MAGNETIC SUPERVISCOSITY OF FERRITES

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A study has been made of the magnetic superviscosity curves, at 78°K, of ferrites of the Ni-Zn series, sintered at temperatures from 1200 to 1400°C. The existence of a second maximum of the viscous emf has been observed in a range of fields less than the coercive force, and corresponding to the rapidly rising part of the curve of differential magnetic susceptibility. After the second maximum there is a hyperbolic decrease of the viscous emf curves. On increase of the field, the second maximum merges with the main maximum, and the viscous emf curve becomes approximately exponential. The observed phenomena can be explained within the framework of Néel's theory.

1. OBJECT AND METHOD OF THE RESEARCH

MAGNETIC superviscosity in nickel-zinc ferrites at a temperature of about 80°K was observed by Kuritsyna in 1952.¹ In 1955, Lednev and Telesnin² studied the temperature dependence of magnetic superviscosity in Ni-Zn ferrites in the temperature interval from 80°K all the way to the Curie point. Kuritsyna observed two maxima of the superviscosity on the back of the hysteresis loop of highpermeability ferrites; these went over to a single maximum on increase of temperature. In lowpermeability ferrites, two maxima were observed at all temperatures, but the duration of the processes was several orders of magnitude smaller.

The object of the present research was to study the actual nature of the process of magnetization change in superviscosity. A series of six specimens was studied. They were of Ni-Zn ferrites of composition 16 mol % NiO, 34 mol % ZnO, the rest Fe_2O_3 , sintered at 1200°C (specimen no. 1), 1250° (no. 2), 1270° (no. 3), 1320° (no. 4), 1350° (no. 5), and 1400° (no. 6), for a period of four hours. The temperature of preliminary sintering was 1000°C.

The superviscosity was studied on the back of the hysteresis loop by recording a curve of emf induced in a coil by the viscous change of magnetization that occurred in the specimen after a sudden change of the magnetic field by an amount ΔH . The apparatus used for the recording was a photo-compensated amplifier, F-16, of B. A. Seliber's system, with automatic recording; the response time of its moving system amounted to 0.4 second, and its maximum sensitivity was 2×10^{-8} volt per division. The toroidal specimens were placed in liquid nitrogen.

2. RESULTS OF THE EXPERIMENT

All the specimens exhibited superviscosity. Typical curves of the decrease of viscous emf in specimen 4 are given in Figs. 1a and b. The duration of the process, defined as the time of decrease of the emf curve to 5% of the maximum residue, ranged for various specimens from 33 to 2.5 seconds. In a number of specimens (high-permeability ferrites) a new phenomenon was observed: after the initial residue and the subsequent decrease of the emf, a second maximum was observed; it appeared at some value of the field less than that that corresponding to maximum differential magnetic susceptibility. This maximum appeared 6 to 7 seconds after the change of field; and upon further increase of the field (approaching the value H_c), it occurred earlier and earlier, until finally it merged with the first residue of the recorder; this occurred near the maximum of the differential susceptibility. The time of appearance of the second maximum after the change of field decreased very rapidly, simultaneously with an increase of its amplitude (curves b and c in Fig. 1). The product of the time of appearance of the second maximum and of its amplitude was almost constant, for each of the specimens, and varied little from specimen to specimen. The curves of dependence on the field H of the time of decrease of the viscous emf to 0.05 and 0.1 of the maximum value (the first residue of the recorder pointer) had two maxima; for specimens



FIG. 1. Curves of the decrease of viscous emf of specimen No. 4. For all curves, $\Delta H = 0.098$ oersted. a - H = 0.557; b - H = 0.588; c - H = 0.618; d - H = 0.630; e - H = 0.704 oersted.

3, 4, and 6, one lay in the neighborhood of the top bend of the hysteresis curve, the other close to the coercive force. For specimen 5, these maxima were shifted further to the right.

