

Tweed in Martensites: A Potential New Spin Glass

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Received January 17, 1992; accepted January 26, 1992

Abstract

We've been studying the "tweed" precursors above the martensitic transition in shape-memory alloys. These characteristic cross-hatched modulations occur for hundreds of degrees above the first-order shape-changing transition. Our two-dimensional model for this transition, in the limit of infinite elastic anisotropy, can be mapped onto a spin-glass Hamiltonian in a random field. We suggest that the tweed precursors are a direct analogy of the spin-glass phase. The tweed is intermediate between the high-temperature cubic phase and the low-temperature martensitic phase in the same way as the spin-glass phase can be intermediate between ferromagnet and antiferromagnet.

The field of disordered systems has been a rich and fascinating branch of condensed matter physics in the last few decades. New ideas, language, and techniques have been developed in order to study problems with intrinsic randomness in their Hamiltonians. Much of the attention has been on relatively new and exotic systems: spin glasses in dilute magnetic systems, charge density waves in complex one-dimensional materials, localization in doped semiconductors. One of the promises of the field is to treat the real, dirty world of materials. So long as our industrial colleagues refrain from making their goods with perfect crystals, the study of disordered systems ought to have ramifications in old-fashioned, practical systems.

One of the old problems in materials physics is that of the "central peak". First-order transitions traditionally have no precursors: water looks like water until 0 °C, at which point it suddenly becomes plain ice. Transitions between different crystalline phases, in contrast, often have rather large precursor effects. In neutron scattering, there is often a large, quasielastic central peak, representing either static or rather long-lived fluctuations in positions. In the martensitic phase transitions (described more fully below) the central peak may be understood to be associated with a tweed morphology in TEM micrographs. The tweed [1], so called because

of its characteristic irregular fabric-like cross-hatched pattern, has stripes aligned along $\langle 110 \rangle$ directions of widths a few atomic spacings. Tweed occurs for tens to hundreds of degrees above the transition temperature.

First, we'll review what a martensitic transformation is. Second, since martensitic materials have high elastic anisotropies, we'll take a limit of infinite elastic anisotropy to explain the tweed morphology. Third, we'll introduce disorder, and show that the tweed regime can be understood (in a two-dimensional model) as a spin-glass phase intermediate between the high and low-temperature phases. Thus, the community's large investment of work on spin glasses is seen to have direct application to a substantial problem in traditional metallurgy. This is a simpler presentation of recently published work [2].

Martensitic phase transformations were first studied in carbon steels. Typically, they involve a cubic high-temperature phase which abruptly changes shape, elongating in one direction and contracting in the other two, with possible intracell, atomic shufflings. The resulting low-temperature phase is usually left with a complicated morphology of micron-sized plates, which in turn can be composed of many parallel twinned regions. The simplest examples of martensitic transitions are the bcc \rightarrow fcc transition or bcc \rightarrow hcp transitions seen in many metals and alloys on cooling. The martensitic transformations of interest to us have (1) little volume change, and (2) no long-range diffusion during the transition. Large changes in volume prevent the nice elastic accommodation that the plates represent: crystals undergoing such transitions may in fact shatter from the resulting strains. The martensitic transformations in carbon steels have too much volume change to be described by our theories. If long-range diffusion is needed in a transition, it usually proceeds rather slowly: in contrast, the growth of a martensitic plate can occur at the speed of sound, and emits a cracking noise.

We model these materials with a two-dimensional simulation. We represent each atomic cell by a quadrilateral: the two types of atoms in the simulation are represented by quadrilaterals with two different energies as a function of shape. The high temperature phase in our simulation is square (grey in Figs 1 and 2), while the low temperature phase has two variants: tall-and-thin (black), and short-and-

