Arch. Mech., 33, 3, pp. 401-414, Warszawa 1981

Dependence of dynamic elasticity moduli of ferrites on magnetic polarization

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THE RESULTS of investigations of dynamic elasticity moduli as functions of magnetic polarization are presented. Nickel ferrites with ionic admixtures of Co ($x = 0$–0.027), Cu ($y = 0$–0.015) and Mn ($z = 0.02$) are investigated. The negative effect ΔE is found to be the strongest (at room temperature) for ferrites with the highest cobalt admixtures content. The negative ΔE effect is connected with a considerable magnetomechanical coupling.

Podano wyniki badań dynamicznych modułów sprężystości ferrytów w zależności od polaryzacji magnetycznej. Badano ferryty niklowe z domieszkami jonów kobaltu ($x = 0-0{,}027$), miedzi ($y = 0-0{,}15$) i manganu ($z = 0{,}02$). Stwierdzono, że ujemny efekt ΔE był najsilniejszy (w temperaturze pokojowej) dla ferrytów o największej zawartości domieszek kobaltu. Ujemny efekt ΔE jest związany dużym sprzężeniem magnetomechanicznym.

Приведены результаты исследований динамических модулей упругости ферритов в зависимости от магнитной поляризации. Исследованы никелевые ферриты с примесями ионов кобальта ($x = 0-0{,}027$), меди ($y = 0-0{,}15$) и марганца ($z = 0{,}02$). Констатировано, что отрицательный эффект ΔE был самым сильным для ферритов (в комнатной температуре) с наибольшим содержанием примесей кобальта. Отрицательный эффект ΔE связан с большим магнитомеханическим сопряжением.

1. Introduction

THE APPLICATION of magnetic fields to magnetic materials produces the process of ordering of magnetization vectors of domains by shifting the Bloch walls and rotating the magnetization vectors. Voluminal changes of domains and rotation of magnetization vectors change the form of crystals [1, 2].

With the magnetostriction is connected ΔE effect which consists in changing the elasticity modulus which accompanies the process of transition from the state of demagnetization to the state of technical saturation [1-4, 12, 13].

The effects of tensile forces (stresses T, σ) are, in addition to normal elongation ε_n according to Hooke's law, the additional elongations produced by the striction ε_m, and so the resultant elasticity modulus E is smaller

$$E = \frac{T}{S} = \frac{\sigma}{\varepsilon} = \frac{\sigma}{\varepsilon_n + \varepsilon_m} = \frac{\sigma}{\varepsilon_m + \sigma/E_n}. \tag{1.1}$$

After transformations,

$$\varepsilon_m = \left(\frac{1}{E} - \frac{1}{E_n}\right)\sigma, \tag{1.2}$$

whence

$$\frac{\Delta E}{E} = \frac{E_n - E}{E} = \frac{\varepsilon_m}{\varepsilon_n}. \tag{1.3}$$

2. The investigated piezomagnetic ferrites

The subject of research were the dynamic elasticity moduli of the piezomagnetic ferrites Ni—Mn, Ni—Mn—Co and Ni—Mn—Co—Cu produced and investigated by R. Wadas. Their main components were nickel ferrites. The admixtures used were contained within the interval 2—20%, the general formula of the ferrites considered having the form

$$Ni_{0.98-x-y}Mn_{0.02}Co_xCu_yFe_2O_4 \tag{2.1}$$

with $y = 0$ and $x = 0$ ($\lambda 1$ ferrites), $x = 0.012$ ($\lambda 2$), $x = 0.015$ ($E2$), $x = 0.027$ ($E1$) and $y = 0.15$, $x = 0.012$ ($\lambda 3$). Density of the ferrites changed from 5.09 g/cm³ ($\lambda 1$) through 5.10 ($\lambda 2$), 5.11 ($E1$), 5.12 ($E2$) up to 5.23 g/cm³ ($\lambda 3$). Testing was performed on toroidal specimens with mechanical resonance frequencies from the 70—110 kHz band, outer diameter 20.7 mm, inner diameter 13.7 mm and thicknesses ranging from 5.3 to 6.0 mm. Tests were performed at room temperatures and also other temperatures, both negative and positive [4, 5, 7, 11, 12]. In this paper only the results obtained at room temperatures are presented, the remaining being left for another publication. The saturation induction of the ferrites investigated (at 4000A/m, i.e. 50 Oe) ranged from 0.24T ($\lambda 1$) to 0.32 T ($\lambda 2$), and the relative initial permeability — from 25 ($\lambda 1$) to 65 ($E1$), [11].

