Elastic and anelastic behaviour of icosahedral quasicrystals

R. E. BOLMARO*

International Centre for Theoretical Physics, (34100) Trieste, Miramare, Strada Costera 11, Italy

F. POVOLO

Comisión Nacional de Energía Atómica, Dpto. de Materiales, Av. del Libertador 8250, (1429) Buenos Aires, Argentina. Universidad de Buenos Aires, Facultad de Ciencias Exactas y Naturales, Dpto. de Física, Pabellòn 1, Ciudad Universitaria, (1428) Buenos Aires, Argentina

A theory is developed, based on theoretical-group analysis, to describe the linear, reversible, time-dependent response of an icosahedral quasicrystal, containing point defects, to a stress field and known as anelastic relaxation. We obtain also anelastic relaxation relationships for the practical Young's, shear and Poisson's moduli.

1. Introduction

Several workers have produced icosahedral and decagonal quasicrystals [1-3] by rapid solidification and many of them have used the composition of, approximately, 14 at % Mn, corresponding to the formula Al₆Mn. In addition, there is now a rapidly expanding list of reports of quasicrystalline phases in different alloy systems produced by different processing methods. Only in Al-Mn and closely related systems, however, has enough work been done to form a reasonably detailed understanding of the physical metallurgy of the quasicrystalline phases. Notwithstanding only some models about the actual atomic distribution have been made [1, 4, 5] and new experimental works will be necessary to decide the ones which are correct. In this way the anelastic behaviour would be one of the phenomena to research in future. In this paper we look for the more interesting elastic and anelastic properties of icosahedral quasicrystals. The point defect anelastic relaxation will be surely used to find the actual location of different kinds of atoms. The model proposed by Guyot and Audier [4] assumed a skeleton building from an inner aluminium icosahedron and an outer manganese icosahedron. Moreover, we have to put some spare aluminium atoms at the middle of the axis of the outer icosahedron and we need to replace some aluminium and manganese atoms by each other. The details of such structure would be clarified by internal friction methods.

The generalized Hooke's law can be expressed, in terms of the commonly used single index notation, as [6]

$$\sigma_i = \sum_{j=1}^{6} c_{ij} \varepsilon_j \quad i, j = 1, ..., 6$$
 (1)

where c_{ii} are the elastic stiffness constants.

In terms of the elastic compliances

$$\varepsilon_i = \sum_{j=1}^6 s_{ij} \sigma_j \quad i, j = 1, \ldots, 6$$
 (2)

The number of different elastic stiffnesses or elastic compliances stays between a maximum of 21 and a minimum of 2 for isotropic solids. Still further simplifications of Hooke's law for crystals can be made if, instead of the usual components of stress and strain, six independent linear combinations of these are chosen, which possess certain fundamental symmetry properties associated with the crystal in question. These linear combinations, which are known as symmetry coordinates of stresses and strains, or as symmetrized coordinates, are obtained by means of group theory [7]. The symmetrized coordinates are classified as Type I and Type II. The special feature of strains of Type I is that a crystal subjected to such a strain is not lowered in symmetry by the deformation. On the other hand, a crystal under a Type II strain is lowered in symmetry. Furthermore, whenever a symmetrized stress is decoupled from all the symmetrized strains, except the one which corresponds to it, Hooke's law reverts to the simple form

$$\varepsilon_{\gamma} = s_{\gamma}\sigma_{\gamma} \tag{3}$$

where γ denotes the symmetry designation and s_{γ} is the appropriate symmetrized compliance. For lower symmetry crystals decoupling occurs less frequently until finally, for the triclinic case, all six stresses and strains are of Type I and a set of completely coupled equations is obtained. The reason is that triclinic crystals show no symmetry and there is no simplification of Hooke's law as a consequence of symmetry considerations.

The icosahedral quasicrystals have point group symmetry $m\overline{35}$ (Fig. 1) which is inconsistent with

^{*}Permanent address: Instituto de Fisica Rosario, Facultad de Ciencias Extas y Ingenieria, CONICET-UNR, Av. Pellegrini 250, (2000) Rosario, Argentina.



