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A simple spin glass perspective on martensitic shape-memory alloys

David Sherrington

Rudolf Peierls Centre for Theoretical Physics, University of Oxford, 1 Keble Road, Oxford OX1 3NP, UK¹,
Center for Nonlinear Studies, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
and
Santa Fe Institute, 1399 Hyde Park Road, Santa Fe, NM 87501, USA

E-mail: D.Sherrington1@physics.ox.ac.uk

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Abstract

A brief qualitative mapping is given between austenite, tweed and twinned phases of martensite alloys and corresponding paramagnetic, spin glass and periodic phases in spin glass alloys.

This paper is dedicated to John Pendry in celebration of his 65th birthday.

1. Introduction

Ferroelastic martensitic alloys show discontinuous phase transitions as the temperature is lowered from a high-temperature high-symmetry phase to a low-temperature phase of twinned lower-symmetry variants: for example from an austenite phase with cubic symmetry to an ordered pattern of stripes of complementarily distorted tetragonal twins [1]. Often these two phases are separated by an intermediate phase, known as 'tweed', in which the tetragonal twins are arranged in a more random way [2]. The lower-temperature phases exhibit interesting shape-memory and aging behaviour [1, 3].

In this paper we present a simple caricaturization of these phases and their transitions and features, in terms of simple spin glass alloy models. The suggestion that tweed is an analogue of a spin glass and a description of these systems in terms of a spin glass model is not new [4–6], but the modelling here is somewhat different and, hopefully, instructive. It also suggests interesting new models (or variations) for study by spin glass physicists.

2. Modelling

The origin of the structural transitions above is viewed as due to the frustrated interplay of competitive interactions of different ranges and anisotropy [8], combined with quenched disorder due to statistical inhomogeneity in the alloy make-up [4]. The origin of the competing forces is elastic but,

rather than retaining the technical complications of a complete elastic analysis, we shall here pursue a much simpler mean-field caricaturization in terms of effective 'spins', with the aim of conceptual simplification and qualitative explanation and prediction.

Our starting point is to separate the local and effective interactive features of the martensitic transition. The transitions from austenite to tetragonal are observed as first order and thus we may view the local propensity to either cubic or tetragonal symmetry as describable by a first order Landau transition.

2.1. Two dimensions

In a three-dimensional cubic system there are three mutually orthogonal tetragonal local distortion orientations. However, for simplicity we shall initially consider a two-dimensional system in which there are only two possible rectangular variants, orientated with their longer axes in the x or the y direction. They may be characterized by a variable ϕ whose magnitude indicates the strength of tetragonal distortion and whose sign indicates which of the two orientations it is in; explicitly, ϕ is the deviatoric strain $\phi = (\epsilon_{11} - \epsilon_{22})/\sqrt{2}$, where ϵ_{ij} is a component of the Lagrangian strain tensor [9]. The local situation can thus be described in terms of a local free-energy function

$$F_0 = \sum_i [A_i(T)\phi_i^2 - B_i(T)\phi_i^4 + C_i(T)\phi_i^6] \quad (1)$$

where B and C are both positive and their temperature dependence is not of qualitative consequence, while the $A(T)$

¹ Permanent address.

change sign from positive to negative as the temperature T is reduced. i labels the local region, and A , B , and C are given subscripts i to allow for quenched heterogeneity. Minimizing the free energy leads to a transition from $\phi_i = 0$ to $\phi_i = \pm\phi_i^*$ as T is reduced. $\phi = 0$ is interpreted as the locally cubic structure and the $\pm\phi_i^*$ as the two tetragonal distortions. The interesting behaviour arises from the inhomogeneity in the passage of the $A_i(T)$ across zero. The signs of B and C are chosen to reproduce the observed first order transitions. The i - and T -dependences of the ϕ_i^* are not of qualitative importance and so will be ignored henceforth.

In a further simplification we may re-write F_0 as an effective ‘spin Hamiltonian’ [7].

$$H_0 = \sum_i D_i(T) S_i^2; \quad S_i = 0, \pm 1 \quad (2)$$

with $S = 0$ corresponding to a local cubic structure, $S = \pm 1$ corresponding to the two orthogonal rectangular distortions and the D_i changing from positive to negative as the temperature T is reduced, emulating the sign-change of $A_i(T)$. The ground state has $S_i = 0$ (cubic) for $D_i \geq 0$ and $S_i = \pm 1$ (tetragonal) for $D_i \leq 0$. Below we shall continue to investigate the effect of the variation of the D_i but drop the explicit reference to their temperature dependence.

