Dislocation motion in quasicrystals and implications for macroscopic properties

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Dislocation mobilities in quasicrystals are calculated from recently developed hydrodynamic equations. The mobilities are expressed in terms of vacancy diffusion constants for the quasicrystal. Estimates of these diffusion constants based on their values in conventional crystals lead to far smaller dislocation mobilities than in conventional crystals. Plastic flow should therefore be effectively prohibited; one dramatic consequence is that quasicrystals should be extremely brittle. A detailed microscopic calculation of vacancy diffusion constants is urged. Implications for icosahedral-order theories of glasses are discussed.

The recent discovery of quasicrystals[1,2]—translationally ordered yet aperiodic systems whose point symmetry groups are not crystallographic—raises a number of extremely interesting questions. One of the most obvious and important is how the macroscopic material properties—elasticity, malleability, etc.—of these exotically ordered systems will differ from those of conventional crystals. Considerable progress towards answering these questions has already been made by formulating the elastic energies,[3–5] hydrodynamic equations of motion,[6] and descriptions[5] of dislocations for quasicrystals. To understand a number of the most important macroscopic properties (e.g., plastic flow) we also need a theory of the *motion* of dislocations. In this paper we present such a theory and use it to show that dislocation mobilities (M_Q) in quasicrystals are dramatically smaller than those (M_c) in conventional crystals.

To state our results in detail, some definitions are in order. Consider a single straight-line dislocation with a Burgers vector of magnitude b in a crystal or quasicrystal sample of typical linear dimension L subject to a shear stress σ. For fundamental—i.e., the smallest possible—dislocations, b is of order the "unit-cell" size ($a \sim 5$ Å). The force per unit length due to the applied stress is $F_D = \sigma b$, and the mobility M is defined by

$$v_D = M F_D \ , \tag{1}$$

where v_D is the velocity with which the dislocation moves in response to the force. Our main result is that for fundamental dislocations in quasicrystals

$$M_Q \sim \frac{\Gamma_w}{a^2} \left\{ \ln \left[\min \left[\frac{L}{a}, \frac{L_d}{a}, \frac{K}{\sigma} \right] \right] \right\}^{-1} , \tag{2}$$

while for gliding dislocations in conventional crystals,

$$M_c \sim \eta_s^{-1} \ , \tag{3}$$

where Γ_w [see Eq. (17)] is a phason kinetic coefficient (expected to be of order D_v/K, where D_v is a vacancy diffusion constant and K a shear modulus), L_d the mean distance between dislocations, and η_s a shear viscosity.

Estimating D_v, η_s, and K by their values in conventional crystals leads to exceedingly small dislocation mobility in quasicrystals—from 10^5 to 10^{10} times smaller than the mobility for gliding dislocations in conventional crystals. To use some jargon, there is *no* glide direction for dislocations in a quasicrystal. Thus, all macroscopic processes which depend on dislocation motion will be far slower in quasicrystals than in conventional crystals. For example, plastic flow[7]—irreversible distortion by an applied stress—will be effectively prohibited.

This fact should have striking consequences for experiments. For example, crack propagation, a process normally impeded by dissipation due to plasticity in the tip of the crack,[7] will probably be quite easy; as a result, quasicrystals (even perfect ones) should be extremely brittle—quite possibly more brittle than *any* conventional crystalline material (since dislocations are less mobile in them than in any conventional crystalline material). Indeed, this brittleness may be so extreme as to render it difficult, if not impossible, to fabricate and maintain macroscopic quasicrystals. Samples of quasicrystals that have been obtained so far are in fact rather brittle, although this may simply be because they are highly polycrystalline.

Equally intriguing is the experimental observation[8] that quasicrystalline Bragg peaks are rather broad and do not narrow upon annealing at a higher temperature, unlike conventional crystals. In conventional crystals, on the other hand, such annealing is normally rather effective. If the translational disordering is due to quenched dislocations, then the failure to anneal can be interpreted as evidence that these dislocations are virtually immobile, in support of our findings here. That this immobility is not caused by, e.g., impurity pinning or some other effect not related to the unique nature of the quasicrystalline state is ruled out by the observation that the conventional crystalline peaks observed in the *same* polycrystalline samples are quite sharp and amenable to annealing. Further ex-

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periments to demonstrate that this failure to anneal is an intrinsic property of quasicrystals and not a "dirt" effect would be of great interest.