Figure 1 Stereographic projection of the symmetry elements of the icosahedral group $m\overline{35}$.

lattice translations. However icosahedral symmetry means that all the average properties, but not the actual locations of the atomic species, are invariant through the icosahedral symmetry operations. If one specimen is rotated through the angles of this point group, selected area electron diffraction patterns clearly display the six five-fold, ten three-fold and fifteen two-fold axes characteristic of icosahedral symmetry [2].

The elastic properties and hydrodynamic modes of icosahedral structures and Penrose lattices have been discussed elsewhere [8]. In principle, the phason modes (or internal rearrangement modes) may be overdamped and not propagate. The phasons might be observable in an inelastic neutron scattering experiment and the existence of three phason branches was discovered independently by Kalugin *et al.* [9]. In this paper we will assume only a simple elastic behaviour and we will deal with the fourth-rank tensor of elastic constants. We will reduce the independent elastic constants by symmetry arguments and we will use them to deal with the anelastic properties due to point defects.

2. Elastic properties

We assume an icosahedral crystal, as we can see in Fig. 2, with a five-fold symmetry axis along the z axis. The x-y plane transforms to a ξ , η complex plane by the following

$$\xi = x + iy \tag{4}$$

$$\eta = x - iy$$



Figure 2 Icosahedral quasicrystal "cell" and Euler's angles.

It is easy to see that the new coordinates transform by

$$\xi \to \exp(2\pi i/5)$$

 $\eta \to \exp(-2\pi i/5)$ (5)
 $z \to z$

As in the hexagonal symmetry

$$c_{zzzz}, \quad c_{\xi\eta\xi\eta}, \quad c_{\xi\xi\eta\eta}, \quad c_{\xi\etazz}, \quad c_{\xiz\eta z} \tag{6}$$

will not be zero.

One of the two-fold axis, perpendicular to the z axis interchange

$$z \rightarrow -z$$

which do not affect the elastic constants.

Now, if we transform the elastic constants by

$$\Theta = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos \theta & -\sin \theta \\ 0 & \sin \theta & \cos \theta \end{bmatrix}$$
(7)

where

$$\cos \theta = \sqrt{\frac{3}{3}} \left(\frac{2+\tau}{2-\tau}\right)^{1/2} \tag{8}$$

$$= 37.377$$
 (9)

and

$$\tau \stackrel{\cdot}{=} \frac{1+\sqrt{5}}{2}$$

is the golden number [10], we obtain the fourth-rank tensor for the new x, y, z axis [Fig. 2].

where $a = \cos \theta$, $b = \sin \theta$ and $c_{66} = (c_{11} - c_{12})/2$.

With a two-fold axis perpendicular to the z' axis the c'_{ijkl} have to have at least the trigonal symmetry, and the following relationships can be obtained

$$(a^{4} - 1)c_{11} + 2a^{2}b^{2}c_{13} + 4a^{2}b^{2}c_{44} + b^{4}c_{33} = 0$$
(11a)

$$a^{2}b^{2}c_{11} - 2a^{2}b^{2}c_{13} + [(a^{2} - b^{2}) - a^{2}]c_{44} + a^{2}b^{2}c_{33} - a^{2}c_{66} = 0$$
(11b)

$$a^{2}b^{2}c_{11} + b^{2}(b^{2} - a^{2})c_{13} - 4a^{2}b^{2}c_{44} + a^{2}b^{2}c_{33} - a^{2}c_{12} = 0$$
(11c)

$$b^{3}a c_{11} + ab (a^{2} - b^{2})c_{13} + 2ab(a^{2} - b^{2})c_{44}$$

$$a^{3}bc_{33} = 0$$
 (11d)

$$a^{3}b c_{11} - 2ba^{3}c_{13} + 2ab (b^{2} - a^{2})c_{44}$$

- $b^{3}ac_{33} + abc_{12} = 0$ (11e)

Starting from Equations 11, it can be shown that

$$c_{11} - c_{13} - c_{33} + c_{12} = 0$$
 (12)

$$c_{13} - c_{44} + c_{66} - c_{12} = 0 \tag{13}$$

$$2 c_{44} - c_{11} + c_{12} = 0 \tag{14}$$

Then, there are only two independent elastic constants as in isotropic solids. We have to take into account that all the average behaviours, but not the local properties, are invariant through the icosahedral symmetry group.

