APPLICATION OF ULTRASOUND IN THE INVESTIGATION OF MAGNETIC RELAXATION EFFECTS IN A FERROMAGNET

P. P. Galenko and V. I. Alekseenko

UDC 538.22:534.321.9

Experimental results are given from an investigation of the magnetic relaxation of samples by the ultrasonic method.

The magnetic state of a ferromagnet is altered under the action of a magnetic field, temperature, mechanical vibrations, and other factors. In the presence of domain structure, thermodynamic equilibrium is not established instantaneously, but requires essentially a finite time varying from 10^{-9} sec to tens of minutes, hours, or even days. The potential causes of such equilibrium may be found in such effects as the movement of dislocations and their Cottrell atmospheres due to magnetostriction stresses; diffusion processes involving impurity atoms; local heat release associated with the shifting of interdomain boundaries, resulting in thermal stresses; etc. It has been shown in a number of papers ([1-12] and others) that several types of time-dependent effects can be discerned in ferromagnets, depending on the structural modifications, the magnetization conditions, the temperature, and other factors. The investigation of these effects associated with relaxation processes in ferromagnets is of practical value at the present time in connection with their consideration in the determination of the magnetic characteristics of materials [13], and it is also of theoretical value in connection with gaining deeper insight into the physical nature of magnetization processes. Time-dependent effects are particularly strong in magnetically soft materials in weak magnetic fields, when processes of domain boundary movements around regions of spontaneous magnetization are prevalent. In the case of movement of the domain boundaries as a result of the magnetostriction effect, internal stresses set in and are subjected to equilibrium redistribution in the boundary layers due to the lag of the magnetization I with variation of the field H. One time-dependent effect is observed immediately after the demagnetization of a ferromagnet by an alternating field with a diminishing amplitude. In this case the permeability decreases when an equilibrium state is established. This magnetic relaxation effect is called disaccommodation of the permeability. The time variation of the permeability can take place for different lengths of time, depending on the particular nature of the redistribution of the boundary layers between regions of spontaneous magnetization in correspondence with the most stable energy state. It is impossible to attain such an equilibrium state of a ferromagnet in demagnetization (when $H \to 0$ and $I \to 0$) for the same particular distribution of the domain boundaries. This demagnetization state, as a rule, can be realized by a large number of means of mutual equilibrium arrangement of the regions. The processes of relaxation to an equilibrium state of a ferromagnet in demagnetization are of a statistical nature, and each of these states will have its own particular distribution of regions. After demagnetization or any other effect acting on a ferromagnet, a shifting of the interdomain boundary will take place, producing larger gradients of the magnetostriction stresses. The arrival at a new thermodynamic equilibrium state involves the relaxation of those stresses and their redistribution in the volume of the boundary layers between the domains. Consequently, together with the diffusion of impurity atoms and other effects, the elastic aftereffect also makes an important contribution to the free energy of the boundary layer. Complete equilibrium of the distribution of internal stresses is not attained instantaneously, but sets in at a definite rate owing to the elastic aftereffect. Naturally, one of the causes of the time variation of the magnetic properties of a ferromagnet is the elastic aftereffect of the stresses, which affects the kinetic magnetic relaxation process in the stabilization of a thermodynamic equilibrium state.

We have investigated the feasibility of studying the evolution of relaxation processes in magnetic materials by means of ultrasonic vibrations. It has been shown [14-19] that when

---

Institute of Applied Physics, Academy of Sciences of the Belorussian SSR, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 48, No. 2, pp. 326-330, February, 1985. Original article submitted December 20, 1983.

