Radiation damage in Nd-Fe-B magnets: temperature and shape effects

1992 J. Phys.: Condens. Matter 4 1007

(http://iopscience.iop.org/0953-8984/4/4/011)

J. Phys.: Condens. Matter 4 (1992) 1007–1014. Printed in the UK

Radiation damage in Nd–Fe–B magnets: temperature and shape effects

O-P Kähkönen†, S Mäkinen†, M Talvitie‡ and M Manninen†

† Department of Physics, University of Jyväskylä, SF-40351 Jyväskylä, Finland
‡ Outokumpu Magnets Oy, SF-28101 Pori, Finland

Received 24 June 1991, in final form 3 September 1991

Abstract. $Nd_2Fe_{14}B$ permanent magnets with different load lines were irradiated with 20 MeV protons at temperatures between 300 and 15 K, and the flux loss was measured as a function of the irradiation temperature. The results show that the magnetic flux loss depends drastically on temperature and on the shape of the sample. A new theoretical model which explains the observed temperature dependence of irradiation effects is introduced.

1. Introduction

The discovery of Nd–Fe–B high performance permanent magnets has increased the interest in the basic research on magnetism and magnetic materials [1]. The lattice structure [2], electronic structure [3] and basic magnetic propeties [4] of these neo-magnetic materials are known. Nd–Fe–B magnets have been shown to be extremely sensitive to irradiation when compared with conventional permanent magnets [5–7]. Technologically this is a drawback since it can prevent the application of these magnets in particle accelerators. In basic research the sensitivity to irradiation opens a new possibility for increasing our understanding of the domain nucleation and demagnetization processes. In earlier papers [8, 9] we showed that the irradiation-induced flux loss is strongly temperature-dependent and suggested that the sensitivity to irradiation is related to the low Curie temperature of the $Nd_2Fe_{14}B$ compound: in the collision cascade the local temperature rises above the Curie temperature, giving the possibility for nucleation of a new magnetic domain in the opposite direction. The new domain can grow to the size of the grain thus slightly decreasing the magnetic flux of the material.

In this paper we study the temperature dependence of the flux loss in the $Nd_2Fe_{14}B$ magnets in more detail. We have observed that the irradiation-induced flux loss is connected to the demagnetizing field created by the magnet itself. This demagnetizing field depends drastically on the shape of the magnet and, therefore, the magnetic damage caused by irradiation has a strong shape dependence. A new theoretical model is introduced for the temperature dependence of the irradiation-induced flux loss.

0953-8984/92/041007+08$04.50

2. Experiment

2.1. Samples

The samples were sintered NdFeB permanent magnets with the main phase a $Nd_2Fe_{14}B$ compound [9]. The grain size of the main phase was 15–30 μm and the maximum energy product of the samples was 290 kJ m^{-3}. The remanence and the intrinsic coercive force of the samples were 1.2 T and 900 kA m^{-1}, respectively. Two kinds of sample sizes were used in order to investigate the geometrical effects: 7.1 mm × 7.1 mm × 1.0 mm platelets and 1.0 mm × 1.0 mm × 1.0 mm cubic samples.

In preparing the samples, Nd–Fe–B-ingots were crushed and attritor milled under an argon atmosphere to a particle size of approximately 3 μm. The powder was aligned in a magnetic field of about 0.8 T and compacted at a pressure of 200 MPa. The direction of the magnetic field was perpendicular to the pressing direction. The compacts were sintered in vacuum at 1340 K for 1 h and heat treated at 870 K. Finally, the samples were cut to appropriate dimensions using a diamond saw.

Three types of samples were studied with completely different permeance coefficient slopes of the load line ($B/\mu_0 H$) depending on the shape and direction of the magnetization [10]. For the sample platelets the value of the $B/\mu_0 h$ ratio was about -0.35 when the magnetization was *parallel* to the shortest dimension, and about -10 when *perpendicular* to this; both measured in open air. In the case of the cubic samples the $B/\mu_0 H$ ratio was about -2.0. It should be noted that the magnetic field inside the samples was not homogeneous, not even in the cubic samples.

