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1990 J. Phys.: Condens. Matter 2 3681

(http://iopscience.iop.org/0953-8984/2/15/023)

J. Phys.: Condens. Matter 2 (1990) 3681–3685. Printed in the UK

LETTER TO THE EDITOR

Unstable magnetisation processes

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Received 14 February 1990

Abstract. The description of magnetisation kinetics in terms of a magnetic constitutive equation has been extended to examine the instability (localisation) of magnetisation. It is shown that there are two separate instability conditions. One requires the magnetisation curve to have regions of negative susceptibility (a re-entrant loop), the other requires anomalous values of the magnetisation rate dependence of the magnetic field.

Unstable magnetisation behaviour was observed extensively by Sixtus and Tonks (1931) who associated the presence of large Barkhausen discontinuities in stressed Fe–Ni wires with localisation of reversed magnetisation. By applying a small local additional field near one end of an otherwise uniformly magnetised wire, it was found possible to initiate and propagate a localised band of reversed magnetisation along the length of the wire. The velocity of band propagation was a linearly increasing function of applied field.

Sixtus and Tonks related the unstable behaviour to the squareness of the magnetisation curve. As discussed by Fiorillo (1986) and others, the observation of a square hysteresis loop in measurements under conditions of magnetic field control may be a misleading artefact of the measuring system and is due to a rapid uncontrolled increase in the rate of change of magnetisation. In such cases, measurements of hysteresis loops under conditions of constant rate of change of intensity of magnetisation or magnetic induction result in re-entrant loop behaviour. While the re-entrant behaviour is clearly indicative of unstable magnetisation, relatively few studies (Damento and Demer 1987, Glaister and Viney 1965) have been directly concerned with spatial localisation of magnetisation reversal since the original measurements of Sixtus and Tonks.

In plasticity the occurrence of re-entrant behaviour in stress–strain curves is referred to as yield drop behaviour which is associated with localisation of plastic flow into one or more deformation bands which propagate along the specimen at a steady state velocity. This is an almost exact analogue of the propagation of a region of reversed magnetisation investigated by Sixtus and Tonks.

Criteria for the localisation of plastic flow which are based on linear stability analysis using mechanical constitutive equations are well established (Hart 1967, Estrin and Kubin 1988). An analogous analysis of the conditions required for localisation of magnetisation does not appear to have been carried out. A constitutive model of magnetisation kinetics has recently been developed (Estrin *et al* 1989, Street *et al* 1989) which provides a function relation between the field, H, acting on a magnetic material, the intensity of magnetisation M and time rate of change of magnetisation $\dot{M}$, which is necessary for a stability analysis to be carried out. In this communication we use this phenomenological model of magnetisation kinetics to derive criteria for localisation of magnetisation in magnetic materials.

Consider a specimen in the form of an ellipsoid of revolution placed in a uniform external field which acts along a principal axis of the ellipsoid. If evaluated over sufficiently large volume elements, the mean values of the intensity of magnetisation M and the internal field H_i will be uniform throughout the specimen. The existence of a domain structure and the polycrystalline or particulate nature of a material will cause considerable variations in M and H_i when computed in volume elements comparable in size to domain and particle volumes. In the following discussion we consider local elements of volume which are sufficiently large for averaging over many domains to be admissible, i.e. it is assumed that the local mean values of M and H_i differ little from the mean values of intensity of magnetisation and internal field computed for the specimen as a whole.

The stability of a magnetic state may be determined by examining if a fluctuation of M within a volume element increases or decreases as a function of time. The magnetic constitutive equation in functional form (Estrin *et al* 1989) is

$$H_i = H_i(M_{irr}, \dot{M}_{irr}) \tag{1}$$

where M_{irr} and $\dot{M}_{irr}$ are the irreversible intensity of magnetisation and its time derivative, respectively.

$$\delta H_i = 1/\chi^i_{irr}\delta M_{irr} + \Lambda\delta \ln \dot{M}_{irr} \tag{2}$$

where

$$\chi^i_{irr} = \partial M_{irr}/\partial H_i|_{\dot{M}_{irr}} \tag{3}$$

and

$$\Lambda = \partial H_i/\partial \ln \dot{M}_{irr}|_{M_{irr}}. \tag{4}$$

In the following analysis the generally complex relation between δH_i and δM_{irr} is simplified by assuming that:

(i) H_i and M_{irr} are in the same direction (which was assumed in writing equation 2);

(ii) the local volume in which δM_{irr} occurs has a shape and orientation with respect to H_i so that the effect of discontinuities in M at the boundaries of the local volume may be expressed in terms of a demagnetising factor, d.

