ON THE THEORY OF DIFFUSION RELAXATION IN POLYCRYSTALS

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The presence of impurities in a crystal lattice is one of the causes of anelasticity of solids. Impurities in single crystals may lead to relaxation of the Snoek [1] and Zener [2] type, and affect the level of dislocation internal friction [3]. In polycrystals there is yet another mechanism of relaxation, i.e., the "uphill" diffusion of impurity atoms in a random elastic stress field produced by the deformation of randomly oriented crystals. This mechanism may contribute to the internal friction, elastic aftereffect and transient creep. Diffusion relaxation of this kind was first analyzed by Zener [4] who, having concluded that the difference between the relaxation and nonrelaxation moduli cannot be accurately calculated, estimated the order of magnitude of this difference; he based this estimation on the calculation of a second-order moment function from the reciprocal of the Young's modulus. An implicit assumption in Zener's model is that the elastic strain of a crystal is determined only by the orientation of its crystallographic axes and on the external applied force field, being independent of the strains of adjacent crystals. The inadequacy of such a model for a polycrystal is evident. Moreover, this approach does not allow to obtain the distribution function of relaxation times. An accurate computation of the intensity of relaxation in polycrystals of an arbitrary crystal symmetry, which takes into account pair correlations between the separate grains, is presented in this article.

1. Formulation of the problem.

Consider a polycrystal which consists of grains of different shape and orientation. If the impurity concentration and strain are assumed to be small, the elasto–diffusion set of equations may be written in the form

$$\nabla_i D_{ik} \nabla_k c - \frac{\partial c}{\partial t} - \frac{V_0}{RT} \nabla_i c D_{ik}^\circ \nabla_k b_{lm} u_{lm} = -q, \quad (1.1)$$

$$\nabla_k \lambda_{iklm} u_{lm} - \nabla_k b_{ik} c = -f_i, \quad (1.2)$$

$$c = n/N, \quad b_{ik} = \partial \sigma_{ik}/\partial c = \lambda_{iklm}\gamma_{lm}, \quad \gamma_{lm} = \partial \varepsilon_{lm}/\partial c,$$

$$D_{ik} = D_{ik}^\circ \left(1 + \frac{\beta V_0}{RT} c\right),$$

$$\beta = \lambda_{iklm}\gamma_{ik}\gamma_{lm}, \quad \varepsilon_{lm} = \tfrac{1}{2}(u_{lm} + u_{ml}). \quad (1.3)$$

Equation (1.1) is the equation of uphill diffusion in a heterogeneous anisotropic medium (for its derivation see appendix), and Eq. (1.2) is the equation of equilibrium taking into account concentration stresses [5, 6]. The summation convention applies, and the symbols appearing in the equation have the following meaning: c is the impurity atom concentration, n and N are the number of, respectively, impurity atoms and all the atoms in unit volume, D_{ik} is the diffusion coefficient, which depends on c and may be quite substantial [7, 8], $D_{ik}^\circ = D_{ik}$ for c → 0, b_{ik} and γ_{ik} are the concentration stress and strain tensors, respectively, V_0 is the molecular volume, R is the gas constant, $u_{ik} = u_{i,k}$ is the distortion tensor, u is the displacement tensor, ε_{ik} is the strain tensor, q characterizes the strength of the source of impurity atoms, and f is the force density.

The elasto–diffusion set of equations (1.1), (1.2) is analogous to the system of thermoelasticity equations, differing from it only in the third term of the left side of the diffusion equation. It would therefore be possible directly to use a method previously developed for the computation of thermoelastic relaxation of polycrystals of cubic symmetry. With lower forms of symmetry, however, direct computation is quite tedious. For this reason the calculation of the frequency dependence of the effective elastic constant tensor is replaced below by the computation of its values at frequencies $\omega \to 0$ and $\omega \to \infty$, which gives directly the relaxation and nonrelaxation values of this tensor and, consequently, the relaxation intensity.

