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A phenomenological model of magnetisation kinetics

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Abstract. A magnetic equation of state or magnetic constitutive equation, in functional form $H = H(M, \dot{M})$, has been developed by analogy with a mechanical constitutive equation of crystal plasticity. It is shown that magnetisation kinetics, i.e. the dependence of magnetisation on magnetic field and time, may be conveniently described in terms of solutions of the magnetic constitutive equation subject to given magnetic field constraints.

Two experimental methods of investigating the phenomenon of magnetic viscosity, in which time variations of intensity of magnetisation are observed, are analysed. The results, from which magnetic viscosity parameters and hence values of the activation volumes involved in irreversible magnetisation processes are determined, are identical with the expressions previously derived in terms of activation energy distributions. The definitions of the magnetic susceptibilities involved in the earlier work are arbitrary; they are precisely defined in the present work.

1. Introduction

The mechanisms responsible for the magnetisation kinetics and the coercivity of ferromagnetic materials are formally similar to the mechanisms that govern the plasticity and strength of solid solutions or two-phase alloys (Haasen 1972). The holomagnetisation of a magnetic material depends on the applied magnetic field and on time, and results from reversible and irreversible movement of domain boundary walls and spontaneous magnetisation vectors.

Néel (1946) developed a general theory of coercivity by considering the constraints imposed on domain wall motion by lattice defects caused by internal stress and inclusions. Stoner and Wohlfarth (1948) showed that high coercivities are exhibited by assemblies of non-interacting single-domain particles due to the inhibiting effect of shape or magnetocrystalline anisotropies on the rotation of domain magnetisation vectors. All magnetic materials exhibit magnetic viscosity—the time dependence of intensity of magnetisation under constant applied field conditions. Preisach (1935), Néel (1949) and Street and Woolley (1949) discussed magnetic viscosity in terms of the thermal activation of magnetisation processes.

The holistic mechanical analogues of the reversible and irreversible changes in intensity of magnetisation are respectively the elastic and plastic components of strain. The latter component is related at the microscopic level to the motion of dislocations.

Siemers and Nembach (1979) developed this analogy and considered a model of thermally activated domain wall mobility using a number of plausible potential functions to describe the interaction of a domain wall with pinning centres. A more elaborate theory taking into account the statistical distribution of pinning centres and the flexibility of domain boundary walls was developed by Gaunt (1983) by analogy with statistical theories of dislocation motion through a random array of obstacles.

The theories of Seimers and Nembach, Gaunt and others led to satisfactory descriptions of the motion of individual domain boundary walls. However, a further step is required to predict the macroscopic magnetisation kinetics as a summation of collective domain wall behaviour. We show in this paper that the procedures already well established to account for macroscopic mechanical stress/strain observations may also be applied by analogy to describe macroscopic magnetisation kinetics.

In dislocation theory extension from the microscopic to the macroscopic level of description is achieved by utilising the Orowan relation between the plastic strain rate $\dot{\epsilon}$ and the average individual dislocation velocity, v_d :

$$\dot{\epsilon} = \varphi b \rho v_d \quad (1)$$

where b and ρ denote respectively the magnitude of the Burgers vector and the density of mobile dislocations, and φ is a geometrical factor. When the deformation rate is limited by the rate of dislocation generation, $\dot{\rho}$, the product ρv_d in (1) may be replaced by $\dot{\rho}L$, where L is the dislocation mean free path (Mecking and Lücke 1970).

The dislocation velocity, v_d , or $\dot{\rho}$ may be represented in the form of an Arrhenius relation which leads to

$$\dot{\epsilon} = \dot{\epsilon}_0 \exp(-\Delta G(\sigma)/kT) \quad (2)$$

$\Delta G(\sigma)$ equals the Gibbs free energy of activation which results from a complex averaging over many thermally activated events; σ is the stress acting on the dislocations, T is the absolute temperature, k equals the Boltzmann constant and $\dot{\epsilon}_0$ is a pre-exponential coefficient which is essentially independent of stress and temperature. In general $\Delta G(\sigma)$ cannot be calculated *ab initio* and has to be considered as some phenomenological average that depends on the state of the material, i.e. it may implicitly depend on some internal variable or variables.

Macroscopic plastic deformation under various conditions of straining is very conveniently described in terms of constitutive relations, or mechanical equations of state. It is assumed that there exists a unique relation between such mechanical quantities as plastic strain rate, stress and the plastic strain, which is independent of previous loading history.

In differential form (Hart 1967) this relation may be written as

$$d\sigma = \nu d\epsilon + \lambda d \ln \dot{\epsilon} \quad (3)$$

where $\lambda = (\partial \sigma / \partial \ln \dot{\epsilon})_{\epsilon}$ is the strain rate sensitivity (SRS) of the stress at constant plastic strain and $\nu = (\partial \sigma / \partial \epsilon)_{\dot{\epsilon}}$ is the strain hardening rate. The SRS characterises the instantaneous mechanical response of a material and the strain hardening rate represents the evolution of mechanical strength with plastic strain.