Let us now add the effective interactions between the rectangular variants at different locations. These are also induced by elastic effects, via compatibility constraints [9], but can be modelled by an effective spin interaction

$$H_I = - \sum_{(ij)} J(\mathbf{R}_{ij}) S_i S_j \quad (3)$$

where (ij) indicates a pair of sites, \mathbf{R}_{ij} is the separation of ‘sites’ (i, j) and $J(\mathbf{R})$ has a short-range attractive part (arising from the energetic costs of strain gradients) and also long-range contributions of \pm sign depending upon the orientation of \mathbf{R}_{ij} , together causing frustration [9]².

Overall the behaviour can be considered as determined by the total effective ‘Hamiltonian’, $H = H_0 + H_I$, together with appropriate boundary and further thermal effects.

2.1.1. Homogeneous case. It is instructive to consider first the homogeneous case, with all D equal. At high temperatures, where the D are all positive, the H_0 term alone would lead to all $S_i = 0$, corresponding to austenite, whereas for all D negative H_0 alone yields an Ising $S_i = \pm 1$ behaviour with the H_I term ordering the rectangular distortions so as to minimize the overall energy. The very experimental existence of twins demonstrates a type of ‘anti-ferromagnetism’; its specific form is a consequence of the combination of the long-range decay and the anisotropic variation of sign of the interactions. More specifically, the long-range interaction in two dimensions behaves dominantly as [8]

$$J(\mathbf{R}_{ij}) \propto -\cos 4\theta_{ij}/|\mathbf{R}_{ij}|^2 \quad (4)$$

² Strictly we should also add short-range terms penalizing the juxtaposition of $S_i = 0$ and $S_i = \pm 1$, for example of the form $K \sum_{mn} S_i^2 (1 - S_j^2)$, and favouring neighbouring $S = 0$, for example $-L \sum_{mn} (1 - S_i^2)(1 - S_j^2)$.

where θ_{ij} is the polar angle of \mathbf{R}_{ij} . The anisotropic variation is thus ferromagnetic along the $\pi/4$ and $3\pi/4$ directions and antiferromagnetic in the 0 and $\pi/2$ directions. Minimization of the two energetic terms immediately suggests the formation of alternating twinned stripes of internally ferromagnetic ‘spins’, the stripes all oriented along either $\pi/4$ or $3\pi/4$ but with the rectangular variants alternating from stripe to stripe between vertical and horizontal long axis. This order is so as to take advantage of the ferromagnetic interactions along the in-plane direction while minimizing the cost of the antiferromagnetic interactions along the x and y directions. If we denote the energy contributions (i) along the direction of the twin stripes, E_1 , (ii) perpendicular to these stripes, E_2 , (iii) along x , E_3 and (iv) along y , E_4 , then we find E_1 and E_2 negative, with $|E_1| \gg |E_2| \approx |E_3| = |E_4|$.³ The balances of gains and losses of energy and entropy, including boundary effects, determine optimal twin widths.

In fact, it is unnecessary for the D to be negative for this twinned order to occur, since the energy gained from H_I by the ordering of the $S_i = \pm 1$ can (and typically does) outweigh the costs associated with H_0 even for positive D , up to a critical value, in analogy with the phenomenon of induced moment behaviour in certain magnetic systems [10]. The transition from ground state $S_i = 0$ to $S_i = \pm 1$ would then be first order. If the D variation were independent of thermal effects and of the inter-site ordering, this qualitative feature would continue as the temperature was increased, until a tricritical point was reached. Note, however, that in the picture being painted here temperature plays two roles, one in effectively tuning D and the other in the cooperative thermal ordering of the effective ‘spins’. There is no reason for the temperature scales, or ‘transition temperatures’, of the two effects to be simply related, but experimental experience suggests that in practice a tricritical temperature is not normally reached while $D(T)$ is negative.

2.1.2. Inhomogeneous case. Allowing for inhomogeneity among the D_i permits the possibility of an intermediate ordered phase between austenite and periodically twinned phase, as will now be shown.