Taking into account that the diffusion coefficient is a kinetic characteristic and does not affect the degree of relaxation, let us replace D_{ik} by its effective value $D\delta_{ik}$ and linearize Eq. (1.1) by substituting for the real concentration c (in the diffusion coefficient D and in the third term) a value c_0 averaged over the polycrystal. Then, for harmonic oscillations this equation may be written in the form

$$Lc = -q + D\chi\nabla^2 b_{lm}u_{lm}$$

$$\left(L \equiv D\nabla^2 - i\omega, \quad \chi = \frac{c_0 V_0 D_0}{RTD}\right). \quad (1.4)$$

Determining the concentration and substituting its value in (1.2), we find

$$L_{il}u_l = -f_i^\circ$$

$$(L_{il} \equiv \nabla_k \lambda_{iklm}\nabla_m + D\chi\nabla_k b_{ik}\nabla^2 g * b_{lm}\nabla_m,$$

$$f_i^\circ \equiv f_i - \nabla_k b_{ik}g * \chi, \quad Lg = -\delta(r)) \quad (1.5)$$

Let us represent tensors λ_{iklm} and b_{lm} as a sum of the average value

$$\langle \lambda_{iklm} \rangle = K\delta_{ik}\delta_{lm} + \mu D_{iklm}, \quad \langle b_{ik} \rangle = b\delta_{ik},$$

$$(D_{iklm} \equiv \delta_{il}\delta_{km} + \delta_{im}\delta_{kl} - \tfrac{2}{3}\delta_{ik}\delta_{lm}),$$

and the fluctuation increment

$$\delta\lambda_{iklm} = \lambda_{iklm} - \langle \lambda_{iklm} \rangle, \quad \delta b_{ik} = b_{ik} - \langle b_{ik} \rangle. \quad (1.7)$$

It should be noted that averaging is carried out over regions whose size is very much larger then the grain size but small in comparison with the distance over which there is a marked variation in the regular part of the function. Then, the operator L_{il} is split into regular Q_{il} and random R_{il} components

$$Q_{il} = \nabla_k \langle \lambda_{iklm} \rangle \nabla_m + D\chi\nabla_k \langle b_{ik}\nabla^2 g * b_{lm} \rangle \nabla_m, \quad (1.8)$$

$$R_{il} = \nabla_k \delta\lambda_{iklm}\nabla_m + D\chi b \times$$

$$\times (\nabla_k \delta_{ik}\nabla^2 g * \delta b_{lm}\nabla_m + \nabla_k \delta b_{ik}\nabla^2 g * \delta_{lm}\nabla_m)$$

$$(Q_{il} + R_{il} = L_{il}). \quad (1.9)$$

Equating expression (1.8) for the regular component

$L_i l$ of the operator to the equilibrium equation

$$\sigma_{ik,k} = -f_i^\circ, \qquad (1.10)$$

we find the tensor operator of the elastic moduli of the polycrystal

$$\Lambda_{iklm}(\omega) = \langle \lambda_{iklm} \rangle + D\chi \langle b_{ik} \nabla^2 g * b_{lm} \rangle. \qquad (1.11)$$

Hence, for the defect of the elastic moduli tensor $\Delta\Lambda_{iklm}$ in the zeroth approximation we find

$$\Delta\Lambda_{iklm}^{(0)} \equiv \Lambda_{iklm}^{(0)}(\infty) - \Lambda_{iklm}^{(0)}(0) = \chi b^2 \delta_{ik}\delta_{lm} + {}^1/_{10}\chi A_{pq}^{pq} D_{iklm}$$

$$(A_{pq}^{pq} \equiv \langle \delta b_{pq}^2 \rangle). \qquad (1.12)$$

Here the first term gives the modulus defect due to diffusion through distances l which are of the order of the strain field heterogeneities. The second term is associated with diffusion through distances a of the order of the grain size.

In the former case the relaxation time is quite large $(\sim l^2/D)$, while in the latter case it is substantially smaller $(\sim a^2/D)$.

