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Classical magnetostriction of nickel in high magnetic field

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Abstract. Thermodynamical description of the classical magnetostriction is proposed. Magnetostriction and magnetization of nickel are measured in fields up to 14 T. The change of the chemical potential of electrons is measured in fields up to 10 T. The experimental results are in a reasonable agreement with the theory.

PACS. 75.80.+q Magnetomechanical and magnetoelectric effects, magnetostriction – 05.70.-a Thermodynamics

The magnetostriction, i.e. the change of the dimensions and shape of the sample under the influence of a magnetic field has been known since its discovery by Joule in 1847 [1]. Terms like "classical" and "atomic" magnetostriction were introduced by famous Russian physicist Kapitza in 1932 [2]. The "classical" concept means that the magnetostriction is dependent only on the macroscopic properties of the sample (magnetization, bulk modulus, etc.). Quite opposite, the "atomic" magnetostriction is determined by microscopic parameters of the sample, i.e. the magnetocrystalline anisotropy, the dependence of the exchange interactions and magnetization on pressure and so on. Starting from the profound work of Akulov [3], the main line of numerous theoretical investigations was just the "atomic" magnetostriction (see [4] and references herein). As for the "classical" magnetostriction we have found only one very old review [5], in which it is considered from purely mechanical idea of interaction between magnetic poles of the sample and the applied magnetic field. A similar approach was used much later for the explanation of the pinning-induced magnetostriction of the high-temperature superconductors [6].

It is rather simple to get thermodynamical description of the classical magnetostriction. First of all, it should be reminded that magnetic field does not perform any work (in other words it is non-potential). Indeed, the Lorentz force acting on an electric charge in magnetic field is directed perpendicular to its velocity. Free spin **s** precesses around the direction of the magnetic field **H** keeping the angle between **s** and **H** independent of the field strength. The net magnetization of the sample is only caused by the "friction" of spins with the environment (relaxation effects). Thus, the magnetic field can not change the total internal energy of the sample, and the magnetic energy $-\mathbf{MH}$ ($\mathbf{M}$ is the magnetic moment of the sample) may originate from other energies of the sample, namely the mechanical energy pV (p is the internal pressure, V is the sample volume), the thermal energy TS (T is the temperature, S is the entropy) and the energy of conductivity electrons μN (μ is the chemical potential of N electrons):

$$-\mathbf{MH} + \delta(pV) + \delta(TS) + \delta(\mu N) = 0. \tag{1}$$

The change of the mechanical energy is equal to

$$\delta(pV) = V\delta p + p\delta V = VB\left(\frac{\delta V}{V}\right) + \frac{3}{2}VB\left(\frac{\delta V}{V}\right)^2 \tag{2}$$

where B is the bulk modulus. Because even a "giant" magnetostriction is of the order of $(\delta V/V) \approx 10^{-3} \div 10^{-2}$ the second term in equation (2) may be neglected.

At constant temperature, $\delta(TS) = T\delta S = Q$, where Q is the extracted (or absorbed) heat.

To fulfil the requirement of electroneutrality, the electron concentration should be constant and we obtain the following expression for the classical magnetostriction:

$$\frac{\delta V}{V} = \frac{\mathbf{MH} - T\delta S - N\delta\mu}{VB}. \tag{3}$$

To test the validity of this thermodynamical description of the classical magnetostriction we have carried out measurements of the magnetostriction and magnetic moment of nickel in fields up to 14 T and we have measured the change of the chemical potential of electrons in fields up to 10 T. This ferromagnetic metal was chosen because its mechanical, magnetic and electronic properties are well investigated and it is used as standard for magnetic measurements. Earlier, the magnetostriction of nickel was measured in much smaller magnetic fields.

