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MAGNETIZATION REVERSAL IN MELT-QUENCHED NdFeB

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ABSTRACT

Melt-quenched NdFeB is an important modern permanent magnet material. However, there still remains doubt as to the magnetization reversal mechanism which controls coercivity in material prepared by this processing route. To investigate this problem a new technique based on measurements of reversible magnetization along recoil curves has been used. This technique identifies the presence of free domain walls during magnetic reversal. For this study samples of isotropic (MQI), hot pressed (MQII) and die upset (MQIII) melt-quenched NdFeB were examined. The results indicate that in MQI free domain walls are not present during reversal and the reversal mechanism is most likely incoherent rotation of some form. Free domain walls are also not present during reversal in the majority of grains of MQII, even though initial magnetization measurements indicate that the grain size is large enough to support them. In MQIII free domain walls are present during reversal. These results are attributed to the reduced domain wall nucleation field in MQIII compared with MQII and the increased dipolar interactions in MQIII.

INTRODUCTION

Melt-quenched NdFeB is an important permanent magnet material which has many technological uses. The magnetization reversal mechanism of material prepared by this processing route is still the subject of doubt, however. Studies of the time dependence of magnetization and measurements of the irreversible susceptibility [1] have concluded that the reversal mechanism is consistent with nucleation. This conclusion is supported by other theoretical and experimental studies of the coercivity [2,3]. At the same time transmission electron microscopy studies performed on thin foils [4] have shown pinning of domain walls at grain boundaries and this supports a domain wall pinning model of magnetization reversal [5].

In this work we use a new technique, developed by Cammarano, McCormick and Street [6] based on measurements of recoil curves from the major hysteresis loop, to investigate magnetization reversal in melt-quenched NdFeB. This technique is based upon the idea that the reversible magnetization (M_{rev}) is not only a function of the field H but also of the irreversible magnetization (M_{irr}), an indicator of the magnetic state of the system. Thus plots of M_{rev} vs. M_{irr} at constant values of field can give insight into the reversal mechanism. These curves will be referred to as M_{rev} curves.

To experimentally determine the M_{rev} curves, a series of recoil curves are measured from the major hysteresis loop to zero internal field as shown in Fig. 1. A value of internal field (H) is chosen and the values of the reversible magnetization (M_{rev}) and the irreversible magnetization (M_{irr}) are calculated for each recoil curve measured. This procedure then defines one M_{rev} curve at a particular field H. By choosing different values of the internal field H a family of such curves may be obtained.

Models of magnetization reversal have shown [7,8] that if domain walls are free to move or bow under the influence of an applied field during reversal of magnetization then the M_{rev} curves exhibit a minimum. This result has been confirmed by experiment on a sample of a confirmed pinning controlled Sm_2M_{17} type magnet [9]. A minimum in the M_{rev} curves only occurs if the domain walls are able to reversibly bow under the action of an applied field. If this is not the case and the domain walls are unable to bow, or if domain walls are not present within the material
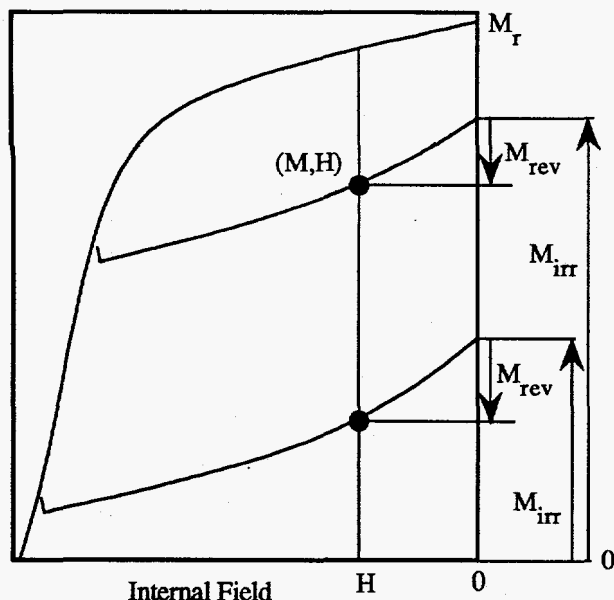
FIG. 1: The definitions of M_{rev} and M_{irr} used in this work plus how the reversible magnetization curves at any field were calculated.

during reversal, then the reversible magnetization behavior observed may only arise from rotation of the magnetization vectors away from the easy axis. In this case the M_{rev} curves are found to be straight lines with slopes that increase with increasing internal field H.

