THE INFLUENCE OF THE MAGNETIC VISCOSITY ON PULSED FIELD MEASUREMENTS

R. Grössinger[*], R. Sato Turtelli, C. Téllez-Blanco

Inst. f. Festkörperphysik, TU Wien, Wiedner Hauptstrasse 8-10, A-1040 Vienna, Austria

The theoretical background and the description of the magnetic viscosity are given. Special emphasis on the effect of the magnetic viscosity on hysteresis loop measurements is laid. Experimental examples as obtained for commercial permanent magnets such as Nd-Fe-B, ferrites and for $Sm(Co,Cu)_5$ are given. The viscosity coefficient S_v determined by pulsed field measurements is generally larger than that determined in static field experiments.

1. Introduction

High quality permanent magnets need for their characterization sufficient high external magnetic fields. Therefore the use of pulsed field magnetometers (PFM) which allows fast and reliable measurements were suggested for scientific and for the industrial uses [1,2]. It was shown that after a careful calibration procedure an absolute accuracy of about $\pm$ 1.5% (in field H and in magnetization M), which is comparable with standard VSMs, can be achieved [3]. The advantage of a PFM lies in offering sufficient high fields (as e.g. 10 T) generated in large bores (up to 100 mm) combined with a fast measurement, which is superior to all superconducting systems [4]. The application of transient fields causes, however, also errors which have to be considered. Two possible errors may arise in pulsed field measurements:

a) Eddy current errors. The application of a transient field causes in metallic samples eddy currents which lead to a dynamic magnetization M_{eddy} proportional to dH/dt [5], the proportionality factor being the specific electrical conductivity. Additionally M_{eddy} scales with R^2 (R – is the radius of a rotational symmetrical sample), which means that the error increases quadratically with increasing sample diameter [5]. Fortunately most of the metallic permanent magnets are sintered materials where the specific resistivity (typically 2.10^{-4} $\Omega \cdot m$) is generally a factor 50 - 100 higher that of Cu. Therefore the error in magnetization measurements due to eddy currents is rather small. These considerations lead to the development of an eddy current correction for the hysteresis loops measured in pulsed fields which is called *f/2f* method. In this case one measures the loop with two different pulse durations and calculates the corrected loop point by point applying an extrapolation procedure [6]. It was shown by finite element calculations that for not too large eddy current errors (less than 20%) the corrected loop agrees with the "true" loop within 2%. This means that the effect of eddy currents is understood and can be corrected in most cases [7].

b) Magnetic viscosity effects. When the hysteresis loop of hard magnetic materials is measured in transient fields the so-called magnetic viscosity causes a difference between the measured loop and the "true" loop. The magnetic viscosity is also observed in nonconducting materials (as e.g. ferrites), therefore it occurs not due to eddy currents. Also it has to be mentioned that the time constant of eddy currents (exponential decay) (in metallic samples) is of the order of typical µs, whereas that of the viscosity (logarithmic decay) lies between ms and s. Additionally eddy currents depend on the geometry of the sample whereas this is not the case for the viscosity.

---

[*] Corresponding author: roland.groessinger@ifp.tuwien.ac.at

The effect of the magnetic viscosity is well known since many years and it was also investigated for many hard magnetic materials [see e.g. 8,9]. It was also shown that the magnetic viscosity coefficient S_v can be used to determine the activation volume that is an important parameter for the understanding of the coercivity mechanism [10]. The viscosity coefficient is usually determined in static field measurement. One measures the loop $M(H)$ and stops in the second quadrant with this measurement at a certain field H. There under the condition H = const. one measures $M(t)$, from which the magnetic viscosity S can be determined. For calculating the magnetic viscosity coefficient S_v one needs also the irreversible susceptibility χ_{irr}. The general question, which will be discussed in the present paper is, if this viscosity parameter determined from quasi-static field experiments can also be used in order to understand the influence of transient fields on the shape of the hysteresis loop in pulsed field measurements. One has here to consider that the typical field sweep rate in pulsed field experiments dH/dt is approximately 1000 T/s which is 106 orders of magnitude bigger than usual in VSM`s using an electromagnet or a superconducting coil.

