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Direct observations of first-order magnetization processes in single-crystal $Nd_2Fe_{14}B$

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Discontinuities in the magnetization curve of a uniaxial single crystal of $Nd_2Fe_{14}B$ along the hard direction are reported. The discontinuities, which have been interpreted as first-order magnetization processes (FOMP's), have been observed below 200 K in high magnetic fields. The critical field at which the FOMP occurs increases with decreasing temperature. The results are discussed in terms of the phenomenological anisotropy-energy description of uniaxial ferromagnets.

There has been much emphasis recently on the magnetic and structural properties of Nd-Fe-B and related compounds.[1,2] This is mainly due to the interesting magnetic properties, specifically the high uniaxial anisotropy[3,4] which is achieved without the use of the expensive and strategic element Co. These two qualities, in particular, make these compounds extremely attractive for applications in the field of permanent magnet materials.

This new family of compounds has been shown to have a tetragonal crystal structure[5] with the easy magnetization direction lying along the *c* axis at room temperature. There has been evidence for a reorientation spin transition from an easy axis to an easy cone at a temperature of 140 K.[4,6] By the use of the singular-point detection (SPD) method[7,8] it has been possible to measure the temperature dependence of the anisotropy field in polycrystalline samples.[3,4,9] A feature of the SPD technique is that it allows a precise determination to be made of the point (critical field H_{cr}) at which a discontinuity in the magnetization curve of a ferromagnet occurs.[10] By the use of the extended SPD technique the presence of a first-order magnetization process[11] (FOMP) has been observed at temperatures below 200 K in polycrystalline $Nd_2Fe_{14}B$ and the temperature dependence of the field (H_{cr}) at which the FOMP occurs has been measured.[3,4,9]

A FOMP is an irreversible rotation of the magnetization vector M_s under the action of an applied magnetic field, which results in a discontinuity of the magnetization curve along the hard magnetization direction of a ferromagnet. Such FOMP's have been also observed in other uniaxial as well as in cubic and trigonal materials.[12–14] A general theory for the conditions governing the existence of FOMP's, in terms of the phenomenological anisotropy-energy constants, in uniaxial[11] and trigonal[14] systems, has previously been reported.

In Fig. 1 is shown an example of the SPD measurement on a random polycrystalline sample of $Nd_2Fe_{14}B$. The discontinuity in the magnetization curve is smeared out to a great extent, whereas in the second derivative of magnetization with respect to time the discontinuity is clearly discernible.

We report here, for the first time, sudden discontinuities in the magnetization curve observed at low temperatures and high magnetic fields in a single crystal of $Nd_2Fe_{14}B$. The same effects can be observed with the use of aligned particles. However, the presence of unavoidable misalignments leads to a broadening of the transition and hence renders difficult a determination of the precise values of the magnetization and magnetic field at which the transition occurs. The observed discontinuities, which are sharp jumps to saturation observed along the hard magnetization direction, are interpreted as being type-1 FOMP's (i.e., a jump of the magnetization, at a critical field value, to saturation[11]).

The single crystal of $Nd_2Fe_{14}B$ was grown by using a Czochralski furnace. The crystal was cut into a sphere, oriented in a 8-kOe magnetic field, and fixed in epoxy resin. The measurements were carried out in pulsed magnetic fields up to 200 kOe, down to 78 K. The experimental apparatus is described in Ref. 8.

In Fig. 2 are shown plots of the magnetization σ versus the applied magnetic field for three temperatures. The

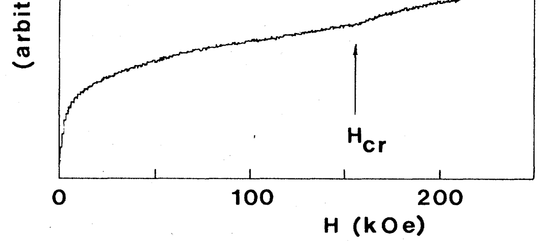

FIG. 1. Magnetization (M) and second derivative ($\ddot{M}$) of the magnetization vs applied magnetic field at 113 K for polycrystalline $Nd_2Fe_{14}B$. The arrow indicates the critical field H_{cr} at which the FOMP transition occurs.

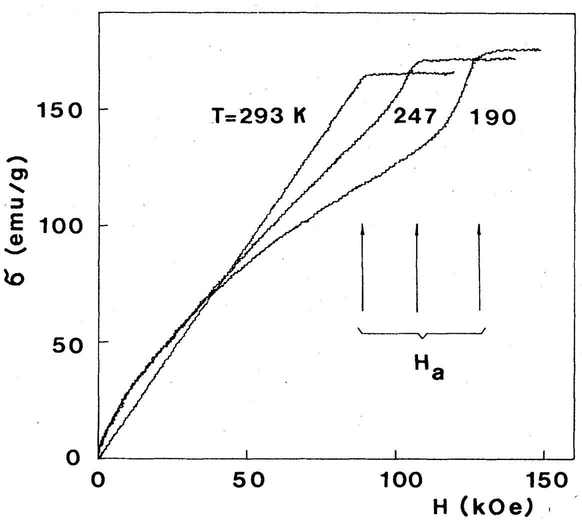

FIG. 2. Magnetization vs applied magnetic field in the hard direction at three different temperatures for single-crystal $Nd_2Fe_{14}B$. The arrows indicate the position of the anisotropy field (H_a).