2.2. Irradiation

The irradiation was performed with 20 MeV protons by using the MC-20 cyclotron of the University of Jyväskylä. The proton energy was chosen so that practically all the protons went through the sample. The penetration depth of the protons was approximated by using the average penetration depth of each component [11] and by using the mass concentration of each component in the alloy. The ion current was kept below 10 nA to make the thermal heating of the sample as low as possible. After the irradiations the relative magnetic changes were measured using a Helmholtz coil and an induction coil.

During the irradiation the sample was kept in a two-stage closed-cycle helium refrigerator [12]. The plate-shaped samples were irradiated at temperatures between 15 and 300 K with a 100 Mrad dose. The cubic samples were irradiated at room temperature with doses of 5–1000 Mrad. The irradiation time was about 31 min for the 100 Mrad dose. The ion current was measured with a Faraday cup between a beam collimator and the sample. The stability of the beam was checked by measuring the proton current after every two minutes. The irradiation temperature was measured by a copper–constantan thermocouple and controlled by a microcomputer.

The direction of the proton beam was always parallel to the shortest dimension of the plate-shaped samples. The cubic samples were irradiated with the beam either parallel or perpendicular to the direction of the magnetization.

The relative flux loss of the irradiated samples was measured with open-circuit methods. The total magnetic flux was measured before and after the proton irradiations and the relative flux losses were determined from these values.

2.3. Results

The results for the plate-shaped samples have already been reported [9]. It was found that the samples with their magnetization perpendicular to the surface were most sensitive to irradiation. At low temperatures (25–50 K) the samples were insensitive to the radiation whereas at room temperature the 100 Mrad dose was enough to reduce the magnetization by about 40%. However, the samples which were magnetized parallel to the big surface (perpendicular to the direction of the ion beam, $B/\mu_0 H = -10$) did not show any decrease in the magnetization at the irradiation temperatures of 25–300 K.

The cubic samples were only studied at room temperature. The flux loss caused by a 1000 Mrad dose was about 70% when the direction of magnetization was *parallel* to the ion beam and about 30% when this direction was *perpendicular* to the ion beam.

The plate-shaped samples with the magnetization parallel to the surface ($B/\mu_0 H = -10$) showed no sensitivity to the proton irradiation. The qualitative explanation is as follows: in these magnets the demagnetizing field is so weak that the net magnetic field inside the sample is in the same direction as the magnetic polarization. In the collision cascade the magnetic spins are free to rotate and the magnetic field orientates them back to their original state during the time when the temperature in the collision cascade is cooled below the Curie temperature. The sample platelets with magnetization perpendicular to the surface ($B/\mu_0 h = -0.35$) had such a strong demagnetizing field inside the magnet that the total magnetic field was in the opposite direction to that of the magnetic polarization. In this case the spins are rotated in the direction of the total magnetic field and a nucleation centre is born. If the size of the nucleation centre is sufficiently large a new domain in the opposite direction might result. This would then be able to grow to the size of the grain and, thus, part of the total magnetization would be lost.

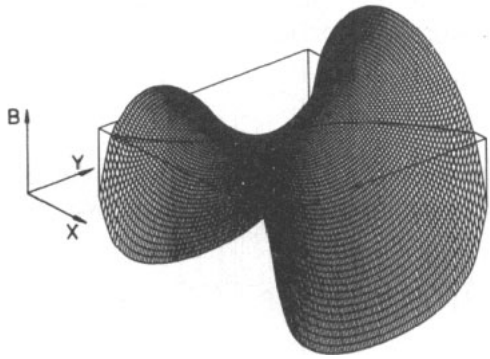

Figure 1. The strength of the magnetic field at the centre plane of the cubic sample. The magnetization is oriented in the x-direction.

In the cubic samples the magnetic field inside the magnet is not homogeneous, as can be seen in figure 1. This is important from the point of view of the magnetic damage, because this occurs more easily in regions where the magnetic field is negative. In the irradiated cubic samples with the magnetic orientation in the same

direction as the ion beam the protons first pass through regions with a negative total magnetic field, which makes magnetic damage very possible. On the other hand when the magnetic orientation was perpendicular to the direction of the ion beam the incoming protons first pass through regions with a positive total magnetic field and damage cannot occur. The energy of the incoming proton decreases when it goes through the sample and therefore it is expected that most of the magnetic damage occurs in the regions which are nearest to the ion source. However, even in this case some protons pass through regions where the total magnetic field is negative (see figure 1), causing a small flux loss in the sample.

