic superconductors of the RNi_2B_2C (R - rare earth) type, first discovered in 1994 [1, 2], is of primary interest due to its exhibition of the interplay between superconductivity and magnetic ordering phenomena. The series displays several arrangements of magnetic ordering temperature and superconducting transition temperature, including no magnetic ordering, which is the case for YNi_2B_2C. This compound is ideal, therefore, to study the fundamental superconducting properties of the series, uninfluenced by magnetic effects. The nonmagnetic members of the series were initially classified as conventional BCS type superconductors having a moderately large density of states at the Fermi level, close to the traditional A-15 superconductors, and a relatively strong electron-phonon coupling constant [3, 4]. In recent years with the availability of high-quality single crystal samples, the situation has changed somewhat as some properties of YNi_2B_2C and $LuNi_2B_2C$ were observed which might be interpreted as indications of unconventional (*d*-wave) superconductivity [5], for example an anisotropy of the upper critical field within the basal plane of $LuNi_2B_2C$, a quadratic flux line lattice at high magnetic fields and an unconventional behaviour of the specific heat [for an overview see Ref. 6. and references therein]. Several groups are involved in the preparation of thin films of these materials by sputtering and pulsed laser deposition techniques [7, 8, 9]. The availability of c-axis oriented or epitaxial thin films enables important experiments to be performed probing the anisotropic properties of the material. Here, we report on the preparation and anisotropic superconducting properties of c-axis fibre textured YNi_2B_2C thin films prepared by pulsed laser deposition.

II. EXPERIMENTAL

Thin films of YNi_2B_2C were deposited using a KrF excimer laser (wavelength 248nm) in an ultra high vacuum system (base pressure $2x10^{-7}$ Pa). The deposition conditions comprised a target-substrate separation of 4cm, a laser pulse repetition rate of 10Hz, and an incident energy density of 8 J/cm^2 onto the target. Polycrystalline YNi_2B_2C targets were obtained by arc melting of a stoichiometric mixture of elemental powders of Y (purity 99.9%), Ni (99.99%), B (99.0%) and C (99.9%) in an inert atmosphere of flowing argon. Single crystal MgO (100) substrates were positioned

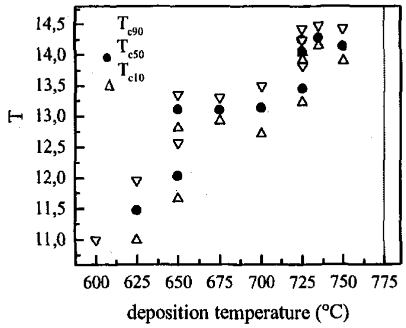

Fig. 1. Inductively measured transition temperature of YNi_2B_2C thin films vs. deposition temperature

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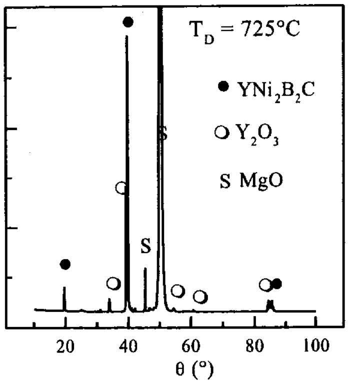

Fig. 2. X-ray diffraction pattern in Bragg-Brentano geometry (CoK_{α} radiation) of an YNi_2B_2C film deposited at 725°C (● YNi_2B_2C, O Y_2O_3 and S substrate reflections)

in an on-axis geometry and heated with a radiation heater. The deposition temperature was estimated using a thermocouple positioned at the rear of the heater. The crystal structure of the films was determined using X-ray diffraction in Bragg-Brentano geometry (CoK_{α} radiation), by grazing incidence diffraction (CuK_{α}) and texture measurements (CuK_{α}). The microstructure was examined with scanning electron microscopy (SEM). Transition temperatures were inductively measured by a magnetic alternating field shielding technique. Transition temperatures measured by this technique are systematically lower than resistively measured transition temperatures. Electrical transport measurements were performed using a four-point contact method on a photolithographically patterned bridge, which was 20μm wide and 600μm long, in a superconducting 8T split-coil magnet system.