3. Dynamic elasticity moduli in magnetic field

In each system of piezomagnetic equations (e.g. [9]) combining the stresses, strains, magnetic induction and magnetic field intensity, in addition to the magnetic coefficient (permeability) and the magnetomechanical constant d, h, e or g, a mechanical coefficient also appears like, for example, the elasticity modulus at a constant magnetic field E_H, or at constant induction E_B, or their reciprocals. This is the reason why these coefficients should be known at each instant of work, that is at the given value of magnetic polarization. The elastic moduli in various fields of application are considered as material constants, while in reality their values change the more (up to 300%), the better their piezomagnetic properties are. Variability of the moduli with the magnetic polarization and temperature is extremely important for possible application of transducers in various ultrasonic or radio-electronic devices (electromechanical and magnetostrictive filters or magnetostrictive delay lines).

Dynamic elasticity moduli for a given temperature are determined at a constant field H or constant induction B

$$E_H = \left(\frac{\partial \sigma}{\partial \varepsilon}\right)_H = \left(\frac{\partial T}{\partial S}\right)_H = \pi^2 d_s^2 \varrho f_H^2 = c_H^2 \varrho \approx \pi^2 d_s^2 f_r^2 \varrho, \tag{3.1}$$

$$E_B = \left(\frac{\partial \sigma}{\partial \varepsilon}\right)_B = \left(\frac{\partial T}{\partial S}\right)_B = \pi^2 d_s^2 \varrho f_B^2 = c_B^2 \varrho \approx \pi^2 d_s^2 f_a^2 \varrho. \tag{3.2}$$

Here f_H, f_B, c_H, c_B are the respective frequencies and velocities of sound at a constant field intensity H and constant induction B. f_r and f_a — frequency of resonance and anti-resonance, d_s — mean diameter of the toroid, ϱ — density, σ, — mechanical stresses, and ε, S — strains.

In practice and, in particular, in the case of the ferrites under investigation, it may be assumed approximately that the maximum of impedance moduli occurs at the mechanical resonance what under sufficiently large quality factors, corresponds to the constant magnetic field (constant current efficiency), i.e. $f_H \approx f_r$; in the case of antiresonance, minimum impedance corresponds to the constant voltage efficiency, and hence the magnetic induction is held constant, $f_B \approx f_a$, [10, 11].

Impedance of the transducer with respect to the nonlinear phenomena and losses occurring in the core and winding of the transducer depends both on the constant and the alternating magnetic field. Figure 1 shows the impedance characteristics of the $E1$ ferrite

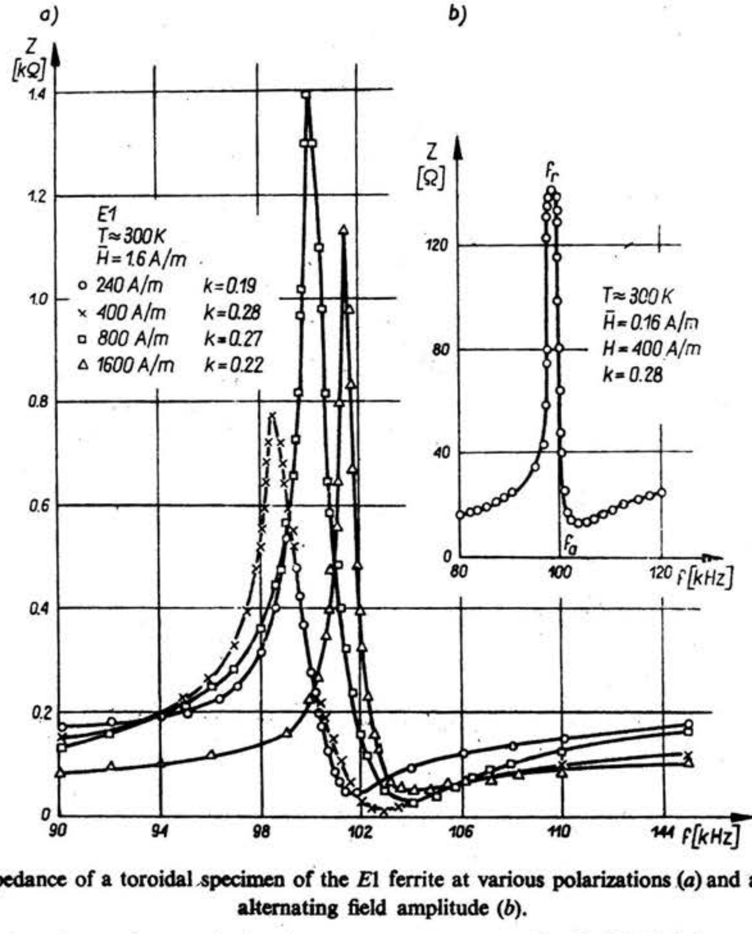

FIG. 1. Impedance of a toroidal specimen of the $E1$ ferrite at various polarizations (*a*) and at 1/10 of the alternating field amplitude (*b*).