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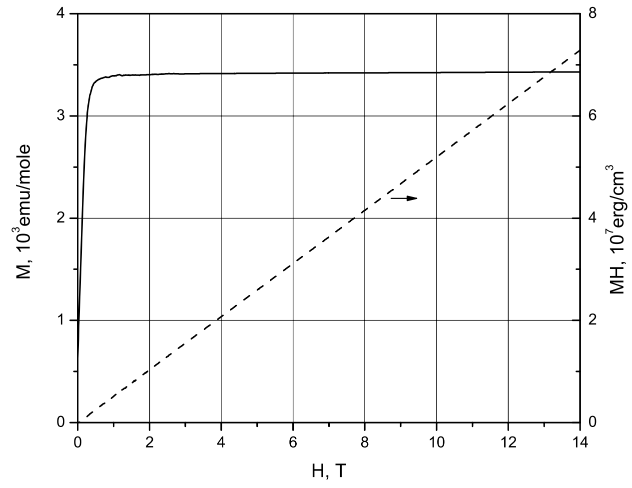

Fig. 1. Magnetization of nickel at $T = 4.2$ K (solid line) and the energy product MH (dashed line).

Magnetostriction and magnetic moment were measured on the polycrystalline sample of nickel which had approximately spherical shape with the diameter of about 0.23 cm. The longitudinal and transverse magnetostriction were measured with a capacitance dilatometer, whereas the magnetic moment was measured with a Foner-type magnetometer.

The method to measure the change of the chemical potential was described in details elsewhere [7,8]. It is based on the determination of the change in charge on the measuring capacitor consisting of the sample under investigation and the reference electrode. The sample was polished plate with a thickness of 0.13 cm made from the same 99.99% purity material as the sample for the magnetostriction and magnetization measurements. The reference electrode with the diameter of 0.85 cm was made from bronze. The measuring capacitor is similar to the one described in reference [7]. The capacitance of the measuring cell was of 31 ± 1 pF. The magnetic field was applied perpendicular to the sample plane. Due to a small value of the charge (less than 20 fC), which was comparable or even smaller than the drift of an electrometer (Keithley 642) during the field sweep, it was necessary to average many up and down runs. All measurements were done in a superconducting magnet at $T = 4.2$ K.

The experimental measured magnetization is shown in Figure 1. It saturates above 1.5 T to $m_{eff} \approx 0.6\ \mu_B$. The magnetic energy increases linearly reaching a value of -7.3×10^7 erg/cm^3 at $H = 14$ T.

The change of the entropy could be estimated from the thermodynamical relation $\partial S/\partial H = \partial M/\partial T$. For this purpose, we have measured the magnetization at $T = 3.8$ K, too. The result was the same as for $T = 4.2$ K with an accuracy better than 10^{-3}, thus the change of the entropy may be neglected.