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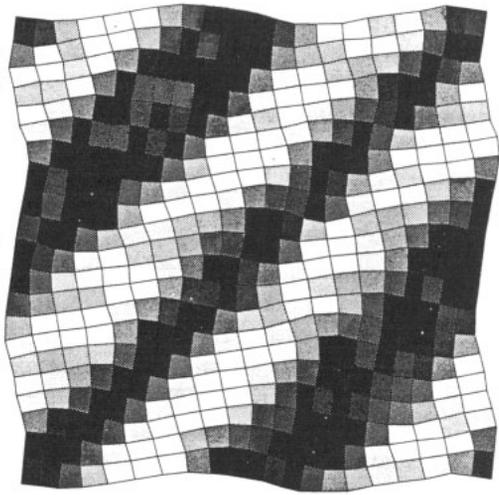


Fig. 1. Martensitic Twins. A typical metastable state of our two-dimensional model, with parameters set to be in the low-temperature rectangular martensitic phase. The free energy is of the same form as given in our earlier publication [2], except that we've added energy terms for bulk compression and diagonal strain (i.e., this is an unconstrained simulation). Parameters are taken from the $\text{Fe}_{0.7}\text{Pd}_{0.3}$ shape memory alloy, with each quadrilateral representing one atom. Naturally, significant simplification was made in going from three dimensions to two. In addition, we don't know yet the coupling to impurities. Details will be published in a longer form.

Notice the twin boundaries between the two variants: they lie along the $\langle 11 \rangle$ diagonals. This nearly one-dimensional variation is of the form $\phi^-(x - y)$ as described in the text, eq. (3). Notice that twin boundaries have arranged to keep the overall shape of the domain the same as that of the original sample: a large chunk of one variant would have produced intolerable strain in the surrounding matrix

fat (white). The martensites we're interested in don't change shape drastically: a couple of percent stretch along one axis is typical. (The shape change in the figures is exaggerated by a factor of 15.)

Because of this, these materials have transitions which are largely reversible. In Fig. 1, we see a typical martensitic region. One sees domains of both variants, separated by twin boundaries along (in this case) the diagonal stretching from lower left to upper right. Notice that the region as a

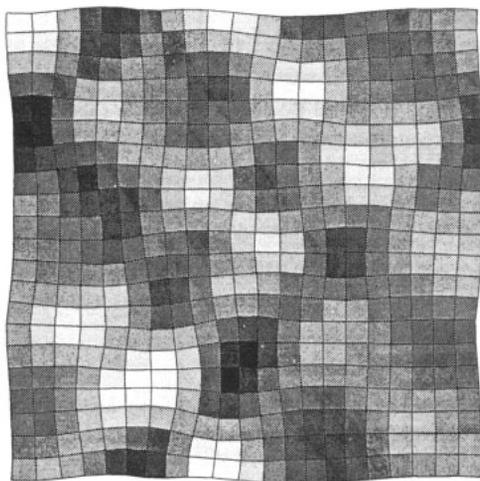


Fig. 2. Tweed. Another metastable state, at parameters set in the tweed regime. Notice the patchy tweed pattern, looking like a superposition of stripes along the two diagonals. Grey is the high-temperature square phase, black is tall-and-thin, white is short-and-fat. Different quenches typically land in different metastable states: the system is glassy

whole has preserved its square shape: the twin boundaries arrange themselves so as to prevent long-range strain from building up. This is for precisely the same reasons that magnetic domains form in ferromagnets: the long range elastic (magnetic) fields introduced from one growing variant lower the energy of formation of the other variant in neighboring regions.

There are three types of materials in which tweed has been seen. First (and most recently), it has been seen in doped YBaCuO [3–5], where our two-dimensional model is particularly appropriate. YBCO goes through a tetragonal \rightarrow orthorhombic phase transition somewhat above its superconducting transition. If one views it in a funny basis, this transition is a square \rightarrow rectangular transition in the copper-oxygen planes. When cobalt or aluminum is doped for copper, it apparently substitutes onto the chains, producing a substantial drop in the superconducting transition temperature and a tweed-like modulation. Second, it has been seen in the A15's [6], which also go through a martensitic transition near the superconducting transition. Third (and the way we got interested), it has been seen in the shape-memory alloys [7–12].