It should be noted that in the case of uniform cyclic deformation the first term in (1.12) should be assumed equal to zero. In fact, the stress tensor in this case is equal to

$$\sigma_{ik} = \langle \lambda_{iklm} \rangle u_{lm} + D\chi u_{lm}(b^2\delta_{ik}\delta_{lm}\nabla^2 g * 1 + b_{lm}^{ik}\nabla^2 g * 1)$$

$$[b_{lm}^{ik}(\mathbf{r} - \mathbf{r}') \equiv \langle \delta b_{ik}(\mathbf{r})\delta b_{lm}(\mathbf{r}')\rangle]. \qquad (1.13)$$

Denoting Fourier transforms by capital letters

$$F(\mathbf{k}) = \int e^{i\mathbf{k}\mathbf{r}} f(\mathbf{r})dV, \qquad f(\mathbf{r}) = \frac{1}{8\pi^3}\int e^{-i\mathbf{k}\mathbf{r}} F(\mathbf{k})dV_\mathbf{k} \quad (1.14)$$

and taking into account relations

$$f * 1 = F(0), \qquad G(\mathbf{k}) = (Dk^2 + i\omega)^{-1}, \quad (1.15)$$

we obtain

$$\nabla^2 g * 1 = -(D + i\omega k^{-2})^{-1}_{k=0} = 0. \qquad (1.16)$$

In the case of nonuniform deformation, e.g., in bending, impurity atoms will diffuse from compressed regions to regions of tensile strains. The corresponding defect of the modulus is obtained if it is taken into account that in these circumstances Δ_g^2 is convoluted with the function $u_{ik}(\mathbf{r})$. In the Fourier space as $\omega \to 0$ this leads to the multiplication of $U_{ik}(\mathbf{k})$ by $-D^{-1}$.

It follows from (1.12) that for uniform deformation the correlation part of the defect of the tensor of elastic moduli is determined in the zeroth approximation by correlations of the second-rank concentration stress tensor. As a result, in the case of cubic crystal symmetry the correlation part of the defect of the elastic moduli tensor in the zeroth approximation is equal to zero; in the case of crystals of lower symmetry, it will be manifested in the presence of a shear component of the stress tensor.

2. Calculation of the degree of relaxation in the first approximation.

Taking into account pair corre-

lations leads to the following expression for the regular part of the displacement vector [9]:

$$Q_{il}\langle u_l \rangle + \langle R_{ip}g_{pq} * R_{ql}\rangle\langle u_l \rangle = -f_i^\circ \qquad (2.1)$$

where the Green's tensor function is given by

$$Q_{ip}g_{pq} = -\delta_{iq}\delta(\mathbf{r}). \qquad (2.2)$$

For high frequencies the Fourier transform of the Green's tensor $G_{pq}(\mathbf{k})$ and the random tensor operator R_{il} are equal to

$$G_{pq}(\mathbf{k}) = \frac{1}{\mu k^2}(\delta_{pq} - \varkappa n_p n_q), \qquad R_{il} = \nabla_k \delta\lambda_{iklm}\nabla_m$$

$$\left(n_p \equiv \frac{k_p}{k}, \quad \varkappa \equiv \frac{K + {}^1/_3\mu}{K + {}^4/_3\mu}\right). \qquad (2.3)$$

Substituting (2.3) in (2.1), we obtain

$$\Lambda_{iklm}^{(1)}(\infty) = -\frac{1}{3\mu}A_{lmpq}^{ikpq} + \frac{\varkappa}{15\mu}(2A_{lmpq}^{ikpq} + A_{lmiq}^{ikpp}). \quad (2.4)$$

The correlation tensors A_{lmpq}^{ikpq} and A_{lmiq}^{ikpp} are related to the binary correlation function of the elastic moduli tensor b_{pqrs}^{iklm} by equations

$$\langle \delta\lambda_{iklm}(\mathbf{r})\delta\lambda_{pqrs}(\mathbf{r}')\rangle = b_{pqrs}^{iklm}(\mathbf{r} - \mathbf{r}') = A_{pqrs}^{iklm}\psi(\mathbf{r} - \mathbf{r}').(2.5)$$