Following Nowick and Heller [7] we will consider a basis vector made up of the components of the stress field which we wish to reduce to appropriate linear combinations known as "symmetry coordinates of stress". Weyl [11] has dealt in a general way with the problem of the transformation of tensor components when they are to be represented as components of a vector in a hyperspace. The conversion from 2-index to 1-index notation is shown to be

$$\sigma_{11} \rightarrow \sigma_1$$

$$\sigma_{22} \rightarrow \sigma_2$$

$$\sigma_{33} \rightarrow \sigma_3$$

$$\overline{2} \sigma_{23} \rightarrow \sigma_4$$

$$\overline{2} \sigma_{31} \rightarrow \sigma_5$$

$$\overline{2} \sigma_{12} \rightarrow \sigma_6$$
(15)

The same conversion applies to the strain components $\varepsilon_{ij} \rightarrow \varepsilon_k$.

In terms of the Weyl components forming a 6-vector $\bar{\sigma}$, we write the transformation into "symmetry coordinates" as

$$\bar{\sigma}' = V\bar{\sigma}$$

 $\bar{\epsilon}' = V\bar{\epsilon}$ (16)

where V is a 6 \times 6 unitary matrix which performs the desired reduction. The specific form of V required to reduce $\bar{\sigma}$ to symmetry coordinates depends on the point group symmetry of the crystal. The result is obtained using the fact that V must be unitary, plus information contained in the usual character tables [12]. The character table of the icosahedral point

TABLE I Character table of the icosahedral group Y

	Ε	12C ₅	$12C_{5}^{2}$	$20C_{3}$	$15C_{2}$	
A	1	1	1	i	1	$x^2 + y^2 + z^2$
T_1	3	τ	τ^{-1}	0	-1	(x, y, z)
T_2	3	τ^{-1}	τ	0	- 1	
G	4	- l	-1	1	0	
H	5	0	0	- 1	1	

group is shown in Table I. In such a way we obtain the V matrix as

The symmetry coordinates are totally decoupled to each other. We obtained them by

$$\sigma_{a,v}^{\prime(y)} = \sum_{k} V_{a,v,k}^{(y)} \sigma_k \qquad (18)$$

where γ , *a* and *v* denote the irreducible representation (i.r.), the degeneracy of γ and a repeat index to cover the case where a given i.r. appears more than once in the reduction to symmetry coordinates. The following six symmetry coordinates of stress and correlated strains are obtained

$$\bar{\sigma}' = \begin{pmatrix} (1/\sqrt{3}) (\sigma_{11} - \sigma_{22} - \sigma_{33}) & A \\ (1/\sqrt{6}) (2\sigma_{11} - \sigma_{22} - \sigma_{33}) \\ (1/\sqrt{2}) (\sigma_{22} - \sigma_{33}) \\ \sqrt{2} \sigma_{33} \\ \sqrt{2} \sigma_{31} \\ \sqrt{2} \sigma_{12} \end{pmatrix} H (19)$$

In mentioning the i.r.'s it is important to give special attention to the particular i.r. which is totally symmetric, i.e. representing complete invariance under the group operations. Such an i.r. occurs for every point group and is the first one listed in the character table. The quantity $\sigma^{\prime(A)}$ is proportional to the hydrostatic stress while $\sigma_a^{\prime(H)}$ represents the shear stresses.

The symmetry coordinates are orthogonal to each other, in view of the unitary property of the V matrix. In a particular coordinate system it is always possible to select a combination of stresses which corresponds to a single symmetry coordinate by choosing the combination of stresses which makes all symmetry coordinates, except the one of interest, equal to zero.