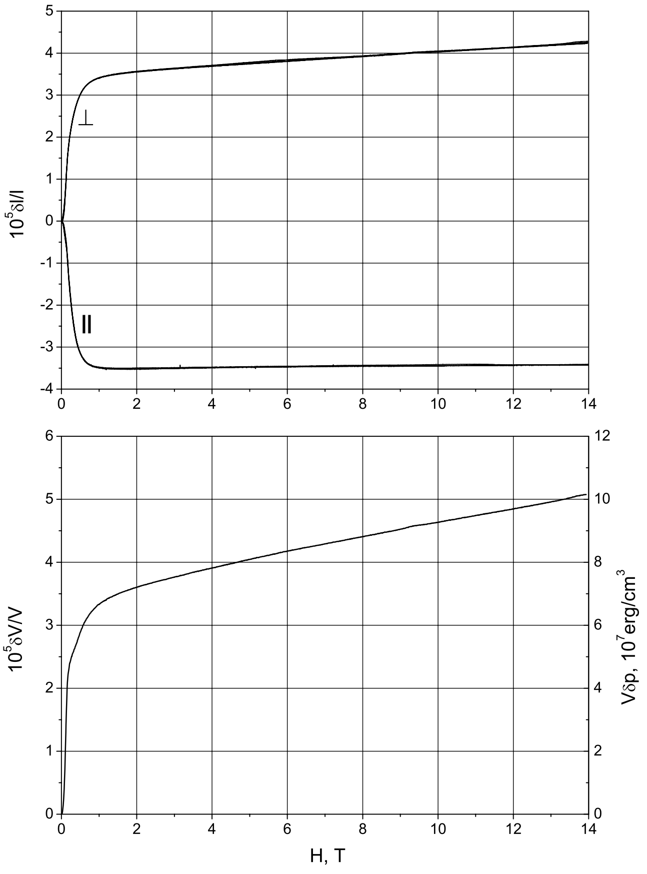

Fig. 2. Magnetostriction of nickel at $T = 4.2$ K.

The experimental results for the linear magnetostriction are shown in the upper part of Figure 2. From these values the volume magnetostriction was calculated according to formula $\delta V/V = (\delta l/l)_{\|} + 2(\delta l/l)_{\perp}$ (Fig. 2, bottom graph).

As it is seen from Figure 2, the linear magnetostriction of nickel is very anisotropic: the longitudinal one is negative, whilst the transverse one is positive. This behavior is well known [4]. The longitudinal magnetostriction seems to saturate with increasing the magnetic field, but it is not the truth, because in field higher than 1.5 T it has small positive slope $(\delta l/l)'_{\|} = 7 \times 10^{-8}$ T^{-1}. The transverse magnetostriction rises with much higher slope $(\delta l/l)'_{\perp} = 5.7 \times 10^{-7}$ T^{-1}. Just the longitudinal magnetostriction of nickel was measured earlier, and its apparent saturation was considered as a clench in favour of the simple theory in which the magnetostriction should be proportional with the square magnetization [9,10].

The volume magnetostriction is positive as it is expected for ferromagnets, but does not saturate with increasing magnetic field as magnetic moment does. Quite contrary, it goes up with a slope $(\delta V/V)' = 1.2\times10^{-6}$ T^{-1}. This value is in a good agreement with the results obtained on single crystals for $H \leq 2.6$ T [11] and polycrystals for $H \leq 6.3$ T [12].

The change of the mechanical energy (right scale in the bottom graph of Fig. 2) was calculated using for the bulk modulus mean value $B = 2 \times 10^{12}$ erg/cm^3 [13,14]. This change is much larger than the magnetic energy (Fig. 1), and if equation (1) is valid, a significant change in the chemical potential of electrons should be observed. The experiment shows that this is the case.

The chemical potential of electrons, μ, may change due to the influence of the magnetic field ($\delta\mu^H$) and due to a striction ($\delta\mu^V$).

The problem of the magnetic field influence on the chemical potential of electrons in ferromagnetic metals was formulated by Dorfman [15]. The formulation in modern terms was given by Walmsley 30 years later [16].

In the ferromagnetic state, the bands of electrons with spins up and spins down are shifted in energy due to the exchange interactions. If we apply a magnetic field, then the energy of spin-up electrons decreases and spin-down electrons energy increases by $m_{\mathit{eff}} H$. The resulting change of the chemical potential may be found from the condition of constancy of the electrons concentration

$$(\delta\mu^H + m_{\mathit{eff}} H)g^+ + (\delta\mu^H - m_{\mathit{eff}} H)g^- = 0 \qquad (4)$$

where g^+ and g^- denote the density of states at the Fermi level of spin-up and spin-down electrons. Thus

$$\delta\mu^H = \alpha m_{\mathit{eff}} H; \quad \alpha = \frac{g^- - g^+}{g^- + g^+}. \qquad (5)$$

The band-structure calculations give gently different values of g^+ and g^-. According to reference [17] $g^+ = 4.5$, $g^- = 22.6$ states/Ry atom and $\alpha = 0.67$, whereas from reference [18], $g^+ = 2.35$, $g^- = 21.2$ states/Ry atom and $\alpha = 0.80$.

