# MIGRATIONAL MAGNETIC VISCOSITY FOR SEVERAL SINGLE TIME CONSTANT RELAXATION PROCESSES

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The equation of motion for a Bloch wall was extended to the case of several simultaneous single time constant relaxation processes, using Néel's theory of the migrational magnetic viscosity.

The solutions of equations of motion show that for the phenomenon of the disaccommodation of magnetic susceptibility one can apply the principle of superposition, while in the case of the tangent of relaxation loss angle this principle is not satisfied. This proves that for the study of complex phenomena of magnetic viscosity, connected with discrete relaxation processes one should apply the method of measurement of the disaccommodation of magnetic susceptibility.

## 1. *Introduction*

Migrational properties of interstitial solution of carbon and nitrogen in $\alpha$ iron and in its substitutional alloys are more and more frequently investigated with the help of the magnetic viscosity methods.

The phenomenon of the disaccommodation of magnetic susceptibility and the temperature dependence of the tangent of the relaxation loss angle are the ones mainly used in these studies. For these alloys one observes several single time-constant relaxation processes as it was shown in Refs [1–6]. Even for binary $\alpha$Fe–C and $\alpha$Fe–N systems apart from the Snoek relaxation, connected with the directional ordering of single atoms of carbon or nitrogen there also exist relaxation processes connected with ordering of pairs and triplets of interstitial atoms [4, 5, 7–9]. For triple alloys of the type $\alpha$Fe–X–AM, where X denotes an atom of substitutional solution and AM is an interstitial atom, there appears in addition a sequence of relaxations caused by the jump of interstitial atoms close to substitute atoms. These relaxations are lying close to the Snoek ones.

At the present time the most commonly applied theory of the migrational magnetic viscosity is the Néel's theory [10]. It was formulated for $\alpha$ iron, containing carbon atoms

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in the solid solution. This theory is also applied to other magnetic materials, containing relaxations subject to directional ordering. However the original theory of Néel was formulated for the case when in the considered sample one relaxation process occurs and is characterized by a single time constant or by continuous spectrum of relaxation times. In connection with this it is proposed to extend the theory of the migrational magnetic viscosity to the case of several simultaneous single time constant relaxation processes. The theory has been modified as to be applicable to the description of experimental results on the disaccommodation of magnetic susceptibility and on the tangent of relaxation loss angle.

## 2. *Equation of motion for the Bloch wall*

Let us assume that for a sample there are observed several independent elementary relaxation processes each of which is characterized by a single time constant. Additionally it is assumed that the domain structure of the sample may be represented by a single effective Bloch wall of 180° or 90°. Néel's equation of motion for the effective Bloch wall for a unit area is built up of following terms:

pressure of applied magnetic field $P_H$;

pressure resulting from perturbation of the lattice $R(u)$, which plays a role of the restoring pressure;

resultant pressure of viscosity field $P_W(u, t)$ induced by directional ordering of relaxation centers interacting with the spontaneous magnetization vector $\vec{I}_s$, which plays a role of a damping factor for the Bloch wall motion:

$$P_H+R(u)+P_W(u, t) = 0. \tag{1}$$

In equation (1) there is no term representing the product of the mass of a unit area of the wall and of its acceleration because of its small value compared to other pressures.

In weak magnetizing fields the restoring pressure moving the wall into the position of equilibrium can be expressed in the form:

$$R(u) = -\alpha u(t) \tag{2}$$

where $\alpha$ is the proportionality coefficient and $u(t)$ denotes deflection of the wall from the position of equilibrium at the time $t$.

Pressure of applied magnetic field equals to

$$P_H = \vec{H}\cdot(\vec{I}_1-\vec{I}_2) \tag{3}$$

where $\vec{H}$ denotes applied magnetic field intensity vector and $\vec{I}_1$ and $\vec{I}_2$ — intensity vectors of spontaneous magnetization in neighbouring domains.

In the case when the applied magnetic field is parallel to the vector $\vec{I}_s$, the pressure acting on the wall 180° equals $P_H = 2HI_s$, while on the wall 90° of the closing domains type $P_H = \sqrt{2}HI_s$.