The representation structure Γ (σ ') of our stress hypervector is

$$\Gamma(\sigma') = A + H \tag{20}$$

where A and H are the stress-active i.r's.

In terms of the Weyl vector components, Hooke's

TABLE II Symmetrized stresses, strains and compliances of icosahedral quasicrystals

A	$\sigma_1 + \sigma_2 + \sigma_3$	$s_{11} + 2s_{12}$	$\varepsilon_1 + \varepsilon_2 + \varepsilon_3$
H	$2\sigma_1 - \sigma_2 - \sigma_3$	$s_{11} - s_{12}$	$2\varepsilon_1 - \varepsilon_2 - \varepsilon_3$
	$\sigma_2 - \sigma_3$	$s_{11} - s_{12}$	$\varepsilon_2 - \varepsilon_3$
	σ_4	$s_{11} - s_{12}$	£ _{4/2}
	σ_5	$s_{11} - s_{12}$	£ _{5/2}
	σ_6	$s_{11} - s_{12}$	$\varepsilon_{6/2}$

law relating the stress and strain components takes the form

$$\bar{\varepsilon} = S\bar{\sigma}$$
 (21)

$$\varepsilon_k = \sum_{l=1}^6 \sigma_{kl} \sigma_l \tag{22}$$

where σ_{kl} are related with the usually defined coefficients s_{kl} [6] by

$$s_{kl} = s_{kl} \quad \text{for } k \text{ and } l \text{ both } \leq 3$$

$$s_{kl} = 1/2s_{kl} \quad \text{for } k \text{ and } l \text{ both } \geq 4 \quad (23)$$

$$s_{kl} = \frac{1}{-1} s_{kl} \quad \text{for } k \leq 3, \ l \geq 4 \text{ or}$$

$$v_{kl} = \frac{1}{\sqrt{2}} s_{kl}$$
 for $k \leq 3, l \geq 4$ or $v_{kl} = 1$

The symmetry coordinates of stress and strain must be also linearly related. Let us express this by

$$\tilde{\varepsilon}' = S \, \bar{\sigma}'$$
 (24)
where $S = VSV^{\dagger}$.

Thus the symmetry elastic compliances are

$$S_{11} = s_{11} + 2 s_{12}$$
 (25)

$$S_{22} = S_{33} = S_{44} = S_{55} = S_{66} = s_{11} - s_{12}$$
(26)

and we obtain the results shown in Table II.

The first symmetrized coordinate is a hydrostatic stress and does not change the symmetry of the crystal. The others belong to the Type II symmetrized coordinates and they change the symmetry of the quasicrystal, reducing it.

3. Point defect anelastic relaxation

The point defects in an icosahedral quasicrystal have to belong to the point groups

$$\mathbf{D}_{5d}, \ \mathbf{D}_{3d}, \ \mathbf{D}_{2h}, \ \mathbf{C}_{2h} \ and \ \mathbf{C}_{i}$$
 (27)

All of which are subgroups of the icosahedral point group Y. Following Nowick and Heller [7] again the point symmetry of the point defects are called the defect groups, designated by \mathscr{G}_d and of order h_d . The order of the defect group must be a submultiple of the order of the crystal h_x and

$$n_{\rm d} = h_{\rm x}/h_{\rm d} \tag{28}$$

represents the number of configurations or orientations of the defect which are physically distinguishable from each other, but are crystallographically equivalent in the original lattice.

In the same way we define

$$n_{\rm t} = h_{\rm x}/h_{\rm t} \tag{29}$$

where h_1 is the order of the tensor group \mathscr{G}_1 determined

by \mathscr{G}_d , \mathscr{G}_x and the type of field under consideration. The point groups (Equation 27) are the tensor groups of the defects. A crystal containing a given defect will undergo relaxation in the presence of a particular type of external field if and only if $n_t > 1$. For an elasticity the n_t criterion can be converted into the following simple rule. An elastic relaxation may be produced by any defect whose symmetry belongs to a lower system than that of the crystal. The icosahedral quasicrystal is isotropic and all the point defects can produce relaxation.