In passing, we can use Fig. 1 to provide a simple illustration of the shape-memory effect. Imagine the large square sample as some object (say a teapot) formed out of the high-temperature austenite phase. We've passed through the phase transition, yet the region is roughly square: twin boundaries have formed to keep the material macroscopically the same shape (square, or teapot). In the martensitic phase, though, the material is much softer, even though the single-crystal elastic constants are stiffer. To plastically deform the teapot, one needn't break any interatomic bonds. One need only push the twin boundaries around! If one takes the sample in Fig. 1 and stretches it vertically, the twin boundaries will shift to shrink the white regions. One can crumple the teapot without undue force, introducing no changes in the underlying cubic lattice. Upon reheating, the material returns to the cubic phase. The twin boundaries, wherever they have moved, will disappear, and the material will reform into its original configuration. The teapot miraculously uncrumples! Shape-memory alloys are used industrially for thermal switches (often in automobiles), and for pipe fittings [13]. They've been used also in more exotic applications, such as satellite antennae which automatically unpack themselves, robotic muscles which flex upon resistive heating, and small bars which bust up rocks [14] in golf courses.

Figure 2 shows another local low-energy state of our model, here in the tweed temperature range. Notice the patchy cross-hatched diagonal checkerboard pattern. There are long-ranged correlations in the two diagonal directions, but only short-ranged correlations horizontally and vertically. Indeed, the dark and light stripes extend in many cases through our whole sample.

Let's define the order parameter for our martensitic transition to be the net stretch in the horizontal direction:

$$\phi = \varepsilon_{xx} - \varepsilon_{yy}, \quad (1)$$

where the strain field $\varepsilon_{ij} = (\partial_i u_j + \partial_j u_i)/2$ and $u(x, y)$ is the displacement field (final position minus initial position, labeled by the initial position x, y). (We have in the analysis ignored the "geometric nonlinearity" $\partial_i u_i \partial_j u_j$, which can be

important if there are large rotations in the problem. Our unconstrained simulations do implement complete rotation invariance.) Thus, $\phi > 0$ in short-and-fat regions, $\phi = 0$ in the square phase, and $\phi < 0$ in the tall-and-thin regions.

Now, writing a free energy in terms of strain fields is tricky. Indeed, most configurations of $\varepsilon(x, y)$ are illegal! Unless

$$\text{inc}(\varepsilon) = \nabla \times (\nabla \times \varepsilon)^T \quad (2)$$

is zero, the strain field describes a material with embedded line dislocations [15]. Doing any calculations in terms of strain fields must either add a term for the energy of dislocations, or must put in some kind of Lagrange multiplier to forbid ε from developing an inc. Working in terms of $\phi(x, y)$ would seem even worse. Since the components of ε are not independent (2), most configurations of $\phi = \varepsilon_{xx} - \varepsilon_{yy}$ will necessarily involve other kinds of strain. If we allow ϕ to vary freely, we must “integrate out” the other elastic degrees of freedom, leaving us with a free energy with long-range forces.

There is another option, which is suggested by the large elastic anisotropy characteristic of martensites. Because the high temperature phase is cubic, it has three independent elastic constants (rather than two for an isotropic material). The elastic constant C' which resists deformations into rectangular shapes is much smaller* than the elastic constant which resists bulk compression ($\varepsilon_{xx} + \varepsilon_{yy} \neq 0$) and the one which resists diagonal strain ($\varepsilon_{xy} \neq 0$); we let these two elastic constants go to infinity, thus constraining the system to forbid any deformation except rotations, translations, and rectangular stretches (measured by ϕ).

These constraints profoundly restrict the configurations. A straightforward computation [2] shows that the two dimensional configurations that are allowed are precisely those which can be written as a sum of two one-dimensional strain modulations [17]

$$\phi(x, y) = \phi^+(x + y) + \phi^-(x - y), \quad (3)$$

ϕ^+ describing a modulation along the (x, y) direction (lower left to upper right), and ϕ^- describing a modulation along the $(x, -y)$ direction. (Equation (3) is derived by first showing that the displacement $u(x, y)$ can be written as a sum of two one-dimensional displacement fields, and then finding the strains.)