To conclude, the magnetic damage was found to depend strongly on the shape of the magnet. This dependence is connected to the demagnetizing field created by the magnet itself. In the next section we introduce a new model for explaining the observed magnetic damage.

3. Theoretical model

The theoretical model is based on the assumption that part of the energy of the incoming particle is transferred to the primary knock-on atom. The energy is then diffused into the lattice raising the temperature of a spherical region. If the temperature rises above the Curie temperature and if this sphere is large enough the demagnetizing field can turn the spins and nucleation of a new domain occurs. This phenomenon is possible if the magnetic field in the sample is in the opposite direction to the direction of magnetization.

The critical size of the nucleation centre is described by a critical radius R which depends on the demagnetizing field, a weaker demagnetizing field requires a larger R. The critical size of the nucleation centre is connected to the magnetic energy of the domain and to the energy of the domain wall.

At first we calculate the minimum energy required to heat the sphere with the radius R to a temperature over the critical temperature T_c. We assume that the primary knock-on atom causes a thermal spike and use the differential equation of heat transfer to describe the energy diffusion to the lattice,

$$\frac{\partial T}{\partial t} = a^2 \nabla^2 T. \tag{1}$$

where a is a constant. Assuming the initial temperature distribution to be a δ-function (1) can be immediately solved (using the Green function) to give

$$T(r,t) = (T_1 - T_0)\left(\frac{d^2}{4\pi a^2 t}\right)^{3/2} e^{-r^2/4a^2 t} + T_0. \tag{2}$$

where T_1 is the initial temperature of the primary knock-on atom, d^3 is the volume of one atom, and T_0 is the initial temperature of the sample. The maximum radius of the region which is heated above the Curie temperature T_c is determined by finding the maximum r which satisfies equation $T(r,t) = T_c$. This gives

$$T_1 = T_0 + (T_c - T_0)\left(\frac{2\pi R^2}{3d^2 e^{-1}}\right)^{3/2}. \tag{3}$$

The relationship of T_1 to the kinetic energy of the primary knock-on atom is

$$E_{\text{kin}} = \tfrac{3}{2} k_{\text{B}} (T_1 - T_0). \tag{4}$$

Equations (3) and (4) determine the minimum kinetic energy needed for a sphere of radius R to be heated above the Curie temperature. Next we have to determine the probability of the collision which can transfer this energy to the primary knock-on atom.

The minimum scattering angle can be determined from the energy transfer of a two-body elastic collision,

$$E_{\text{kin}} = E_{\text{i}} \left(1 - \frac{(\cos \Theta + [(m_2/m_1) - \sin^2 \Theta]^{1/2})^2}{[(m_2/m_1) + 1]^2} \right) \tag{5}$$

where E_{i} is the energy of the incoming particle, Θ the scattering angle, m_1 is the mass of the incoming particle and m_2 is the mass of the target atom. Equation (5) determines the minimum angle, Θ_{min}, which results in an energy transfer of more than E_{kin}. The scattering in the energy region of interest is basically Coulomb scattering. The cross section, σ, for events resulting in an energy larger than a certain E_{kin} can now be obtained by integrating the Coulomb scattering cross section over angles larger than Θ_{min}:

$$\sigma = 2\pi q^2 \left(\frac{1}{1 - \cos \Theta_{\text{min}}} - \frac{1}{2} \right) \tag{6}$$

where

$$q = \frac{zZe^2}{2 \times 4\pi\epsilon_0 (\frac{1}{2} m v_0^2)}.$$

The total cross section is $\Sigma = n\sigma$, where n is the ion density of the sample and the probability of one collision is

$$P = L\Sigma = n\sigma L \tag{7}$$

where L is the sample thickness.