For preliminary mental orientation, let us first consider a situation in which the D_i take just two values randomly, one large and positive and the other large and negative. The model Hamiltonian,

$$H = \sum_i D_i(T) S_i^2 - \sum_{(i,j)} J(\mathbf{R}_{ij}) S_i S_j : \quad S_i = 0, \pm 1 \quad (5)$$

is then recognized as essentially analogous to that for a site-disordered Ising spin glass of magnetic and non-magnetic atoms,

$$H = - \sum_{(ij)} c_i c_j J(\mathbf{R}_{ij}) S_i S_j : \quad S_i = \pm 1, \quad c_i = 1, 0 \quad (6)$$

³ Along a stripe all the interactions are ferromagnetic and so add coherently; perpendicular to the stripe planes the alternating-variant planes interfere destructively and the long range of the interaction leads to a significant reduction of the binding energy in this direction as compared with in-plane; in the 0 and $\pi/2$ directions there is a combination of both long-range antiferromagnetic interaction and plane spin sign alternation; the short-range ferromagnetic interaction favours planes of more than single spin width.

where the $c_i = 1$ indicate magnetic sites, $c_i = 0$ non-magnetic; ignoring bootstrap effects, the magnetic sites correspond to those with D_i negative and the non-magnetic sites to D_i positive. The detailed make-up of attractive and repulsive forces is rather different from those of either conventional metallic or conventional semiconducting spin glass systems [13], but experience has taught that these details are not normally crucial to the existence of a spin glass phase. (The energy scale of the present pseudo-spin system is also different from that of real spin systems: much higher.) Based on this analogy one is led to conclude that the ground state will be ‘antiferromagnetic’/twin-stripe-ordered, ‘spin glass’/tweed or ‘paramagnetic’/austenite depending upon the relative concentration of sites with the two signs of D : twin ordered for a sufficient concentration of negative D ; spin glass beneath this critical concentration and above a lower percolation–frustration threshold, beneath which no cooperative order is possible⁴.

A more realistic distribution of D would be a continuous but bounded one: say of width ΔD around a mean of D_0 , with D_0 effectively decreasing monotonically (across zero) with reducing temperature T of the underlying martensitic alloy. At high enough D_0 the ground state would be non-magnetic $S_i = 0$ (austenite). Clearly, any site whose D -value is negative would contribute an effective magnetic site to (6) and consequently at low enough D_0 all sites would be ‘magnetic’ and twin ordered. But for sufficient finite ΔD an intermediate spin glass (tweed) state may be expected.

Let us first consider the case in which bootstrapping effects are ignored. Then the effective magnetic concentration would be equal to the number of sites with negative D and cooperative order would onset at the corresponding lower critical concentration of a cooperative phase of (6). For a continuous D -distribution this concentration would grow continuously as D_0 is reduced, so that the transition from non-magnetic (austenite) will be to spin glass via a continuous transition. Eventually, as D_0 is reduced further, the concentration will reach that at which twin order becomes preferable to spin glass and a further transition would occur.

However, when the effects of bootstrapping of moments are included both these transitions could become first order, the gain in interaction binding energy overcoming the cost of finite S_i on certain sites even when their corresponding D_i is still positive, up to a self-consistently determined D_c . This is possible for austenite to tweed whenever the low concentration limit for tweed order of (6) is non-zero. The tweed–twin transition will be first order when at the critical concentration separating tweed and twin phases of (6) the slope of the transition temperature versus concentration of the twin phase is sufficiently greater than that in the tweed phase that a jump in effective concentration (due to discontinuous moment formation) becomes energetically advantageous. Of course,

⁴ Note that the existence of a finite lower threshold to paramagnetism depends on the character and range of the longer-ranged interactions. If ferromagnetic and extending to infinite range there would be no zero-temperature threshold while with isotropic antiferromagnetic interactions it is believed that there is no long-range order in the limit of small concentration, even when these interactions are long ranged [11, 12]. Of course, at finite temperature (6) always has a lower concentration ordering threshold.

if the statistical inhomogeneity, and therefore ΔD , is too small, the tweed phase could be bypassed in the discontinuous ‘moment’ jump.

2.2. Symmetry-breaking fields

Above we have not discussed explicitly the inclusion of effects breaking the symmetry between alternative variants. These can occur due to either external or internal stresses and would add terms of the form

$$H_{\text{stress}} = - \sum_i h_i S_i \quad (7)$$

with contributions to the h_i either externally imposed or a consequence of internal inhomogeneities favouring particular local orientations. Thus one could extend the Hamiltonian to include these terms and model with

$$h_i = h_i^{\text{imposed}} + h_i^{\text{random}} \quad (8)$$

with the h_i^{random} randomly chosen from some distribution $P(h^{\text{random}})$.