Now, it's been known for a long time that large elastic anisotropies are associated with strain fields which extend long distances along the diagonals. We are encouraged for three reasons to think that the infinite anisotropy limit is a natural starting point for studying tweed. First, the form (3) describes the behavior of our simulation well. The displacement variation in Fig. 1, for example, is nearly purely of the ϕ^- form. The variation of Fig. 2 is well described as a superposition (3). Second, it plausibly describes the experimental tweed morphology. The experimentalists agree with us that tweed is composed of $\{110\}\langle 1\bar{1}0 \rangle$ shears, and

that it looks cross-hatched.* Third, it describes the modulation patterns seen in earlier and later simulations, with different driving mechanisms for the modulations [18, 5]. Thus, in the limit of infinite elastic anisotropy, cross-hatched patterns becomes the only allowed patterns.

So far, we have explained how the tweed morphology is a natural modulation for materials with large elastic anisotropy. We now must address why these materials choose to modulate at all. Experimentally, in closely related systems, neutron scattering experiments in nearly pure materials have illuminated the important role of disorder [19]. Moreover recent simulations have found tweed in a disordered alloy but not in an ordered one [20]. We consider here the disorder introduced by concentration variations.

Because the shape memory alloys are alloys, static statistical concentration variations provide an intrinsic source of randomness. First, the martensitic transition temperature varies drastically with concentration η in most of these systems: in $\text{Fe}_{1-\eta_0}\text{Pd}_{\eta_0}$, the fcc \rightarrow fct transition is at room temperature for $\eta_0 = 0.29$, and drops to 0K for $\eta_0 = 0.32$. This will produce a free energy difference $F_A(\eta(x, y)) - F_M(\eta(x, y))$ (or more concisely, $(F_A - F_M)(\eta)$) which varies with position. At high temperatures, this will always be negative, at low temperatures always positive, but near the transition temperature at the average concentration, this difference will vary in sign from place to place. Second, the local configurations of Fe and Pd atoms can introduce local distortions, favoring one or another of the martensitic variants [8, 4, 20]. This will produce a free energy difference $(F_- - F_+)(x, y)$ which will depend on gradients of η [2].

The local elastic free energy can in general have three minima: two at $\pm\phi_0$ for the two martensitic variants, and one at $\phi = 0$ for the austenite.† If we define $i = x + y$ and $j = x - y$, then each ϕ_i^+ and each ϕ_j^- will represent a contribution to the local order parameter for all the sites along some diagonal. The order parameter at a certain site (x, y) will be given by the sum of ϕ_i^+ and ϕ_j^- for appropriate i and j . In our constrained system (3), its possible to arrange to have the local order parameter modulate solely between the three states by letting ϕ^+ and ϕ^- each take on two possible values, $-\phi_0/2$ or $+\phi_0/2$. If we then think of the ϕ_i^+ and ϕ_j^- as spins that either point down or up, then we have a convenient spin representation for conceptualizing the tweed problem. Either of the two antiparallel configurations produces an undeformed, square, austenite region ($\uparrow\downarrow$ or $\downarrow\uparrow$ give $\phi = 0$), and the parallel configurations produce the two rectangular martensitic variants ($\uparrow\uparrow$ gives $+\phi_0$, $\downarrow\downarrow$ gives $-\phi_0$).

The tweed model, in spin language, is precisely equivalent to an infinite range spin glass. The spins in the spin glass are analogous to the diagonals which connect the sites in the martensite. In the spin glass, each spin is coupled to every other spin by a particular bond with a random sign and

* Actually, the measured elastic constants in the tweed and martensitic regime will have contributions from rearrangements of the domains, as in the A15's [16]. How much the tweed is due to the lowering of C' and how much the lowering of C' might be due to the response of the tweed morphology is a subject we plan to explore.

* Actually, the experimentalists haven't for sure decided that the tweed really is a superposition. Since TEM measures through a rather thick slab, there is no direct evidence that the observed cross-hatched patterns aren't plain old twins pointing in different directions at different depths. In two dimensional systems, at least, we unambiguously predict modulations in both $\langle 11 \rangle$ directions at once.