From equation (2)

$$\lambda = kT/V^* \quad (4)$$

where $V^* = (-\partial(\Delta G)/\partial \sigma)_{\dot{\epsilon}}$ is the activation volume.

Values of λ may be derived from the results of experiments involving rapid changes in strain rate and ν -values may be calculated from the results of experiments carried out under constant strain rate conditions. In general both ν and λ are found to be functions of σ and $\dot{\epsilon}$. The mechanical behaviour of a specimen under various modes of straining, in particular under constant stress conditions (creep), may be predicted from the functional dependences of ν and λ (Mecking *et al* 1988).

2. The magnetic constitutive relation

The development, by analogy with the mechanical case outlined above, of a magnetic equation of state or constitutive relation is based on the assumption that there is a unique relationship between magnetic field, magnetisation and rate of change of magnetisation with time. The magnetic field acting within a specimen H_i is related to the applied magnetic field H_a by the expression

$$H_i = H_a - DM_{\text{tot}} \quad (5)$$

where D equals the demagnetisation factor of the specimen and M_{tot} is the total intensity of magnetisation. H_i and M_{tot} are uniform within specimens in the form of ellipsoids of revolution.

The total intensity of magnetisation has reversible and irreversible components, M_{rev} and M_{irr} , respectively (Street *et al* 1952). This distinction parallels the division of total strain into (reversible) elastic and (irreversible) plastic parts. It is the plastic strain and its time derivative that enter the mechanical constitutive equation (equation (3)). Analogy with plastic deformation requires the magnetic constitutive relation to be expressed in terms of the *irreversible* intensity of magnetisation, M_{irr} , and its time derivative, \dot{M}_{irr} :

$$H_i = H_i(M_{\text{irr}}, \dot{M}_{\text{irr}}) \quad (6)$$

or, in differential form,

$$dH_i = (1/\chi_{\text{irr}}^i) dM_{\text{irr}} + \Lambda d(\ln \dot{M}_{\text{irr}}) \quad (7)$$

where

$$1/\chi_{\text{irr}}^i = (\partial H_i / \partial M_{\text{irr}}) |_{\dot{M}_{\text{irr}}} \quad (8)$$

and

$$\Lambda = [\partial H_i / \partial (\ln \dot{M}_{\text{irr}})] |_{M_{\text{irr}}} \quad (9)$$

χ_{irr}^i is the intrinsic irreversible magnetic susceptibility (i.e. corrected for demagnetisation effects) determined at constant time rate of change of intensity of magnetisation. Λ gives the dependence of the rate of change of intensity of magnetisation on internal field and may be determined by measuring the change in \dot{M} ($\dot{M}_1 \rightarrow \dot{M}_2$) produced by a change in H_i from H_1 to H_2 from

$$\Lambda = (H_2 - H_1) / \ln(\dot{M}_2 / \dot{M}_1). \quad (10)$$

As in the mechanical case Λ may be related to the activation volume involved in the process of magnetisation. The individual domain wall velocity and the rates of nucleation

of domains of reverse magnetisation and of unpinning domain walls from pinning centres are controlled by an Arrhenius relation such that

$$\dot{M}_{\text{irr}} = \dot{M}_0 \exp(-\Delta G(H_i)/kT). \quad (11)$$

Combining equations (9) and (11) yields:

$$\Lambda = -kT/(\partial \Delta G(H_i)/\partial H_i)|_{M_{\text{irr}}}. \quad (12)$$

As in the mechanical case $\partial \Delta G(H_i)/\partial H_i$ is related to v , the volume involved in the activation process (Siemers and Nembach 1979, Street et al 1987) and may be written as

$$q = -(\partial \Delta G(H_i)/\partial H_i)|_{M_{\text{irr}}} = \psi M_s v \quad (13)$$

where ψ is a numerical factor dependent on the type and geometry of the microscopic magnetisation process involved and M_s is the spontaneous magnetisation of the material.

3. Applications

The constitutive relation (7) may be applied to describe magnetisation behaviour under various imposed conditions. As an example we consider the phenomenon of magnetic viscosity and the results of the analyses of the time dependence of magnetisation under various applied field conditions given by Street and Woolley (1949), Street et al (1952) and Street et al (1987).

When the magnetic field applied to a ferromagnet is changed discontinuously at time $t = 0$ the observed variation of intensity of magnetisation varies approximately as

$$M(t) = \text{constant} + S(D) \ln t \quad (14)$$

when $H_a = \text{constant}$ for $t > 0$. The coefficient $S(D)$ has been shown to be a function *inter alia* of the demagnetisation factor, D , of the specimen (Street et al 1952, Barbier 1954).

