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Abstract. – Magnetothermal effect associated heat flow and eddy current loss serve as an additional entropy source in the magnetization process. Based on simplified models, it is shown that, though these effects are included, losses and the entropy production in a hysteresis cycle still obey Warburg's law or its simple version.

The energy loss associated with a hysteresis loop is equated to the area enclosed by the loop (Warburg's law [1]). The loss divided by the temperature of the body is considered to be an entropy production in each cycle:

$$\oint \mathrm{d_i}S = (1/T_0) \oint H \; \mathrm{d}M_H, \tag{1}$$

where M_H is the component of magnetization in the direction of applied field $\mathbf{H}$ and T_0 is a temperature. The present author discussed, on a domain theoretical basis, the entropy production in the isothermal magnetization processes of ferromagnetic insulators; he showed that an irreversible heat loss was given by the decrement of thermodynamic potential and derived equation (1) in some of simple cases [2]. In reality, the magnetothermal effect involves a heat flow so as to eliminate the temperature difference between the body and an environment. This serves as an additional entropy source. In ferromagnetic metals, eddy currents also contribute to the energy loss. The purpose of this report is to show that even in these cases the total entropy production in the hysteresis cycle is given by equation (1).

Consider a magnetizing process of a ferromagnetic insulator, where temperature, T, may differ from that of the environment, T_0. The heat transferred to the environment is denoted by $\mathrm{d}Q'$, then the change in the internal energy is given by

$$\mathrm{d}U = \mathbf{H} \cdot \mathrm{d}\mathbf{M} - \mathrm{d}Q', \tag{2}$$

where uniform magnetization $\mathbf{M}$ is assumed. Two sorts of heat induce the temperature change: one which compensates for the entropy of the spin system, S_m, is given by

$$\mathrm{d_e}Q = -T \; \mathrm{d}S_\mathrm{m}. \tag{3}$$

The other, $\mathrm{d_p}Q$, which is a remainder of the evolved heat is proper to the irreversible change. On the other hand, $\mathrm{d}Q'$ is the excess heat over the amount used for raising the temperature of the body:

$$\mathrm{d}Q' = \mathrm{d_e}Q + \mathrm{d_p}Q - C_M \mathrm{d}T. \tag{4}$$

Here, C_M is a heat capacity at constant $\mathbf{M}$. Apart from the temperature close to the Curie point, C_M is practically independent of $\mathbf{M}$. Since $\mathrm{d_e}Q/T$ counterbalances $\mathrm{d}S_\mathrm{m}$, a net entropy change of the body (lattice and spins) is given by $\mathrm{d_p}Q$ and $\mathrm{d}Q'$ as in the form

$$\mathrm{d}S = (1/T) \left(\mathrm{d_p}Q - \mathrm{d}Q'\right), \tag{5}$$

or

$$= \mathrm{d}S_\mathrm{m} + (C_M/T)\, \mathrm{d}T. \tag{5$'$}$$

Under the condition of constant field, equations (2) and (5) together with the thermodynamic potential $\Psi = U - TS - \mathbf{H} \cdot \mathbf{M}$ yield an expression

$$\mathrm{d_p}Q = -\mathrm{d}\Psi - S \; \mathrm{d}T. \tag{6}$$

The entropy source in the whole system (body and environment) consists of two parts: the first part $\mathrm{d_p}Q/\mathrm{d}t$ is due to irreversible heat evolution (intrinsic loss), while the second one given by

$$[(1/T_0) - (1/T)] \left(\mathrm{d}Q'/\mathrm{d}t\right)$$

is a consequence of heat flow resulting from the temperature difference between the body and the environment. Thus, by using equations (3), (4) and (6), the entropy production of the whole system becomes

$$\begin{aligned} \mathrm{d_i}S/\mathrm{d}t = &-(1/T_0)\,\{(\mathrm{d}\Psi/\mathrm{d}t) + S\,(\mathrm{d}T/\mathrm{d}t) + \\ &+\Delta T\;[(\mathrm{d}S_\mathrm{m}/\mathrm{d}t) + (C_M/T)\,(\mathrm{d}T/\mathrm{d}t)]\} \end{aligned} \tag{7}$$

with $\Delta T = T - T_0$.

Expressidons of the entropy and the thermodynamic potential of the body at a state (T, $\mathbf{H}$; $\mathbf{M}$) will be derived. Here, $\mathbf{M}$ may not be in equilibrium under $\mathbf{H}$ and at T. We assume a state (T_0, $\mathbf{H}$; $\mathbf{M}$) whose magnetization is the same as that of (T, $\mathbf{H}$; $\mathbf{M}$) but the temperature is remained at T_0. The entropy of the spin system of these two states are equal, so that the difference in the entropy of the body is given by the difference in the lattice entropy. Therefore the entropy of the state (T, $\mathbf{H}$; $\mathbf{M}$) is

$$S = S_{l0} + S_\mathrm{m} + \int_{T_0}^{T} (C_M/T)\, \mathrm{d}T, \tag{8}$$

where S_{l0} is the lattice entropy of the state (T_0, **H**; **M**). By using equations (2), (4) and (8), the difference in the thermodynamic potential of two states, $\Delta\Psi = \Delta U - \Delta(TS)$, can be obtained in the explicit form. Thus, the thermodynamic potential of the state (T, **H**; **M**) is

$$\Psi = \Psi_0 - (S_{l0}+S_{\rm m})\,\Delta T + \int_{T_0}^{T} C_M \mathrm{d}T - T\int_{T_0}^{T}(C_M/T)\,\mathrm{d}T, \quad (9)$$

where Ψ_0 is the thermodynamic potential of the state (T_0, **H**; **M**). By substituting equations (8) and (9) into equation (7), the entropy production in the whole system is obtained as

$$\mathrm{d_i}\mathcal{S}/\mathrm{d}t = -(1/T_0)\,[(\mathrm{d}\Psi_0/\mathrm{d}t) + (C_M/T)\,\Delta T\,(\mathrm{d}T/\mathrm{d}t)]\,. \quad (10)$$