The change of the chemical potential due to the magnetostriction itself may be estimated in the free-electrons model. In this model $\mu \sim (N/V)^{2/3}$, so

$$\delta\mu^V = -\frac{2}{3}(\delta V/V)\mu. \qquad (6)$$

Using equations (5), (6) and (3) we obtain the following expression for the slope of magnetostriction in high magnetic fields

$$\left(\frac{\delta V}{V}\right)' = \frac{N_A(1-\alpha)m_{\mathit{eff}}}{V_M B - \frac{2}{3}N_A\mu} \qquad (7)$$

where N_A is the Avogadro number, V_M is the molar volume.

For nickel $V_M = 6.592$ cm^3, $\mu \simeq 0.6$ Ry [17], $B \approx (1.9 \div 2.1) \times 10^{12}$ erg/cm^3 [13,14], $\alpha \approx 0.7 \div 0.8$ (see above) and the estimated value of $(\delta V/V)' \approx (0.8 \div 1.3) \times 10^{-6}$ T^{-1}. Thus, the experimental value of the slope ($(\delta V/V)' = 1.2 \times 10^{-6}$ T^{-1}) is within this interval.

The change of the chemical potential observed from experiment (average of 14 runs) is shown in Figure 3 by the solid line. The dashed line shows the behavior of the chemical potential as expected from equation (1). Quantitative difference between the curves may be explained by the fact that the measurements of the chemical potential give information about the very thin surface layer of a metal (of order of the Debye screening length). The bulk modulus at the surface is reduced, whereas the effective magnetic moment is enhanced. Smaller bulk modulus results in a deeper minimum at $H \approx 1.5$ T, and a higher magnetic moment leads to a larger slope of the chemical potential for $H > 2$ T. The difference in small magnetic fields $H < 0.5$ T may be ascribed to the difference in the demagnetizing factors of the spherical and flat samples.

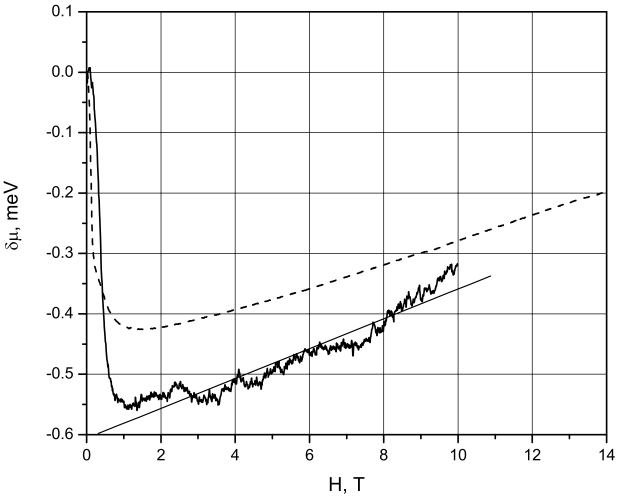

Fig. 3. Change of the chemical potential of electrons in nickel with a magnetic field.

Earlier, we claimed that the chemical potential of electrons in bulk sample is independent on the magnetic field [7,19]. This wrong conclusion resulted from the experiments performed with low-sensitivity electrometer. The same situation appears during the experimental search for the change of the chemical potential of electrons at transition to the superconducting state: low-sensitivity measurements did not allow to observe any change in the chemical potential of tin [20], whereas after significant improvement of the sensitivity such a change was observed in niobium [8].

In conclusion, the thermodynamical description of the magnetostriction was proposed and confirmed by experiments on ferromagnetic nickel. It was experimentally shown that electronic contribution to the magnetostriction is decisive. This is in agreement with the microscopical theoretical investigations of the magnetostriction, those taking into account a spin-orbit coupling [21] or a strain dependence of the magneto-crystalline anisotropy [22,23].

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