We assume that each such collision causes domain nucleation in the opposite direction and that the domain immediately grows to the size of the grain. The relative loss of magnetization can thus be calculated as

$$\frac{\Delta M}{M} = 2 \frac{V_{\text{grain}}}{V_{\text{sample}}} P N_{\text{p}} \tag{8}$$

where N_{p} is the number of incoming particles and V_{grain} and V_{sample} are the grain and sample volumes, respectively.

In calculating the relative flux loss of magnetization we have two parameters: the critical radius of the nucleation centre (R) and the average volume of the grains, described by the grain diameter D_{g}. The retardation of the incoming protons inside the sample was calculated by assuming that the sample was pure iron [13]. The flux

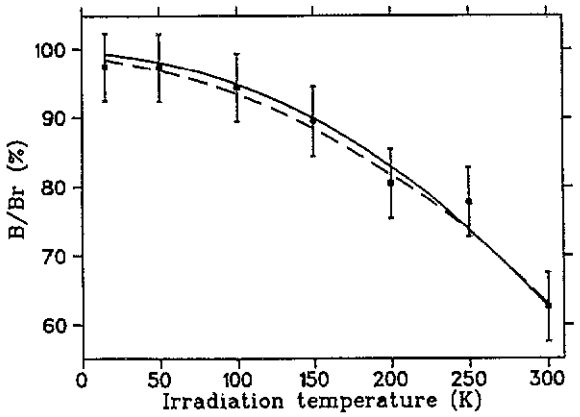

Figure 2. The measured and calculated temperature dependence of the magnetic flux loss. The broken and full curves are the result of models where the primary knock-on atom is assumed to be boron and iron, respectively.

loss was calculated as a function of temperature for the plate samples, inside which the magnetic field could be approximated as constant.

The calculated temperature dependence of irradiation-induced demagnetization is shown in figure 2. The calculations were made using either boron or iron atoms as target atoms. In both cases we were able to explain the temperature dependence of the experimental data. When the target atom was assumed to be boron the best fit was obtained with $R = 425$ Å and $D_g = 16$ μm. When the target atom was iron the values of R and D_g was 263 Å and 2.5 μm, respectively. This gives us two possible ways to explain the temperature dependence of the irradiation-induced flux loss: either the new domain with a radius above 425 Å grows to the size of the grains in the irradiated samples (about 15–30 μm) or the domain growth is interrupted at the size of 2.5 μm by, for example, defects or impurities. In this latter case the required critical size of the new domain (nucleation centre) is smaller.

The obtained critical radius (425 or 263 Å) is much larger than those of the nucleation volumes obtained from the magnetic viscosity measurements [14]. However, in our case the demagnetization field is much smaller and requires a larger nucleation centre.

It should be noted that in our model only the collision between the incoming proton and the primary knock-on atom is important. If it is a collision with a boron atom, the critical radius R is so large that collisions with iron atoms cannot provide enough energy to the lattice to heat such a large volume. If we assume that the iron atoms dominate (R smaller) then the cross section with lighter boron atoms is vanishingly small compared with that of the iron atoms.

The size of the critical radius of a nucleation centre cannot be determined exactly due to the thermal fluctuations and because the magnetic field inside the magnet is changing during the irradiation due to the magnetic flux loss. The magnetic energy connected to the domain and the domain walls change during the irradiation which will also change the critical radius. These effects have not been taken into account in our model. Macroscopically the critical size of the nucleation centre could be determined by equallizing the magnetic energy of the domain with the domain wall energy.

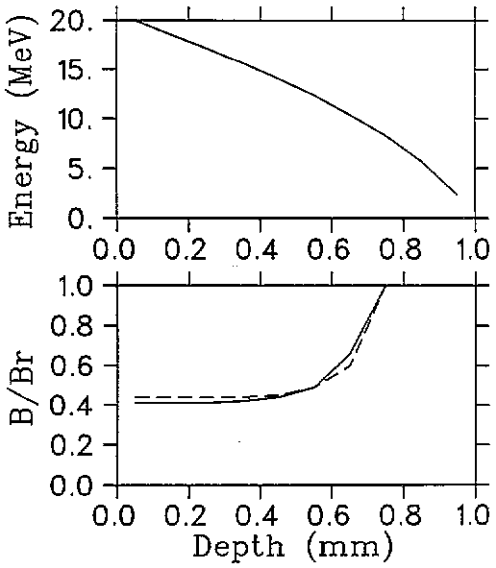

Figure 3. The calculated energy loss of the incoming proton and the relative magnetic flux loss as a function of the sample depth. The broken curve denotes for the B target atom and the full curve for the Fe target atom.