In the calculation of the pressure of the viscosity field $P_W(u, t)$ one assumes, according to Néel [10], that the $i$-th kind of relaxator ordered directionally under the influence of vector $I_s$, which makes the angle $\varphi_i$ with it, has an additional energy:

$$E_i = w_i \cos^2 \varphi_i \tag{4}$$

where $w_i$ denotes the energy of interaction with the spontaneous magnetization field vector when $\varphi_i = 0$.

Let us assume that the stabilization energy of the $i$-th kind of relaxator equals $E_{si}$. By analogy to Néel's theory of single relaxation process [10], the total stabilization energy at the time $t$ can be expressed in the form

$$E_s(t) = \sum_{i=1}^{n} E_{si}(t) = \sum_{i=1}^{n} \int_0^t E_{di}(\vartheta) \exp\left(-\frac{t-\vartheta}{\Theta_i}\right) d\vartheta \tag{5}$$

where $E_{di}(\vartheta)$ is the stabilization energy just after the change of direction of spontaneous magnetization vector and $\vartheta$ is an integration variable from the interval $0 \leqslant \vartheta \leqslant t$. The dependence of the time constant $\Theta_i$ on absolute temperature $T$ is given by the Arrhenius law:

$$\Theta_i = \Theta_{0i} \exp\left(\frac{Q_i}{RT}\right) \tag{6}$$

where $Q_i$ and $\Theta_{0i}$ denote activation energies and preexponential factors for single relaxation processes, respectively and $R$ is the gas constant.

In Refs [5, 11] the stabilization energy for the solutions αFe–C and αF–X–C was calculated using the equations describing the kinetics of jump of the interstitial atoms.

The total pressure of the viscosity field, which is the derivative of stabilization energy with respect to the wall shift in conformity with its bounds is given by

$$P(u, t) = -\sum_{i=1}^{n} \int_0^t \frac{p}{d\Theta_i} [u(t)-u(\vartheta)] W_{0i} \exp\left(-\frac{t-\vartheta}{\Theta_i}\right) d\vartheta \tag{7}$$

where $p$ is a coefficient dependent on the kind and position of the Bloch wall and $d$ is the wall thickness. For the wall 90° situated in plane (100) one has $p = 2/3$, while for the wall 180° in the (100) plane $p = 4/3$. $u(t)$ and $u(\vartheta)$ are the deflections of wall at the time $t$ and $\vartheta$, respectively. Between $E_{di}(\vartheta)$ and $W_{0i}$ there exists the relationship:

$$E_{di}(\vartheta) = W_{0i}(\alpha^2\alpha'^2 + \beta^2\beta'^2 + \gamma^2\gamma'^2) \tag{8}$$

where $\alpha$, $\beta$, $\gamma$ and $\alpha'$, $\beta'$, $\gamma'$ are the direction cosines for spontaneous magnetization vector just after demagnetization and at the time $\vartheta$; the cosines are calculated in a coordinate system with axes parallel to the direction $\langle 100 \rangle$. Energy $W_{0i}$ of the $i$-th relaxation depends on concentration of relaxation centers $C_i$, on absolute temperature $T$ and on the Boltzmann constant $k$ in the following way:

$$W_{0i} = \frac{w_i^2 C_i}{3kT}.$$

Thus, in the case of flat wall 180°(100) and field parallel to $I_s$ the equations of motion has the form:

$$\frac{2I_sH(t)}{\alpha}-u(t)-\sum_{i=1}^{n}\frac{\eta_i}{\Theta_i}\int_0^t[u(t)-u(\vartheta)]\exp\left(-\frac{t-\vartheta}{\Theta_i}\right)d\vartheta=0 \tag{9}$$

where

$$\eta_i=\frac{pW_{0i}}{\alpha d}=\frac{pw_i^2C_i}{3d\alpha kT}.$$

### *3. Solution of equation of motion for the phenomenon of disaccommodation of magnetic susceptibility*

In general, Eq. (9) for single relaxation process is the integral equation of Volterra of the second kind. In the case of measurement of disaccommodation of magnetic susceptibility with the help of the ballistic method Eq. (9) becomes an ordinary equation, because $u(\vartheta)=0$. If we assume that the time needed for deflection of a ballistic galvanometer spot is very small compared to the time of measurement $t$, the solution of equation (9) has the form

$$u(t)=\frac{2I_sH}{\alpha}\cdot\frac{1}{1+\sum_{i=1}^{n}\eta_i\left[1-\exp\left(-\frac{t}{\Theta_i}\right)\right]}. \tag{10}$$