Our results may also have some bearing on recent theories of the glass transition.[9] If we imagine, following Shockley's ideas[10] for conventional crystals, that a quasicrystal could melt via a proliferation of "unbound"[11] dislocation loops, the viscosity of the resultant liquid crystalline phase (which would have residual icosahedral bond orientational order[11]) would be related to the dislocation mobility by[10]

$$\eta \sim \frac{1}{ML_D b^2} , \tag{4}$$

where L_D is the average length of unbound dislocation line per unit volume, and b was defined earlier. Using our results (2) for the mobilities, we see that the viscosity of the liquid crystal obtained by saturating a *conventional* crystal by fundamental ($b \sim a$) dislocation lines (i.e., $L_D a^2 \sim 1$) is comparable to that of the unmelted crystal,[12] while that of the phase ("icosahedratic") obtained in the same manner from a quasicrystal is far larger ($\eta > 10^6$ poise). Thus, this icosahedratic might well be so viscous as to be considered a glass. The relevance of this to recent theories of the glass transition,[9] which view glasses as heavily dislocated and *disclinated* quasicrystals, needs elaboration (since we have not considered the effect of disclinations) but is clearly great.

An important distinction must be made here. We are *not* asserting that the yield stress for dislocations in quasicrystals is unusually large. In fact, we do not even address the issue of the yield stress. To do so would compel us to go beyond the continuum elastic theory used here, and specifically to take detailed account of the spatially varying potential in which dislocations—in quasicrystals as in ordinary crystals—move. We have not attempted such a treatment; however, we see no reason to expect the resulting yield stress in quasicrystals to be particularly larger than in ordinary crystals. Nonetheless, our results imply that once the yield stress—whatever it is—is exceeded, the velocity with which dislocations move in response to this stress is lower, by a factor of M_Q/M_c, than it would be in a conventional crystal subjected to a stress comparably larger than its own yield stress.

It should further be emphasized that our numerical estimates are based upon the assumption that the phason and vacancy diffusion constants D_P and D_V in a quasicrystal are comparable to the vacancy diffusion constant in a conventional, simple periodic crystal. While this seems reasonable, it need not be the case. In particular, because vacancy diffusion is a thermally activated process with a large activation energy (i.e., $D_V \propto e^{-E_a/K_B T}$), relatively small changes in activation energy E_a could lead to enormous changes in D_V. E_a and D_V could quite accurately be estimated for the quasicrystal *i*(Al-Mn-Si) (Ref. 13) using well-developed *ab initio* techniques,[14] by calculating E_a for the cubic crystal α(Al-Mn-Si),[15] which has an almost identical local structure.[16] Such a calculation would be of interest in light of our results. We do not, however, expect D_v to be so large that M_Q becomes comparable to the glide mobility M_c of dislocations in simple conventional crystals.

Our conclusions are based upon two calculations: First, we show that the force on a dislocation due to a uniform externally applied stress is linear in that stress, using the elastic energy presented in Ref. 3. Then we calculate the mobility of the dislocation in response to this force, by applying techniques[17,18] developed for defect motion in liquid crystals and hydrodynamic instabilities to the hydrodynamic equations of motion for quasicrystals presented in Ref. 6.