In figure 3 we show the calculated energy loss of the incoming proton and the magnetic flux loss as a function of the depth. Most of the magnetic flux loss occurs in the regions where the proton energy is over 10 MeV, whereas, in the regions where the proton energy is below 8 MeV no flux loss is obtained. This calls for flux loss measurements as a function of the proton energy.

In the cubic samples the internal magnetic field is very inhomogeneous as shown in figure 1. In this case we did not perform computations in detail with our model since it would have required spatial integration over the whole cube. Furthermore, in the case of a cube we cannot neglect the fact that the actual critical nucleation radius is dependent on the internal magnetic field. However, *qualitatively* our model explains the dependence of the flux loss on the direction of the magnetization. When the magnetization is parallel to the beam, the high-energy protons hit the face with the largest demagnetizing field. It is then expected that the domain nucleation is more probable in that direction than when the magnetization is perpendicular to the ion beam. This was also the experimental result.

4. Conclusions

The irradiation-induced flux loss was found to depend on the irradiation temperature as well as on the direction of the magnetization and on the shape of the sample. The temperature dependence of the magnetic flux loss can be explained with a simple model based on local heating by the primary knock-on atom. The model fits the experimental results and it also gives an explanation for the shape effects of the magnetic flux loss. By assuming that the collisions with boron atoms dominate, the estimated grain size equals the experimentally observed value. This suggests that the high sensitivity of the Nd–Fe–B magnets on radiation could be due to the existence

of light atoms in the material. This suggestion is still premature but it can be further studied by measuring the flux loss with different proton energies and also by studying the temperature dependence of the irradiation-induced flux loss in other permanent magnet materials.

Acknowledgments

We are grateful to J Timonen, J Kumpulainen and E Kautto for useful discussions.

References

[1] Sankar S G, Herbst J F and Koon N C (ed) 1987 *High Performance Permanent Magnet Materials (Materials Research Society Symposia Proceedings 96)* (Pittsburgh, PA: Materials Research Society)
[2] Herbst J F, Croat J J, Pinkerton F E and Yelon W B 1984 *Phys. Rev.* B **29** 4176
[3] Jaswal S S 1990 *Phys. Rev.* B **41** 9697
[4] Sagawa M, Fujimura S, Yamamoto H, Matsuura Y and Hirosawa Y 1985 *J. Appl. Phys.* **57** 4094
[5] Blackmore E W 1985 *IEEE Trans. Nucl. Sci.* **32** 3669
[6] Zeller A F and Nolen J A 1986 *Annual Report 1986, National Superconducting Cyclotron Laboratory, Michigan State University, USA* p 161
[7] Cost J R, Brown R D, Giorgi A L and Stanley J T 1987 *Materials Research Society Symposia Proceedings* vol 96, ed S G Sankar, J F Herbst and N C Koon (Pittsburgh, PA: Materials Research Society) p 321
[8] Kähkönen O-P, Mäkinen S, Talvitie M, Rajainmäki M and Manninen M 1990 *Europhys. Lett.* **12** 413
[9] Talvitie M, Kähkönen O-P, Mäkinen S, Rajainmäki H, Manninen M and Lindroos V 1991 *Magn. Magn. Mater.* at press
[10] Parker R J 1990 *Advances in Permanent Magnetism* (New York: Wiley)
[11] Littmark U and Ziegler J F 1980 *Handbook of Range Distributions for Energetic Ions in All Elements* (New York: Pergamon)
[12] Hansen H E, Talja R, Rajainmäki H, Nielsen H K, Nielsen B and Nieminen R M 1985 *Appl. Phys.* A **36** 81
[13] Ziegler J F, Biersack J P and Littmark U 1985 *The Stopping and Ranges of Ions in Matter* (New York: Pergamon)
[14] Givord D, Tenaud P and Viadieu T 1988 *IEEE Trans. Magn.* **24** 1921