† ϕ_0 will generally depend upon temperature, and at high enough temperatures there will be no martensitic minima. This just contributes to the effective temperature dependence of the bonds, described below.

strength. In the tweed, each diagonal is “coupled” to every other diagonal by a particular site, which prefers austenite ($\phi = 0$, $\uparrow\downarrow$ or $\downarrow\uparrow$) or martensite ($\phi = \pm\phi_0$, $\uparrow\uparrow$ or $\downarrow\downarrow$) at random. In each case, the couplings represent the frozen-in disorder.

Figure 3 illustrates the analogy between the two systems. Each letter (A, F; A, M) represents a term in the energy: the spins, squares and rectangles show one of the possible metastable configurations of each of the systems. Both the spin glass and the tweed are frustrated: not all the connections can be made happy at the same time. In both cases, any closed loop with an odd number of A’s is frustrated: one of the bonds in the spin glass must be broken, and one of the sites in the martensite must locally be in the wrong phase.

To complete the analogy, one must notice three more correspondences. First, just as the effective bond strength J_{ij} between two diagonals is given by $(F_A - F_M)(\eta)$ at the corresponding (x, y) , there is a contribution to the effective “magnetic field” H_{ij} on the two diagonals given by $(F_- - F_+)$. Second, the free energy difference $F_A - F_M$ is strongly temperature dependent. In the spin glass, the Hamiltonian really represents all the degrees of freedom: the parameters are the bare ones, and do not depend on temperature (although they might depend, for example, on pressure). In our problem, the martensitic transition is (probably) driven by vibrational entropy: the phase space of small oscillations is not represented by the degrees of freedom $\phi(x, y)$ we’re

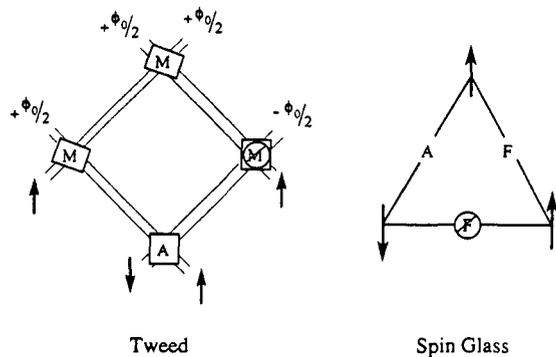


Fig. 3. Frustration. Our model of martensites is frustrated in precisely the same way as are spin glasses.

Spin Glass. On the right, we have three spins which can either point up or down. They are connected by bonds which prefer parallel (Ferromagnetic) or antiparallel (Antiferromagnetic) alignment. In a spin glass, there are many such spins, and the bonds are chosen F or A at random. If we start at the lower left corner of the triangle, starting (arbitrarily) down, and move around clockwise, we can satisfy each bond in turn. However, for any loop with an odd number of antiferromagnetic bonds, one bond must be broken (in its high-energy state): here the ferromagnetic bond connecting the bottom two spins is not satisfied.

Tweed. On the left, we have four regions in the material, forming a diamond. Starting at the bottom, we have a region where the local alloy concentration makes the square, Austenite phase lower in energy. In the limit of infinite elastic anisotropy, the order parameter at the bottom site is the sum of two contributions, one for each diagonal. By choosing the contribution for the lower left diagonal $+\phi_0/2$ (represented by a plus sign \uparrow), and the contribution for the lower right diagonal $-\phi_0/2$ (\downarrow), we satisfy the local free energy in the bottom site: $\phi = 0$, represented by the square box surrounding the A. Again, moving clockwise, we can satisfy the free energy in the left-hand site (which prefers Martensite) by choosing $+\phi_0/2$ (\uparrow) for the upper left diagonal, leading to a net order parameter ϕ_0 and a short-fat rectangular deformation. We can continue this process through the top site, but when we reach the final, right-hand site, the order parameter is already determined to be $0 = +\phi_0/2 - \phi_0/2$ ($\uparrow\downarrow$): the local free energy, which prefers M, is not satisfied