The difference in the M_{rev} curves measured from samples that reverse via free domain wall motion versus those that reverse by rotation of the magnetization permits the use of this technique to discriminate between different reversal mechanisms.

EXPERIMENT

In this study three samples of melt-quenched NdFeB, subject to different post-solidification processing steps, were used. The first sample, MQI, was optimally quenched ribbon which had been compacted with a resin binder to form a material with an isotropic distribution of easy axis orientations. The second sample, MQII, consisted of ribbons compacted with hot pressing, producing a very small amount of grain alignment. The last sample, MQIII, was comprised of quenched ribbons which had been hot pressed and then die upset to produce a material which was highly anisotropic and well aligned. All samples were supplied by Magnequench.

MQI and MQIII were ground and polished into spheres using the sphere polisher described by Folks et al. [10]. The final polishing medium was 0.25 μm diamond paste. Corrections to the magnetic measurements were made to account for the non-zero demagnetization factor of these samples. The MQII sample was cut into the shape of a square-based prism with a height-to-width ratio greater than 4. The surface finish was 300 grit fine sandpaper and the demagnetization factor was small enough that it could be neglected in this study.

Each of the samples was measured from the virgin magnetic state, i.e. after thermal demagnetization. Both the initial magnetization curve was recorded as well as the isothermal remanent magnetization (IRM) curve [11] which is an indication of irreversible processes which occur along the initial magnetization curve. After saturation in a 50 kOe positive field, recoil curves from the major hysteresis loop were measured on each sample. The recoil curves were measured in the second quadrant of the hysteresis loop and so all measurement fields were negative. The instrument used was either a vibrating sample magnetometer (MQI and MQIII) or a SQUID magnetometer (MQII). All measurements were performed at room temperature.

The data was analyzed using the method described above to obtain a series of M_{rev} curves at different values of field H for each sample.

RESULTS

The M_{rev} curves for MQI are shown in Fig. 2. These data consist of approximately straight lines which increase in slope as the internal field H at which they are measured increases. There is no evidence that a minimum occurs anywhere in this data.

The curves for MQII, shown in Fig. 3, are similar to those for MQI for most of the range of M_{irr} but exhibit a small, sharp minimum in M_{rev} near positive remanence for low values of field. These curves are similar to those measured previously in a Sm_2M_{17} pinning type magnet [9]. The magnitude of the reversible magnetization present in MQII is larger than that measured in MQI and over the majority of the range of M_{irr} there is less curvature as compared with MQI. The

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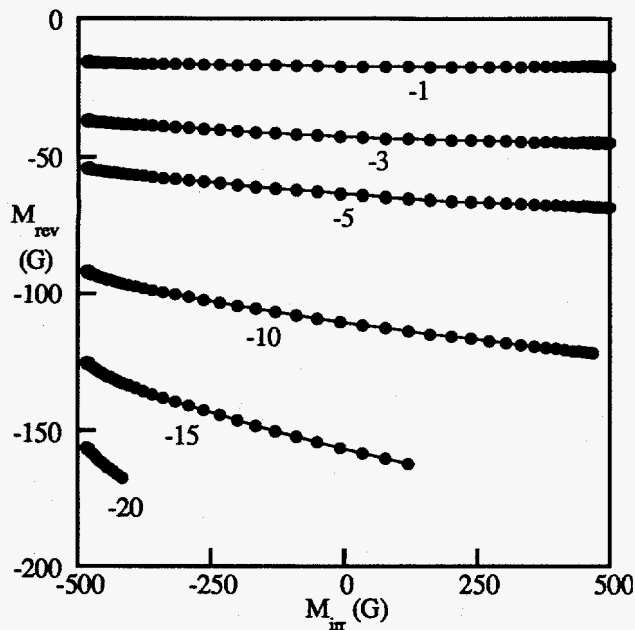
FIG. 2: Reversible magnetization curves for MQI. The fields at which the curves were calculated are shown in the figure in kOe.
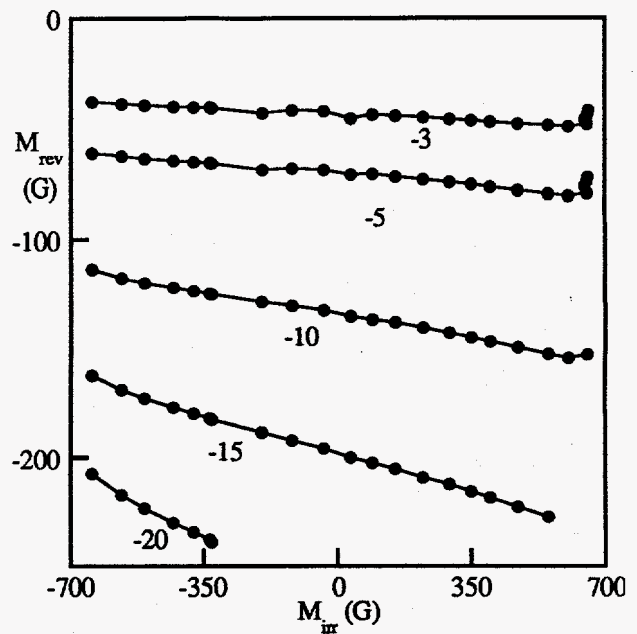
FIG. 3: Reversible magnetization curves for MQII. The fields at which the curves were calculated are shown in the figure in kOe.

