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Effect of intergrain exchange interaction on magnetic viscosity of nanocrystalline isotropic NdFeB magnets

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Abstract. The coefficient of magnetic viscosity, *S*, of NdFeB alloys with varied intergrain exchange interaction constant, K_{IEI}, was measured. The observed dependences of fluctuation field, H_f, on the applied field, *H*, are strongly correlated with K_{IEI} value. Cooperative magnetization reversal of single domain interacting particles leads to improve the temperature stability of permanent magnets and magnetic systems based on them.

1. Introduction

Hard magnetic materials play an important role in modern technology and science developments in energy converters. Magnetized permanent magnets are in a thermodynamically metastable state and, therefore, their magnetization relaxes as a result of chemical changes or partial demagnetizations under internal or external magnetic fields and thermal fluctuations. In most cases of ferromagnetic or ferrimagnetic materials, the relaxation of the magnetization owing thermal fluctuations with time can be described by

$$M(t) = M(0) - S \ln(1+t/t_0), \quad (1)$$

with $M(t)$ – magnetization depending on time *t*, $M(0)$ – magnetization at $t = 0$ after a certain magnetic history of the sample (usually a negative field jump is applied to a magnetically saturated sample), *S* – the coefficient of magnetic viscosity, t_0 – some reference time depending on the measuring procedure.

For many materials, the field dependence of the irreversible part of the magnetic susceptibility, χ_{irr}, and of *S* are similar, with a maximum near the coercivity H_c [1]. Therefore, term fluctuation field, H_f, is introduced and defined as

$$S = \chi_{irr} H_f \quad (2)$$

is expected to depend only weakly or even not at all on the applied field, *H*. As an example, H_f of a commercial sintered Nd-Fe-B magnet has been found to be field-independent [2]. However, it is obvious that the field independence of H_f implies that the considered material has narrow distributions of coercivities. Strongly heterogeneous materials, such as mixtures, are expected to show a variation of H_f with *H*. Thus it has been shown, experimentally as well as theoretically, that H_f of certain magnetic systems varies remarkably with *H* [3 – 6].

The correlation of magnetic viscosity with magnetic hysteresis had been experimentally investigated by Barbier who found a universal relationship,

$$H_f \sim H_c^{\alpha}, \tag{3}$$

being approximately fulfilled for a large class of materials, where both quantities, H_f and H_c, vary over several orders of magnitude, and the exponent is $\alpha = 1.35$ [7]. In a simplified theoretical approach to coercivity and viscosity, Liu and Luo found a relation like (3), however with $\alpha = 1$, even considering various coercivity mechanisms [8]. A more realistic theoretical model relating viscosity to coercivity i.e. S to H_c, respectively, H_f to H_c, in permanent magnets, has been developed in [9].

The exchange interaction in rapidly quenched alloys of the NdFeB leads to a narrowing of the distribution of critical fields at which magnetization reversal of the crystallites of the hard magnetic phase occurs [10]. Studies on magnetization reversal processes in sintered permanent magnets of the NdFeB system [11] indirectly show that even in the presence of a paramagnetic intergranular layer, the exchange interaction between neighboring grains is partially preserved which leads to a decrease in the coercive force of such a magnet relative to magnets with a fully suppressed exchange interaction.
For this reason, it is relevant to establish the effect of intergrain exchange interaction on the magnetic aftereffect for the potential use of permanent magnets with intergrain exchange interaction in magnetic systems.

2. Experiment

The NdFeB alloys with compositions $Nd_{11.7}(Fe_{1-x}Co_x)_{82.5}B_{5.8}$ (x = 0, 0.2, 0.25, 0.5) and $Nd_{22.7}(Fe_{0.8}Co_{0.2})_{70.5}B_{6.7}$ were prepared by arc melting of appropriate masses of pure elements and subsequent rapidly quenching procedure. Strips of alloys were annealed in vacuum for 1 hour at different temperatures from the range of 773 – 1273 K.

Two infiltrated samples were produced by milling of 80 % wt. of prepared MQP-B and 20 % wt. of arc melted $Pr_{75}(Cu_{0.25}Co_{0.75})_{25}$ alloys with a vibration mill. An infiltration routine was carried out at temperature of 773 K for 4 hours. Preparation of first MQP-B alloy included annealing at 1000 °C for 1 hour to provide wide distribution of grain coercivities. Preparation of second MQP-B alloy included hand grinding.