In Fig. 2 are shown the curves of the time τ of occurrence of the second maximum, its amplitude A, the viscosity curves on the basis of a decrease of the emf to 0.1 and to 0.05, the χ_d curve, and the back of the hysteresis loop for specimen 4. In fields corresponding to the maximum of the differ-



FIG. 2. 1, the back of the hysteresis loop, I; 2, curve of the differential susceptibility, χ_d ; Δ and \bullet —curves of viscosity, $\tau_{0.05}$ and $\tau_{0.01}$; \odot —curve of the time of appearance of the second maximum of τ ; \Box — amplitudes of the second maximum A for specimen 4.

ential magnetic susceptibility, the second maximum of the viscous emf vanished, and at high fields it did not appear. The curve of the viscous emf after the second maximum is a hyperbola. After the disappearance of the second maximum, the emf curve became transformed to an exponential, E = $E_0 \exp(-t/\tau)$; the value of E_0 was found by extrapolation of the curve to its intersection with the axis of ordinates. The values of the relaxation time ranged between 2 and 3 seconds.

At small values of the magnetic field, corresponding to the beginning of the rise in the differential susceptibility curve, the second maximum either does not yet exist or is still very small (curve a, Fig. 1); the recorded curve of the decrease of emf is nearly exponential, with the exception of the very first part. Upon further increase of the field, the second maximum appears, then gradually increases and approaches the initial residue (Fig. 1, c and d). The curve of emf decrease after the second maximum is a hyperbola (Fig. 1 c, d, e). The hyperbolic character of the curve is retained until the merging of the two maxima; after this the curve gradually changes to an exponential (Fig. 3, a), again with the exception of the very beginning of the curve.

An interesting matter is the influence of the size of the jump ΔH in magnetic field intensity that is responsible for the viscous change of magnetization. When ΔH exceeds 0.055 oersted and the value of the field intensity H is constant, the time of appearance of the second maximum and its magnitude are almost independent of ΔH ; the duration of the decrease of the emf curve is also almost independent of ΔH [the second law of magnetic viscosity (Figs. 3 b and c)]. On decrease of ΔH below 0.055 oersted, the second maximum rapidly decreases and then vanishes (Figs. 3 d and e). For $\Delta H \ge 0.01$ oersted, no viscous process is in general observed, possibly because of the smallness of the viscous emf.

If after commutation from a positive to a negative value of the field, and before switching off of an auxiliary field of less than 0.055 oersted, a delay of about a minute is allowed, the magnitude of the first residue diminishes, and the second maximum vanishes, although the duration of the whole process actually increases (Fig. 3, f). When ΔH exceeds 0.055 oersted, an even larger delay after commutation does not decrease the second maximum and lowers the first residue hardly at all. In Fig. 3 g is given the curve of emf decrease for a five-minute delay after commutation. Figure 4 shows, for specimen 4, the curves of dependence of the viscous emf on the reciprocal of the duration



FIG. 4. Dependence of the viscous emf of specimen 4 on the reciprocal of the duration of the process. O - H = 0.630; $\Delta - H = 0.636$; $\Box - H = 0.643$ oersted.

FIG. 5. Dependence of the logarithm of the viscous emf on the duration of the process. o - H = 0.643; $\Delta - H = 0.704$; $\Box - H = 0.857$ oersted.

FIG. 3. Curves of the decrease of viscous emf when there is a time delay between commutation of the basic field H and removal of the auxiliary field ΔH . a - H =0.857 oersted, $\Delta H = 0.098$ oersted, $\Delta t \rightarrow 0$; b - H = 0.566, $\Delta H = 0.098$, $\Delta t \rightarrow 3$ min; c - H = 0.566, $\Delta H = 0.098$, $\Delta t \rightarrow$ $\rightarrow 0$; d - H = 0.566, $\Delta H = 0.042$, $\Delta t \rightarrow 0$; e - H = 0.597, $\Delta H = 0.024$, $\Delta t \rightarrow 0$; f - H = 0.566, $\Delta H = 0.042$, $\Delta t = 5$ min; g - H = 0.566, $\Delta H = 0.098$, $\Delta t = 5$ min.

of the viscous process. As is evident, at fields of 0.63 and 0.636 oersted the experimental points lie exactly on a single straight line, which corresponds to a hyperbolic dependence $E = (\alpha/t) + b$ (curve 1). On increase of the field to 0.643 oersted, the hyperbolic dependence breaks down (curve 2). On further increase of the magnitude of the field to 0.857 oersted, as is evident from Fig. 5, the dependence of E on t becomes exponential.