data of Fig. 3 were collected using a SQUID magnetometer and are thus more sparse than those of Fig. 2. Nevertheless the trend of the curves in Fig. 3 is clear.

The curves for MQIII, Fig. 4, exhibit a pronounced minimum in the vicinity of $M_{irr} = 0$. This is not a sharp minimum like that seen in MQII. The magnitude of M_{rev} is much smaller than that seen in either MQI or MQII.

The initial magnetization curves, as manifest in the IRM data, are shown in Fig. 5. In low fields MQI evidences little magnetization change, requiring a field near 12 kOe before substantial magnetization is induced. MQII shows two steps in the curve, each of approximately the same magnitude, the first finishing at approximately 10 kOe, the other starting at fields of 17 kOe. Upon careful inspection MQIII shows two steps as well [1], similar to MQII, but the steps are much closer together.

DISCUSSION

MQI

The straight lines seen in the reversible magnetization data for MQI in Fig. 2 suggest strongly that there are no free domain walls present during reversal and that rotation of magnetization vectors away from the easy axis is responsible for the reversible magnetization seen in this material. This conclusion is consistent with the measured initial magnetization curve which shows a long flat region at low field. These results from MQI indicate that domain walls, if present in the thermally-demagnetized state, are not free to move. Additionally, if domain walls are nucleated in a saturated specimen under the action of a reverse field they will be pinned and unable to bow reversibly. The estimation by Folks, Street and Woodward [1] that only 6% of grains in MQI are multi-domain is consistent with this conclusion.

The grain structure of MQI consists of very fine grains (30 nm diameter) which are equiaxed and randomly oriented [12]. This grain size is below the theoretical limit for single domains, calculated for an isolated non-interacting particle to be 300 nm [1]. As the grains in isolation cannot support a domain wall within them, one possible mechanism for reversal in MQI is incoherent rotation such as curling. Such a mechanism would explain both the lack of free domain walls seen in the reversible magnetization measurements and the initial flat region in low fields of the IRM data. It is also possible, however, that magnetostatic and exchange interactions among
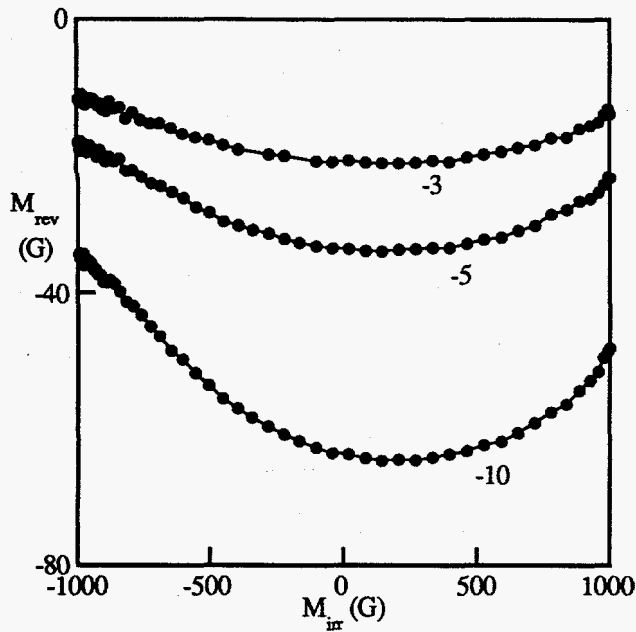
FIG. 4: Reversible magnetization curves for MQIII. The fields at which the curves were calculated are shown in the figure in kOe.
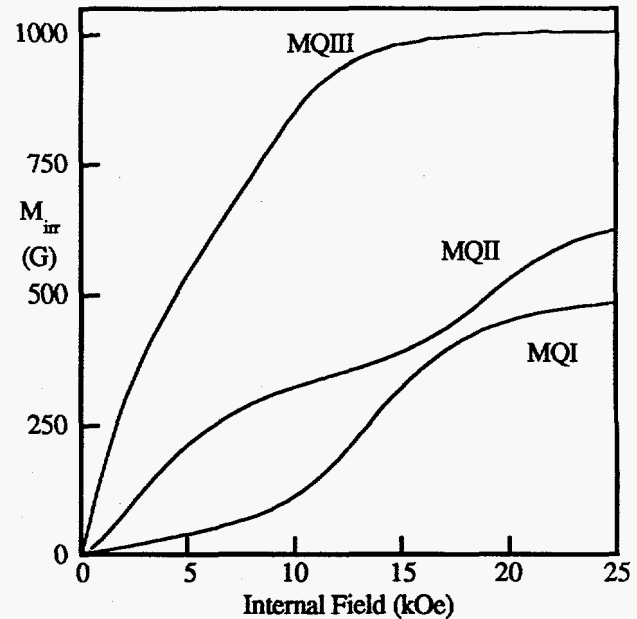
FIG. 5: Isothermal Remanent Magnetization (IRM) curves for MQI, MQII and MQIII showing the steps present in the curves for MQII and MQIII compared with MQI.

