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MAGNETIC VISCOSITY IN NdFeB MAGNETS

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Abstract. - The relaxation of the magnetization is calculated for isotropic and anisotropic magnets. For NdFeB magnets, the dependence of \( S_v \) on texture, above room temperature, is roughly consistent with the model, while the NdDyFeB magnets show no dependence upon texture.

1. Introduction

The study of time dependent effects in permanent magnets should provide insight into the nature of the coercivity mechanisms. In many systems it is found that the magnetic viscosity \( S = -\mathrm{d}M/\mathrm{d}(\ln t) \) can be used to characterize the data. Early work by Street and Wooley [1] and Néel [2] considered thermally activated transitions between two energy levels separated by a barrier to describe this time dependence. It is the logarithmic behavior of \( M \) which suggests that the activation energies \( E \) for the system possess a broad spectrum. Using this hypothesis, Gaunt [3] related both \( S \), the magnetic viscosity, and \( \chi_{irr} \), the irreversible susceptibility, to the distribution function of the activation energies. This provides a relation between \( S \) and \( \chi_{irr} : S = S_v \chi_{irr} \) where \( S_v = kT/(\partial E/\partial H)_T \). The quantity \( (\partial E/\partial H)_T \) is the magnetic field derivative of the activation energy or effective barrier energy. \( S_v \) is independent [4] of the sample shape, unlike \( S \) and \( \chi_{irr} \). It is useful [2, 4] furthermore, to define an activation volume \( v = (\partial E/\partial H)_T/M_s \), where \( M_s \) is in the saturation magnetization.

Recently, Givord et al. proposed that the magnetization reversal is determined by the formation of a domain wall of width \( \delta \) and is initiated in a volume equal to the activation volume [5]. In reference [5] the angular dependence of the coercive field was also studied after making an assumption about the dependence of the effective barrier energy \( E \) upon the angle \( \theta_{e,H} \) between the external field and the c-axis for an individual grain. Assuming the field dependence of \( E \) is due only to a term which represents the energy of the moments in the applied field, then the \( H \)-dependence of \( E \) is due to the term \(-vM_s H \cos \theta_{e,H} \). This hypothesis results in the well known \( 1/\cos \theta_{e,H} \) dependence for the coercive field of a single grain. The authors of reference [5] obtained good agreement between measured and calculated values for the angular dependence of \( H_c \). The angular dependence of \( H_c \) was interpreted assuming that the anisotropy in the activation volume is not strongly reduced in relation to that of the main magnetic phase, in contrast to the usual assumption.

The present work represents an attempt to test the above hypothesis about the angular dependence of \( E \) in another manner. If the effective barrier energy has the angular dependence assumed above, then the contribution of one grain to the magnetic viscosity parameter \( S_v \) will be \( kT/(\partial E/\partial H)_T = kT/(vM_s \cos \theta_{e,H}) \) and the result for the whole magnet will be determined by an average of this quantity over the angular distribution of grain orientations. We have measured \( S_v \) for magnets with different textures or degrees of orientations, i.e., for isotropic and anisotropic magnets. The distribution of grain orientations was then determined using metallographic techniques to allow the calculation of average values of \( S_v \) for each magnet.

2. Experiment

The magnets used in this work were prepared by powder metallurgy techniques. After grinding, part of the powder was compacted in the presence of a magnetic field of about 1.2 T, thus producing an anisotropic sample. The rest of the powder was compacted without field orientation. The compacts were sintered in vacuum at a temperature of 1 100 °C for one hour, but underwent no further annealing treatment. Needle shaped samples were then cut from the sintered magnets. Thus, samples A and B were produced from the same powder, under the same conditions, and differ only in their degree of orientation. The same holds true for samples C and D.