The constitutive relation (7) may be written for constant H_a conditions (i.e. $\dot{H}_a = 0$) as:

$$\dot{H}_i = (1/\chi_{\text{irr}}^i) \dot{M}_{\text{irr}} + \Lambda \ddot{M}_{\text{irr}}/\dot{M}_{\text{irr}} \quad (15)$$

with

$$\dot{H}_i = -D \dot{M}_{\text{tot}} \quad (16)$$

(from equation (5)). Now

$$\dot{M}_{\text{irr}} = \dot{M}_{\text{tot}} - \dot{M}_{\text{rev}} = \dot{M}_{\text{tot}} - \chi_{\text{rev}}^i \dot{H}_i = \dot{M}_{\text{tot}}(1 + D\chi_{\text{rev}}^i) \quad (17)$$

where χ_{rev}^i is the reversible component of intrinsic susceptibility. Substituting from (16) and (17) in (15) gives

$$[(1 + D\chi_{\text{tot}}^i)/\chi_{\text{irr}}^i] \dot{M}_{\text{tot}} + \Lambda \ddot{M}_{\text{tot}}/\dot{M}_{\text{tot}} = 0 \quad (18)$$

using the relation $\chi_{\text{tot}}^i = \chi_{\text{rev}}^i + \chi_{\text{irr}}^i$. Equation (18) may be integrated, assuming the susceptibilities and Λ are independent of M during a magnetic viscosity experiment, to give

$$\dot{M}_{\text{tot}} = \dot{M}_{\text{init}}/(1 + K\dot{M}_{\text{init}}t) \quad (19)$$

and

$$M_{\text{tot}} = M_{\text{init}} + (1/K) \ln(1 + KM_{\text{init}}\dot{M}) \quad (20)$$

where $K = (1 + D\chi_{\text{tot}}^i)/\Lambda\chi_{\text{irr}}^i$. For values of $t \gg 1/K\dot{M}_{\text{init}}$ equation (20) becomes

$$M_{\text{tot}} = \text{constant} + (1/K) \ln t. \quad (21)$$

Comparison of equations (21) and (14) shows that $S(D) = \Lambda\chi_{\text{irr}}^i/(1 + D\chi_{\text{tot}}^i)$ or, transforming to directly observable susceptibilities, χ^a (measured at constant \dot{M}_{irr}), using expressions of the form $\chi^i = \chi^a/(1 - D\chi^a)$ gives

$$S(D) = \Lambda(\chi_{\text{tot}}^a - \chi_{\text{rev}}^a)/(1 - D\chi_{\text{rev}}^a). \quad (22)$$

Equation (22) is the same expression as that derived by Street *et al* (1987) utilising a different model for describing magnetic viscosity.

Equation (22) (Street *et al* 1987, Shi *et al* 1987) and a truncated form omitting the $(1 - D\chi_{\text{rev}}^a)$ factor (Givord *et al* 1987, Street *et al* 1952) have been used extensively to calculate Λ and hence the activation volumes (equations (12) and (13)). However, the susceptibilities have not been precisely defined. The derivation given above of equation (22) shows that the appropriate values of the susceptibilities are those obtained from magnetisation versus magnetic field curves taken at constant \dot{M}_{irr} .

Street *et al* (1952) obtained an expression for Λ in terms of parameters observed during field jump experiments in which the applied field is changed discontinuously by ΔH_a at a time $t_1 \gg S/\dot{M}_{\text{init}}$ during a magnetic viscosity experiment.

We denote the time variation of magnetisation for times $t < t_1$ by $\dot{M}_1(t)$ and that for $t > t_1$ by $\dot{M}_2(t)$. Equation (19) may be written as

$$\dot{M}_2 = S/[S/\dot{M}'_{\text{init}} + (t - t_1)] \quad (23)$$

where \dot{M}'_{init} is the initial value of \dot{M}_2 at $t = t_1$. Equation (23) may be transformed to $\dot{M}_2 = S/(t_0 + t)$ defining t_0 as $S/\dot{M}'_{\text{init}} - t_1$. Thus the rate of change of magnetisation immediately after ΔH_a is applied is

$$\dot{M}_2 = S/(t_0 + t_1). \quad (24)$$

The rate of change of magnetisation immediately before ΔH_a occurs is

$$\dot{M}_1 = S/t_1. \quad (25)$$

Substituting from equations (25) and (24) in (10) gives $\Lambda = \Delta H_a/\ln[t_1/(t_1 + t_0)]$ which is exactly the result derived by Street *et al* (1952).

