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A domain size effect in the magnetic hysteresis of NiZn-ferrites

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The domain size and the ac-hysteresis of NiZn-ferrites have been investigated as a function of grain size. It is found that the ac-hysteresis or energy dissipation at MHz frequencies is substantially reduced in fine-grained, monodomain NiZn-ferrites, which is attributed to the absence of intragranular domain walls. © 1996 American Institute of Physics. [S0003-6951(96)00245-8]

Magnetic hysteresis (or the delayed response of magnetic material to changes in applied field) is a key factor determining the possible applications of magnetic materials. Generally, magnetic hysteresis is related to domain wall motion.¹ In permanent magnets, due to inhibited domain wall nucleation or strong pinning, the magnetization is not reversed easily, hence the coercivity, H_C , is high. Whereas in soft magnetic materials, domain walls are formed easily and move without much impediment, hence H_C is low. Ferrites with the spinel structure form an industrially important class of soft-magnetic materials, and because their low conductivity enables high frequency applications such as transformer cores or inductors. Since H_C decreases with increase in grain size,² conventional wisdom is to use large-grained ferrites as their hysteresis or dissipation of energy (which is equal to the area enclosed in the hysteresis loop) should be lowest. Surprisingly, we have found that at MHz frequencies *below* a certain grain size NiZn-ferrites exhibit ten times lower dissipation. Our interpretation is that ac-hysteresis is dominated by *intragranular* domain wall movement. At sufficiently small size the grains become monodomain, and this domain wall motion is inhibited.

The polycrystalline NiZn-ferrites used in our study were prepared by standard ceramic techniques. By adjusting the sinter temperature between 1000 and 1300 °C the grain size was varied between 1.2 and 6.8 μm , while maintaining the composition $\text{Ni}_{0.49}\text{Zn}_{0.49}\text{Co}_{0.02}\text{Fe}_{1.90}\text{O}_{3.85}$. For this composition the spontaneous magnetization, $\mu_0 M_s$, is 0.50 ± 0.05 T at 18 °C. The average grain size of these ferrites was determined from scanning electron micrographs. Figure 1 shows such a micrograph for a typical NiZn-ferrite with $D = 1.4$ μm . As a measure of the grain size, D , the mean linear intercept³ was taken, because it gives an unequivocal definition of D , independent of the grain shape. The magnetic domain size, Δ , was determined at ambient temperature in the ac-demagnetized state by neutron depolarization (ND).^{4,5} The advantage of this technique over many other domain imaging techniques is that domain sizes *within* the bulk of a material are determined. As neutrons traverse the grains in a bulk material on random chords, the domain size, Δ , determined in a ND experiment also corresponds to a linear mean intercept value. The ac-hysteresis (or ac-energy dissipation)

was determined from the complex permeability of a ferrite core.⁶

Figure 2 gives the results of a study at zero magnetic field of the domain size, Δ , versus the grain size, D , for the NiZn-ferrite, $\text{Ni}_{0.49}\text{Zn}_{0.49}\text{Co}_{0.02}\text{Fe}_{1.90}\text{O}_{3.85}$. For $D \leq 2.8$ μm , the domain size is equal to D , as can be seen by comparison to the solid line, for which $\Delta/D = 1$. The dashed line corresponds to the ratio $\Delta/D = 0.63$ calculated for the two-domain state⁷ and confirmed by previous experiments on MnZn-ferrite.⁸ Hence, for $D \geq 3$ μm the NiZn-ferrites appear to be in a state with two magnetic domains per grain i.e. the grains contain an *intragranular* domain wall. The transition from the two- to monodomain state of uniform magnetization appears to occur quite suddenly at $D \approx 2.9$ μm .

The striking result is that data on the energy dissipation or losses in these ferrites support the idea of an abrupt transition from the two- to monodomain state. Figure 3 gives the (ac-) hysteresis or dissipation per unit volume, P , versus grain size, D , at a frequency of 3 MHz and an induction level, B , of 10 mT for the same NiZn-ferrite. Around $D = 3.0$ μm a dramatic change in the dissipation by a peak order of magnitude is seen, leading to low dissipation in fine-grained NiZn-ferrites. Comparing the data with Fig. 2 one concludes that this reduction occurs at the same grain size $D = 2.8$ μm at which the NiZn-ferrites become monodomain. Apparently, the absence of intragranular domain walls in monodomain NiZn-ferrites reduces the ac-hysteresis at 3 MHz and a modest induction level, because reversal of magnetization can only occur through rotation.

FIG. 1. Scanning electron micrograph of the microstructure of a representative NiZn-ferrite. The mean linear intercept $D = 1.4$ μm .

FIG. 2. The domain size Δ as a function of the grain size D for $\text{Ni}_{0.49}\text{Zn}_{0.49}\text{Co}_{0.02}\text{Fe}_{1.90}\text{O}_{3.85}$. For the solid line $\Delta/D=1$, the ratio corresponding to monodomain grains; the dashed line corresponds to $\Delta/D=0.63$, the ratio calculated for grains in the two-domain state.

Our finding of the role of the intragranular domain walls as prime source of dissipation in NiZn-ferrites (at 3 MHz and modest induction levels) is further underscored by Fig. 4. In this figure the dissipation, P , at a 10 mT induction level is plotted versus frequency, f , for two NiZn-ferrites with different grain sizes, $D=2.6$ and $3.3 \mu\text{m}$, respectively. For the NiZn-ferrite with $D=2.6 \mu\text{m}$, $P \propto f^{1.0}$, which is the expected, trivial scaling with the measurement frequency, whereas for the NiZn-ferrite with $D=3.3 \mu\text{m}$, $P \propto f^{1.3}$. This superlinear increase with frequency points to an additional, intragranular domain wall, contribution to the dissipation. Finally, we note that, when the induction level B at which the dissipation measurements are made is increased, the dissipation difference between the mono- and two-domain state is reduced. An expected result, since this difference should vanish when B reaches the spontaneous induction, $\mu_0 M_S$.