Due to the fact that the magnetic susceptibility is proportional to the wall shift $u$, the inverse of magnetic susceptibility is given by formula:

$$\frac{1}{\chi(t)}=\frac{1}{\chi_0}+\sum_{i=1}^{n}\frac{1}{\chi_i}\left[1-\exp\left(-\frac{t}{\Theta_i}\right)\right]. \tag{11}$$

where $\chi_0$ is the magnetic susceptibility of the time $t=0$ and $\frac{1}{\chi_i}=\frac{\eta_i}{\chi_0}$ denotes the intensity of the $i$-th relaxation. Then in turn, since the susceptibility is given by: $\chi_0=\frac{4J_s^2}{\alpha^l}$ [12, 13] where $l$ is the cell thickness, the intensity of the $i$-th relaxation is equal:

$$\frac{1}{\chi_i}=\frac{pw_i^2lC_i}{12dkTI_s^2}. \tag{12}$$

As it is seen from formula (12) the intensity of the $i$-th relaxation is proportional directly to the number of relaxation centers in 1 cm³ ($C_i$) and inversely to the absolute temperature. The intensity of relaxation can be expressed in the following form

$$\frac{1}{\chi_i} = f_i(T)C_i \tag{13}$$

where

$$f_i(T) = \frac{pw_i^2 l}{12kTdI_s^2}\,. \tag{13a}$$

In the case when one measures the disaccommodation of magnetic susceptibility using the method of alternating current, in solution of integral equation (9) there appear additional terms connected with each relaxation process. One should estimate each of them, as they can influence the final result. With this aim let us write equation (9) in the following form:

$$u(t) = \frac{2I_s H(t)}{\alpha} \cdot \frac{1}{1+\sum\limits_{i=1}^{n} \eta_i \left[1-\exp\left(-\frac{t}{\Theta_i}\right)\right]} +$$

$$+\sum_{i=1}^{n} \frac{\eta_i}{1+\sum\limits_{k=1}^{n} \eta_k \left[1-\exp\left(-\frac{t}{\Theta_k}\right)\right]} \int_0^t \Theta_i u(\vartheta) \exp\left(-\frac{t-\vartheta}{\Theta_i}\right) d\vartheta. \tag{14}$$

To facilitate the estimation of each individual term we shall solve Eq. (14) with the help of the Liouville-Neumann method of series [14]. In the case of sinusoidal elementary field applied $H = H_m \sin \omega t$, we assume the solution in the form of the following series:

$$u(t) = \sum_{n=0}^{\infty} \lambda^n u_n(t) \tag{15}$$

where

$$\lambda = \sum_{i=1}^{n} \lambda_i\,. \tag{15a}$$

Substituting (15) into (14) and comparing terms corresponding to the equal powers of $\lambda$ we obtain individual terms of the series. For the first one we get

$$u_0(t) = \frac{2I_s H_m \sin \omega t}{\alpha} \cdot \frac{1}{1+\sum\limits_{i=1}^{n} \eta_i \left[1-\exp\left(-\frac{t}{\Theta_i}\right)\right]}\,. \tag{16}$$

While measuring the disaccommodation of magnetic susceptibility in an alternating field, the relaxation times $\Theta_i$ are greater than 15s and the applied frequency is of the order of 1000 Hz. Thus further terms in phase with the applied field can be neglected as they are about $10^{10}$ times smaller than (16). So, a very good approximation to the solution represents the term $u_0(t)$ and the magnetic susceptibility can be expressed by the formula (11).