δH_i may then be expressed as

$$\delta H_i = -d(\delta M_{irr} + \delta M_{rev}) \tag{5}$$

where δM_{rev} is the incremental change in the reversible component of intensity of magnetisation which accompanies the incremental change of internal field δH_i and

$$\delta M_{rev} = \chi^i_{rev}\delta H_i \tag{6}$$

where χ^i_{rev} = intrinsic reversible susceptibility.

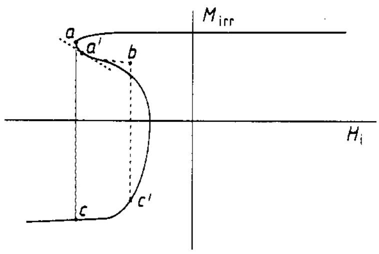

Figure 1. Re-entrant M, H curve under constant $\dot{M}$ constraint. The full curve is an idealised constant $\dot{M}$ curve which would be expected in the absence of localisation. The branch ac will be followed when magnetisation takes place under constant $\dot{H}_i$ conditions. The branch bc' will be observed when localisation occurs under conditions of constant average $\dot{M}$. Points a and a' correspond to the onset of localisation where $\chi^{i}_{irr} \to \infty$ (i.e. $d = 0$) and $\chi^{i}_{irr} = -(1/d + \chi^{i}_{rev})$ respectively.

Substituting (6) into (5) gives

$$\delta H_i = [-d/(1 + d\chi^{i}_{rev})]\delta M_{irr} \tag{7}$$

and from (2) and (7)

$$\delta \dot{M}_{irr}/\delta M_{irr} = -(\dot{M}_{irr}/\Lambda)[1/\chi^{i}_{irr} + 1/(1/d + \chi^{i}_{rev})]. \tag{8}$$

Equation (8) has solutions of the form

$$\delta M_{irr} = (\delta M_{irr})_0 \exp(\lambda t)$$

where

$$\lambda = -(\dot{M}_{irr}/\Lambda)[1/\chi^{i}_{irr} + 1/(1/d + \chi^{i}_{rev})]. \tag{9}$$

If $\lambda < 0$ a small local perturbation δM_{irr} will decay whereas if $\lambda > 0$ the perturbation will grow in time. The instability criterion $\lambda > 0$ gives rise to two instability conditions:

$$\text{(i)} \qquad \dot{M}_{irr}/\Lambda > 0 \qquad 1/\chi^{i}_{irr} < -1/(1/d + \chi^{i}_{rev}) \tag{10}$$

$$\text{(ii)} \qquad \dot{M}_{irr}/\Lambda < 0 \qquad 1/\chi^{i}_{irr} > -1/(1/d + \chi^{i}_{rev}). \tag{11}$$

The instability conditions (10) and (11) are applicable to all points on the magnetisation curves.

For virgin magnetisation and the ascending branch of a hysteresis loop $\dot{M}_{irr} > 0$ whereas for the descending branch $\dot{M}_{irr} < 0$.

The instability condition (i) of equation (10) requires a negative intrinsic irreversible susceptibility. Magnetisation curves obtained under conditions of constant $\dot{M}_{irr}$ for which $\chi^{i}_{irr} < 0$ exhibit re-entrant characteristics with a region where M_{irr} decreases with increasing H_i, shown diagrammatically in figure 1.

From (10) it follows that the onset of instability depends on the demagnetising factor, d, and hence on the shape of the volume in which the perturbation δM_{irr} occurs. If conditions allow homogeneous fluctuations in long thin cylindrical volumes for which $d \to 0$ the onset of localisation of magnetisation will occur at point a of the curve in figure 1 where $\chi^{i}_{irr} \to \infty$. However if $d \neq 0$ the onset of localisation will be delayed to point a', at which the slope is $-(1/d + \chi^{i}_{rev})$.

As the region of localised magnetisation grows it is impossible to maintain conditions of constant $\dot{M}_{irr}$ uniform over the entire specimen and the observed magnetisation curve will depend on the degree of localisation and the experimental conditions under which the measurements are made. An idealised curve appropriate to constant uniform $\dot{M}_{irr}$ conditions (i.e. in the absence of localisation) is indicated in figure 1. The experimentally observed constant $\dot{M}_{irr}$ curve is displaced to higher values of H_i relative to the idealised curve due to the higher local values of $\dot{M}_{irr}$ associated with localisation.