III. RESULTS AND DISCUSSION

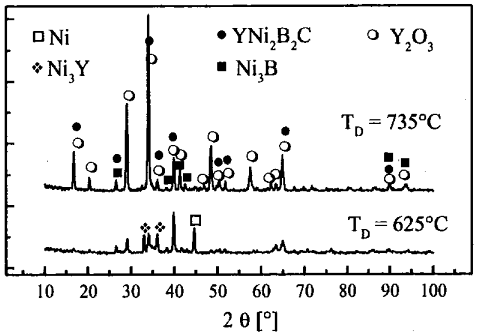

Fig. 3. Grazing incidence x-ray diffraction patterns of YNi_2B_2C films deposited at 625°C and 735°C (CuK_{α} radiation, incident angle ω=4°)

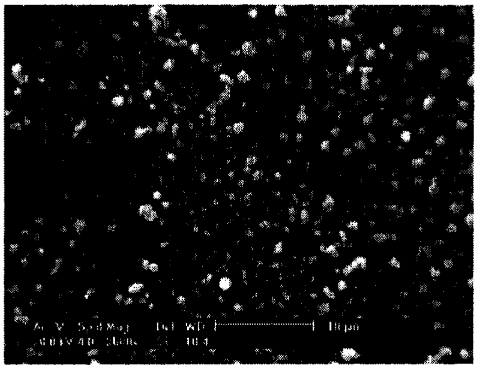

Fig. 4. SEM image of a YNi_2B_2C film deposited at 725°C

Optimisation of the borocarbide phase formation was achieved by varying the deposition temperature. The superconducting transition temperature T_c increases with increasing deposition temperature (Fig. 1) and the highest superconducting transition temperature was found at 735°C. At higher temperatures, films began to crack and peel away from the substrate upon cooling. X-ray diffraction patterns in Bragg-Brentano geometry (Fig.2) show sharp *(00l)* borocarbide peaks [10], indicating a strong c-axis orientation, as well as peaks corresponding to secondary phases. We identified the secondary phases Y_2O_3 and Ni_2B by grazing incidence X-ray diffraction from a film deposited at 735°C (Fig.3). At lower temperatures, for instance at 625°C, no borocarbide peaks were observed (Fig.3). In the YNi_2B_2C films the Y_2O_3 phase formation has been shown to take place at the substrate-film interface [11]. X-ray pole figures of the (112) peak did not reveal any in-plane texture for these films. In-plane textured films with comparable superconducting properties have been prepared using lower energy densities (3J/cm^2), a reduced substrate-target separation and repetition rates of 30Hz.

The microstructure of the films was investigated by SEM. In the YNi_2B_2C films a majority of small round borocarbide grains are visible, as well as a number of larger, irregularly shaped impurity phase regions (Fig. 4). The grain size is about 400nm at a deposition temperature of 725°C (Fig.4).

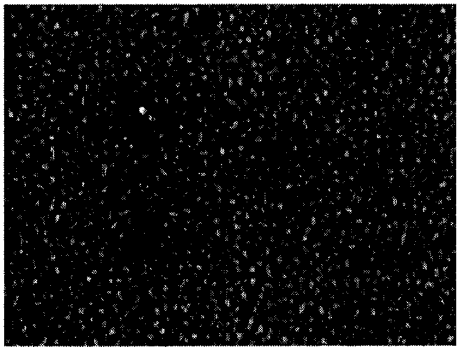

Fig. 5. SEM image of an YNi_2B_2C film deposited at 600°C; scan area 11.5μm x 9.5μm

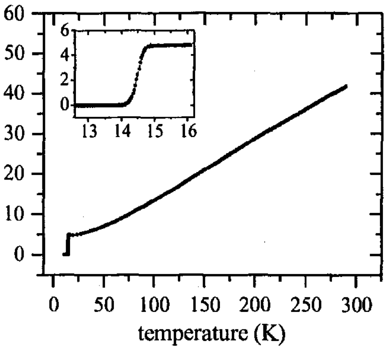

Fig. 6. Temperature dependence of the resistivity for an YNi_2B_2C film deposited at 725°C; inset: superconducting transition in detail

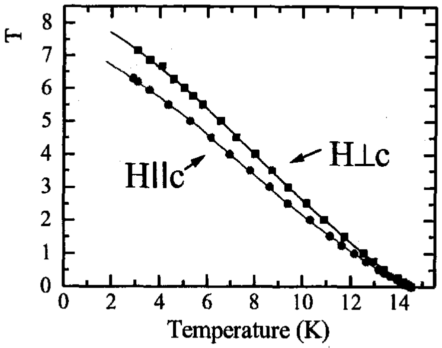

Fig. 8. Temperature dependence of the upper critical field H_{c2} for an YNi_2B_2C film deposited at 725°C

At lower deposition temperatures, 600°C in Fig. 5, the grains are significantly smaller and less impurity phase regions are visible. Fig.6 shows the temperature dependence of the resistivity of an YNi_2B_2C thin film. The film shows a superconducting transition at 14.7K with a transition width of 0.3K and a residual resistance ratio ($R_{300K}/R_{\text{just above }T_c}$) of 9. The decrease in resistivity is linear between 300K and 75K; in the low temperature range the temperature dependence changes to a quadratic behavior. The resistivity just above T_c is 5μΩcm (best single crystal values: 1.3 μΩcm [12]).