Hysteresis of a single domain particle of insulator will be treated. A complete cycle consists of a number of irreversible and reversible rotations. The direction of the field is fixed and its stregth is changed sufficiently slowly. In other words, if once a critical field is reached and an irreversible rotation starts, the strength of the field is kept constant until the rotation is completed and the temperature of the body recovers its initial value T_0. The entropy produced during individual irreversible rotation, $\Delta_{\rm i}\mathcal{S}$, is given by the integration of equation (10) with respect to t from $t = 0$ to ∞. C_M is a function of the temperature only, so that integration of the second term on the right-hand side of equation (10) vanishes. Hence, we have

$$\Delta_{\rm i}\mathcal{S} = -(1/T_0)\,\Delta\Psi_0, \quad (11)$$

where $\Delta\Psi_0$ is a difference in Ψ_0 between the initial and the final state of irreversible rotation. Since $\mathrm{d}\Psi_0 = \mathrm{d}F_0 - H\ \mathrm{d}M_H$ holds (F_0 is the Helmholtz free energy when the temperature of the body is assumed to be T_0), then

$$\Delta_{\rm i}\mathcal{S} = (1/T_0)\,(H\ \Delta M_H - \Delta F_0)\,. \quad (12)$$

Furthermore, for a reversible rotation, $\mathrm{d}\Psi_0 = 0$, namely,

$$0 = (1/T_0)\,(H\ \mathrm{d}M_H - \mathrm{d}F_0) \quad (13)$$

holds. By adding equations (12) and (13) for all elementary processes and bearing in mined that $\oint \mathrm{d}F_0 = 0$, equation (1) is obtained for a complete cycle.

Now we treat wall displacements in ferromagnetic metals and examine the effect of eddy currents on hysteresis loss. For simplicity, a plane 180° wall is considered and a constant temperature, T_0, throughout the medium (i.e. infinitely large heat conductivity) is assumed. The field, H, is applied in parallel or antiparallel to the domain magnetization (intensity: M), and its strength is changed sufficiently slowly. Besides H the wall experiences an eddy current counterfield $H_{\rm B}$. Hence, the thermodynamic potential of the wall [2] is given by

$$\Psi = \Psi_0 + 2MH_{\rm B}x \quad \text{with} \quad \Psi_0 = \sigma(x) - 2MHx, \quad (14)$$

where x is a position of the wall. Therefore the intrinsic loss becomes

$$\mathrm{d_P}Q/\mathrm{d}t = -(\mathrm{d}\Psi_0/\mathrm{d}t) - 2MH_{\rm B}\,(\mathrm{d}x/\mathrm{d}t)\,. \quad (15)$$

To evaluate the eddy current loss, we invoke the energy theorem following from Maxwell's equations which relate the eddy current **i** and electromagnetic fields $\mathbf{E}_{\rm I}$ and $\mathbf{H}_{\rm I}$ produced by induction [3]. At the wall position $|\partial\mathbf{M}/\partial t| = 2M\ \mathrm{d}x/\mathrm{d}t$, otherwise $|\partial\mathbf{M}/\partial t| = 0$. $H_{\rm I}$ at the wall position is replaced by $-H_{\rm B}$. By taking into account these specifications, the Joule heat induced by a unit area of the moving wall is found to be

$$\rho\int_V i^2\mathrm{d}V = 2MH_{\rm B}\mathrm{d}x/\mathrm{d}t + -(\mathrm{d}/\mathrm{d}t)\left[\int_V\left(\varepsilon_0E_{\rm I}^2+\mu_0H_{\rm I}^2\right)\mathrm{d}V\right], \quad (16)$$

where integration is made over a sufficiently large volume as compared with the unit area of the wall, ρ is resistivity, ε_0 and μ_0 being permittivity and permeability of vacuum. A net energy loss associated with wall motion is given by the sum of equations (15) and (16), where the counterfield term disappears. Since $E_{\rm I}$ and $H_{\rm I}$ vanish before and after the irreversible wall motion, the second term on the right-hand side of equation (16) integrated with respect to t from $t = 0$ to ∞ becomes zero. Thus, the energy loss per individual irreversible wall jump is simply $-\Delta\Psi_0$. In a hysteresis cycle, walls go and back with succession of reversible and irreversible motions. A procedure similar to that made in the last part of the previous paragraph can be applied. Thus, it turns out that equation (1) holds again in this case.

The summary is as follows: so far as the static hysteresis is concerned, it has been shown, in some simplified cases, that, though eddy currents or heat flow associated with magnetothermal temperature change contributes to the energy loss or/and the entropy production, total loss or total entropy production in the hysteresis cycle strictly obeys Warburg's law or its entropy version.

[1] Warburg, E., *Ann. Phys. Lpz.* **13** (1881) 141.

[2] Iwata, T., *J. Magn. Magn. Mater.* **59** (1986) 215.

[3] Becker, R. and Döring, W., *Ferromagnetismus* (Springer, Berlin) 1939, p. 360.