If measurements are made under constant $\dot{H}_i$ control, a step from *a* to *c* in figure 1 will be observed, giving square loop behaviour since H_i increases monotonically during the measurement.

Re-entrant magnetisation curves have been observed in specimens of single crystals (Mazzetti and Soardo 1966, Grosse-Nobis 1977) and of polycrystalline materials (Glaister and Lee 1965, Wolfe and Haszko 1967).

Re-entrant behaviour is observed with specimens having low demagnetising factors of materials with a magnetisation nucleation field that is large compared with the field required for domain wall motion (Wolfe and Haszko 1967). Sixtus and Tonks (1931) decreased the nucleation field for reverse magnetisation of Fe–Ni wires by applying tensile stress. Square loop curves were observed due to the field control adopted and macroscopic localisation occurred as discussed in section 1 above. Glaister and Lee (1965) studied re-entrant behaviour in toroids of polycrystalline barium ferrite. Annealing at temperatures of 200 °C led to diffusion processes which had the effect of increasing the field required to initiate domain wall motion.

In studies of plastic flow it has been shown that re-entrant stress–strain behaviour is similarly associated with diffusion effects, in this case of impurity atoms to dislocations, resulting in dislocation pinning. The stress required to initiate dislocation motion is thus higher than the stress necessary to keep dislocations moving (Hall 1970). This negative initial hardening, i.e. softening of the material, is directly analogous to the negative irreversible susceptibility regime of re-entrant magnetisation curves.

We now consider the anomalous magnetisation rate dependence of H_i: instability condition (ii), equation (11), is associated with anomalous signs of Λ. For specimens which do not exhibit localisation of magnetisation, $\Lambda > 0$ for the initial magnetisation curve and for the ascending branch of the hysteresis loop whereas $\Lambda < 0$ for the descending branch. Thus the condition $\dot{M}_{irr}/\Lambda < 0$ will be satisfied, for example, on the ascending branch of the hysteresis loop when H_i increases with decreasing $\dot{M}_{irr}$ for constant M_{irr}. The existence of anomalous Λ regimes has yet to be established experimentally. However, by analogy with crystal plasticity it is evident that such behaviour may result from the occurrence of diffusion controlled interaction of domain walls with solute atoms. For example, if during magnetisation the waiting time of temporarily pinned domain walls is such that diffusion of solute atoms occurs and causes additional pinning then the resistance to boundary wall motion will increase. In this regime H_i will increase with waiting time and therefore with decreasing $\dot{M}_{irr}$ leading to an anomalous value of Λ.

Evidence for the existence of magnetisation instabilities associated with anomalous Λ values is found in the measurements of Ferro *et al* (1965) and Martin *et al* (1986). Ferro *et al* reported the occurrence of large Barkhausen discontinuities which arose from discontinuous domain wall motion in experiments carried out under constant $\dot{M}$ conditions and at elevated temperatures using Fe–Al and Fe–Ni alloy specimens. The Barkhausen discontinuities disappeared at sufficiently high values of $\dot{M}$ and the threshold values of $\dot{M}$ below which discontinuities were observed increased with increasing temperature. Similar results were obtained by Martin *et al* (1986). In both studies the Barkhausen discontinuities were associated with diffusion controlled effects on domain wall motion.

In plastic deformation the phenomenon analogous to the appearance of anomalous values of Λ described above is known as dynamic strain aging. In the strain-rate–temperature regime of dynamic strain aging, solute atoms diffuse to dislocations during the waiting time of dislocations at obstacles in the slip path which results in a negative strain rate sensitivity of the flow stress. Experimental measurements of the strain rate sensitivity of the flow stress, made by discontinuously changing the strain rate during a series of measurements, exhibit transient behaviour in the dynamic strain aging regime (Van den Brink *et al* 1975). The instantaneous strain rate sensitivity measured at the point of change is always positive while the steady state strain rate sensitivity measured in the post transient regime becomes negative prior to the development of localised flow. Yield stability analysis (McCormick 1988) shows that a negative strain rate sensitivity is a necessary but not sufficient condition for the localisation of plastic flow due to dynamic strain aging. Strain localisation sets in only if the strain rate sensitivity acquires a value smaller than a certain negative value. It is expected that similar concepts will be applicable to unstable magnetisation behaviour caused by diffusion effects.

One of us (YE) gratefully acknowledges financial support from the Deutsche Forschungsgemeinschaft and The University of Western Australia through the award of a Gledden senior visiting fellowship.

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