determined under various polarizations at room temperatures [10]. With increasing polarization the resonance amplitude increases, reaches its maximum between 800 and 1600 A/m, and then decreases. The resonance frequency at small polarizations decreases and at higher magnetic field increases again. This phenomenon is closely connected with the negative ΔE effect. The antiresonance frequency in the cases under consideration always increases. Decreasing of the alternating magnetic field amplitude decreased also the impedance, while no changes in the resonance and antiresonance frequencies and in the coefficient of magnetomechanical coupling could be observed (Fig. 1b). The presented characteristics are referred to the ferrites of the lowest mechanical quality factor. In other ferrites the characteristics are steeper (in the same scale) and the accuracy of evaluating the moduli is higher. Figure 2 shows the characteristics of impedance Z and reactance X of the ferrite transducer $E2$. Values of the moduli are determined more exactly from the

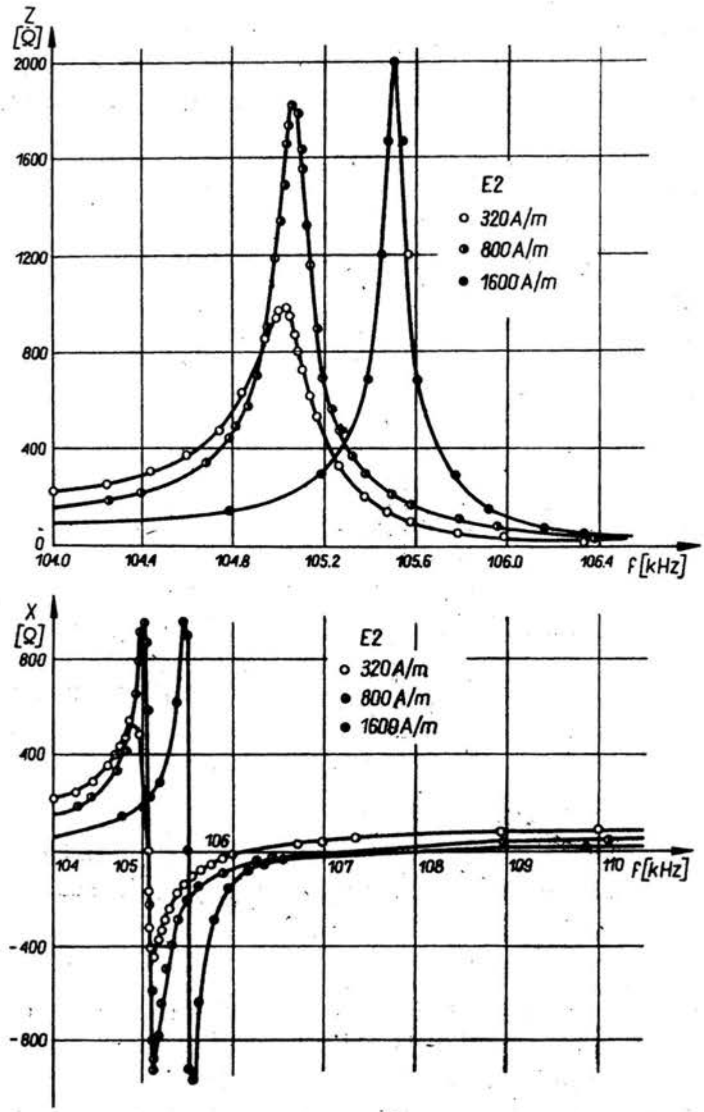

FIG. 2. Impedance and reactance of a ferrite $E2$ specimen at various polarizations.

impedance or admittance circles. The impedance circle of an unloaded transducer is shown, as an example, in Fig. 3, while Fig. 4 demonstrates the frequency characteristic of impedance Z, resistance R and reactance X of the transducer (a), resonance (b) and antiresonance ranges (c) being taken into account [11]. The maxima and minima of $|\mathbf{Z}|$

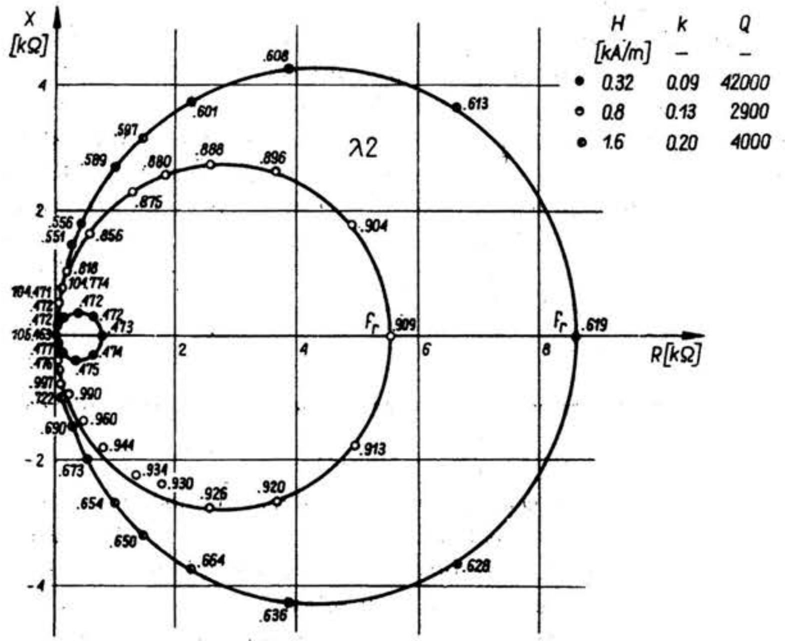

FIG. 3. Impedance circle of a ferrite λ2 specimen.