In the following the theoretical background as well as experimental examples for the magnetic viscosity will be given. As examples, measurements on permanent magnets as well as on selected samples such as $Sm(Co,Cu)_5$ will be shown.

2. Models

Magnetic viscosity in magnetic materials may occur due to the diffusion effect (migration of impurity atoms), tunneling effect (at low temperatures in highly anisotropic materials) and thermal activation of metastable states over energy barriers. The thermal activation process is specific for permanent magnets. Thermal activation is a time-dependent process and this leads to time-dependence of the magnetization reversal. An example of the practical importance of this effect is the frequency-dependence of the coercivity in recording media which has important consequences for the field required to write information and the storage life of that information.

For a flat distribution of energy barriers at a fixed field the magnetization decays with time, t, as [11]:

$$M(t) = A + S \ln(t + t_0),$$ (1)

where $t_0 = S / \dot{M}(0)$ and $\dot{M}(0)$ is the initial rate of change of magnetization which depends on the experimental conditions. Street et al [12] showed that the magnetic viscosity S is given by:

$$S = \Lambda . \chi_{irr} \text{ respectively } S_v = \Lambda = S / \chi_{irr}$$ (2)

Here both S and χ_{irr} have been corrected for demagnetization effects. χ_{irr} is that part of the magnetic susceptibility which reflects irreversible processes and is associated with a state activating over an energy barrier. Eq. 2 defines the magnetic viscosity coefficient S_v derived from the viscosity S given by Eq. 1. Assuming that each successful activation event results in the same change in magnetization, Street et al [12] showed that $\Lambda = S_v$ was related to the energy barriers of magnetization reversal within a material by:

$$S = -\frac{k_B T \chi_{irr}}{(dE/dH)\big|_{M_{irr}}},$$ (3)

where T is the absolute temperature, k_B is the Boltzmann's constant, E is the energy barrier and H is the field. It is important that measurements of dE/dH should be taken at constant M_{irr} so that the same energy barrier is being probed as the field is changed. The viscosity parameter S_v is analyzed considering [13]

$$S_v = S(1 - N\chi_{rev}) / \chi_{irr},$$ (4)

where N is the demagnetization factor of the material and χ_{rev} is the reversible susceptibility. Generally $N\chi_{rev} << 1$, then Eqs. 1 and 3 for $J = \mu_0 M$ can be rewritten respectively by:

$$J = const + \frac{S_v}{\chi_i} \ln t$$ (5)

$$S_v = -k_B T / \frac{dE}{dH}$$ (6)

S_v is related to the so-called activation volume v (volume within which the magnetization is reversed by one individual activation process) by the following relation:

$$S_v = \frac{k_B T}{M_s v} \tag{7}$$

Givord *et al.* [8] deduced for various permanent magnets that S_v varies with H_C as: $S_v \propto H_c^{1.5}$. For pulsed field measurements it was considered that the time dependence of the coercivity is related to a thermal activated process where the relaxation time is associated with the energy barrier $dE/dH|_{H \approx H_C}$. Under this assumption an expression for the temperature dependence of the coercivity in permanent magnets with narrow domain walls (high anisotropy) was deduced [14]:

$$H_C = \frac{8\pi AKD}{k_B T M_S} \cdot \frac{1}{\ln t/\tau_0}, \tag{8}$$

where K, M_S, A and D are the anisotropy energy, the saturation magnetization, the exchange energy and the density of defects participating in the relaxation phenomenon, respectively. The relaxation time t follows the classical Arrhenius law: $t = \tau_0 \exp(E/k_B T)$, where E is the energy barrier. The constant τ_0 depends on the exact nature of the magnetization reversal process and lies between 10^{-12} and 5×10^{-7} s [8]. According to [14] the time dependence of the coercivity was determined from loop measurements as a function of dH/dt, where for t approximated values were obtained according to:

$$t \cong \frac{S_v}{dH/dt} \tag{9}$$

Using pulsed field measurements and analyzing the time dependence of the coercivity of $SmCo_{5-x}Co_x$ as a model-material an exponential dependence between the field sweep rate dH/dt and the coercivity was found [14]:

$$\frac{dH}{dt} = \frac{dH_0}{dt} \cdot \exp(H_C/S_v) \tag{10}$$

From this, finally, an estimate for the viscosity coefficient S_v was obtained [15]:

$$S_v = \frac{H_{C1} - H_{C2}}{\ln[(dH_1/dt)/(dH_2/dt)]} \tag{11}$$

where H_{c1} and H_{c2} are the coercive fields obtained from the hysteresis loops measured at field rates, dH_1/dt and dH_2/dt, respectively. Grössinger *et al.* have shown that in $Sm(CoCu)_5$ crystalline alloys, the relation above is valid for a large range of dH/dt [15]. Therefore it is suggested that using formula (11) allows to determine the viscosity coefficient S_v using the dependence of H_C from the field sweep rate dH/dt.

3. Experimental procedure

Three sets of samples are shown:

a) Nanocrystalline Ba-ferrite: Hexagonal Bariumferrite ($BaO\ 6Fe_2O_3$) is a well known permanent magnet material. There exist many publications describing the physical properties of common Bariumferrite [16,17]. In a previous work it was tried to produce nanocrystalline Ba-ferrite using various methods of high energy ball milling [18]. To obtain small grains (about 100 nm), two ways of production were applied: Intensive Milling (sample name: IM) and Mechanical alloying (sample name: MA) [18]. In both cases, a high energy ball mill was used to reduce the grain size of commercial Bariumferrite powder or of Barium carbonate and Iron oxide (mechanical alloying) respectively. For further reduction of the grain size a small amount of niobium was added to Barium carbonate and Iron oxide (sample name: MAN). After milling the powder in air, it was pressed in a steel press and annealed at 1273 K for 1 hour.

b) Standard permanent magnets like Nd-Fe-B from VAC (270/95h).

c) *$SmCo_{5-x}Cu_x$* (x= 0, 1, 1.5, 2, 2.5, 3, 4): Polycrystalline $SmCo_{5-x}Cu_x$ were prepared by induction melting appropriate amounts of the raw materials of a purity of at least 99.9 wt%. The

samples were investigated in the as "cast" state as well as after annealing. The annealing was done at 1273 K for three weeks.

The time dependence of the polarization $J(t)$ and the coercive field H_c applying different rates of change in magnetizing field were measured by using both a vibrating sample magnetometer (VSM) at room temperature and an extraction (at low temperatures) magnetometer with external field strength up to 4.8 and 12 MA/m, respectively. The pulsed field measurements were performed in a PFM at T.U.Vienna which allows pulse durations between 10 and 50 ms.

4. Results and discussion

4.1. Ba-ferrite

Performing hysteresis measurements with the pulse field system, we have seen that the measured coercive field differs slightly, if we apply different field rates. To allow a comparison measurements were performed using a static field system (VSM) and a pulsed field magnetometer. The measured coercivity increased with increasing field rate. These results were attributed to the magnetic viscosity. The viscosity coefficient can be determined either by using formula (4) for the quasistatic measurements or formula (11) for the pulsed field measurements. Fig. 1 shows that the measured magnetization of different samples as a function of time measured with the VSM. Fig. 2 shows the H_C versus dH/dt dependence for these samples. In the figures one can also see the viscosity coefficients S_v obtained using formula (11). The values differ depending on the method used (see table I). The general magnitude of S_v agrees well with microcrystalline ferrites [19]. However, the disagreement between the S_v values determined by the two different methods is not yet clear.
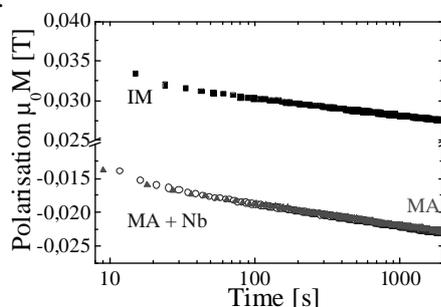