Table I. Correspondence between spin glass and tweed parameters

Spin glass	i	j	s_i	s_j	J_{ij}	H_{ij}	T
Tweed	$x + y$	$x - y$	ϕ^+	ϕ^-	$F_A - F_M$	$F_- - F_+$	$\langle -J \rangle_{ij}/L$

including in our free energy. Tracing over these small vibrations makes the bonds J_{ij} in our free energy temperature dependent: roughly speaking, $-J_{ij}(T) = -J_{ij}^0 + LT$, where L is the vibrational latent heat per site. Third, one must notice that every lower-left–upper-right diagonal crosses every upper-left–lower-right diagonal. The interactions are of infinite range!* Table I shows the correspondences, up to things like factors of 2 and ϕ_0^2 .

We can now write a spin-glass like Hamiltonian for the martensitic tweed system:

$$\mathcal{F} = \sum_{i,j} \{-J_{ij}\phi_i^+\phi_j^- - H_{ij}(\phi_i^+ + \phi_j^-)\}. \quad (4)$$

Every site i interacts with every site j : it corresponds to an infinite range spin glass [21], on which we expect replica theory to give an exact solution [22]. Unlike the SK model, there are two classes of spins, each of which interacts only with the other class: it is a bipartite infinite-range spin glass [23].

Figure 4 shows the phase diagram we expect for the free energy equation (4), setting the random field term to zero. What do I mean by “expect”? While this particular model has never been completely solved, we can guess the answer from the partial solutions and from the behavior of the SK model. Korenblit and Shender [23] solved this model in the replica symmetric approximation. The phase diagram they found was exactly that of the SK model with varying fractions of ferromagnetic bonds, except that the bipartite model phase diagram was doubled by reflection through $\langle J \rangle = 0$. The replica-symmetry breaking solution to the SK model [22] has a vertical phase boundary between the spin-

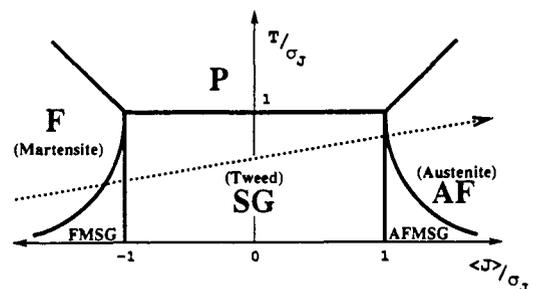


Fig. 4. Phase Diagram. The phase diagram expected [23] for the infinite-range bipartite spin-glass model, eq. (4). A given sample will traverse a sloped path in this phase diagram as temperature is increased, because of the temperature dependence in the effective coupling. It will pass from the martensitic phase through the tweed regime into the austenite phase, as shown by the arrow. The two magnetized-spin-glass phases (FMSG and AFMSG) are supposed to have glassy dynamics and metastability, even though they have long-range order. We ignore here the random field term H_{ij}

* This happens only for infinite elastic anisotropy, of course.

glass phase and a glassy phase with long-range order (called the magnetized spin-glass, or MSG phase). We expect, thus, to find vertical phase boundaries in the bipartite model too; hence Fig. 4. The problem with the random field has been studied too [24].

What can we see from this phase diagram? Tweed is a spin-glass phase intermediate between the two ordered phases. The antiferromagnetic phase on the right has the two sublattices pointing in opposite directions: apart from thermal fluctuations, the entire region has $\phi = \phi^+ + \phi^- = 0$. This is the high-temperature square austenite phase for our model. The ferromagnetic phase on the left has the two sublattices pointing in the same direction: the region has transformed entirely into the low-temperature martensitic phase. The paramagnetic phase on the top also has square symmetry on average, but is a thermal mixture of square regions together with both rectangular variants. Conceivably, this could represent the austenite phase too. (Could bcc not be the familiar lattice plus small oscillations, but really sometimes be an equal mixture of various other variants? Recent experiments on zirconium suggest that this actually might be the case! [19]) Finally, the spin-glass phase corresponds to tweed. It is a frustrated attempt to accommodate the concentration fluctuations: a patchy, glassy mixture of square and rectangular regions. Because our effective bonds are temperature dependent, heating the physical martensite moves us along the dotted line in the figure: both the average concentration of antiferromagnetic bonds and the temperature of the "spin" system increases.