neighboring grains in MQI creates interaction domains and thus allows reversal by the movement of domain walls, a reversal route that many not be energetically favorable for an isolated single-domain size grain. If domain wall movement is operative in this material, however, the measured reversible magnetization data provide no evidence of domain wall bowing and thus requires the walls to be trapped at the grain boundaries by pinning sites that are too closely spaced to allow such reversible bowing to occur. This latter mechanism would resemble nucleation in form and indeed these two mechanisms of reversal are difficult to distinguish [13].

MQII

The first step in the IRM curve in Fig. 5 for MQII, in which a rapid increase in magnetization occurs for low fields, suggests that there is a significantly larger proportion of multi-domain grains in this material than were present in MQI. Folks, Street and Woodward [1] estimated that 44% of grains in a similar material were multi-domain in the thermally demagnetized condition. The reversible magnetization data however is only partly consistent with a reversal mechanism which relies upon movement of domain walls. Over the majority of the range of the M_{irr} data, the reversible magnetization curves are straight with a negative slope. This form for the M_{rev} curves is what would be expected if rotation was the dominant mechanism responsible for reversible magnetization. It is likely, therefore, that the reversal mechanism in this material is similar to that of MQI, i.e. incoherent rotation of some form or domain wall movement by domain walls trapped at grain boundaries (and hence unable to bow reversibly). Again the latter reversal mechanism is difficult to distinguish from a nucleation controlled process.

It has been observed that there is an intergranular phase present in this material [14] which may provide exchange coupling between the grains. If the grains are exchange coupled then it is expected that large multi-grain interaction domains will form. This in turn would mean that the most likely reversal mechanism will be domain wall movement or nucleation at the peripheries of reversed regions.

The small minimum in M_{rev} observed near positive remanence in MQII implies that there are, during this part of the reversal, free domain walls able to bow reversibly upon application of an external field. The form of the curves in Fig. 3 are similar to those observed in an Sm_2M_{17} type magnet although in MQII there is no evidence of a coherent intragranular network of pinning

sites. It is logical therefore to attribute the small minimum in M_{rev} observed to the estimated 15% of grains which have been observed by transmission electron microscopy to be large and probably uncoupled [14] to the smaller-sized matrix grains. These large grains form at the positions of former ribbon boundaries during the hot-pressing process [14]. It is feasible therefore that these grains are large enough so that a nucleated domain wall is energetically stable and may bow reversibly upon application of a field. The fact that the minimum in the M_{rev} curve is so sharp indicates that the nucleation field of these grains is low but that little irreversible change occurs until the domain walls are driven from these grains by a higher field. The reason that a higher field is required to remove domain walls from these large grains is due to localized dipolar interactions which hinder the complete annihilation of reverse areas of magnetization. In this way these large grains in MQII behave in a similar manner to large grains of sintered NdFeB and PrFeB on the initial magnetization curve [15].

