



# The peak in anomalous magnetic viscosity

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## ABSTRACT

Anomalous magnetic viscosity, where the magnetization as a function of time exhibits non-monotonic behaviour, being seen to increase, reach a peak, and then decrease, is observed on recoil lines in bulk amorphous ferromagnets, for certain magnetic prehistories. A simple geometrical approach based on the motion of the state line on the Preisach plane gives a theoretical framework for interpreting non-monotonic behaviour and explains the origin of the peak. This approach gives an expression for the time taken to reach the peak as a function of the applied (or holding) field. The theory is applied to experimental data for bulk amorphous ferromagnet alloys of composition  $\text{Nd}_{60-x}\text{Fe}_{30}\text{Al}_{10}\text{Dy}_x$ ,  $x = 0, 1, 2, 3$  and  $4$ , and it gives a reasonable description of the observed behaviour. The role played by other key magnetic parameters, such as the intrinsic coercivity and fluctuation field, is also discussed. When the non-monotonic behaviour of the magnetization of a number of alloys is viewed in the context of the model, features of universal behaviour emerge, that are independent of alloy composition.

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## 1. Introduction

The time dependent behaviour of the magnetization, termed the magnetic viscosity, is well known in ferromagnetic materials, and is a consequence of thermally activated processes that excite transitions over energy barriers [1,2]. The field history used to observe magnetic viscosity (see Fig. 1) is a single-step process, namely a single-step change from a large positive field that magnetically saturates the material to a holding field, usually close to the coercive field,  $H_c$ , where the magnetic polarization,  $J$ , is seen, over a limited range of times, to decay with time,  $t$ , according to the relation

$$J(t) = S_v \ln(t) + \text{const}, \quad (1)$$

where  $S_v$  is the magnetic viscosity coefficient [1].

For a more complicated field history, as is the case for the two-step process, the magnetic polarization as a function of time displays a distinctly different behaviour. In this case, shown diagrammatically in Fig. 1, a magnetic field,  $H_s$ , is applied in the forward direction to magnetically saturate the material, the field is then ramped in a negative direction, through zero, to a field in the third quadrant, the turning point denoted  $H_1$ , where it is paused momentarily, and then ramped in a positive direction along a recoil line to the final or holding field  $H_2$ , where it is held constant

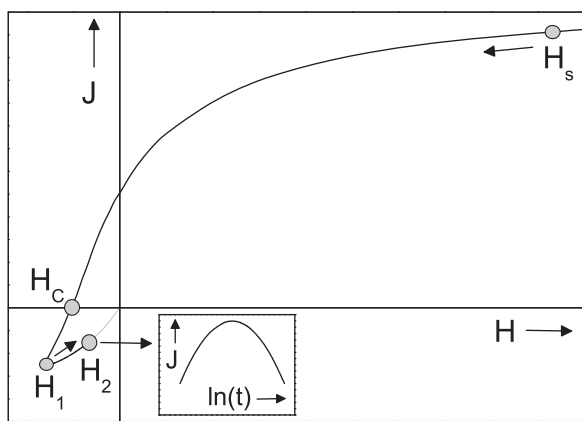
and the magnetic polarization is measured as a function of time. When this two-step field history is followed, for certain values of  $H_1$  and  $H_2$ , the magnetic polarization as a function of time (as shown in the inset in Fig. 1) exhibits non-monotonic behaviour, being seen to increase, reach a peak, and then decrease. The non-monotonic behaviour of the magnetic polarization is often termed anomalous magnetic viscosity. There is the special case for the two-step process, where  $H_1$  is chosen to give the recoil line that leads to the dc demagnetized state i.e. zero magnetic polarization in a zero field.