Before presenting either of these calculations, however, we will briefly review the description of quasicrystals and dislocations therein developed in Refs. 3–6. This point of view considers an icosahedral quasicrystal Q as a system with a spatially inhomogeneous mass density:

$$\rho(\mathbf{r}) = \sum_{\mathbf{G}\in L_R} \rho_{\mathbf{G}} e^{i\mathbf{G}\cdot\mathbf{r}} = \sum_{\mathbf{G}\in L_R} |\rho_{\mathbf{G}}| e^{-i\phi_{\mathbf{G}}+i\mathbf{G}\cdot\mathbf{r}} , \tag{5}$$

where L_R is the reciprocal lattice for Q and $|\rho_G|$ and ϕ_G are, respectively, the amplitude and phase of the density wave at $\mathbf{G}$. L_R can then be constructed by taking linear combinations with integer coefficients of the vectors $\mathbf{G}_n$ in a minimal set B (the basis) containing six elements. The six phases ϕ_n of the complex order parameters ρ_G provide a complete description of the long-wavelength, low-energy mechanical deformations of Q, as can be seen for example from Landau theory.[3–5]

The six vectors $\mathbf{G}_n$, $n = 1,2,\ldots,6$, are conveniently taken as the vectors pointing to the six vertices in the upper half space of an icosahedron. The magnitude G of all six vectors (which are of course equal in length) is of order a^{-1}, where $a \sim 5$ Å is the "unit-cell" size. The six independent phases ϕ_n can be parametrized by two three-component fields $\mathbf{u}$ and $\mathbf{w}$ according to

$$\phi_n = \mathbf{G}_n \cdot \mathbf{u} + \mathbf{H}_n \cdot \mathbf{w} , \tag{6}$$

where

$$\begin{aligned} &\mathbf{H}_1 = -\mathbf{G}_1, \quad \mathbf{H}_2 = \mathbf{G}_2, \quad \mathbf{H}_3 = \mathbf{G}_5 , \\ &\mathbf{H}_4 = \mathbf{G}_3, \quad \mathbf{H}_5 = \mathbf{G}_6, \quad \mathbf{H}_6 = \mathbf{G}_4 . \end{aligned} \tag{7}$$

$\mathbf{G}_1$ is chosen to point through the vertical fivefold axis and $\mathbf{G}_2 \rightarrow$ are arranged counterclockwise around $\mathbf{G}_1$. The fields $\mathbf{u}$ and $\mathbf{w}$ transform under different three-dimensional representations of the symmetry group of the icosahedron, with $\mathbf{u}$ transforming like a vector. $\mathbf{u}$ is just the familiar displacement field and $\mathbf{w}$ the phason field.

The transformation properties of $\mathbf{u}$ and $\mathbf{w}$ under the icosahedral group, along with constraints of translational and rotation invariance and the fact that the ϕ_n's are hydrodynamic allow us to construct the harmonic elastic free energy $F(\mathbf{u},\mathbf{w})$ for the quasicrystal. In Fourier space we find

$$\begin{aligned} F(\mathbf{u},\mathbf{w}) = \tfrac{1}{2} \int_{\mathbf{q}} q^2 [& A_{ij}(\hat{\mathbf{q}}) w_i(\mathbf{q}) w_j(-\mathbf{q}) \\ & + K_{ij}(\hat{\mathbf{q}}) u_i(\mathbf{q}) u_j(-\mathbf{q}) \\ & + C_{ij}(\hat{\mathbf{q}}) u_i(\mathbf{q}) w_j(-\mathbf{q})] , \end{aligned} \tag{8}$$

where $\underline{A}$, $\underline{K}$, and $\underline{C}$ are elasticity tensors which are independent of the magnitude of the wave vector $\mathbf{q}$ and are

nonzero for all directions $\hat{\mathbf{q}}$. Their detailed structure, given in Ref. 5 and the first of Ref. 6, is irrelevant for us here.