Young people giving their first talks often fear that someone will ask the key questions which they know will expose the gaping holes in their theories. I would like to conclude with my attempt to answer three such questions.

Q.: I thought the martensitic transition was first order. Also, does your theory work in three dimensions?

A.: Indeed, the Ising spin-glass transition is second order. I guess in two dimensions (i.e., extremely thin high- T_c samples) we do predict that the first-order martensitic transition will be replaced by two second-order transitions in the presence of sufficient disorder. (Imry and Wortis [25] have discussed the effects of random-field disorder, which can destroy the transition entirely.) In three dimensions, the system is clearly not an Ising spin glass. First, there are at least three martensitic variants: the order parameter ϕ becomes a two component object with three local minima stretching in the three orthogonal directions. Second, in the limit of infinite elastic anisotropy, there are six scalar one-dimensional functions corresponding to ϕ^\pm . Even changing from an Ising to a Potts glass [26] makes for a first-order spin-glass transition (albeit with no latent heat). What kind of transitions we will find once we identify the right spin model for 3D martensites is completely open at this point.

Before mindlessly generalizing our work from two to three dimensions, we want to get a better physical picture of what is important. We're exploring effective-medium simulations with Karsten Jacobsen and TEM experiments with John Silcox in order to get a handle on the impurity couplings, and on the important long-range correlations. If I have to guess, I'd say that there is a better chance of a real spin-glass phase in 3D. In 2D, tweed is a true phase only in mean-field (infinite elastic anisotropy): in 3D, a sufficiently frustrated system ought to have a true glassy phase.

Q.: Why haven't you taken parameters from the 2D high- T_c materials, instead of the uncontrolled extrapolation from FePd? Also, do you have any predictions for strontium-doped LaCuO? (Mac Beasley actually asked the second question, and pointed out some experimental data that we should pursue.)

A.: We were, oddly enough, more interested in the shape-memory alloys, and it took us a while to realize how hard 3D was going to be. High T_c is next on the agenda.

Q.: Do you have any experimental predictions?

A.: We don't have definite predictions for the statics in 3D. We'd really like to predict a diverging nonlinear elastic susceptibility. As one crosses the phase diagram in Fig. 4 from the paramagnetic phase to the tweed spin-glass phase, the nonlinear susceptibility diverges. That is, if we put an external force F which stretches the sample coupling to ϕ , and measure the response $\phi = \chi_1 F + \chi_3 F^3$, then χ_3 will diverge at the phase boundary. There are three problems, though. First, the susceptibilities which diverge along the side phase boundaries of the tweed apparently aren't elastic constants [22]. Unless the high-temperature phase is a melt of the various low-temperature variants (as discussed above), so the austenite corresponds to a paramagnet, we don't get an effect. Second, in two dimensions the mean-field transition will be rounded whenever the elastic anisotropy isn't infinite. Third, the predictions in three dimensions are likely to be quite different. If I were an experimentalist, I'd certainly try looking for phase boundaries, using elastic probes. We can't say yet, though, exactly what to look for.

The dynamical predictions are much more straightforward. All of the glassy systems have interesting, slow dynamics. Even if our spin-glass identification is wrong, and random-field effects dominate the problem, there should be memory, hysteresis, and logarithmic decays in response functions. While this may seem old hat, in metallurgy this ought to be more exciting. I'm not saying, of course, that slow elastic relaxation in dirty metallurgical systems is news: what ought to be exciting is that there may be interesting, quantitative science to be done with the dirt.

Acknowledgements

We acknowledge the support of DOE Grant #DE-FG02-88-ER45364. T. C. wishes to thank the Laboratory of Atomic and Solid State Physics for their hospitality, and the D. G. I. C. Y. T. (Ministry of Education, Spain) for financial support. J. P. S. would like to thank the Technical University of Denmark and NORDITA for support and hospitality.

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