0022-0841/85/4802-0245 $09.50

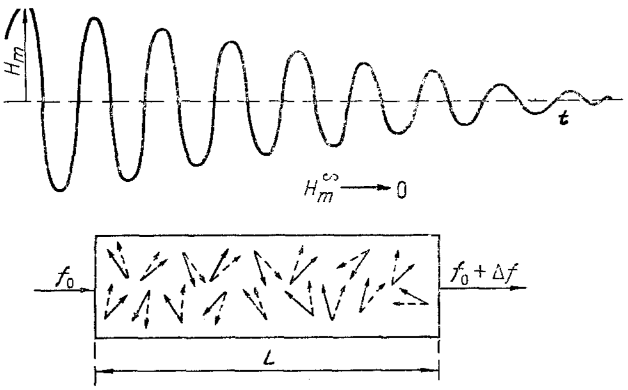

Fig. 1. Scheme of the distribution of the regions of spontaneous magnetization after demagnetization of the sample.

a wave is transmitted through a medium with variables properties, its frequency and phase grow with time. This effect is called the transient Doppler effect, for which a detailed theory was first presented in [17, 18].

If a ferromagnet is subjected to the action of a magnetic field with a diminishing amplitude $H_m^{\infty} \to 0$, its magnetic equilibrium state ($H \to 0$, $I \to 0$) sets in only after a finite time. At a certain time t_0 immediately after demagnetization an unstable redistribution of the regions of spontaneous magnetization takes place (this is illustrated schematically by the dashed arrows in Fig. 1). However, this distribution is energetically unfavorable, and as $t \to \infty$ the system enters into a thermodynamic equilibrium state, when all relaxation processes terminate. In this case the domain boundaries occupy a stable position, and the distribution of the magnetization regions stabilizes; this state is represented by the solid arrows in the diagram of Fig. 1. In the transition of the ferromagnet from an unstable to a stable state, the domain boundaries are continuously involved in a certain motion, "sticking" in certain "fresh" sites of the crystal lattice. This process continues until the regions of spontaneous magnetization are redistributed in correspondence with an energetically favorable state.

When ultrasonic vibrations propagate through such a ferromagnet with time-variable properties, the parameters of the wave vary both in frequency and in phase. It has been shown [17] that the frequency shift induced by time variation of the properties of the medium is given by the relation

$$\Delta f(t) = \frac{f_0 L}{v^2(t)} \, \frac{\Delta v}{\Delta t}, \tag{1}$$

where Δf is the frequency increment of the oscillations due to the time-dependent properties of the ferromagnet.

The variation of the propagation velocity of the vibrations in a ferromagnet produces not only a frequency shift, but also a secondary phase difference $\Delta\varphi$ equal to

$$\Delta\varphi(t) = 2\pi \int_0^t \Delta f(t)\, dt = \frac{2\pi f_0 \Delta v(t)}{v_0 [v_0 + \Delta v(t)]}, \tag{2}$$

where $t = 0$ is teken as the zero-reference time and v_0 is the average initial velocity in the sample. It has been shown [17, 18] that relations (1) and (2) are valid for any waves, regardless of their nature and for either mechanical or electromagnetic oscillations.

In the general case the wave propagation velocity can influence a number of physical parameters that vary with time. In our experiments the parameter associated with the relaxation of a ferromagnetic sample to an equilibrium state following demagnetization is the variation of the permeability (disaccommodation), which characterizes the behavior of magnetic relaxation processes leading to thermodynamic equilibrium.

The experimental investigations of magnetic relaxation were carried out on samples of iron containing carbon in fractions of tenths of a one percent to one percent with an identi-

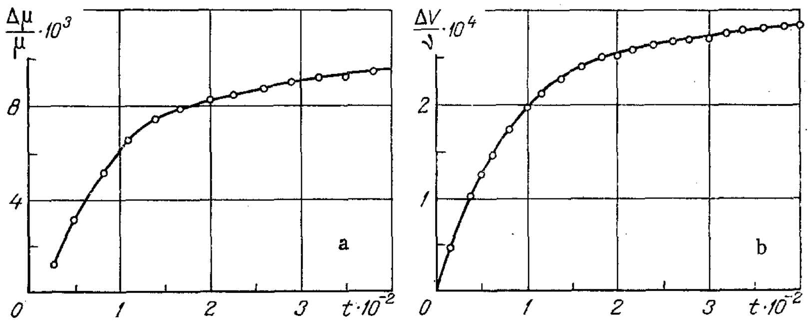

Fig. 2. Relaxation curves. a) Disaccommodation magnetic relaxation curve; b) relative variation of the velocity of ultrasound in a ferromagnetic sample vs duration of the relaxation processes. t, sec.