Anomalous magnetic viscosity has been observed in  $\text{Nd}_4\text{Fe}_{77}\text{B}_{19}$  flakes prepared by melt-spinning [3] and magnetic tapes [4,5]. However, in these materials it is a very small effect, hampering its detailed study. It has been found that anomalous magnetic viscosity is much larger in bulk amorphous alloys with hard magnetic properties, in particular the multicomponent alloy system  $\text{RE-Fe(Co)-Al}$ ,  $\text{RE} = \text{Nd or Pr}$  [6–9], making them ideal materials for its study. The Preisach model of hysteresis, with its concept of the Preisach plane, provides a theoretical framework for understanding the non-monotonic behaviour of the magnetization, with its associated peak [4,7,10–12].

The prime interest of this study of anomalous magnetic viscosity is the relationship between the time taken to reach the peak,  $t_{pk}$ , and  $H_2$ , and the exploration of the roles played by other parameters that relate to the field history. It is shown how a very simple geometrical approach based on the motion of the state line on the Preisach plane can be used to explain non-monotonic behaviour, and an expression relating  $H_2$  and  $t_{pk}$  deduced. This approach is then expanded

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**Fig. 1.** Magnetic field history for single and two-step magnetic viscosity experiments:  $H_s$  is the applied field to magnetically saturate the material,  $H_c$  is the coercive field,  $H_1$  is the holding field in the single-step experiment for observation of magnetic viscosity, and in the two-step experiment for observation of anomalous magnetic viscosity  $H_1$  is the turning point and  $H_2$  is the final (or holding) field lying on the recoil line for the special case leading to the dc demagnetized state (zero magnetic polarization in zero field). When held at  $H_2$  the magnetic polarization,  $J$ , is seen to vary in a non-monotonic fashion with time,  $t$ , as is shown in the inset.

through consideration of the mathematical equation that governs the behaviour of the state line, without the need to resort to the more detailed mathematics of the Preisach model, including the Preisach distribution. (As a general comment, studies have tended to overlook the rather unique circumstances that apply when  $t_{pk}$  occurs, and the simplifications that result.) The approach is tested using experimental data from bulk amorphous ferromagnet alloys of composition  $\text{Nd}_{60-x}\text{Fe}_{30}\text{Al}_{10}\text{Dy}_x$ ,  $x = 0, 2$  and  $4$ ,  $\text{Nd}_{60}\text{Fe}_{20}\text{Co}_{10}\text{Al}_{10}$ , and  $\text{Pr}_{58}\text{Fe}_{24}\text{Al}_{18}$ . Also, the roles of key magnetic parameters, i.e. intrinsic coercivity, fluctuation field and the turning point ( $H_1$ ), are explored. A secondary aim of this study is to explore the effect of small additions of Dy on the anomalous magnetic viscosity. The partial substitution of Nd by Dy in  $\text{Nd}_{60-x}\text{Fe}_{30}\text{Al}_{10}\text{Dy}_x$  bulk amorphous ferromagnets is known to substantially increase the coercivity [13], but its effect on the behaviour of the anomalous magnetic viscosity is little explored. The application of this Preisach based geometrical approach to alloys of differing composition allows features to emerge that have a universal character.

## 2. Preisach model and theory

For a general overview of the Preisach model the reader is referred to the monographs of Bertotti [11] and Della Torre [12], and [14–16] (and references therein) which bring together the Preisach model and the ideas of Néel relating to thermal activation over energy barriers, particularly in the context of a single-step field history. Here, only key elements of the Preisach model relevant to the non-monotonic behaviour of the magnetization are discussed, with the focus on the peak in the anomalous magnetic viscosity, which is a consequence of the two-step field history.

In the Preisach model the magnetic polarization arises from the sum of contributions from square-loop bi-stable units or hysterons, which can switch between two states, an ‘up’ state and a ‘down’ state. To change from one state to another, switching over an energy barrier takes place, and this may arise from thermal activation or be driven by an external field, as is the case with magnetic hysteresis. Many hysterons make up a system, and at any given time there exist two subsets of units,  $S_+$  and  $S_-$ , denoting those units in the ‘up’ or ‘+’ state and those in the ‘down’ or ‘−’ state, respectively. Each elementary hysteron is characterized by a

half-width or critical field,  $h_c$ , and a mid-value or interaction field,  $h_u$ , of its M–H hysteresis curve, and is linked with a point in the  $(h_c, h_u)$  plane, termed the Preisach plane. For a certain field history a state line  $b(h_c)$  is formed in the Preisach plane separating the  $S_+$  and  $S_-$  subsets (see Fig. 2) [10,17].