2.3. Three dimensions

Let us now turn to three dimensions, the more normal dimensionality of martensitic systems. Again, first order transitions from high to lower local symmetry are observed experimentally: in classic cubic systems with the two types of rectangular lower-symmetry local variants of the two-dimensional example replaced by three tetragonal variants, orientated orthogonally along the x , y and z axes. Again, the interaction terms lead to cooperative spatial ordering of the variants. At low temperatures this ordering is again of complementary twins, implying again long-range interactions of oscillating sign as a function of angular orientation [9]. Both of these features can be emulated by extensions of the simple spin model used above or by an axis-director analogue of it. For example, the three tetragonal variants can be described analogously to liquid-crystal (axis-)‘directors’, with interactions between them analogous to those in quadrupolar glasses [14]. Local inhomogeneities again lead to different local propensities to cubic (zero director) or tetragonal (finite director) and hence to random variations in the effective concentration of directors. This in turn can be anticipated to permit the insertion of a quasi-random three-dimensional tweed phase between austenite and twinned phases. A possible model formulation would be to employ the effective Hamiltonian

$$H = \sum_i D_i(T) |S_i|^2 - \sum_{(i,j)} |S_i| |S_j| J(\mathbf{R}_{ij}) (2(\mathbf{S}_i \cdot \mathbf{S}_j)^2 - 1) \quad (9)$$

where the spins are restricted to $S_i = 0, \hat{x}, \hat{y}$ or \hat{z} and the last three options are respectively unit vectors in the x , y and z directions. Another possible description would be in terms of a four-state Potts spin glass [15] in a field and with anisotropic exchange: one of the Potts dimensions corresponding to austenite, the other three to the orthogonal variants, with the exchange interactions only among these latter three dimensions.

3. Properties

Among the most interesting properties of martensitic systems is that of shape memory. In so-called ‘one-way shape memory’ a system which is forged into a particular shape at a high temperature, in the austenite phase, cooled and then distorted, will regain the original shape on reheating. This can be explained easily from twinning alone along with the feature of ease of distortion in the twinned phase [1]. The initial distortion corresponds to the imposition of a boundary (at whatever necessary energy cost). Cooling alone retains this shape, with the twin stripe-widths accommodating it. In the twinned phase, however, the shape is easily modified at low energy cost by adjusting the individual twin section widths by moving the intertwin boundaries. Reheating removes the twins and their boundaries and the sample regains the originally imposed shape. There is however often observed another type of shape memory, ‘two-way-shape memory’, in which the system remembers its history both on heating and on cooling [16]. This is not readily explainable by twinning alone. However, such two-way memory is a characteristic feature of spin glasses [18, 27] and hence is attributable to tweed [17]. The present model suggests a way to model it minimally, but further pursuit of its study is deferred to a future paper.

Another of the characteristic features of a spin glass is preparation dependence, as illustrated in a difference between the susceptibility as measured by first cooling from the paramagnetic state and then applying a field (ZFC: zero-field cooling) and that observed by first applying the field and then cooling (FC: field cooling), demonstrating non-ergodic behaviour over experimental times. There should be a corresponding difference in uniaxial compliances (or, inversely, compressibilities) in tweed measured along principal axes⁵. This effect is a consequence of the multiplicity of metastable states of spin glasses, their non-trivial evolution under changes of control parameters (such as temperature or applied field) and consequential slow dynamics in exploring all of phase space [31, 32]. It should however also be noted that the normal fluctuation–dissipation relation does not hold for spin glasses [18] and a simple measurement of the sound velocity will correspond more to the ZFC situation. However, variations can be expected as a function of frequency.

We might also note in passing that the full Gibbs susceptibility of a mean-field spin glass (and, at least to a good approximation, the FC susceptibility of a real spin glass) is independent of temperature, and hence one might anticipate a similar independence in a martensitic alloy throughout the tweed regime.

Aging behaviour in glasses has a long history [19] and similar aging features should exist in tweed. More recent work on spin glasses has shown how many aging quantities can be expressed as a function of the ratio of times t/t_w , where t_w is the time following a rapid quench and t is time

of measurement, for both t and t_w large. A corresponding systematic exploration of tweed would seem to be called for.

Another intriguing characteristic of conventional spin glasses is rejuvenation [27], whereby a system relaxing/aging in an external dc field appears to start relaxing/aging anew when this field is suddenly changed, ignoring its previous aging; this feature is, for example, apparent in an observation of the ac out-of-phase susceptibility. Again, one would expect an analogue for tweed.