Dislocations in this picture are line defects that occur at the boundaries of an integer number of semi-infinite layers inserted into one or more of the six fundamental density waves. Mathematically, this means

$$\oint_P \nabla\phi_\alpha \cdot d\mathbf{l} = 2\pi n_\alpha \ , \tag{9}$$

where the closed path of integration P surrounds the line defect and the n_α's are all integers. Using (6) to express (9) in terms of the fields $\mathbf{u}$ and $\mathbf{w}$, Ref. 3 showed that

$$\nabla \times \nabla \underline{u} = \underline{R}_B \delta(\mathbf{r} - \mathbf{r}_B(s,t)) \ , \tag{10}$$

where the six-component vector

$$\underline{u} \equiv \begin{bmatrix} \mathbf{u} \\ \mathbf{w} \end{bmatrix}$$

and the set of six-component vectors $\underline{R}_B$ form a hypercubic lattice with lattice spacing $\sim a$ (~ 5 Å in real materials) in the six-dimensional space. This hypercubic lattice has the very important property that none of its vectors lies in the $\mathbf{w}=0$ subspace. As a result, all dislocations have some nonzero $\mathbf{w}$ field associated with them. We shall see later that it is because dislocations must "carry" this $\mathbf{w}$ field with them when they move that they are so immobile, since the $\mathbf{w}$ field itself responds very slowly (diffusively, to be precise, and more importantly with a very small diffusion constant).

The exact forms of the fields $\mathbf{u}_D(\mathbf{r})$ and $\mathbf{w}_D(\mathbf{r})$ surrounding a straight, infinitely long dislocation line depend only on the two-component projection $\mathbf{r}_\perp$ of $\mathbf{r}$ perpendicular to the dislocation line and can be found in the usual way by minimizing the free energy [Eq. (8)] subject to the constraint [Eq. (10)].

We will not explicitly do this calculation here, but rather simply note that elementary power counting arguments imply that the spatial Fourier transforms (over $\mathbf{r}_\perp$) $\mathbf{u}_D(\mathbf{q})$ and $\mathbf{w}_D(\mathbf{q})$ can be estimated as

$$u_D(\mathbf{q}) \sim w_D(\mathbf{q}) \sim b/q^2 \ , \tag{11}$$

where $b \sim |R|$. To see this, we note that F has no soft directions; i.e., anisotropic though it is, it involves the same number of gradients for all directions. Thus we expect that up to factors of order 1, $\nabla\mathbf{u}$ and $\nabla\mathbf{w}$ are both roughly constant on a circle centered on the dislocation line. Choosing a circle of radius r as the path P then gives

$$(\nabla u) r \sim b \sim (\nabla w) r$$

or

$$\nabla u \sim \nabla w \sim \frac{b}{r} \ , \tag{12}$$

where we have assumed that the projections of $\underline{R}$ onto the $\mathbf{u}$ and $\mathbf{w}$ subspaces are comparable in magnitude (which is true for the smallest and hence lowest energy and most mobile dislocations). Fourier transforming this result leads immediately to (11), which can also be verified by an explicit calculation.

We will now turn to the calculation of the Peach-Koehler force. Using similar simple power counting arguments, we can show that, as in conventional crystals, the Peach-Koehler force per unit length F_D on a straight dislocation line is proportional to the applied stress and the (six-dimensional) Burgers vector $\underline{R}$, i.e.,

$$F_D = \underline{A} \, \underline{\sigma} \, \underline{R} \ , \tag{13}$$

where the tensor $\underline{A}$ depends only on the direction $\hat{\mathbf{n}}$ along the dislocation line and is dimensionless with entries of order unity. Only uninteresting details of the form of $\underline{A}$ distinguish quasicrystals from conventional crystals in this respect.

The reason for this similarity is that in both quasicrystals and conventional crystals the elastic energies are quadratic functions of gradients of the fields (with no "soft" directions), and furthermore, dislocations are defined in essentially the same way in both [as comparison of Eq. (10) to the definition of a Burgers vector in conventional crystals should make clear].

Dislocation mobilities, on the other hand, depend on the dynamics of quasicrystals, which differ fundamentally from those of conventional crystals. We turn now to the calculation of these mobilities.