The set of concentrations of defects, C_p , in the various orientations, $p = 1, 2, \ldots n_d$, are the components of a hypervector \bar{C} in a space of dimension n_d . This basis vector can be reduced by an $n_d \times n_d$ unitary transformation matrix, which takes \bar{C} into the vector \bar{C}' whose components are symmetry coordinates

$$\bar{\boldsymbol{C}}' = \boldsymbol{W}\bar{\boldsymbol{C}} \tag{30}$$

In a crystal containing defects of a given symmetry a symmetrized compliance constant can undergo relaxation only if there exist symmetry coordinates of concentrations belonging to the same i.r. y. In view of this selection rule, it is only necessary to know the representation structure of the concentration, $\Gamma(C)$, in order to determine which coefficients of the crystal may undergo relaxation due to the presence of the defects. A simpler method, making use of the "correlation theorem" [12], avoids the necessity to examine the effect of each symmetry operation on the defect orientations. The theorem states that if γ is an i.r. of \mathscr{G}_{x} , the quantity n_{γ} , the number of times that the i.r. γ appears in $\Gamma(C)$, is equal to the number of times that the A_1 representation appears when γ is reduced in \mathcal{G}_d . In order to use the correlation theorem one could carry out the reduction of γ in \mathscr{G}_d with the aid of the character tables for both groups \mathscr{G}_x and \mathscr{G}_d [\mathscr{G}_t]. Table III shows the results for the different point defect groups. We can see that, as we hoped, all the possible point defects undergo relaxation. There is one amendment to these selection rules if only a simple species of defects is present in a total concentration ΣC_{p} that remains constant. This condition implies that we do not permit diffusion of defects into and out of the crystal, or a "reaction" whereby ΣC_p may change at expense of the concentration of another type of defect. Accordingly, an auxilliary condition is imposed on the problem namely

$$\sum_{p} C_{p} = C_{0} = \text{constant}$$
(31)

where C_0 is the total defect concentration in the crystal. This auxilliary condition forbids any change in the symmetry elastic compliance $s_{11} + 2 s_{12}$ as the result of an applied field. Due to the particular spatial structure of the icosahedral quasicrystals it is slightly probable to find near-neighbour sites with the same point group symmetry. We may expect that the $s_{11} + 2 s_{12}$ compliance undergoes relaxation as a result of atom jumps between different species of sites. Moreover we expect the number of configurations of each defect, n_d , to be different in an actual icosahedral quasicrystal. It is clear that more work is necessary to elucidate this fact.

TABLE III The quantity n_i for various defect symmetries and results of the "correlation theorem"

	D _{5d}	D_{3d}	D _{2h}	C _{2h}	\mathbf{C}_{i}
h _d	20	12	8	4	2
n _t	6	10	15	30	60
A	A_{lg}	A_{lg}	A_{g}	$A_{ m g}$	$A_{ m g}$
Н	$E_{lg} + E_{2g} + A_{lg}$ $A + H$	$2\tilde{E}_{g} + A_{lg}$ A + H	$\frac{2A_g}{A} + \frac{B_{1g}}{B} + \frac{B_{2g}}{B} + \frac{B_{3g}}{B}$	$\frac{3A_g + 2B_g}{A + 3H}$	$5A_g$ A + 5H

4. Anelastic relaxation of practical moduli

In order to generalize the equations of elasticity of crystals to allow for time-dependent effects, the validity of the standard anelastic solid model will be accepted for each symmetrized coordinate decoupled one for another (13). In this case Equation 3 leads to

$$\varepsilon_{\gamma} + \tau_{\sigma\gamma}\dot{\varepsilon}_{\gamma} = s_{\gamma r}\sigma_{\gamma} + \tau_{\sigma\gamma}s_{\gamma u}\dot{\sigma}_{\gamma}$$
 (32)

where r and u denote relaxed and unrelaxed compliances, respectively, and $\tau_{\sigma\gamma}$ is the relaxation time at constant stress.