MQIII

In MQIII the minimum observed in the M_{rev} curves is much broader than that seen in MQII. Folks, Street and Woodward [1] estimate that 49% of the grains in MQIII are multi-domain, a similar proportion to that found in MQII. The occurrence of a notably broad minimum in the M_{rev} curves implies that there are free domain walls present in MQIII during much of the reversal process. The fact that free domain walls are present during reversal over a large range of M_{irr} in MQIII while domain walls are only present in a small proportion of grains in MQII is most likely due to the differences in alignment of these materials. Alignment differences can create changes in the magnitude of the relative dipolar interactions which will alter the reverse domain nucleation and unpinning fields. Additionally, the die-upsetting process can produce subtle chemical and topographical differences in the grain boundary environment.

In MQII the isotropic orientation of the grains means that the dipolar interactions are relatively low when compared with the nucleation or unpinning field, which is influenced by the grain boundary environment. Thus when a domain wall nucleates on the demagnetization curve, or equivalently is unpinned from a grain boundary, the external field is already high enough to overcome dipolar interactions and the domain wall is free to sweep across the grain and to become pinned in the grain boundary on the other side or be annihilated altogether. This process guarantees that there is never a free domain wall and the reversible magnetization data displays characteristics most reminiscent of rotational processes. In MQIII the enhanced grain alignment increases the strength of dipolar interactions and decreases the nucleation field, as evidenced by the lower coercivity of MQIII (12.3 kOe) compared with MQII (18.7 kOe). Additional support for this hypothesis is found in the decreasing trend in coercivity observed in a series of samples as the degree of die-upsetting, and hence alignment, increases [16]. Thus it is reasonable to suppose that when a domain wall is nucleated or unpinned in MQIII, the external applied field is not large enough to overcome dipolar interactions and the domain wall remains within the grain, free to move and bow reversibly. It is possible to observe the effects of the domain walls in MQIII through reversible magnetization measurements. The dipolar interactions in MQIII retard the pinning and annihilation of a domain walls in a similar manner to the mechanism proposed by Taylor et al. [15] to explain the correlation observed between coercivity and initial magnetizing field in sintered NdFeB and PrFeB magnets .

The change in the nature of the dipolar interactions between MQII and MQIII type magnets has been investigated by Gavigan and Givord [17] using measurements of the temperature dependence of coercivity. In their work they concluded that dipolar interactions in MQII type magnets reinforce coercivity, because domain walls are pinned in regions which minimize the divergence of magnetization. In MQIII type magnets they found that the dipolar interactions result in a decrease in the coercivity. It is not inconceivable therefore that this change in the nature of the dipolar interactions between MQII and MQIII could affect the manifestation of free domain walls in these materials.

CONCLUSION

It has been shown that measurements of the reversible magnetization can determine the existence of free domain walls within a material during magnetization reversal. This information then puts constraints upon the possible reversal mechanism.

In MQI, the reversible magnetization measurements point to a reversal mechanism which does not involve free domain walls during reversal. The small grain size of this material and its isotropic orientation favors incoherent rotation or nucleation, although domain wall movement from grain boundary to grain boundary through well coupled grains cannot be discounted.

In MQII a similar process must occur to that operative in MQI. An intergranular phase has been observed in this material which may provide an avenue for exchange coupling of the grains which makes domain wall motion more likely. The initial magnetization curve indicates that the grain size in this material is large enough to support domain walls although only a small minority of grains are found to be multi-domain during demagnetization, which gives rise to a sharp minimum in the reversible magnetization data.

In MQIII the minimum in the reversible magnetization data is much broader than that of MQII which indicates that domain walls are free to move in a large majority of grains during reversal. This result is attributed to well-coupled, aligned grains that may support a domain wall together with dipolar interactions which are large and able to prevent a domain wall from being driven to a grain boundary and annihilated once nucleated.

For both MQII and MQIII it is possible that either nucleation of a reverse domain, most likely at the periphery of already reversed regions, or domain wall unpinning and motion is responsible for the demagnetization process. The differences between the materials is that free domain walls are present in a large number of grains in MQIII while in the majority of grains in MQII free domain walls are not present. These differences are most likely due to differences in both the nucleation fields and the dipolar interactions within grains produced upon deformation processing.

ACKNOWLEDGEMENTS

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