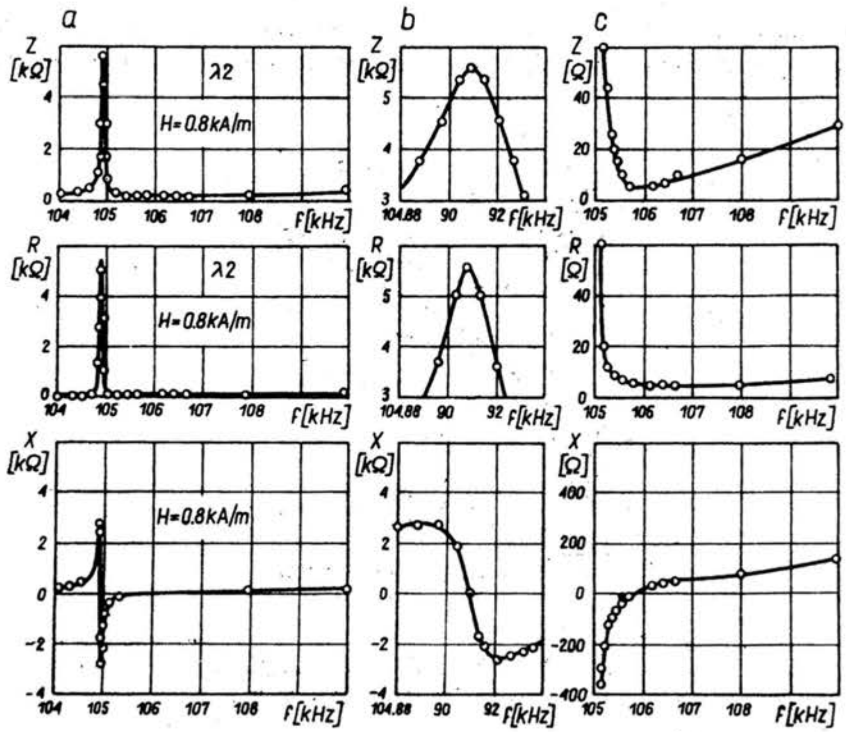

FIG. 4. Frequency characteristics of impedance *Z*, resitance *R*, reactance *X* of a toroidal λ2 ferrite specimen (a), with marked resonance range (b) and antiresonance (c).

and R coincide with the zero values of X and with the corresponding frequencies (cf. Figs. 1 and 2) the dynamic moduli E_H and E_B are determined. The moduli are found from the impedance circles by drawing separate characteristics of X, R, Z (Fig. 4) or by calculating the frequencies corresponding to the points of intersection with the circles: of the diameters parallel to the R-axis (in case of small losses or high quality factors the diameters lie almost on the R-axis — cf. Fig. 3) or, finally, of the chords drawn from the origin of the coordinate system to the points on the circle which are the most remote from 0. The relative differences between the frequencies f_r, f_H and f_a, f_B in the case of ferrites are usually less than one percent, and sometimes reach even the order of 10^{-5} [11].

4. Investigation of the E_H-modulus in the range of the initial magnetization curve and the negative ΔE-effect

Up to the early fifties of this century it was believed that the moduli of elasticity increase monotonically with the magnetic polarization and stabilize their values in the range of magnetic saturation, see e.g. [1, 3]. In 1955 it was observed by Ochsenfeld that in the case of nickel and its alloys the modulus of elasticity initially decreases with increasing polarization, and after reaching a minimum it begins to increase so as to reach a stable value in the range of saturation [13]. The range in which the elasticity modulus E_H is smaller than the value in the state of demagnetization E_0 is called the negative increment range of E, and the effect itself — the negative ΔE-effect. A similar phenomenon in E1 ferrites was observed by the author [4-7, 12]. Further investigations confirmed the existence of the

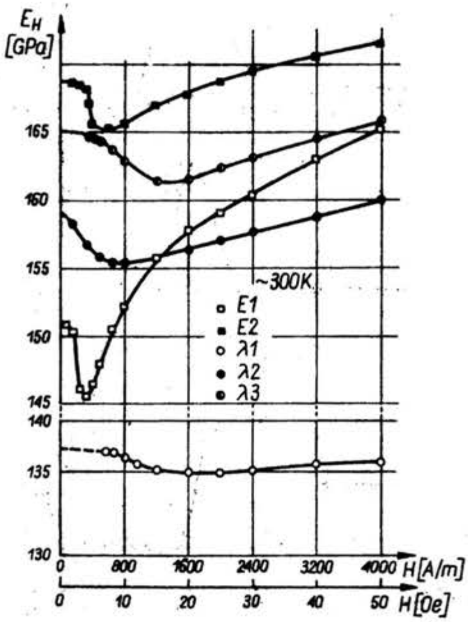

FIG. 5. Dependence of elastic moduli E_H of the considered ferrites on the magnetic polarization in the case of initial curve of magnetization.

negative effect in numerous types of ferrites in a wide range of temperatures [8, 11]; some of the results are presented in this paper.