As an illustration of the above analysis we have calculated Λ and χ_{irr} from magnetic viscosity data obtained under constant H_a conditions by Street *et al* (1989) using a sample of hot-pressed isotropic NdFeB. The $(M_{\text{irr}})_{\dot{M}_{\text{irr}}}$ versus H_i curves shown in figure 1 were constructed from these data. An example of the variation of $\chi_{\text{irr}}^i|_{\dot{M}}$ versus M_{irr} is shown in figure 2. Two sets of data corresponding to two rates \dot{M}_{irr} differing by a factor of 40 are plotted on this graph. The two sets lie on a common curve, indicating that $\chi_{\text{irr}}^i|_{\dot{M}}$ is a function of M_{irr} only and is independent of \dot{M}_{irr} . In contrast, $\chi_{\text{irr}}^i|_t$ versus H_i curves depend on t (see, e.g., Street *et al* 1987, Viadieu *et al* 1989). However, when plotted as a function of M_{irr} all values of $\chi_{\text{irr}}^i|_t$ lie on a common curve independent of t .

Values of Λ are plotted as a function of M_{irr} in figure 3. Also shown in this figure are values of $\Lambda (= kT/q)$, from equations (12) and (13) above, calculated from the same data, using equation (22) with χ_{tot}^a computed from $M_{t=30s}$ versus H_a curves. With this choice of constant time the two sets of variations in Λ are in good agreement. However, the agreement becomes worse for choices of constant time of say 10 s or 100 s. The good

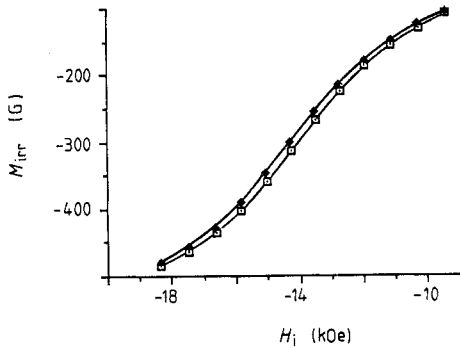


Figure 1. M_{irr} versus H_i for two different rates of change of M_{irr} (in $G s^{-1}$): \blacklozenge , 1.0; \square , 0.025.

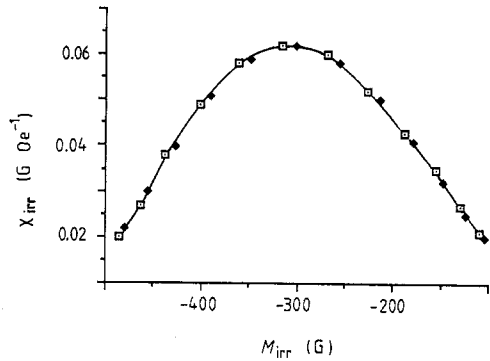


Figure 2. χ_{irr} versus M_{irr} for two different rates of change of M_{irr} (in $G s^{-1}$): \blacklozenge , 1.0; \square , 0.025.

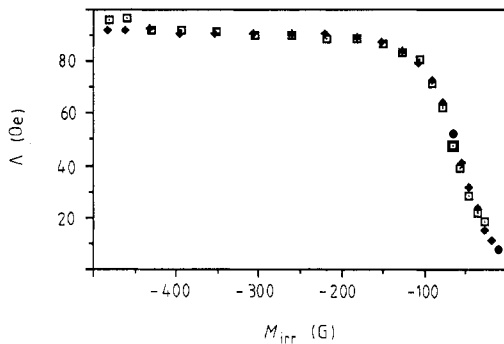


Figure 3. The variation of Λ as a function of M_{irr} : \square , calculated using equation (11); \blacklozenge , calculated using equation (24).

agreement between the two methods for $t = 30$ s is due to the dependence on M_{irr} of S (Street *et al* 1987), i.e. the M_{irr} versus $\ln(t)$ curves are not strictly linear. As a consequence, the value of S calculated from a particular curve is associated with the mean of the range of relaxation times used in the experiment, which was 30 s (Street *et al* 1987). Of course this problem does not arise in the direct determination of Λ from equation (10). These effects will be discussed more fully elsewhere.

4. Conclusions

We have shown that the postulate of a magnetic constitutive equation of the functional form $H_i = H_i(M_{irr}, \dot{M}_{irr})$, analogous to a mechanical constitutive equation of crystal plasticity, makes it possible to predict magnetisation kinetics under diverse imposed magnetic field conditions. From this it is possible to define experimental procedures involving the traversal of magnetisation curves at constant \dot{M} from which values of the quantity Λ may be determined directly. The magnitudes of the activation volumes that are involved in irreversible magnetisation processes at each point of a magnetisation curve may then be measured.

The response of the intensity of magnetisation to other applied magnetic field regimes, e.g. involving continuous or alternating variations, may also be analysed using the magnetic constitutive equation.

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