Equation (2.4) coincides with the fluctuation increment to the elastic moduli tensor of a polycrystal calculated in [10]. In the second limiting case, setting $\omega = 0$, we find

$$G_{pq}(\mathbf{k}) = \frac{1}{\mu_0 k^2}(\delta_{pq} - \varkappa_0 n_p n_q)$$

$$R_{il} = \nabla_k \delta\lambda_{iklm}\nabla_m - \chi b \nabla_k(\delta_{ik}\delta b_{lm} + \delta b_{ik}\delta_{lm})\nabla_m \quad (2.6)$$

$$\left(\mu_0 \equiv \mu - \frac{1}{10}\chi A_{pq}^{pq}, \quad \varkappa_0 \equiv \frac{K_0 + {}^1/_3\mu_0}{K_0 + {}^4/_3\mu_0}, \quad K_0 \equiv K - \chi b^2\right).(2.7)$$

Substituting (2.6) in (2.7) and (2.1), and performing the appropriate transformations, we find the following expression for the correlation increment to the averaged value of the elastic moduli tensor:

$$\Lambda_{iklm}^{(1)}(0) = -\frac{1}{3\mu_0}\left(1 - \frac{2}{5}\varkappa_0\right)A_{lmpq}^{ikpq} +$$

$$+ \frac{\varkappa_0}{15\mu_0}A_{lmqq}^{ikpp} + \frac{2\chi b}{3K_0 + 4\mu_0}A_{lmpp}^{ik}. \qquad (2.8)$$

Tensor A_{lmpq}^{ik} is given by

$$\langle \delta b_{ik}(\mathbf{r})\delta\lambda_{lmpq}(\mathbf{r}')\rangle = A_{lmpq}^{ik}\psi(\mathbf{r} - \mathbf{r}'). \qquad (2.9)$$

Subtracting (2.8) from (2.4), we find the correlation increment of the first approximation to the defect of the elastic moduli tensor.

$$\Delta\Lambda_{iklm}^{(1)} = \frac{\chi b^2}{15(K + {}^4/_3\mu)^2}(2\Omega_1 A_{lmpq}^{ikpq} + \Omega_2 A_{lmqq}^{ikpp}) -$$

$$- \frac{2\chi b}{3(K + {}^4/_3\mu)}A_{lmpp}^{ik},$$

$$\Omega_1 \equiv 1 + \frac{2}{5b^2}\left(1 + \frac{K}{\mu} + \frac{3K^2}{8\mu}\right)A_{pq}^{pq},$$

$$\Omega_2 = 1 - \frac{4}{15b^2}\left(\frac{1}{6} + \frac{K}{\mu} + \frac{3K}{8\mu^2}\right)A_{pq}^{pq}. \qquad (2.10)$$

For a cubic crystal $\Omega = 1$. For lower symmetry $\Omega > 1$. In the case of hexagonal symmetry for $b_{33} = 1.5b$

$$\Omega_1 = 1 + \frac{4}{15}\left(1 + \frac{K}{\mu} + \frac{3K^2}{8\mu^2}\right), \qquad \Omega_2 = \frac{31}{27} - \frac{2}{3}(\Omega_1 - 1).$$

In the case of zinc, for instance, having calculated K and μ from data in [10], we obtain $K = 6.71$ and $\mu = 2.75 \times 10^{11}$ dyne/cm², which gives $\Omega_1 = 2.5$ $\Omega_2 = 4/27$. The binary correlation tensors in (2.10) are easily calculated with the aid of relations

$$A^{ik}_{lmpp} = \frac{1}{10} A^{rs}_{rspp} D_{iklm}, \qquad A^{ikpp}_{lmqq} = \frac{1}{10} A^{rspp}_{rsqq} D_{iklm},$$

$$A^{ik\,pq}_{lmpq} = \frac{1}{9} A^{rrpq}_{sspq}\delta_{ik}\delta_{lm} + \frac{1}{10}\left(A^{rspq}_{rspq} - \frac{1}{3}A^{rrpq}_{sspq}\right)D_{iklm}. \quad (2.11)$$

The scalar values in the right sides of these equations are easily calculated in the crystallographic coordinate system.