Figure 5 presents the family of initial curves of elasticity moduli of the ferrites considered. The characteristics are established at an amplitude not exceeding 1.6 A/m (20 mOe). The negative ΔE effect appeared in all ferrites tested. To simplify the analysis, the characteristics of relative variation of the moduli are shown in Fig. 6. The occurrence of the

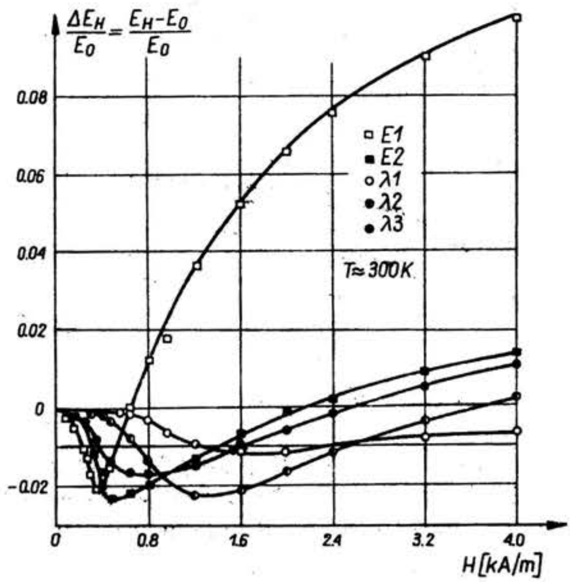

FIG. 6. ΔE_B-effect of the considered ferrites.

ΔE-effect is connected with magneto-mechanical coupling, and so in the $E1$ ferrite (in which the coupling coefficient $k > 0.3$) the effect was the most significant, its negative value reaching 2%, and positive—about 10% [5, 11, 12].

A complete mathematical description of the negative ΔE-effect is still lacking; it will be made possible by introducing to the relations the corresponding piezomagnetic coefficients.

5. Investigation of the E_B-modulus

The elastic moduli at a constant induction E_B increase their values with increasing polarization until the state of magnetic saturation is reached, when further field increments do not influence the modulus E_B. The corresponding characteristics are shown in Fig. 7. The largest relative variations of E_B occur in the $E1$ ferrites and reach 13% [5, 11, 12]. From the characteristics it follows that the magnetic field commonly considered as sufficient for technical saturation (e.g. in various companies producing the ferrites the field intensities of 30 Oe, i.e. 2400 A/m are considered as sufficient for saturation) does not

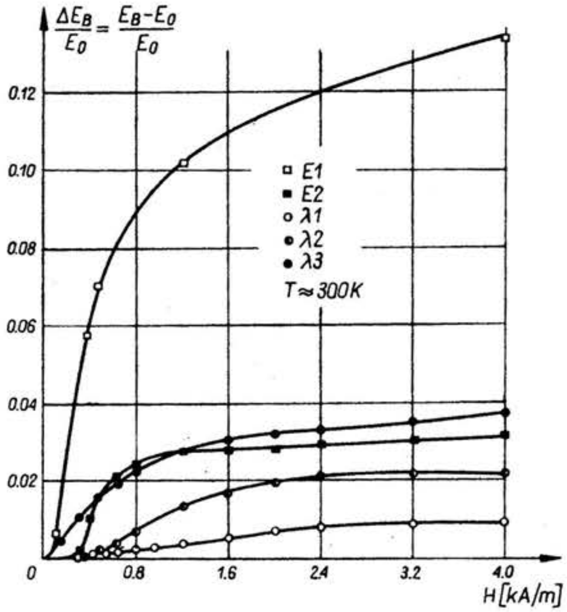

FIG. 7. Relative changes of E_B.

ensure the stabilization of the modulus. A more precise determination of the state of magnetic saturation is possible by means of mechanical investigations rather than by many other classical methods. Of course, such accurate measurements are not always necessary.