We will do this using the approach developed in Refs. 17 and 18. This essentially involves equating the work done by the force on the dislocation to the rate of energy dissipation in the field of the moving dislocation. It can be verified *a posteriori* that this energy dissipation is dominated by the direct dissipation of the elastic energy in the $\mathbf{u}$ and $\mathbf{w}$ fields. Thus, we have

$$\mathbf{F}_D \cdot \mathbf{v}_D = \frac{d}{dt} \int d^2r \, H_{el} = \int d^2r \left[\frac{\delta H}{\delta \mathbf{u}} \cdot \partial_t \mathbf{u} + \frac{\delta H}{\delta \mathbf{w}} \cdot \partial_t \mathbf{w} \right] \ , \tag{14}$$

where the left-hand side is the rate at which work is done by the Peach-Koehler force $\mathbf{F}_D$, the right-hand side is the rate at which it is dissipated in the elastic fields, and the integral is over the two-dimensional plane orthogonal to the (presumed straight) dislocation line.

Note that here we are only calculating the contribution to the drag on the dislocation of the fields far from the dislocation core (where hydrodynamics is valid). This far-field contribution—which is comparable for climb and glide—provides a lower bound on the total drag and hence an upper bound on the mobility since it ignores additional nonhydrodynamic processes taking place in the dislocation core. The latter are presumably responsible for the empirical fact that climb is *much* more difficult than glide in *crystals*. This point of view is supported by the experimental observation[19] that, in convective structures, where no slow process analogous to atomic rearrangements in the core is required for climb, the mobilities for climb and glide of dislocations are comparable, as a theory like that presented here predicts. We believe that this far-field contribution provides a good estimate for the total drag on a *gliding* dislocation since glide does not require elaborate atomic rearrangements within the core.

The fields of a *moving* dislocation must be determined from the linearized equations of motion[6] which read, ignoring inessential complications,

$$\partial_t \mathbf{g} = -\frac{\delta H}{\delta \mathbf{u}} + \underline{\eta}\nabla^2 \mathbf{g}/\rho_0 , \tag{15}$$

$$\partial_t \mathbf{u} = \frac{\mathbf{g}}{\rho_0} - \Gamma_u \frac{\delta H}{\delta \mathbf{u}} , \tag{16}$$

$$\partial_t \mathbf{w} = -\Gamma_w \frac{\partial H}{\partial \mathbf{w}} , \tag{17}$$

coupled with the condition (10), where $\mathbf{r}_B(t)=v_D t$, $\mathbf{g}$ is the momentum density, ρ_0 the equilibrium mass density, Γ_u and Γ_w are kinetic coefficients whose magnitudes we will estimate later, and $\underline{\eta}$ the (isotropic) viscosity tensor.

If one considers the case in which the velocity $\mathbf{v}_D$ of the dislocation is small (compared to the sound speeds in the quasicrystal), the inertial ($\partial_t \mathbf{g}$) term in Eq. (15) can be neglected. We thus arrive at the condition

$$\frac{\eta}{\rho_0}\nabla^2 \mathbf{g} = \frac{\delta H}{\delta \mathbf{u}}$$

or

$$\frac{\mathbf{g}}{\rho_0} = \frac{1}{\eta\nabla^2}\frac{\delta H}{\delta \mathbf{u}} . \tag{18}$$

Thus, (16) and (17) become

$$\partial_t \mathbf{u} = -\left[\Gamma_u - \frac{1}{\eta\nabla^2}\right]\frac{\delta H}{\delta \mathbf{u}} , \tag{19}$$

$$\partial_t \mathbf{w} = -\Gamma_w \frac{\delta H}{\delta \mathbf{w}} . \tag{20}$$

Now if we seek a solution to the coupled sets of Eqs. (10), (19), and (20) of the form

$$\mathbf{u}(\mathbf{r},t)=\mathbf{u}(\mathbf{r}-\mathbf{v}_D t)$$

and

$$\mathbf{w}(\mathbf{r},t)=\mathbf{w}(\mathbf{r}-\mathbf{v}_D t) ,$$

we get

$$\partial_t \mathbf{u} = -(\mathbf{v}_D\cdot\nabla)\mathbf{u} = -\left[\Gamma_u - \frac{1}{\eta\nabla^2}\right]\frac{\delta H}{\delta \mathbf{u}} , \tag{21}$$

$$\partial_t \mathbf{w} = -(\mathbf{v}_D\cdot\nabla)\mathbf{w} = -\Gamma_w \frac{\delta H}{\delta \mathbf{w}} . \tag{22}$$