To determine the spectrum of grain orientations in our samples, a surface containing the orientation direction was prepared for metallographic examination. Photographs of the domain patterns for thermally demagnetized samples were obtained using the Kerr effect and the angles between the alignment direction and the domain wall direction were measured at each point of a grid placed over the photograph. A Gaussian curve was fitted to the data points and the width for a three-dimensional Gaussian distribution was then calculated [6]. For sample A, with composition \( \text{Nd}_{16.8}\text{Fe}_{75.4}\text{B}_{8.8} \), we obtained \( \sigma = 52° \); and for sample C, with composition \( \text{Nd}_{17.2}\text{Dy}_{2.8}\text{Fe}_{73.8}\text{B}_{6.4} \), \( \sigma = 35° \). Samples B and D are, respectively, the isotropic versions of samples A and C.
Measurements of the magnetic viscosity at temperatures above room temperature were made using a VSM coupled to a resistive furnace. Magnetic fields up to 2 T were provided by an electromagnet.

3. Results and discussion

Values of the magnetic viscosity parameter $S_v$ as a function of temperature are shown in figure 1 for all samples. We note that the values of $S_v$ for sample B (isotropic) are consistently larger than those for sample A, but we observe that the curves $S_v$ vs. $T$ for these two samples have the same slope. The identical values of the slope may reflect the fact that the samples A and B were prepared from the same powder under nearly identical conditions. Larger values of $S_v$ for the isotropic sample are to be expected if $S_v$ for a single grain is given by $S_v = kT/vM_s \cos \theta_{g,H}$ as supposed previously.

One could try to calculate an average value for $S_v$ for the whole sample by integrating the above expression for $S_v$ over the entire probability distribution, i.e.

$$S_v = \frac{kTN^{-1}}{vM_s} \int_0^\beta P(\theta_{g,H}) \sin \theta_{g,H} \cos \theta_{g,H} d\theta_{g,H}$$

(1)

where the normalization factor $N$ is given by integrating $P$ over the entire probability distribution. For the isotropic case, $P(\theta_{g,H}) = 1$, while, for the anisotropic case, we used the Gaussian function $P_a(\theta_{g,H}) = \exp(-\theta_{g,H}^2/2\sigma^2)$. The upper limit of the integral was chosen to include some large percentage of the total number of grains, but the final result is not too sensitive to this value as long as $\beta$ is sufficiently far from $\pi/2$. Using values of $\beta = 52^\circ/60^\circ/78^\circ$, corresponding to fractions $f = 0.7/0.8/0.9$ of the total number of grains, we obtained $(S_v)_{iso}/(S_v)_{aniso} = 1.26/1.35/1.47$. This is in reasonable agreement with the experimental value $1.6 \pm 0.3$. The model suggests, however, that the curves for the isotropic and anisotropic samples should be proportional instead of having the same slope.

Turning our attention now to samples C and D, which differ from A and B in that their composition includes a small amount of Dy, we discover in figure 1 that the curves $S_v$ vs. $T$ coincide for these two samples. Calculations similar to those detailed above with values of $\beta = 49^\circ/60^\circ/68^\circ$, corresponding to fractions $f = 0.7/0.8/0.9$ of the total number of grains result in $(S_v)_{iso}/(S_v)_{aniso} = 1.47/1.67/1.89$. The discrepancy observed here is much larger than the errors estimated for these measurements.

To understand these results, it is useful to consider the integral of equation (1). The authors of reference [5] are careful to distinguish between grains which can be reversed in a field $H$ and those which cannot be reversed because their $c$-axis makes a large angle with the field direction. Indeed, it has been suggested [7] that the upper limit $\beta$ of the integral of equation (1) might be interpreted as corresponding to the angle above which the grain magnetization cannot invert. Then for magnets with larger values of $H_c$, grains at high angles will be less likely to invert $M_s$, resulting in smaller values of $\beta$ and smaller values of the ratio $(S_v)_{iso}/(S_v)_{aniso}$. For magnets with smaller values of $H_c$, we expect more grains to invert at large angles and larger values of the ratio $(S_v)_{iso}/(S_v)_{aniso}$. This is in qualitative agreement with our results.

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