6. Elastic moduli and the magneto-mechanical coupling

The work done during elastic deformation is completely transformed, under ideal reversible conditions, into the potential energy of internal elasticity forces. The specific elastic energy in the case of materials subjected to the magnetic field is changed due to the changes of the modulus of elasticity. The value of the modulus measured at constant field intensity E_H and constant induction E_B are equal to each other in the state of demagnetization, but differ in the case of polarization. The formulae for the specific elastic energy must also reveal corresponding differences. Elastic strain energy at a constant field intensity H is defined by the following formula:

$$W_H = \frac{1}{2} TS_H = \frac{1}{2} \frac{T^2}{E_H}, \tag{6.1}$$

and the elastic strain energy at constant induction

$$W_B = \frac{1}{2} TS_B = \frac{1}{2} \frac{T^2}{E_B}. \tag{6.2}$$

The difference of these two energies may be transformed into the magnetic energy:

$$W_\mu = \Delta W = W_H - W_B = \frac{1}{2} T^2 \left(\frac{1}{E_H} - \frac{1}{E_B}\right). \tag{6.3}$$

As in the case of a piezomagnetic transmitter, in the case of a receiver the definition of the coefficient of magneto-mechanical coupling k may be introduced (cf. e.g. [9]). The ratio of mechanical energy transformed into magnetic energy to the total amount of energy stored in the transducer is called the square of the magneto-mechanical coupling coefficient.

$$k^2 = \frac{W_\mu}{W_H} = \frac{W_H - W_B}{W_H} = 1 - \frac{W_B}{W_H} = 1 - \frac{E_H}{E_B}. \tag{6.4}$$

Values of k for the $E1$ and $\lambda 2$ ferrites is given in Figs. 1 and 3.

In such states in which the magneto-mechanical coupling does not occur, and namely in the state of demagnetization and magnetic saturation, the elastic moduli at constant field intensity and at constant induction are equal to each other. At the instant when such a coupling occurs, the values of E_H and E_B diverge, and they attain their maxima usually in the range of polarization larger than that at which the minimum of E_H occurs, and less than that at which the magnetization curve bends markedly. Figure 8 shows examples of the

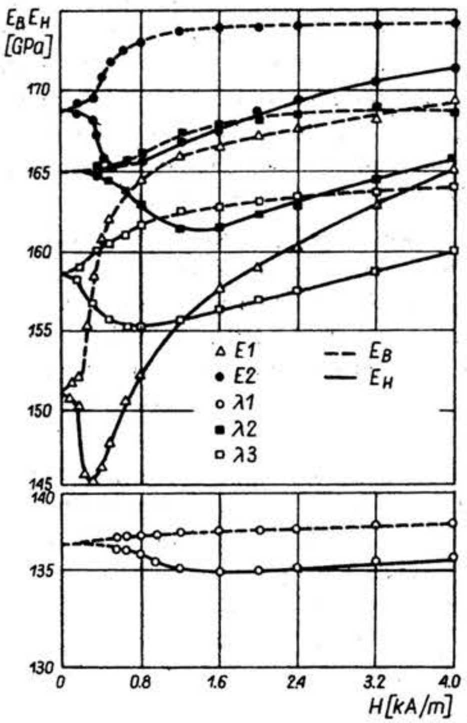

FIG. 8. Primary curves of the E_H and E_B moduli of $E1$, $E2$, $\lambda 1$, $\lambda 2$ and $\lambda 3$ ferrites.

initial curves of the E_H and E_B moduli of the investigated ferrites. In the vicinity of the state of demagnetization the moduli E_H and E_B differ only slightly and it was possible to determine their common values by means of extrapolation. The actual saturation occurred, however, outside the measurement range. Basing upon several additional tests and the extrapolation technique it was established that $E_H = E_B$ at polarizations of the order of 10–25 kA/m. Thus the actual saturation may occur at the field intensities three or even ten times stronger than those generally accepted as sufficient for technical saturation.

7. Discussion of results and conclusions

Research on the variation of mechanical parameters is of both the cognitive and practical values; the variations are connected with the changes of sound velocity and the resonance frequencies of the transducers (Eqs. (3.1) and (3.2)).

Once the variation of elastic moduli is known, the characteristics of ultrasound velocities or resonance frequencies can also be determined. However, the course of research in our case is different: from the resonance frequencies which are characteristic of the given transducer and given polarization, more general parameters of the investigated material are determined, like the elasticity moduli or the sound velocities, which are strongly dependent not only on the magnetic polarization (Figs. 5–8, ΔE effect) but also on the temperature [5, 8]. The temperature coefficients of the elastic moduli of ferrites, in particular temperature ranges, may be of the order of $0.1-200.\ 10^{-6}/°C$.