We can now use these conditions to rewrite (14) entirely in terms of $(\mathbf{v}_D\cdot\nabla)\mathbf{u}$ and $(\mathbf{v}_D\cdot\nabla)\mathbf{w}$,

$$\mathbf{F}_D\cdot\mathbf{v}_D = \int d^2r \left[\frac{1}{[\Gamma_u - (1/\eta\nabla^2)]}[(\mathbf{v}_D\cdot\nabla)\mathbf{u}]^2 + \frac{1}{\Gamma_w}[(\mathbf{v}_D\cdot\nabla)\mathbf{w}]^2\right] , \tag{23}$$

or, Fourier transforming in space,

$$\mathbf{F}_D\cdot\mathbf{v}_D = \int d^2q \left[\frac{\eta q^2}{1+\eta\Gamma_u q^2}|\mathbf{u}_q|^2 + \frac{1}{\Gamma_w}|\mathbf{w}_q|^2\right](\mathbf{v}_D\cdot\mathbf{q})^2 . \tag{24}$$

Note that the right-hand side of this equation is proportional to v_D^2, while the left is proportional to v_D. Choosing, for simplicity, a direction (call it x) in which $\mathbf{F}_D$ and $\mathbf{v}_D$ are parallel, we can solve for v_D in terms of F_D, obtaining Eq. (1) with

$$M^{-1} = \int d^2q \left[\left[\frac{\eta q^2}{1+\eta\Gamma_u q^2}\right]|\mathbf{u}_q|^2 + \frac{1}{\Gamma_w}|\mathbf{w}_q|^2\right]q_x^2 . \tag{25}$$

Consider only the term involving w. Equation (22) tells us that for length scales smaller than $L_v=\Gamma_w K/v_D$, where K is a typical shear modulus in F, $\mathbf{w}_q$ can be approximated by its static value which is $\sim a/q^2$ for an elementary dislocation. Thus, apart from dimensionless factors of order unity, the contribution M_w^{-1} of $\mathbf{w}$ to the dissipation M^{-1} is

$$M_w^{-1} = \frac{a^2}{\Gamma_w}\ln\left[\frac{L_v}{a}\right] .$$

This is readily shown to dominate completely the contribution of the u term, which is $M_u^{-1}\sim\eta\sim 1$ poise, while estimating Γ_w by $D_v/K\sim 10^{-22}$ cm^{3}sec/g we obtain $M_w^{-1}\sim 10^6$ poise. We shall therefore ignore M_u^{-1}, so that the mobility

$$M \approx M_w = \frac{\Gamma_w}{a^2}\left[\ln\left[\frac{K}{\sigma}\right]\right]^{-1} , \tag{26}$$

where in the argument of the logarithm we have used this mobility to express the dislocation velocity in terms of $\mathbf{F}_D$ and hence, ultimately, the applied stress σ.

The infrared cutoff in the wave-vector integral (25) should clearly be the inverse of the smallest length at which the w field departs appreciably from its configuration in the presence of a single, isolated stationary dislocation line. We have already pointed out that this length is bounded above by L_v. Should either the sample dimension L or the mean distance to the nearest dislocation L_d be smaller than L_v, the static, single dislocation approximation to w will clearly first break down at the smaller of those lengths, which would therefore become the infrared cutoff. Thus, the appropriate infrared cutoff in any given situation is the smallest of these three lengths; Eq. (2) follows immediately.

The glide mobility of *ordinary* crystals is just given by the u part of Eq. (25) since there is no w field in conventional crystals. (Climb motion, as mentioned before, is much slower because of complex processes in the core.) The $\mathbf{q}$ integral for this part diverges in the ultraviolet, cutting it off at $q\sim a^{-1}$ and recalling that $b\sim a$ for fundamental dislocations leads immediately to our expression for the glide mobility, Eq. (3).

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