3. Calculating total defects of the omnidirectional compression and shear moduli. Using the relation

$$\triangle K = \frac{1}{9}(\triangle\Lambda^{(0)}_{iill} + \triangle\Lambda^{(1)}_{iill}), \qquad (3.1)$$

we find

$$\triangle K = \chi b^2 + \frac{2}{135}\frac{\chi b^2 \Omega_1}{(K + \frac{4}{3}\mu)^2} A^{rrpq}_{sspq}. \qquad (3.2)$$

Here the first term describes relaxation which takes place only in the case of nonuniform deformations. This term will be nonzero for crystals with a symmetry lower than cubic.

The defect of the shear modulus is found from

$$\mu = \frac{1}{10}(\Lambda_{ikik} - 3K). \qquad (3.3)$$

This gives

$$\triangle\mu = \frac{1}{10}\chi A^{pq}_{pq} - \frac{1}{15}\frac{\chi b}{(K + \frac{4}{3}\mu)} A^{rs}_{rspp} + \frac{1}{75}\frac{\chi b^2}{(K + \frac{4}{3}\mu)^2}\left(\Omega_1 A^{rspq}_{rspq} + \frac{1}{2}\left(\Omega_2 - \frac{1}{3}\Omega_1\right)A^{rrpq}_{sspq}\right). \quad (3.4)$$

For a cubic structure, (3.4) simplifies to

$$\triangle\mu = \frac{\chi b^2}{75(K + \frac{4}{3}\mu)^2} A^{rspq}_{rspq} = \frac{18}{125}\frac{\chi K^2\gamma^2 c_3^2}{(K + \frac{4}{3}\mu)^2}. \qquad (3.5)$$

The values γ and c_3 for cubic crystals are given by expressions

$$\lambda_{iklm} = c_1\delta_{ik}\delta_{lm} + c_2(\delta_{il}\delta_{km} + \delta_{im}\delta_{kl}) + c_3\sum_j \delta_{ij}\delta_{kj}\delta_{lj}\delta_{mj},$$

$$\gamma \equiv \frac{1}{3}\gamma_{ii}. \qquad (3.6)$$

4. Calculating the relaxation time distribution functions.
To a given degree of relaxation $\triangle\mu/\mu$ there corresponds an internal friction peak whose height and width are determined by the relaxation time distribution function $f(\tau)$. To calculate $f(\tau)$ it is necessary to consider (1.11) and (2.1) without assuming $\omega = 0$ or $\omega = \infty$.

As shown in [9], the relaxation time distribution function is determined by a coordinate dependence of the corresponding binary correlation functions. Func-

tion $f(\tau)$ is expressed through a function $\Psi(r)$, previously introduced into the Fourier transform $\psi(k)$, in the following way:

$$f(\tau) = \frac{D^{-3/2}}{4\pi^2}\Psi\left(\frac{1}{\sqrt{D\tau}}\right)\tau^{-5/2}, \qquad \int_0^\infty f(\tau)\,d\tau = 1. \qquad (4.1)$$

If the coordination dependence of all the binary correlation functions is taken in the form

$$\psi(r) = \exp(-r/a), \qquad (4.2)$$

where a is of the order of the crystal size, the distribution function is given by

$$f(\tau) = \frac{2a^3 D^{-3/2}}{\pi}\frac{\tau^{-5/2}}{(1 + a^2/D\tau)^2}. \qquad (4.3)$$