### *4. Solution of equation of motion for the tangent of the relaxation loss angle*

The formula for the tangent of angle of phase shift between the vectors of magnetic induction and of magnetic field intensity can be obtained from the stationary solution of Eq. (9). For the fixed conditions we have $t \gg 0$, $\exp(-t/\Theta_i) \to 0$ and formula (14) takes the form:

$$u(t) = \frac{2I_s H(t)}{\alpha} \frac{1}{1+\sum\limits_{k=1}^{n} \eta_k} + \sum_{i=1}^{n} \frac{\eta_i}{1+\sum\limits_{k=1}^{n} \eta_k} \int_0^t u(\vartheta) \exp\left(-\frac{t-\vartheta}{\Theta_i}\right) \Theta_i^{-1} d\vartheta. \tag{17}$$

Let us assume that the applied field is a periodic function of the form $H = H_m \exp(j\omega t)$. Substituting the series (15) into Eq. (17) and comparing terms with equal powers of $\lambda$ we get for the $n$-th term the following expression:

$$u_n(t) = \frac{2I_s H_m \exp(j\omega t)}{\alpha(1+\sum\limits_{i=1}^{n} \eta_i)} \cdot \left(\sum_{i=1}^{n} \frac{1}{1+j\omega\Theta_i}\right)^n. \tag{18}$$

Thus, the amplitude for the wall shift is given by

$$u_m = \frac{2I_s H_m}{\alpha} \cdot \frac{1}{1+\sum\limits_{i=1}^{n} \frac{\eta_i j\omega\Theta_i}{1+j\omega\Theta_i}}, \tag{19}$$

and the tangent of the relaxation loss angle by

$$\operatorname{tg} \delta_r = \frac{4\pi\chi_0' \sum\limits_{i=1}^{n} \frac{\eta_i\omega\Theta_i}{1+\omega^2\Theta_i^2}}{M+4\pi\chi_0' \left[1+\sum\limits_{i=1}^{n} \frac{\eta_i\omega\Theta_i}{1+\omega^2\Theta_i}\right]} \tag{20}$$

where

$$M = \left[1+\sum_{i=1}^{n} \frac{\eta_i\omega^2\Theta_i^2}{1+\omega^2\Theta_i^2}\right]^2 + \left[\frac{\eta_i\omega\Theta_i}{1+\omega^2\Theta_i^2}\right]^2. \tag{20a}$$

In the above expression $\chi_0'$ is the magnetic susceptibility for angular frequency $\omega \to 0$.

For Snoek relaxation for carbon and nitrogen in αFe and for additional relaxations in αFe–X–C(N) one has: $\eta_i < 1$, $4\pi\chi_0' \gg 1$, $M/4\pi\chi_0' \ll 1$. Dividing the expression (20) by $4\pi\chi_0'$, one gets as a good approximation:

$$\operatorname{tg}\delta_r \approx \frac{\sum_{i=1}^{n} \frac{\eta_i \omega \Theta_i}{1+\omega^2\Theta_i^2}}{1+\sum_{k=1}^{n} \frac{\eta_k \omega^2 \Theta_k^2}{1+\omega^2\Theta_k^2}}. \tag{21}$$

Formula (21) can be also expressed in the form

$$\operatorname{tg}\delta_r = \sum_{i=1}^{n} A_i \operatorname{tg}\delta_{ri} \tag{22}$$

where

$$\operatorname{tg}\delta_{ri} = \frac{\omega\Theta_i}{1+\omega^2\Theta_i^2}, \tag{22a}$$

$$A_i = \frac{\eta_i}{1+\sum_{k=1}^{n} \frac{\eta_k \omega^2 \Theta_k^2}{1+\omega^2\Theta_k^2}}. \tag{22b}$$

One can see from Eqs (22), (22a) and (22b) that in the case when $\eta_i$'s are small compared to unity, the tangent of the total relaxation loss angle is the sum of single partial tangents.

## 5. *Discussion*

In the case of a single time constant, the solutions (11) and (20) for disaccommodation of magnetic susceptibility and for the tangent of the relaxation loss angle get transferred into the expressions obtained in Refs [2] and [15]. It is seen from Eq. (11) that for several discrete relaxation processes the phenomenon of disaccommodation of magnetic susceptibility is consistent with the superposition rule. Total disaccommodation is the sum of partial disaccommodations connected with particular relaxation processes. The intensity of a particular relaxation (formula (12)) is directly proportional to the concentration of relaxation centers $C_i$ responsible for its initiation. It is worth mentioning that these quantities do not depend on the reactive force coefficient $\alpha$. This is of great importance as *e.g.* in the interstitial solutions, the concentrations of relaxators can be decreased by precipitation of the solute from supersaturated solution. As a rule this process leads to the change of the coefficient $\alpha$.

