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Thermal diffuse scattering in polycrystalline materials with a texture

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A simplified method of calculating thermal diffuse scattering (TDS) for materials at or above their Debye characteristic temperature is presented. This method is based upon the properties of the lattice Green's function and eliminates the need of solving the equations of lattice dynamics. This simplifies the calculation procedure. The general scheme of evaluating the diffuse scattering for polycrystalline materials with a texture is discussed. A sample calculation of the TDS from a polycrystalline material with a fiber texture is given using the simplified procedure just presented.

I. INTRODUCTION

X-ray diffuse scattering is an important technique for investigating local atomic arrangements and defects in crystals. Previous quantitative analyses are all based upon single crystals or ideally random powder samples. In reality, most polycrystalline materials have a certain degree of texture or preferred orientation due to sample preparation. Therefore, it is still necessary to develop a quantitative theory to access the importance of texture on the diffuse scattering from these materials. One important kind of diffuse scattering that exists in all materials is the thermal diffuse scattering (TDS). In short range order studies TDS must be subtracted, as a correction, from the total diffuse scattering before other forms of scattering can be analyzed. Traditionally, TDS is calculated from the eigenvalues and eigenvectors of the equations of lattice dynamics,¹ which are cumbersome to compute. In this paper, a simplified method of calculating the TDS is described, based on interatomic force constants and the lattice Green's function. This procedure recognizes the properties of the lattice Green's function and eliminates the need of solving the many equations of lattice dynamics. It can be applied to single crystals at or above the Debye characteristic temperature with only one kind of atom. It can also be used approximately for a random mixture of different kinds of atoms using average parameters. As in Debye's theory, the material is treated as a pure element with average parameters such as an average mass distributed at each lattice point. A calculation of the TDS in a polycrystalline copper beryllium solution having a fiber texture is given using these procedures. Our approach is compared with a random powder calculation using additional simplifying assumptions.² A more general treatment for other kinds of diffuse scattering, using the inverse pole density function,³ is also discussed.

II. TDS FROM LATTICE GREEN'S FUNCTION

Based upon the harmonic approximation, the motion of atoms in a crystal is described by a set of normal modes of vibration. The thermal motion of atoms influences the x-ray diffraction pattern in two ways: (i) Bragg reflections are at-

tenuated; and (ii) this loss of intensity produces diffuse scattering that strongly depends upon the position of the diffraction vector relative to the Bragg positions.

In Born-von Karman theory, the interatomic force acting on an atom is given by

$$F^i = \sum_m \sum_{j=1}^3 D^{ij}(\mathbf{r}_m) u^j, \quad (1)$$

where $D^{ij}(\mathbf{r}_m)$ is the interatomic force constant, representing the force component in the direction i ($=1,2,3$) on this atom caused by a unit motion in the j direction of another atom that is \mathbf{r}_m away. The summation over m goes over all possible atomic positions in the crystal. Force constants are typically obtained experimentally from neutron scattering. The eigenfrequency ω_{gj} and eigenvectors e_{gj} for a normal mode designated by wave vector \mathbf{g} and polarization j ($j = 1, 2, 3$) can be obtained from the following eigenequations:

$$\sum_{v=1}^3 \tilde{D}^{uv}(\mathbf{g}) e_{gj}^v = m_a \omega_{gj}^2 e_{gj}^u, \quad u = 1, 2, 3, \quad (2)$$

where $\tilde{D}^{uv}(\mathbf{g})$ is defined by

$$\tilde{D}^{uv}(\mathbf{g}) = - \sum_m D^{uv}(\mathbf{r}_m) \cos 2\pi \mathbf{g} \cdot \mathbf{r}_m, \quad u, v = 1, 2, 3, \quad (3)$$

and m_a is the mass of the atoms. The allowed values of \mathbf{g} depend on the boundary conditions of the crystal and are confined to the first Brillouin zone. An explicit form of $\tilde{D}^{uv}(\mathbf{g})$ can be found, for example, in the book by Warren¹ for a FCC crystal using force constants up to the third nearest neighbors.

The diffracted intensity for a crystal, in electron units, with thermal motion having only one kind of atom at or above the Debye characteristic temperature is given by¹

$$I = Nf^2 \sum_m \sum_n e^{2\pi i S_\lambda \cdot (\mathbf{r}_m - \mathbf{r}_n)} \times \exp \left(-\frac{4\pi^2 kT}{Nm_a} \sum_{\mathbf{g}} \sum_{j=1}^3 \frac{(S_\lambda \cdot \mathbf{e}_{gj})^2}{\omega_{gj}^2} \times [1 - \cos 2\pi \mathbf{g} \cdot (\mathbf{r}_m - \mathbf{r}_n)] \right), \quad (4)$$

where the summations extend over all possible wave vectors and all atomic positions m and n . The quantity k is Boltzmann's constant, T is the absolute temperature, N is the total number of atoms in the crystal, f is the atomic scattering factor, S_λ is the diffraction vector given by

$$\mathbf{S}_\lambda = \frac{\mathbf{s} - \mathbf{s}_0}{\lambda}, \quad (5)$$

where \mathbf{s} and \mathbf{s}_0 are unit vectors in the directions of the diffracted and incident beams, respectively, and λ is the wavelength of the radiation.