To determine the temperature dependence of the upper critical field H_{c2}, the R(T) transitions were measured under various magnetic fields applied both parallel and perpendicular to the c-axis. The expected shift in T_c with increasing field is observed as well as a successive broadening of the transition width from 0.3K at 0T to 1.5K at 6T (Fig. 7). The temperature dependence of the upper critical field H_{c2} was obtained from these resistivity curves using the midpoints of the transitions. The general behavior of the temperature dependence of H_{c2} (Fig.8) is comparable with single crystal data [12] and can be described by the multi-band Eliashberg theory [13]. An anisotropic behavior of H_{c2} was found in YNi_2B_2C for the two different applied field directions (Fig.8). Measurements of the angular dependence of H_{c2} were carried out to determine the nature of the anisotropic behavior. The magnetic field dependence of the resistivity was measured at temperatures of 2K, 6K and 10K for various angles between the c-axis and the applied field (0°...180°). The angular dependence of H_{c2} is shown in Fig. 9 and its general form was found to be independent of temperature, showing a slight broadening of the curve as the

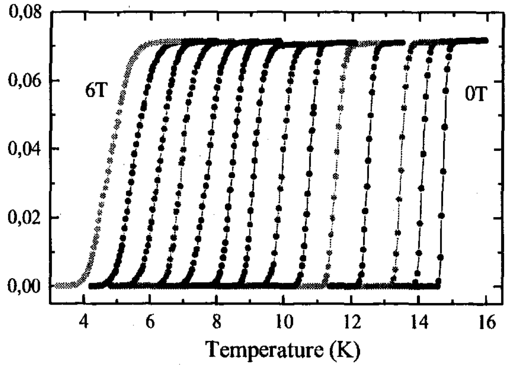

Fig. 7. Temperature dependence of the resistivity for various applied magnetic fields H||c (0, 0.25, 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5, 5, 5.5 and 6T) for an YNi_2B_2C film deposited at 725°C

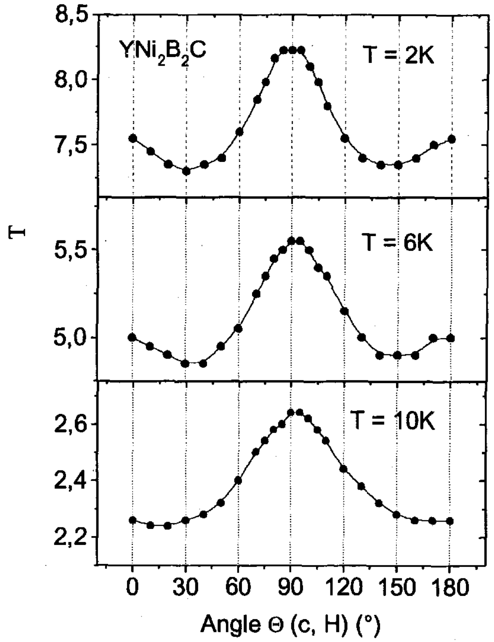

Fig. 9. Angular dependence of the upper critical field H_{c2} for an YNi_2B_2C film at various temperatures (Θ - angle between c-axis and applied field)

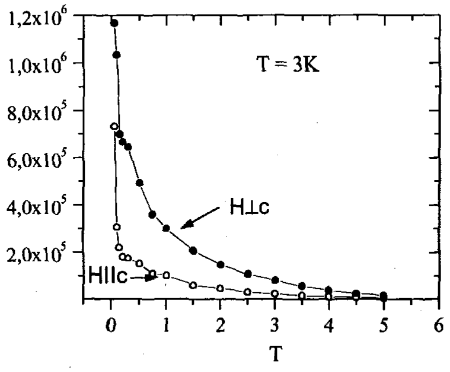

Fig. 10. Magnetic field dependence of the critical current density for an YNi_2B_2C film at 3K for the magnetic field parallel and perpendicular to the c-axis

temperature increased. There is a clear temperature independent minimum at 20°-30°. The complex and anisotropic Fermi surface of RNi_2B_2C calculated by Singh [14] and Drechsler *et al.* [15], can account for this unconventional angular dependence of H_{c2}.

The critical current density j_c was measured at a fixed temperature of 3K under an applied magnetic field (Fig. 10). In zero field a high critical current density of $1.2x10^6$ A/cm^2 was found although j_c decreases severely in the increasing magnetic field. Both the upper critical field and the critical current density show higher values if the magnetic field is applied perpendicular to the c-axis of YNi_2B_2C.

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