cal, practically insignificant quantity of other impurities. Prior to testing, the samples were subjected to vacuum annealing at a temperature of 900°C, with the virtual absence of surface oxidation. The base of the investigated samples was L = 0.35 m, and their diameter $\phi = 2\cdot 10^{-3}$ m. The samples were demagnetized in an alternating magnetic field at a frequency of 50 Hz from a maximum amplitude $H_{max} = 8\cdot 10^3$ A/m to a minimum value $H_{min} \to 0$ ($H_{min} < 10^{-1}$ to 10^{-2} A/m) for 5-8 sec. The magnetic field along the length of the demagnetizing sample was practically uniform. This demagnetization regime was maintained for all of the tested samples. The time variations of the magnetic properties of the samples due to disaccommodation were measured by a procedure similar to that in [20, 21]. Magnetic relaxation curves $\Delta\mu/\mu = \Phi(t)$, which characterize the disaccommodation, were recorded at room temperature immediately after demagnetization according to the above-indicated regime. One of the disaccommodation curves, showing the time variation of the magnetic relaxation in the sample, is given in Fig. 2a.

It follows from an examination of these experimental data that the time variation of the magnetic properties of the investigated ferromagnetic sample is more intense in the initial stage of relaxation, slows down in the later stages of the process, and, beginning with a certain time, it practically stabilizes. The relaxation rate increaes rapidly in the first 10-15 sec, and the time variation of the magnetic properties of the samples amounts to more than 80% of the steady-state value of the relaxation parameter. This kind of evolution of the relaxation process was also observed in the other investigated samples.

Ultrasound was transmitted through the same samples after their demagnetization, and the variation of the frequency of the vibrations due to the time variation of the properties of the ferromagnet was recorded by means of suitable instrumentation operating in an automatic mode. With the onset of thermodynamic equilibrium the domain boundaries are in continuous motion until the redistribution of the regions of spontaneous magnetization in correspondence with the energy minimum of the system is completed. Curves of $\Delta v/v = f(t)$, which characterize the different velocities of ultrasound in the ferromagnetic materials as a result of magnetic relaxation processes in them, can be obtained from Eq. (1) by measuring the time variation of the vibration frequency at the receiver.

Figure 2b shows an experimental curve of the relative variation of the velocity of ultrasound in an investigated sample during the magnetic relaxation process. It is seen that the propagation velocity varies more rapidly in the initial stage, and after a certain time the processes subside and begin to stabilize. The behavior of the given curve exhibits the same tendency as the disaccommodation curves in Fig. 2a. This result confirms the fact that these two curves, both $\Delta\mu/\mu = \Phi(t)$ and $\Delta v/v = f(t)$, are associated with the inception and evolution of the same relaxation process of redistribution of the domain boundaries around the regions of spontaneous magnetization in such a way as to make the system occupy an energetically favorable state. During relaxation to thermodynamic equilibrium, the domain boundaries are in continuous motion, generating internal elastic stresses. Their subsequent space—time redistribution in the ferromagnet induces an elastic aftereffect, which, in turn, affects both the variation of the velocity of propagation of the ultrasonic probe vibrations and the evolution of the magnetic relaxation processes. The ultrasonic power used in the investigation of the magnetic relaxation kinetics must be chosen at such a level as not to affect the inception and evolution of the process itself in the sample.

The reported experimental investigations show that the ultrasonic method based on the transient Doppler effect can be used to study magnetic relaxation phenomena. This procedure in conjunction with conventional magnetic techniques will afford the possibility of penetrating more deeply into the physical essence of time-dependent effects in ferromagnetic materials.

NOTATION

t, time; H, magnetic field intensity; I, magnetization; μ, permeability; v, velocity of wave propagation in the medium; f, frequency (f_0 and f are the frequencies of the radiated and received waves, respectively); L, length of the investigated sample; φ, phase of the vibrations.