In the presence of a distribution function, the height of the internal friction peak is reduced by a factor of two (in comparison with the peak of a standard linear body) and becomes

$$Q^{-1}_{max} = \frac{1}{4}\frac{\triangle\mu}{\mu}.$$

For the purpose of a quantitative estimate let us consider the system Fe − C (of cubic symmetry) with 4.5% C at 1250° C. The elastic constants of α-iron (expressed in 10^{11} dyne/cm²) are: $c_1 = 11.4$; $c_2 = 8.9$; $c_3 = 13.6$. The dimensionless parameter $\gamma = 0.2$ (see [11]). This gives $\Delta\mu/\mu = 1.5 \times 10^{-3}$, hence $Q^{-1}_{max} = 3.7 \times 10^{-4}$. This value corresponds to a frequency $v_m = 0.6Da^{-2}$. Using data cited in [12], we find $D = 3.2 \times 10^{-6}$ sec/cm² at $T = 1250°$ C. For a grain size $a \simeq 10^{-3}$ cm, the frequency corresponding to the internal friction peak will be 1 cps.

These results indicate that the diffusion internal friction of polycrystals can be determined by experiment. The precise magnitude of the internal friction peak may be slightly different, because the peak at 1250° C was estimated on the basis of elastic constants at room temperature and without taking into account the effect of carbon. However, the order of the magnitude of the estimated internal friction peak should be correct.

5. **Appendix.** (Derivation of the equation of uphill diffusion in a heterogeneous anisotropic medium.) For small strains u_{ik} and impurity atom concentration c in a heterogeneous medium, the free energy of unit volume F can be expanded in a series in the parameters u_{ik} and c.

If the expansion is limited to the quadratic approximation, we obtain

$$F = F_0 + \frac{1}{2}\lambda_{iklm}u_{ik}u_{lm} - \lambda_{iklm}u_{ik}\gamma_{lm}c + \frac{1}{2}\lambda_{iklm}\gamma_{ik}\gamma_{lm}c^2$$

$$(c \equiv n/N). \qquad (5.1)$$

Hence we find the chemical potential μ

$$\mu \equiv \frac{\partial F}{\partial n} = \mu_0 - \frac{1}{N}b_{ik}u_{ik} + \frac{1}{N}\beta c,$$

$$\left(\mu_0 \equiv \frac{\partial F_0}{\partial n}, \quad b_{ik} \equiv \lambda_{iklm}\gamma_{lm}, \quad \beta \equiv \lambda_{iklm}\gamma_{ik}\gamma_{lm}\right). \qquad (5.2)$$

The particle flux density J is expressed in the usual way [13] by the chemical potential

$$J_i = -\frac{n}{kT}D_{ij}{}^0\nabla_j\mu, \qquad (5.3)$$

where D^0_{ij} is the diffusion coefficient as $c \to 0$.

Substituting (5.2) in (5.3) and taking into account that for dilute solutions

$$\mu_0 = kT\ln n, \qquad (5.4)$$

we find

$$-J_i = D_{ij}{}^0 \left(1 + \frac{c\beta V_0}{RT}\right) \nabla_j n - \frac{D_{ij}{}^0 c}{kT} \nabla_j b_{lk} u_{lk} \qquad (5.5)$$

Here V_0 is the molecular volume.

The change in the number of particles in unit volume per unit time is

$$\frac{\partial n}{\partial t} = -\nabla_i J_i = \nabla_i D_{ij} \nabla_j n - \nabla_i D_{ij}{}^0 \frac{c}{kT} \nabla_j b_{lk} u_{lk}$$

$$(D_{ij} \equiv D_{ij}{}^0 [1 + c\beta V_0/RT]). \qquad (5.6)$$

Relating this expression to the total number of particles in unit volume, we obtain

$$\frac{\partial c}{\partial t} - \nabla_i D_{ij} \nabla_j c + \frac{V_0}{RT} \nabla_i D_{ij}{}^0 c \nabla_j b_{lk} u_{lk} = 0. \qquad (5.7)$$

In the presence of sources of impurity atoms the equation of uphill diffusion (5.7) will be nonuniform, as in (1.1) in the text. Equation (1.2) is obtained from (5.1) by differentiating F with respect to the strain tensor and substituting the result in (1.10).

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