Samples for magnetometry were prepared by fixing the pieces of alloy in paraffin wax in a container with an inner diameter 6.3 mm and height 2.5 mm. The filling height of the samples in the container was about 0.3 – 0.5 mm. All samples were isotropic.

Measurements of the magnetic viscosity were performed using Quantum Design MPMS-XL-7 EC with SQUID as a detector at temperature T = 300 K. After saturating the sample at an applied magnetic field of 7 T, the field was decreased to a specified negative value at which the magnetic moment was measured from approximately 30 – 1500 s after reaching a stable field. Irreversible magnetic susceptibilities were measured by the next technique. For DC measurements, a negative external magnetic field $H = H_1$ was applied to the previously saturated sample and then the magnetization M (H_1) was measured. After that the magnetic field was turned off and then the magnetization M_d (H_1) was measured. The irreversal magnetic susceptibility χ_{irr} was obtained by calculating the derivative $d\sigma_d(H)/dH$, where σ_d – remanence magnetization after partitial demagnetization of previously saturated sample at magnetic field H.

Effects of demagnetization have been neglected (N ~ 0).

3. Results and discussion

Figures 1 – 3 show major demagnetization curves and dependencies of χ_{irr} (H), S (H) and H_f (H) of $Nd_{11.7}(Fe_{0.8}Co_{0.2})_{82.5}B_{5.8}$, $Nd_{22.7}(Fe_{0.8}Co_{0.2})_{70.5}B_{6.7}$ rapidly quenched alloy and infiltrated MQP-B samples correspondingly. The first alloy is characterized by intergrain exchange interaction with the constant K_{IEI} ~ 8 erg/cm^2 [12]. Similar dependencies (the results are not given in this article) are observed for other stoichiometric alloys with different iron substitution by cobalt. The intergrain exchange interaction in such alloys leads to the formation of exchange domains in the process of magnetization [13]. A paramagnetic layer enriched with Nd or Pr is formed in over stoichiometric alloys or alloys after infiltration procedure.

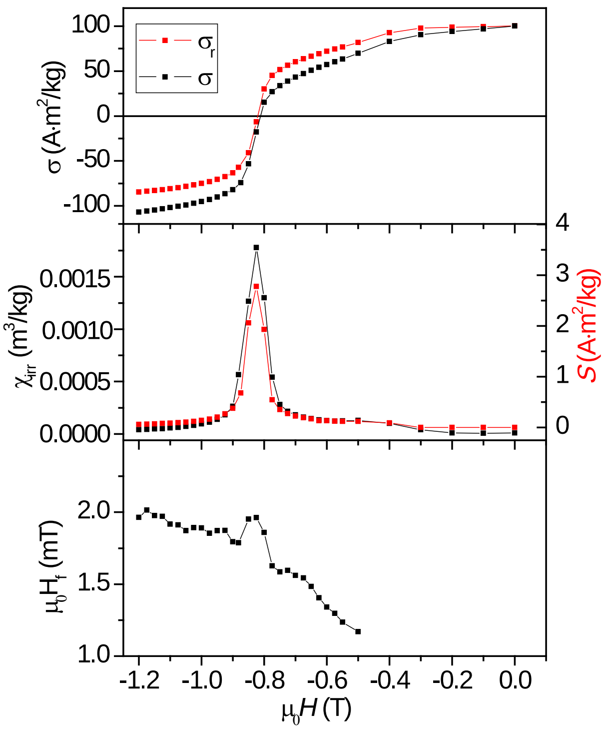

Figure 1. Major demagnetization curve, χ_{irr} *vs*. *H*, *S vs*. *H* and H_f *vs*. *H* of the rapidly quenched alloy $Nd_{11.7}(Fe_{0.8}Co_{0.2})_{82.5}B_{5.8}$.