According to the theory of AKULOV and KONDORSKY [1], the ΔE-effect should be associated with the domain structure of magnetic materials. If a single-domain ball were cut out of the crystal in the temperature higher than the Curie point and cooled afterwards, it would be transformed, due to spontaneous magnetization, into an ellipsoid. Such an ellipsoid reflects the anisotropy, and the positions of its principal axes depend on the directions of spontaneous magnetization. It could be imagined that each domain corresponds to a certain ellipsoid of striction, and the mechanical forces applied from outside to the crystal transform the ball into the so-called Hooke's ellipsoid. Minimum of the crystal energy corresponds to the state in which the major axes of the striction and Hooke's ellipsoids coincide. Action of forces on the magnetic material, even in absence of the external field, changes the magnetic properties. In a multi-domain material subject to the external forces, the volume of those striction ellipsoids whose major axes make small angles with the major axes of Hooke's ellipsoids will increase, while those making large angles will decrease. Consequently, additional changes due to the rotation and expansion of the striction ellipsoids are superposed on the changes produced by elastic (Hooke) strains. This additional change of form of magnetic crystals which does not result from Hooke's law was called by Akulov and Kondorsky mechanostriction, and its theory was developed by BROWN [2]. Since it was assumed that each domain corresponds to a striction ellipsoid, the voluminal changes of domains and rotations of their magnetization vectors change the forms of the crystals. If the striction ellipsoids were placed with their major axes parallel to the field direction, the body would be elongated and the magnetostriction would be positive, and if the axes were perpendicular to the field, the body would be contracted (negative magnetostriction). It follows that major axes of the ellipsoids may be either parallel to the magnetization vectors of the domains ($\varepsilon > 0$), or perpendicular ($\varepsilon < 0$).

In soft magnetic materials and in the range of weak fields, the stresses produced by external forces contribute to the shifting of the domain walls, and in stronger fields to the rotation of the magnetization vectors. In hard magnetic materials the stresses produce mainly the rotations of the magnetization vectors. The direction of changes depends on the directions of the forces and magnetic field applied, and on the sign of magnetostriction.

In the materials with negative magnetostriction, tensile external forces produce, in addition to Hooke's deformations, also the striction deformations, and due to the mechanostriction principle major axes of the ellipsoids will be parallel to the forces applied, and so the magnetization vectors will be perpendicular (Fig. 9).

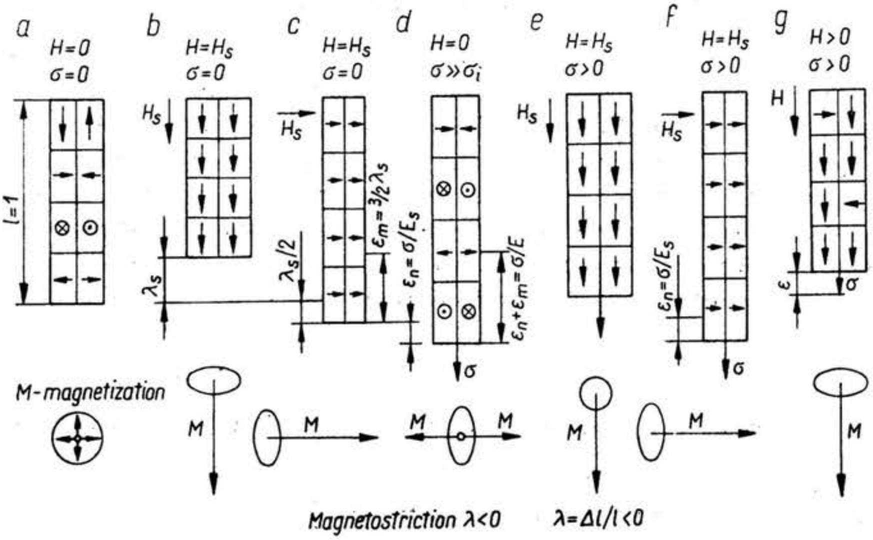

FIG. 9. Distribution of magnetization vectors and changes of dimensions in the piezomagnetic material with negative magnetostriction as a function of the applied magnetic field and external forces.

In the materials with positive magnetostriction the situation will be reversed. Tension makes the magnetization vectors tend to become parallel to the forces, and compression — perpendicular. This mechanism explains numerous phenomena known from practice such as material processing in a magnetic field, magnetization of materials subjected to stresses, properties of materials with inclusions, etc.

In the case of compression, minor axes of the striction ellipsoids will be elongated, and major axes shortened, and hence the mechanostriction will be negative. Mechanostriction is, like Hooke'a deformation, of the same sign as the force applied. In the case of compression, similarly to the case of tension, the elasticity modulus will decrease. If the positions of the magnetization vectors and the striction ellipsoids were frozen in space so as to eliminate their rotations, the mechanostriction would be zero and the elasticity modulus would assume its normal value $E_n = E_s$. Such a stiffening of the magnetization vectors may be achieved by providing a sufficiently high magnetic field intensity.

The elasticity modulus as a function of the magnetic field intensity increases and reaches its normal value in the state of saturation. The phenomenon is sketched in Fig. 9. In a demagnetized material free of stresses, the distribution of magnetization vectors is disordered and the resultant magnetization vanishes (a). Spontaneous magnetization disturbs the lattice symmetry. After saturation the material contracts by the value corresponding to the saturation magnetostriction, that is by $\varepsilon_s l$ in the case of magnetization and measurements taken along the specimen (b), and by $\varepsilon_s l/2$ in the case of transversal saturation (c). In both cases the imagined ball deformes to identical (but perpendicular to each other) ellipsoids.