For a time dependent sinusoidal longitudinal stress along the X' axis and an analogue response for the strain, that is

$$\sigma_1' = \sigma_{01} \exp(i\omega t) \tag{33}$$

$$\varepsilon_j = (\varepsilon_j^{(1)} - \varepsilon_j^{(2)}) \exp(i\omega t)$$
 (34)

it can be shown that

$$\varepsilon_{1}' = \frac{1}{3} \left(\frac{s'' + (s'' - \delta_{s'}) \omega^{2} \tau_{\sigma s'}^{2} - i\omega \tau_{\sigma s'} \delta_{s'}}{1 + \omega^{2} \tau_{\sigma s'}^{2}} + \frac{s' + (s' - \delta_{s'}) \omega^{2} \tau_{\sigma s'}^{2} - i\omega \tau_{\sigma s'} \delta_{s'}}{1 + \omega^{2} \tau_{\sigma s'}^{2}} \right) \sigma_{1}'$$
(35)

$$\varepsilon_{2}' = \frac{1}{3} \left(\frac{s'' + (s'' - \delta_{s'}) \, \omega^{2} \tau_{\sigma s'}^{2} - i\omega \tau_{\sigma s'} \, \delta_{s'}}{1 + \omega^{2} \tau_{\sigma s'}^{2}} - \frac{1}{2} \frac{s' + (s' - \delta_{s'}) \, \omega^{2} \tau_{\sigma s'}^{2} - i\omega \tau_{\sigma s'} \, \delta_{s'}}{1 + \omega^{2} \tau_{\sigma s'}^{2}} \right) \sigma_{1}'$$
(36)

where $s'' = s_{11} + 2 s_{12}$, $s' = 2 (s_{11} - s_{12})$ and δ_s is the relaxation of the compliance s.

We can not observe orientation dependence as we hoped. Moreover, the Young's modulus is defined by

$$E_{(\omega)}^{-1} = \frac{\varepsilon_1'}{\sigma_1'} \tag{37}$$

and, if $\delta_{s^*} = 0$, $\tau_{\sigma s'} = 0$, we can obtain the dynamic Poisson's ratio [13–15]

$$v(\omega) = v_1 - iv_2 = -\frac{\varepsilon'_2}{\varepsilon'_1}$$
$$= \frac{v_r + v_u \,\omega^2 \tau^2_{\varepsilon s'} - i\omega \tau_{\varepsilon s'} \,\delta(v)}{1 + \omega^2 \tau^2_{\varepsilon s'}}$$
(38)

where $v_r = (E_r/2G_r) - 1$, $v_u = (E_u/2G_u) - 1$, $\delta(v) = (\frac{1}{2} - v_r)\delta_{(1/E)}$, $\tau_{\varepsilon} = (E_r/E_u)$ τ_{σ} is the relaxation time at constant strain, G is the shear modulus. For a shear stress σ_{23} we obtain

$$G^{-1}(\omega) = J_{1} - iJ_{2}$$

= $\frac{s' + (s' - \delta_{s'}) \omega^{2} \tau_{\sigma s'}^{2} - i\omega \tau_{\sigma s'} \delta_{s'}}{1 + \omega^{2} \tau_{\sigma s'}^{2}}$ (39)

In longitudinal vibration we could compare the experimental results with Equations 37 and 38. In shear tests we may hope a behaviour to be adjusted by Equation 39.

5. Discussion and conclusions

We have shown, by simple arguments from the symmetry of the icosahedral group, that the icosahedral quasicrystals are isotropic under stress fields and the anelastic behaviour can be reduced to the classical properties for isotropic materials.

Also we have shown that all the possible point defects can undergo elastic relaxation and we can expect internal friction phenomena under periodic stresses from them all.

However we have to work out the problem of the actual location and symmetry of the defects in the spatial structure of the whole icosahedral quasicrystal.

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