Fig. 1. Time dependence of the magnetization of nanocrystalline Ba-ferrite.
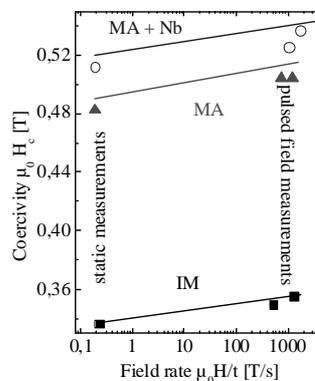
Fig. 2. Dependence of H_c on the sweep rate of nanocrystalline Ba-ferrites.

Table 1. S_v values determined by pulsed and static field measurements.

Samples	Pulsed field $\mu_0 S_v$ (mT)	Static field $\mu_0 S_v$ (mT)
IM	1.9	1.1
MA	2.6	1.3
MA+Nb	2.2	1.3

4.2. Nd-Fe-B magnet

For a systematic comparison with standard viscosity experiments, a set of magnetization curves was drawn at different rates of change in magnetizing field using a quasistatic VSM measurement. Details of these measurements are shown and discussed in [20]. Typical results as obtained for a commercial sintered Nd-Fe-B magnet from VAC (270/95h) in a VSM are shown in Fig. 3. This demonstrates that even at low sweep rats the viscosity effect is clear detectable.
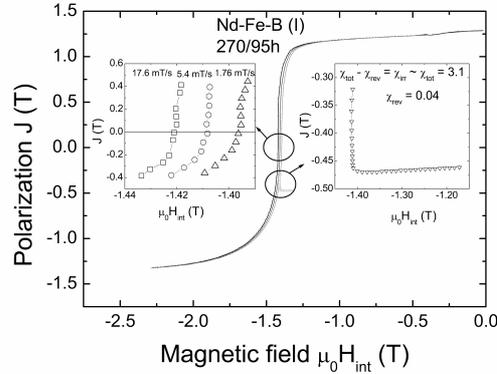
Fig. 3. Part of the hysteresis cycle for a NdFeB-magnet at 300 K by using a VSM for different rates of change in magnetizing field strength: 17.6, 5.4, and 1.76 mT/s. χ_{irr} is approximately equal to the total susceptibility, χ_{tot}.

4.3. SmCo$_{5-x}$Cu$_x$ alloys

Sm(Co,Cu)$_5$ magnets show giant intrinsic magnetic hardness irrespective of the mode of preparation [21]. The coercivity at room temperature in Sm(Co$_{1-x}$Cu$_x$)$_5$ materials were measured by using a pulsed field magnetometer with maximum applied fields of H = 12 MA/m and fast pulses of 4 ms [21]. H_C increases linearly with the Cu concentration up to x = 0.5.

In Fig. 4 the room temperature coercive field as obtained in pulsed field measurements with different dH/dt measured in SmCo$_{5-x}$Cu$_x$ as-cast and annealed magnets is shown. The coercivity increases with dH/dt evidencing the existence of a strong magnetic after effect from which the viscosity coefficient S_v can be determined.
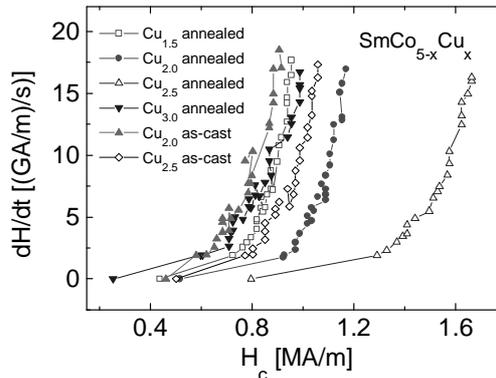

Fig. 4. *Hc* as function of *dH/dt* for SmCo$_{5-x}$Cu$_x$ alloys.