When a demagnetized specimen is loaded by external tensile forces counteracting the internal stresses, its length increases by $[(\varepsilon_s/2)+\varepsilon_n]l$ (d) in comparison to the initial state (a) or by $\left(\frac{3}{2}\varepsilon_s+\varepsilon_n\right) l$ in comparison to the state of longitudinal saturation (b). Only with respect to the state of transversal saturation (c) will the deformation be the usual one, i.e. $\varepsilon_n l$, and the additional ralative elongation $\varepsilon_m = 0$. Similar processes may also be observed in the case of smaller tensile force acting on a specimen transversally saturated up to the value of $\sigma(f)$, when the deformation follows Hooke's law. Some differences appear in the values of the resulting magnetization. Under small field intensities and moderate tensile stresses the deformations will follow partly from Hooke's law, and partly from the striction (g). Under certain conditions both phenomena may compensate each other. A similar compensation may also be achieved in the case of longitudinal saturation and tension (e).

In all cases the tensile forces do not exced the elastic limit. The greatest additional relative deformation $\varepsilon_m = \frac{3}{2}\varepsilon_s$ is reached in the specimen when the direction of saturation is changed from parallel to perpendicular, and such a maximum correction should be taken into account under strong tension parallel to the field H_s. For $H_s \perp \sigma$ the correction vanishes $\varepsilon_m = 0$. Under small tensile stresses only partial reorientation of the magnetization vectors occurs, and the mechanostriction is strongly influenced by the internal stresses. The applied forces make the magnetization vectors assume the perpendicular positions, and hence in a demagnetized material the numbers of vectors assuming paralled and opposite directions are equal. The magnetic field leads to parallel ordering of the vectors. If the direction of the field and the tensile stresses coincide, the mechanism of changes is highly complicated since under the action of the field the vectors will tend to parallel positions and forces will try to make the perpendicular. Thus the energy of the saturating must in this case be sufficiently great. The above considerations enable us to determine the character of deviations from Hooke's law.

As it was mentioned before, the magnetic field in the state of saturation immobilizes all the magnetization vectors of the domains so that their reorientation cannot occur; no striction deformations take place and the elasticity modulus does not change. The ΔE-effect is an "even" phenomenon and does not occur in the case of 180° — displacements of the Bloch walls; that is why the application of a perpendicular field to a specimen under tension (Fig. 9d) does not, in principle, change its dimensions. The ΔE-effect is connected with rotation of the magnetization vectors and non-180° displacements of the Bloch walls (e.g. 90°, 109° and 71°). The specific strain energy E_σ under constant external stresses is equal to

$$E_\sigma = \sigma \int_0^{\varepsilon_s} d\varepsilon = \frac{3}{2} \varepsilon_s \sigma \sin^2\theta. \tag{7.1}$$

This means that in the materials with negative magnetostriction the energy reaches its minimum under tension for 90°, that is the stresses place the magnetization vectors in perpendicular positions, while in the materials with positive magnetostriction the angle is 0°. In the case of compressive forces the situation will be reversed. This author suggested [11] that the negative ΔE-effect was connected with the fulfilling of the condition

$$k^2 > \frac{\Delta E_B}{E_B}. \tag{7.2}$$

At small values of coupling the negative ΔE-effect may not occur at all.

The corresponding theory of the effect should be associated with the variations of the piezomagnetic coefficients. Value of the moduli change with temperature and the changes depend mainly on the variations of magnetocrystalline anisotropy constants. In the case of $E1$ ferrites at $-70°C$, $E_0 = 1.78 \cdot 10^{12}$ dynes/cm^2, i.e. 178 GPa. If the anisotropy constant K_1 is compensated and changes its sign at the temperature characteristics, the minima of resonance frequencies and elastic moduli occur in this range [5–8, 11, 12]. This problem will be discussed in detail in another publication.

The moduli of elasticity of ferrites are strictly dependent on the magnetic polarization. Most considerable changes occured in the case of the $E1$ ferrite containing large amounts of cobalt ions admixtures which lead to compensation of the magnetocrystalline anisotropy constants in the range of room temperatures [8].

Slight changes of cobalt contents (order of per mill) produce considerable variations of the moduli and their dependence on the magnetic intensity (and also temperature). Technology of production may also exert an influence on the moduli since it determines the density, porosity, defects, grain sizes etc. Once the dependence of the moduli on temperature and ferrite composition is known, the corresponding materials for piezomagnetic transducers may be programmed; one might design the materials with a constant temperature coefficient f_r in a definite temperature range, or with a variable (positive or negative) coefficient if compensation of piezomagnetic coefficients of other elements of the system were required.

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Received August 8, 1980.