It can be seen from Eq. (20) that the tangent of the total relaxation loss angle is not a superposition of partial tangents corresponding to the particular elementary relaxations.

In the case of magnetic measurements, the partial tangents attain considerable values. *E.g.* in Ref. [16] it was shown, that for the αFe–C alloy containing 0.007% of C we have $\text{tg}\,\delta_{r\,\max} = 0.3$. This value corresponds to the parameter $\eta = 0.72$ in accordance with the considerations presented in Ref. [1]. It is then seen that the analysis of complex relaxation phenomena is practically impossible with the help of measurements of the tangent of relaxation loss angle. Similar situation can be observed in the case of measurements of internal friction, where the curve of total attenuation cannot be represented as the sum of Debye curves, as it was shown in Ref. [17]. The above discussion shows that for the investigation of complex relaxation processes a basic role is played by the curves of disaccommodation of magnetic susceptibility.

In practice, one uses most frequently the isothermic curves of disaccommodation with the help of which one can find the time constants and relaxation intensities, applying the method given by Bosman and coworkers [18]. One can also use the isochronic curves of disaccommodation, analysing them with the help of method given by Seeger and Rieger [19]. From Eq. (11) follows that the difference of the inverse magnetic susceptibilities found at the times $t_1$ and $t_2$ after the disaccommodation was started is equal

$$\frac{1}{\chi(t_1)} - \frac{1}{\chi(t_2)} = \sum_{i=1}^{n} \frac{1}{\chi_i} \left[ \exp\left(-\frac{t_2}{\Theta_i}\right) - \exp\left(-\frac{t_1}{\Theta_i}\right) \right]. \tag{23}$$

It is seen that curve $\frac{1}{\chi(t_1)} - \frac{1}{\chi(t_2)} = f(T)$ is the sum of curves corresponding to the particular elementary relaxations. Intensity of particular elementary process can be expressed in terms of the heights of partial curves $A_i$ in the following way

$$\frac{1}{\chi_i} = \frac{A_i}{\left[ \exp\left(-\frac{t_2}{\Theta_i}\right) - \exp\left(-\frac{t_1}{\Theta_i}\right) \right]_{\max}}. \tag{24}$$

If we assume that the relaxation intensities do not depend on temperature, the maxima of partial curves are observed at the temperatures $T_i$ for which the relaxation times fulfil the condition:

$$\Theta_i = \frac{t_2 - t_1}{\ln \frac{t_2}{t_1}}. \tag{25}$$

In the case when a given relaxation is caused by clusters of interstitial or substitutional and interstitial atoms, the intensities of these relaxations depend strongly on temperature. *E.g.* the relaxation intensity caused by pairs of interstitial atoms is given by the expression [8, 20]:

$$\frac{1}{\chi_p} = \text{const} \cdot f_p(T) \exp\left(\frac{\Delta H_p}{RT}\right) C_s^2 \tag{26}$$

where $C_s$ is the concentration of single interstitial atoms and $\Delta H_p$ denotes the pair binding energy. For relaxation caused by a cluster of the type X–AM, the intensity changes with temperature according to the dependence:

$$\frac{1}{\chi_{X-AM}} = \text{const} \cdot C_X C_{AM} f(T) \exp\left(\frac{\Delta H_{X-AM}}{RT}\right) \tag{27}$$

where $C_x$ denotes the concentration of substitutional atoms, $C_{AM}$ — concentration of interstitial atoms and $\Delta H_{X-Am}$ is the cluster binding energy.

Isothermic and isochronic curves of disaccommodation of magnetic susceptibility were successfully applied for the analysis of binary αFe–C(N) alloys [2, 21] and of triple alloys αFe–Al–C [3–5] and αFe–V–C [6]. In the case of αFe–0.1% Al–C alloy three single time constant elementary relaxation (I, II and III) were obtained from the distribution of isothermic curves. Relaxation I corresponds to clusters of the type Al–C and their binding energy is estimated to be equal to 5200 cal/mole. Relaxation II is the carbon Snoek relaxation. Process III is caused by the pairs of carbon atoms. C–C carbon pair binding energy is equal to 2600 cal/mole. For the αFe–0.5% V–C alloy, the isochronic curves for the inverse magnetic susceptibility can be splitted into the three elementary curves.

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