The sequence of time events for both the single-step and two-step magnetic viscosity experiments shown in Fig. 1 are detailed in Fig. 2(a). In the single step experiment the field is ramped from  $H_s$  to  $H_1$ , with the time at which  $H_1$  is attained  $t_0$ . In the two-step experiment, there is an additional step, with the field being reversed and then ramped to  $H_2$ . In real experiments the field does not reverse instantaneously at  $t_0$ , rather there is a short dwell time of order 10 s, where  $H_1$  remains constant, before the field ramp from  $H_1$  to  $H_2$  commences at  $t_1$ . The field  $H_2$  is attained at  $t_2$  and remains constant. At a time that may vary from a few tens of seconds to many hours, the peak in the magnetization is reached at  $t_{pk}$ .

The Preisach description for the simple case of a single-step change from positive saturation,  $H_s$ , to a field,  $H_1$  (the field-history of the magnetic viscosity experiment shown in Fig. 1), is shown in Fig. 2(b). The state line separates the regions of ‘−’ hysterons and ‘+’ hysterons, and it has a component that moves to the right, with logarithmic speed [17]. As time progresses, the state line moves to the right, as indicated by the arrow in Fig. 2(c), the ‘+’ denoted region decreases, and the magnetic polarization decreases. This is the well known behaviour observed experimentally in magnetic viscosity experiments [1].

For the two-step field-history shown diagrammatically in Fig. 1, required for the observation of anomalous magnetic viscosity, there is an additional field step, with the field increased from the turning point  $H_1$  to the holding field  $H_2$ . The state line is now composed of two portions which relax on two fronts: a front of negative gradient, emanating from the field step from  $H_1$  to  $H_2$ , shown in Fig. 2(d), that bounds the small triangular area of ‘−’ hysterons, denoted Area A (shaded grey), in Fig. 2(e); and a front of positive gradient emanating from the field step from  $H_s$  to  $H_1$  that bounds the triangular area of ‘+’ hysterons denoted Area B (shaded grey), shown in Fig. 2(g). The motion of these two fronts give opposite contributions to the observed magnetic polarization i.e. as time progresses Area A contains fewer ‘−’ hysterons (it decreases in area), so the magnetic polarization will increase, whilst for Area B the number of ‘+’ hysterons decrease, as its area decreases, and there will be a corresponding decrease in the magnetic polarization. As has been pointed out by Bertotti [11], the two fronts relax with very different time constants, with the front that defines Area A relaxing much faster than the front that defines Area B. For this reason the reduction in Area B is shown in Fig. 2 as not commencing until Area A has disappeared.

This is the origin of the peak in the anomalous magnetic viscosity. The observed magnetic polarization is the sum of contributions from Area A and Area B. There is an increase in the magnetic polarization as those hysterons that make-up Area A relax (Fig. 2(e)), and at a point in time the hysterons in Area A will be exhausted. This is the time at which the peak,  $t_{pk}$ , is reached, shown in Fig. 2(f). Finally, as shown in Fig. 2(g), there will only be contributions from hysterons in Area B, and the magnetic polarization will be seen to decrease with time. What is also apparent is that the hysterons that make up Area A have exhausted (Area A in Fig. 2(e) reaches zero area) long before there is any significant contribution from Area B, as the respective time constants are significantly different. To a very good approximation Area B can be ignored, and only the motion of the portion of the state line that defines Area A need be considered, with the condition that the peak occurring when Area A is zero which occurs at  $t = t_{pk}$ .

The recognition that Area B can be ignored, with only Area A considered in determining when  $t_{pk}$  occurs, enables simplification of

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