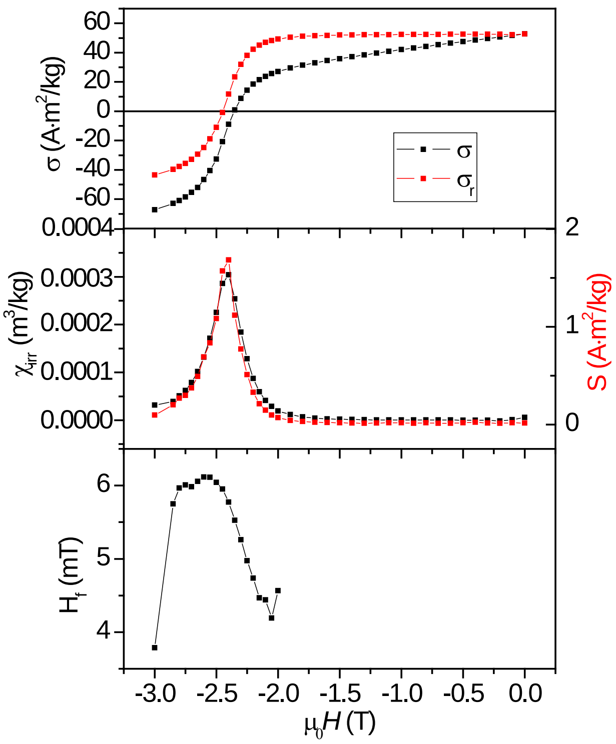

Figure 2. Major demagnetization curve, χ_{irr} *vs*. *H*, *S vs*. *H* and H_f *vs*. *H* of the rapidly quenched alloy $Nd_{22.7}(Fe_{0.8}Co_{0.2})_{70.5}B_{6.7}$.

The major demagnetization curves show that the coercivity in samples without intergrain exchange interaction exceeds one in comparison with a sample with a strong exchange interaction.

The axes of easy magnetization of crystallites are randomly distributed in the sample. Demagnetizing field forms regions in which the magnetic moments of the grains have a negative projection on the direction of the residual magnetization. Due to the intergrain exchange interaction of the ferromagnetic type the height of the energy barrier between the different orientations of the magnetic moment along the easy magnetization axis of the grain with surrounding of demagnetized grains decreases.

The dependence χ_{irr} *vs*. *H* is equal to the distribution of the grains of the sample in terms of coercivity up to a constant. Common to all alloys is the identity of the dependences χ_{irr} *vs*. *H* and *S vs*. *H*. The identity is due to the relatively narrow value of 25 kT in comparison with the height of the energy barrier separating the positions of global minima of the energy of the spatial orientation of the magnetic moment of grains. A slight difference in the forms of these dependences is of the most interest object because of namely both the features of the magnetization reversal process and the interactions of different nature in the sample are manifested.

The H_f *vs*. *H* dependences of samples with a negligibly small value of K_{IEI} are characterized by a rectangular shape with a sharp increase of the H_f value at $H < H_c$, followed by a plateau and a decrease at $H > H_c$. To eliminate the influence of the width of the distribution of grains by coercivity one of the infiltrated MQP-B was previously annealed. All samples with supressed exchange interaction demonstrate identical H_f *vs*. *H* dependence.

In the case of exchange-coupled samples, a gradual increase in the H_f value is observed with increasing intensity of the demagnetizing field since the beginning of the irreversible processes of magnetization reversal. For such samples H_f *vs*. H can be approximated in accordance with relation

$$\ln H_f = \beta \ln |H| + \gamma. \tag{4}$$

with β, γ – approximation constant. The value of β is 0.55 – 0.65, which is less than the slope of the original or modified Barbier plot.