3. DISCUSSION OF RESULTS

The observed phenomena can be interpreted within the framework of Néel's theory³ regarding the existence of two types of magnetic viscosity: reversible and irreversible. According to Néel, reversible viscosity is characterized by the presence of a field H_r opposing the change of magnetization, and which in the case of 180° walls becomes equal to zero at sufficiently large displacement of the wall.

Irreversible viscosity is characterized by an opposing field

$$H_1(t) = S\left(Q + \log t\right),$$

where S and Q depend only slightly on log t; they may be considered constant over the whole range of the hysteresis loop.

The viscous change of magnetization is

$$I_1(t) = \chi_d H_1(t) = \chi_d SQ + \chi_d S \log t$$

At a given point of the hysteresis loop, $\chi_d SQ$ may be set equal to a constant; then

$$I_1(t) = a \log t + \text{const.}$$

This gives a hyperbolic dependence of the viscous emf on time, which was observed in our experiments after appearance of the second maximum (Fig. 4). The decrease of viscous emf after the first residue and its subsequent increase, giving the second maximum, can be explained by the decrease of the opposing field of reversible viscosity, which occurs in our specimens in the course of a few seconds. The later process is explained on the basis of the process of irreversible viscosity.

The physical mechanism of the process under consideration can be described as follows. On passage to a point that lies near the maximum of the differential susceptibility, some of the domains undergo magnetization reversal; this process proceeds with a speed determined by the diffusion of impurities (reversible viscosity). Under the influence of the reversed domains, other domains, for which there was previously not enough energy for magnetization reversal, begin to reverse. These are "captured" domains, in the terminology of Street and Woolley.⁴ This viscous process is determined by energy fluctuation, which gives irreversible viscosity. With approach to the maximum of the differential susceptibility, the number of such "captured" domains becomes smaller and smaller, and a larger and larger number of domains reverse by the reversible process; this leads to coalescence of the maxima, and the change of emf becomes exponential.

An important fact is that after repeated immersion of the specimens in liquid nitrogen, the second maximum decreased considerably, and the whole process described became almost imperceptible. This is attributable to a kind of hardening of the specimen upon repeated violent cooling, which was observed previously by one of us and Lednev.² Through such hardening, internal stresses appear in the material, and the energy fluctuations that occur are no longer sufficient for magnetization reversal of "captured" domains.

The disappearance of the second maximum upon continued exposure to the magnetic field after commutation can be explained by the fact that the field of reversible viscosity is essentially created on commutation of the field, but not on switching off of a field ΔH . Consequently, if we allow a sufficiently long delay after commutation, the impurity atoms responsible for this form of viscosity have time to diffuse into positions determined by the new position of the walls; and a change of field by a small amount ΔH , less than 0.055 oersted, gives only elastic wall displacements, not accompanied by diffusion of atoms. If the magnitude of the change ΔH exceeds 0.055 oersted, for example if it is of

FIG. 6. Analysis of the experimental curve of decrease of viscous emf into two curves, and curves of magnetization obtained by graphical integration. H = 0.60 oersted. a — experimental curve; b — exponential component; c — second component; d — magnetization determined by the second component; e — magnetization determined by the exponential component; f — total magnetization.

order 0.1 oersted, then this process is accompanied by diffusion of impurity atoms; and, independently of the delay time after commutation, the second maximum appears, but the time of its appearance is earlier — about two seconds.

In Fig. 6 the experimental viscous emf curve a is represented as the sum of two emfs — a fast one that decreases according to an exponential law (curve b), and another that first rises and then falls (curve c). The change of magnetization with time is shown in curves d, e, f. Curve e is the magnetization whose change gives the exponentially decreasing emf. Curve d corresponds to curve c, and curve f shows the complete change of magnetization.

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