The results reported here show that intragranular domain

FIG. 3. The energy dissipation per unit volume, P , of $\text{Ni}_{0.49}\text{Zn}_{0.49}\text{Co}_{0.02}\text{Fe}_{1.90}\text{O}_{3.85}$ vs grain size D at a driving field of 10 mT and a frequency of 3 MHz for two different temperatures, 20 and 100 °C.

FIG. 4. A double logarithmic graph of the energy dissipation P versus frequency f at an induction level B of 10 mT for two different grain sizes, $D=2.6 \mu\text{m}$ and $3.3 \mu\text{m}$. Note that for the NiZn-ferrite of which the grain size is in the two-domain range the increase of P with f is superlinear.

wall movement is a prime contribution to the ac-hysteresis (in particular, at frequencies around 1 MHz) and that this hysteresis is eliminated in fine-grained, monodomain ferrites. This is *opposite* to the situation encountered for dc-hysteresis. In that case, the energy dissipated by going around the major hysteresis loop (i.e. the area enclosed in the hysteresis loop) is approximately given by $\approx 4 \mu_0 M_S \times H_C$.⁹ For ferrites of a given composition, $\mu_0 M_S$ is independent of D in the range of D studied here. However, it is well established that H_C , and hence the dissipation, scales as $H_C \propto D^{-1}$.^{2,10} This latter relation has been the basis for the traditional practice, to reduce H_C and suppress the effect of dc-hysteresis by employing ferrites composed of large grains. In clear contrast to the ac-hysteresis data presented here; thus suggesting that a clear distinction should be made between dc- and ac-hysteresis. The difference in grain size dependence between dc- and ac-hysteresis and the fact that *intragranular* domain wall movement appears to be responsible for the ac-hysteresis at modest induction levels (see Fig. 4), suggest that if dc-hysteresis is related to domain wall movement, this should be *intergranular* domain wall movement.

A second surprising aspect of our data is the abruptness of the transition between the two- and monodomain state. One would expect that the distribution in grain sizes inherent to ceramic materials, and in these ferrites some 30%, should smear out this transition. In addition, according to advanced micromagnetic theory¹¹ only upper and lower limits for the critical grain size, D_C , at which particles become monodomain can be derived. Theoretically, a transition to a monodomain state is expected with decreasing grain size for an isolated grain, as the domain wall energy scales with the

domain wall area, approximately as D^2 , whereas the magnetostatic energy decreases more rapidly with D^3 .¹² A fundamental complication in comparing the experimentally determined D_C to that theoretically expected is that the micromagnetic theory^{11,12} deals with an isolated grain. In the bulk ferrite discussed here this condition is not met. Hence, the soft-magnetic environment of the grain should be taken into account. This requires correlations between the magnetization directions in neighboring grains such that the condition $\text{div } \mathbf{M} \approx 0$ between neighboring grains is met. This is, for instance the case, when four neighboring grains form a diamond-like magnetization structure. However, in a first approximation for the calculation of D_C (mean intercept value), one can account for this correction due to the soft-magnetic environment, which reduces the magnetostatic energy of the grain^{8,13} by the isotropic permeability correction proposed earlier.⁸ Applying this correction to the upper and lower limits derived by Brown¹¹ yields:

$$D_{C \text{ lower}} = 3.6 \sqrt{\frac{A \frac{2}{3} \mu_i}{\frac{1}{2} \mu_0 M_S^2}}, \quad (1)$$

$$D_{C \text{ upper}} = 28 \sqrt{\frac{A \frac{2}{3} \mu_i}{\frac{1}{2} \mu_0 M_S^2}} \sqrt{\pi + \frac{K \frac{2}{3} \mu_i}{\frac{1}{2} \mu_0 M_S^2}} \quad (2)$$

with A the exchange constant, μ_i the initial permeability at D_C and K the anisotropy constant. Using the numerical values pertaining to this NiZn-ferrite, $A = 3.78 \times 10^{-12}$ J/m, $\mu_i \approx 100$, $K = 330$ J/m³ and $\mu_0 M_S = 0.50$ T, yields then a lower limit of $0.18 \mu\text{m}$ and an upper limit of $2.6 \mu\text{m}$ for D_C . In view of the approximations made in accounting for the soft-magnetic environment⁸ the experimentally found D_C of $2.8 \pm 0.1 \mu\text{m}$ seems reasonable.

Although presently no explanation is available for the abruptness of the two- to monodomain transition (which suggests some form of collective behavior) the data on the dissipation clearly confirm this observation. In the latter case a partial explanation for the abruptness may be given in that a double effect occurs: The suppressing of the intragranular domain walls is accompanied by a change in eddy currents, because domain wall displacement gives a larger contribu-

tion to eddy current loss than rotation of magnetization.^{14,15} This is the well-known eddy current anomaly.¹⁴ The conductivity of NiZn-ferrites within the grains is still appreciable as it is a factor of 10^3 higher than the bulk conductivity which is mainly limited by grain boundary resistance.¹⁶ Hence, elimination of intragranular domain walls may also decrease the dissipation due to eddy currents within the grains.

In conclusion, we have shown that the ac-hysteresis or dissipation of energy in polycrystalline NiZn-ferrites can be reduced substantially by using fine-grained materials in which intragranular domain walls are absent. The resulting low-loss monodomain NiZn-ferrites should allow further miniaturization of ferrite cores used in, for instance, power supplies. Finally, our results may form an important clue for the study of hysteresis in magnetic materials.

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