For example, for the samples SmCo$_{2.5}$Cu$_{2.5}$ and SmCo$_3$Cu$_2$ values of S_v of 0.11 and 0.17 MA/m were obtained covering the whole dH/dt range. From these results one comes to the conclusion that S_v is independent of dH/dt, i. e. it does not change between very low and very high values of dH/dt. This result is extremely important for two reasons:

a) S_v of the permanent magnets can be determined by a pulsed field method; therefore, this method is very practical;

b) To estimate the errors in a pulsed field magnetometer due to the magnetic viscosity, the value S_v as obtained by standard experiments can be used also for large dH/dt ranges. The change in H$_C$ gives an upper boundary for this effect.

Generally, at a given temperature, a higher coercivity corresponds to a higher S_v value. S_v versus the coercive field obtained at a sweep rate of 16.3 GA/m·s in alloys with x = 2.0 and 2.5 is shown in Fig. 5.
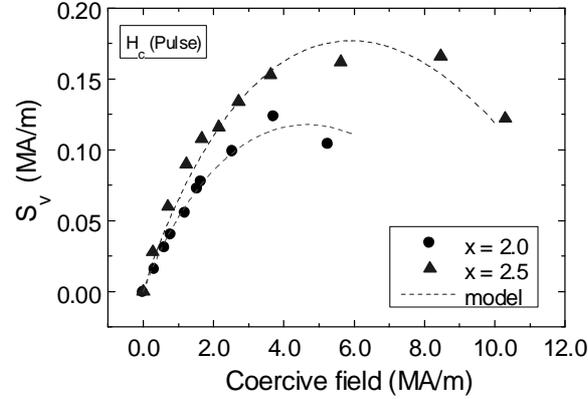
Fig. 5. Coercive field dependence of S_V obtained for x = 2 and 2.5. The dashed line indicates the fitting of the equation $S_v = aH_c - b(H_c)^{1.5}$ to the experimental data.

The well known Barbier relations, which is based on the assumption that H_C is proportional to the anisotropy field and S_V is proportional to $1/\delta_W{}^3$ [22]:

$$H_C \approx S_v{}^{2/3} \qquad \text{or} \qquad S_v \approx H_C{}^{1.5} \qquad\qquad (12)$$

have been tested for various materials. However, we found for $Sm(Co,Cu)_5$ an extension of the Barbier plot leading to a semi-empirical relation of the type:

$$S_v = aH_c - b(H_c)^{1.5}, \qquad\qquad (13)$$

where a and b are constants. Excellent agreement was obtained between this relation and the experimental data. This result shows also that for describing the magnetic viscosity over a broad temperature range, one needs to include thermal activation processes.

5. Summary

The magnetic viscosity is an important property which is generally found for all hard magnetic materials. From the viscosity coefficient S_v the activation volume can be determined which is important for the understanding of the coercivity mechanism. The magnetic viscosity influences also the shape of the hysteresis loop and changes consequently the hysteresis parameters (as e.g. the coercivity). This effect becomes very important for pulsed field measurements. The dependence of the coercivity H_C on the field sweep rate dH/dt can be used to estimate S_v. However the determined values for S_v are generally larger than those classically determined.

Although equation (1) has been used for decades to describe the magnetic relaxation process, it is necessary here to consider an approximation to be applicable to a limited time interval, because it diverges for both at $t \rightarrow \infty$ and $t \rightarrow 0$. The large-time failure of equation (1) is related, in reality, when it is used for systems with a complicated energy landscape and barriers widely distributed in amplitude causing a distribution of the time needed to reach equilibrium. To date, there is only short discussion on the magnetization decay at $t \rightarrow 0$. In fact, it is impossible by means of VSM to measure the magnetization decay in times less than a few milliseconds. However, our pulse-field instrument can measure a loop in several milliseconds with very high dH/dt. We speculate that the pulse-field set might provide an experimental tool for the measurements of magnetization decay at $t \rightarrow 0$, if we could know exactly to what extent the sweep-rate experiments are able to describe the magnetic relaxation process.