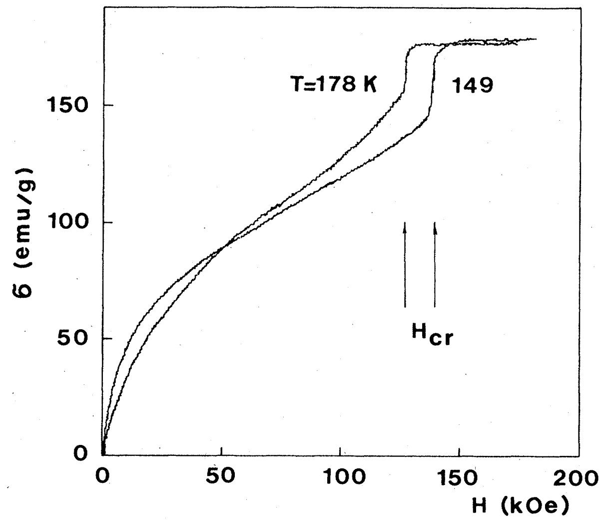

FIG. 3. Magnetization vs applied magnetic field in the hard direction at two different temperatures for single-crystal $Nd_2Fe_{14}B$. At these two temperatures, the easy direction lies along the c axis. The arrows indicate the position of the FOMP transition (H_{cr}).

shape of the curves can be seen to change rapidly with decreasing temperature. The magnetization curve can be described by the familiar phenomenological expression for the total energy of a uniaxial crystal. Neglecting basal plane anisotropy, the effects of which are significant only at low fields,[6] the expression for the total energy is

$$E = K_1 \sin^2\theta + K_2 \sin^4\theta + K_3 \sin^6\theta - HM_s \sin\theta$$

(where K_1, K_2, and K_3 are second-, fourth-, and sixth-order anisotropy constants, θ is the angle between the c axis and the magnetization vector M_s, with the magnetic field H applied perpendicular to the c axis). From this description it can be seen that while the room-temperature magnetization curve can be adequately described using only K_1, higher-order constants K_2 and K_3 have to be taken into account at lower temperatures. Furthermore, K_1 decreases rapidly with decreasing temperature as seen from the initial slope of the magnetization curve (Figs. 2 and 3). At temperatures lower than 200 K the discontinuity in the magnetization curve becomes evident as seen in Fig. 3. Below 140 K the easy magnetization direction lies along a cone.[6] With decreasing temperature the value of the cone angle, the critical field value, and the amplitude of the discontinuity all increase as can be seen in Fig. 4.

It has already been shown that the Nd ion gives the main contribution to the anisotropy, even if the Fe contribution is unusually high.[6,15] Both the observed evolution of the magnetization curve shape and the existence of FOMP's in a uniaxial crystal imply significant contributions to the magnetic anisotropy coming from higher-order anisotropy constants K_2 and K_3. This can be accounted for by considering competing contributions to the anisotropy that may originate from the two inequivalent Nd sites. It has indeed been shown that in the case of strong competing anisotropies, the resultant anisotropy is no longer the simple algebraic sum of the individual sublattice contributions, and higher-order anisotropy constants are found.[16]

The compound $Nd_2Fe_{14}B$ undergoes a FOMP transition below 200 K and a spin reorientation (easy axis to easy cone) at 140 K. From these observations and from an

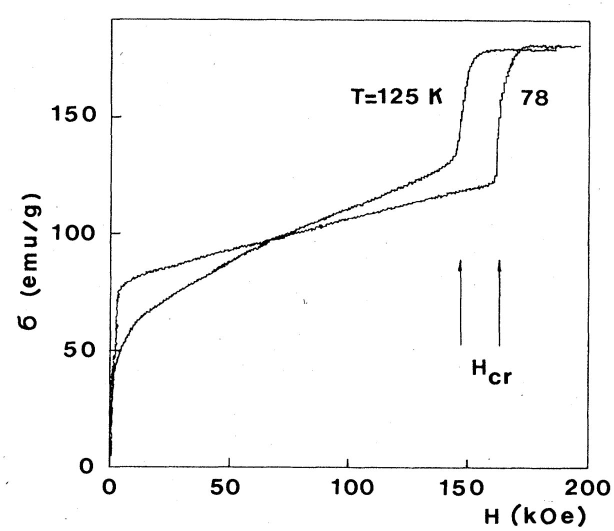

FIG. 4. Magnetization vs applied magnetic field in the basal plane at two different temperatures for single-crystal $Nd_2Fe_{14}B$. In contrast with Fig. 3, the easy direction in this case lies along a cone. The arrows indicate the position of the FOMP transition (H_{cr}).

analysis of the magnetic phase diagram obtained by considering the conditions necessary for the existence of a FOMP transition (Fig. 5 of Ref. 11), one can deduce the sign and the relative amplitude of the anisotropy constants for $Nd_2Fe_{14}B$ at low temperatures. In particular, the following conditions have been obtained: $|K_3| > (\frac{6}{15})K_2 + (\frac{1}{15})K_1$ with $K_1, K_2 > 0$ and $K_3 < 0$ for 140 K $< T <$ 200 K (easy-axis region) and $K_1, K_3 < 0$, $K_2 > 0$, $K_2 > |K_1|$ for $T <$ 140 K (easy-cone region).

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