LITERATURE CITED

1. R. Bozorth, Ferromagnetism, Van Nostrand, New York (1951).
2. R. V. Telesnin, "Rate of change of the magnetization of iron on different parts of the hysteresis loop," Dokl. Akad. Nauk SSSR, 20, No. 9, 649-659 (1938).
3. S. V. Vonsovskii, Magnetism [in Russian], Nauka, Moscow (1971).
4. P. P. Galenko, "Phenomenological theory of magnetic relaxation in constant magnetizing fields," in: New Physical Methods and Means for the Inspection of Industrial Products [in Russian], Nauka i Tekhnika, Minsk (1978), pp. 382-386.
5. D. Ratenau, "Transient effects in magnetization," in: Magnetic Properties of Metals and Alloys [Russian translation], IL, Moscow (1961), pp. 227-240.
6. Y. Tahara and T. Sugeno, "A theory of magnetic aftereffect for the irreversible magnetization process in Rayleigh region," Phys. Status Solidi, 55, 385-398 (1973).
7. E. Kneller, Ferromagnetism, Springer-Verlag, Berlin—New York (1962), pp. 422-673.
8. J. Przybyta and J. W. Moron, "Migracyjne opoznienia magnetyczne w martenzycie w weglowym zelaza," in: Prace Naukowe Uniwersytetu Slaskiego, No. 165, Fizyka i Chemia Metali, Vol. 2, Katowice (1977), pp. 32-52.
9. P. P. Galenko, "Magnetic relaxation of Fe—Co alloy in constant magnetizing fields," Vestsi Akad. Navuk, BSSR, Ser. Fiz.-Tekh. Navuk, No. 3, 99-102 (1982).
10. P. P. Galenko and I. M. Morozov, "Theory of the influence of dislocations on magnetic relaxation in ferromagnets," Vestsi Akad. Navuk BSSR, Ser. Fiz.-Tekh. Navuk, No. 1, 90-96 (1981).
11. P. P. Galenko, "Stability of the magnetic properties of Fe—Co alloy," Vestsi Akad. Navuk BSSR, Ser. Fiz.-Tekh. Navuk, No. 1, 83-86 (1982).
12. P. P. Galenko, "Influence of magnetic annealing on the stability of the magnetic properties of Fe—Co alloy," Inzh.-Fiz. Zh., 41, No. 6, 966-971 (1982).
13. V. V. Druzhinin, Magnetic Properties of Electrical Steel [in Russian], Gosénergoizdat, Moscow—Leningrad (1962), pp. 245-246.
14. W. Michelson, "On the question of the correct application of Doppler effect," Astrophys. J., 13, No. 3, 192-197 (1901).
15. N. V. Antonishin and V. I. Krylovich, "Feasibility of using the acoustic Doppler effect in heat physics and other areas," in: Heat and Mass Transfer [in Russian], Vol. 7, Énergiya, Moscow (1966), pp. 75-82.
16. V. I. Krylovich and A. D. Solodukhin, "Acoustical method for the investigation of transient heat convection in cylindrical layers of gases and liquids," Inzh.-Fiz. Zh., 31, No. 6, 1105-1112 (1976).
17. V. I. Krylovich, "Transient Doppler effect and frequency—phase methods of investigation and inspection," Inzh.-Fiz. Zh., 36, No. 3, 487-492 (1979).
18. V. I. Krylovich, "Frequency shift of received waves," Inzh.-Fiz. Zh., 41, No. 3, 508-513 (1981).
19. V. I. Krylovich and A. D. Solodukhin, "Distinctive features of the evolution of transient heat convection in cylindrical gas layers," Inzh.-Fiz. Zh., 37, No. 3, 429-432 (1979).
20. J. W. Moron, "Zastosowanie pomiarow dezakomodacji przenikalnosci magnetycznej w fisice metali," in: Prace Naukowe Uniwersytetu Slaskiego, No. 165, Fizika i Chemia Metali, Vol. 2, Katowice (1977), pp. 32-52.
21. F. Walz, "Ein vollstandig automatisiertes Messverfahren zur Bestimmung der magnetischen Nachwirkung in ferromagnetischen Kristallen," Appl. Phys., 3, 313-319 (1974).