Acknowledgement

References

[1] R. Grössinger, M. Küpferling, A. Wimmer, M. Taraba, W. Scholtz, J. Dudding, P. Lethuillier, B. Enzberg-Mahlke, W. Fernengel, G. Reyne, Proc. of the 16th Internat. Workshop on Rare-Earth Magnets and Their Applications, Ed by H. Kaneko, M. Homma, M. Okada; Sendai, Japan, 2000, pp. 1129.

[2] J. Dudding, P. A. Knell, R. N. Cornelius, B. Enzberg-Mahlke, W. Fernengel, R. Grössinger, M. Küpferling, P. Lethuillier, G. Reyne, M. Taraba, J. Magn. Magn. Mater. **242-245**, 1402 (2002).

[3] R. Grössinger, M. Taraba, A. Wimmer, J. Dudding, R. Cornelius, P. Knell, P. Bissel, B. Enzberg-Mahlke, W. Fernengel, J. C. Toussaint, D. Edwards; IEEE Trans. Magn. **38**, 2982 (2002).

[4] R. Grössinger, E. Wiitig, M. Küpferling, M. Taraba, G. Reyne, C. Golovanov, B. Enzberg-Mahlke, W. Fernengel, P. Lethuillier, J. Dudding; IEEE Trans. Magn. **35**, 3971 (1999).

[5] R. Grössinger, M. Küpferling, P. Kasperkovitz, A. Wimmer, M. Taraba, W. Scholz, J. Dudding, J. C. Toussaint, B. Enzberg-Mahlke, W. Fernengel, G. Reyne; J. Magn. Magn. Mater. **242–245**, 911 (2002).

[6] G. W. Jewell, D. Howe, C. Schotzko, R. Grössinger, IEEE Trans. Magn. **28**, 3114 (1992).

[7] C. Golovanov, G. Reyne, G. Meunier, R. Grössinger, J. Dudding, IEEE Trans. Magn. **36**, 1222 (2000).

[8] D. Givord, M. F. Rossignol, V. Villas-Boas, F. Cebollada, J. M. Gonzales, in: F. P. Missel et al., (Eds.), Rare Earth Trans. Metal Alloys, Publ. World Scientific, Sao Paulo, Brasil, 1996, pp. 21.

[9] R. Street, R. K. Day, J. B. Dunlop, J. Magn. Magn. Mater. **69**, 106 (1987).

[10] D. Givord, P. Tenaud, T. Vadieu, IEEE Trans. Magn. **MAG-24**, 1921 (1988).

[11] R. Street, J. C. Woolley, Proc. Phys. Soc., **62A**, 562 (1949).

[12] R. Street, J. C. Woolley, P.B. Smith; Proc. Phys. Soc. **65A**, 679 (1952).

[13] R. Street, R. K. Day, J. P. Dunlop. J. Magn. Magn. Mater. **69**, 106 (1987).

[14] J. C. Téllez-Blanco, R. Sato-Turtelli, R. Grössinger, E. Estévez-Rams, J. Fidler, J. Appl. Phys. **86**, 5157 (1999).

[15] R. Grössinger, J. C. Téllez-Blanco, R. Sato Turtelli, R. R. Hauser, K. Reiterer, H. Sassik, G. Chouteau, Physica B **294–295**, 194 (2001).

[16] K. Kojima, Ferromagnetic Materials, Vol. 3, Edited by. E.P. Wohlfahrt, North-Holland Publishing Company, 1982, pp. 307.

[17] J. Smit, H. P. J. Wijn, Ferrites, Hexagonal ferrites, Philips Tech. Library, 1959, pp. 547.

[18] P. Kerschl, R. Grössinger. C. Kussbach, R. Sato-Turtelli, K. H. Müller, L. Schulz, J. Magn. Magn. Mater. **242–245**, 1468 (2002).

[19] David Crew; Thesis Univ. of Western Australia (1997).

[20] J. C. Tellez Blanco, R. Sato-Turtelli, R. Grössinger, D. Eckert, A. Handstein, K. H. Müller, to be published.

[21] J. C. Téllez-Blanco, R. Grössinger, R. Sato Turtelli, J. Alloys Compd. **281**, 1 (1999).

[22] J. C. Barbier, Ann. Phys. **9**, 84 (1954).