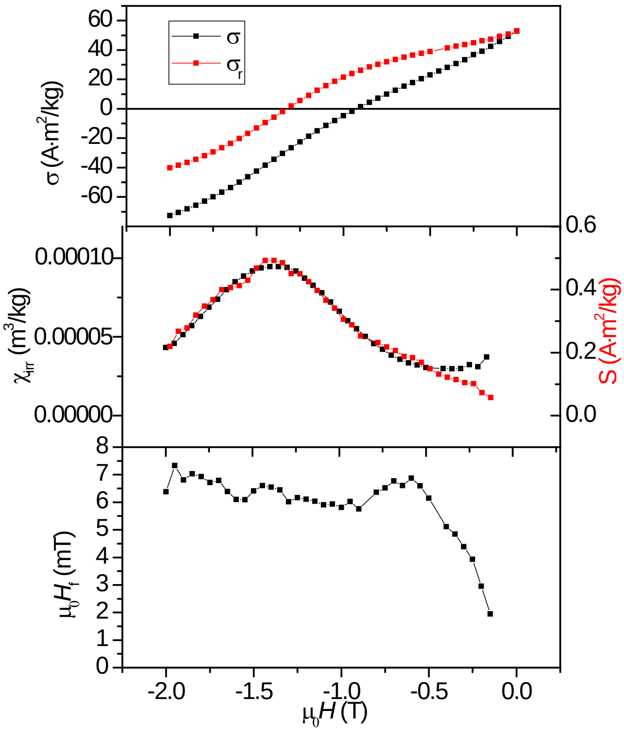

Figure 3. Major demagnetization curve, χ_{irr} *vs*. H, S *vs*. H and H_f *vs*. H of the infiltrated sample of annealed MQP-B with addition 20 % wt. of alloy $Pr_{75}(Cu_{0.25}Co_{0.75})_{25}$.

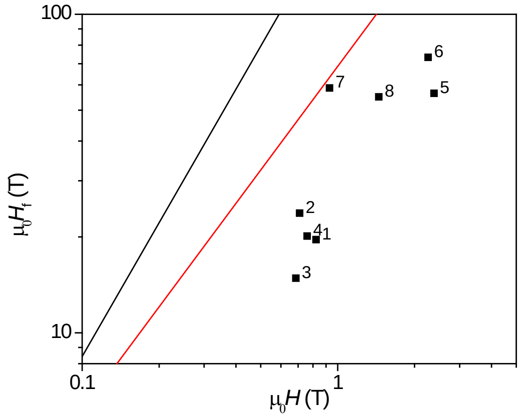

Figure 4. Fitting of our experimental data with Barbiers relation (3) with the original (black) [7] and the modified (red) [8] line, respectively: 1 – rapidly quenched alloy $Nd_{11.7}(Fe_{0.8}Co_{0.2})_{82.5}B_{5.8}$; 2 – rapidly quenched alloy $Nd_{11.7}Fe_{82.5}B_{5.8}$; 3 – rapidly quenched alloy $Nd_{11.7}(Fe_{0.75}Co_{0.25})_{82.5}B_{5.8}$; 4 – rapidly quenched alloy $Nd_{11.7}Co_{82.5}B_{5.8}$; 5 – rapidly quenched alloy $Nd_{22.7}(Fe_{0.8}Co_{0.2})_{70.5}B_{6.7}$; 6 – permanent magnet Sm-Co; 7 – infiltrated sample of annealed MQP-B with addition 20 % wt. of alloy $Pr_{75}(Cu_{0.25}Co_{0.75})_{25}$; 8 – infiltrated sample of as received MQP-B with addition 20 % wt. of alloy $Pr_{75}(Cu_{0.25}Co_{0.75})_{25}$.

Values of the fluctuation field of the samples in a magnetic field equal to their coercive force are presented in figure 4. Samples can be divided into two groups according to the magnitude of the fluctuation field. The first group includes samples with intergranular exchange interaction and the value $\mu_0 H_f \approx 15 – 25$ mT, the second group includes samples with independent magnetization reversal of individual elements and the value $\mu_0 H_f \approx 55–80$ mT. This means that when considering the effect of thermal fluctuations on magnetization reversal processes it is necessary to take into account not only the energy of magnetocrystalline anisotropy but also the energy of intergrain exchange interaction. The contribution of the last one is inversely proportional to the ratio of the crystallite volume to its surface area and increases with decreasing size and transition to the amorphous structure.

4. Conclusion

Functional dependencies of the fluctuation field *vs*. the demagnetizing field were established for anisotropic nanocrystalline alloys NdFeB with different constants of intergrain exchange interaction.

The qualitative difference between the dependences of the fluctuation field and the demagnetizing field is shown in the case of exchange-coupled and exchange-decoupled hard magnetic materials based on the NdFeB system.

The phenomenon of cooperative magnetization reversal of an ensemble of particles on the one hand leads to a decrease in the coercive force but on the other hand in the future it can be used to improve the temperature stability of permanent magnets.

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