3.1. Relation to prior models

The models considered above can be viewed as extensions of the Blume–Capel [20] or Blume–Emery–Griffiths (BEG) models [21]; in two dimensions to allow for randomness of the local anisotropy term and long-range and orientational sign variation of the exchange interaction with separation \mathbf{R}_{ij} , and in three dimensions also to allow for Potts-like director replacement of the BEG $S = 1$ Ising spins. A complementary discrete random field Potts modelling of elastic systems has recently been proposed by Cerruti and Vives [22].

4. Soluble models

Although the physical conclusions indicated above seem inevitable, the actual models introduced are almost certainly not completely soluble, as neither is the corresponding equation (6) of an even-simplified conventional site-disordered spin glass. In theoretical and simulational studies of conventional spin glasses one usually replaces site disorder with bond disorder $P(J_{ij})$, as suggested by Edwards and Anderson in their classic seminal paper [23]. Taking the mean J_{ij} to be non-zero [24] permits competition (and transition) between two types of frozen phase, spin glass and ferromagnet (or anti-ferromagnet). An exactly soluble version is the infinite-range extension of Sherrington and Kirkpatrick⁶ [5, 25]. A tempting analogue in the present case is a variant of the Ghatak–Sherrington model [28] (see also [29, 30]), with

$$H_{GS} = \sum_i D_i S_i^2 + \sum_{(i,j)} J_{ij} S_i S_j : \quad S_i = 0, \pm 1 \quad (10)$$

where the J_{ij} are independently distributed according to some $P(J_{ij})$, including allowance for non-zero mean, and the D_i also independently according to some other distribution $P_D(D)$. Such a model should be straightforwardly soluble by the methods developed for the SK model and its extensions [31, 32] but it would miss many of the crucial features of the martensitic systems (including the feature of easily malleable twin planes). Consequently computer simulation of the model system seems to be the sensible next direction to pursue at the theoretical level.

Conclusions

A simple spin-glass-like modelling has been presented, emulating key ingredients of martensitic alloy transformations.

⁵ Note that uniaxial compression is needed so that the effective field has a different energetic consequence for the different types of tetragonal distortion; isotropic compressibility would not have the same effect since it is an ‘irrelevant field’ for separating tetragonal twins. Neither would compressibility at $\pi/4$ to the tetragonal variant long directions.

⁶ SK can be extended to cover antiferromagnetic ordering via the introduction of two sublattices as in [26].

Qualitative considerations and analogies have been used to suggest that tweed should be viewed as a spin glass, echoing a message originally expressed, via a different formulation, by Kartha *et al* [4]. Analogies with aging, rejuvenation and memory of spin glasses suggest explanations and experimental investigations of the behaviour of martensitic alloys. The study also exposes a new type of spin glass Hamiltonian worthy of further investigation.

This brief paper has concentrated on devising simple models and considering their likely thermodynamic phases and transitions, together with some anticipated properties by analogy with more conventional spin glasses. These need further more rigorous consideration, but so also does the dynamics of these systems viewed within the simple spin-Hamiltonian modelling. There has already been some study of the dynamics within a fuller elastic strain formulation without alloy inhomogeneity [8] (i.e. without imposed disorder quenched into the controlling equations) and for a different simple model with quenched disorder [33]. A start has also been made in terms of computer dynamical iteration of the mean-field solution for the two-dimensional homogeneous-Hamiltonian case [34]. However, a fuller examination along the lines of those employed for more conventional spin glasses is now called for, with special attention to the metastabilities that characterize systems with first order transitions; note that conventional spin glasses have thermodynamically continuous transitions even though soluble model systems with $p > 2$ -spin interactions or with interactions lacking symmetry of definiteness (such as Potts or quadrupolar) spins have discontinuous replica symmetry breaking (DRSB) spin glass onset. We might also note that structural glasses, which have no imposed quenched localized disorder in their Hamiltonians, have quasi-transition temperatures T_g (glass transition high viscosity) and Kauzmann temperatures T_K (temperature at which the entropy difference between liquid and glass extrapolates to zero) analogous to the dynamical and thermodynamic glass transitions of a DRSB model. Hence it is also possible that an analogous dynamically self-generated quasi-disorder could occur in martensites [34]. Simple modelling such as introduced here might provide a useful laboratory within which to probe the physics.

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