Traditionally, the TDS is calculated from Eq. (4) by solving Eq. (2) to obtain the eigen-frequencies, ω_{gj} , and eigenvectors, \mathbf{e}_{gj} . Because this approach requires considerable computing resources, a simplified procedure is presented utilizing the properties of the lattice Green's function and thereby eliminates the need of solving the eigen-equations. It can be shown that $1/m_a \omega_{gj}^2$ is the eigenvalue of $\tilde{G}^{uv}(\mathbf{g})$, the inverse matrix of $\tilde{D}^{uv}(\mathbf{g})$, with the same eigenvectors, i.e.,

$$\sum_{v=1}^3 \tilde{G}^{uv}(\mathbf{g}) \mathbf{e}_{gj}^v = \frac{1}{m_a \omega_{gj}^2} \mathbf{e}_{gj}^u, \quad u=1,2,3. \quad (6)$$

And furthermore, the three polarization vectors \mathbf{e}_{g1} , \mathbf{e}_{g2} , and \mathbf{e}_{g3} form an orthonormal set of vectors for every \mathbf{g} . Because

$$\begin{pmatrix} \mathbf{e}_{g1}^1 & \mathbf{e}_{g1}^2 & \mathbf{e}_{g1}^3 \\ \mathbf{e}_{g2}^1 & \mathbf{e}_{g2}^2 & \mathbf{e}_{g2}^3 \\ \mathbf{e}_{g3}^1 & \mathbf{e}_{g3}^2 & \mathbf{e}_{g3}^3 \end{pmatrix}$$

is a coordinate transformation matrix from an orthonormal set of vectors to another, and for every \mathbf{g} , it must satisfy⁴

$$\begin{pmatrix} \mathbf{e}_{g1}^1 & \mathbf{e}_{g2}^1 & \mathbf{e}_{g3}^1 \\ \mathbf{e}_{g1}^2 & \mathbf{e}_{g2}^2 & \mathbf{e}_{g3}^2 \\ \mathbf{e}_{g1}^3 & \mathbf{e}_{g2}^3 & \mathbf{e}_{g3}^3 \end{pmatrix} \begin{pmatrix} \mathbf{e}_{g1}^1 & \mathbf{e}_{g1}^2 & \mathbf{e}_{g1}^3 \\ \mathbf{e}_{g2}^1 & \mathbf{e}_{g2}^2 & \mathbf{e}_{g2}^3 \\ \mathbf{e}_{g3}^1 & \mathbf{e}_{g3}^2 & \mathbf{e}_{g3}^3 \end{pmatrix} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (7)$$

or simply

$$\sum_{j=1}^3 \mathbf{e}_{gj}^u \mathbf{e}_{gj}^v = \delta^{uv}, \quad u,v=1,2,3, \quad (8)$$

where δ^{uv} is 1 when $u=v$ and 0 when $u \neq v$. The function $\tilde{G}^{uv}(\mathbf{g})$ is called the lattice Green's function in Fourier space and is defined by

$$\sum_{v=1}^3 \tilde{G}^{uv}(\mathbf{g}) \tilde{D}^{vw}(\mathbf{g}) = \delta^{uw}, \quad u,w=1,2,3. \quad (9)$$

From the properties of the Green's function and its eigenvectors, the summation over j in Eq. (4) can be evaluated as follows:

$$\begin{aligned} \sum_{j=1}^3 \frac{(\mathbf{S}_\lambda \cdot \mathbf{e}_{gj})^2}{m_a \omega_{gj}^2} &= \sum_{j=1}^3 \sum_{u=1}^3 \sum_{v=1}^3 \frac{S_\lambda^u e_{gj}^u S_\lambda^v e_{gj}^v}{m_a \omega_{gj}^2} \\ &= \sum_{j=1}^3 \sum_{u=1}^3 \sum_{v=1}^3 \sum_{k=1}^3 S_\lambda^u \tilde{G}^{uk}(\mathbf{g}) e_{gj}^k S_\lambda^v e_{gj}^v \\ &= \sum_{u=1}^3 \sum_{v=1}^3 S_\lambda^u \tilde{G}^{uv}(\mathbf{g}) S_\lambda^v, \end{aligned}$$

giving

$$\begin{aligned} I &= Nf^2 \sum_m \sum_n e^{2\pi i S_\lambda \cdot (\mathbf{r}_m - \mathbf{r}_n)} \\ &\times \exp \left(-\frac{4\pi^2 kT}{N} \sum_{\mathbf{g}} \sum_{u=1}^3 \sum_{v=1}^3 S_\lambda^u \tilde{G}^{uv}(\mathbf{g}) S_\lambda^v \right. \\ &\quad \left. \times [1 - \cos 2\pi \mathbf{g} \cdot (\mathbf{r}_m - \mathbf{r}_n)] \right). \quad (10) \end{aligned}$$

The Green's function in real space is related to \tilde{G}^{uv} through

$$G^{uv}(\mathbf{r}) = \frac{1}{N} \sum_{\mathbf{g}} \tilde{G}^{uv}(\mathbf{g}) \cos 2\pi \mathbf{g} \cdot \mathbf{r}, \quad u,v=1,2,3. \quad (11)$$

Substituting Eq. (11) into Eq. (10) gives

$$\begin{aligned} I &= Nf^2 \sum_m \sum_n e^{2\pi i S_\lambda \cdot (\mathbf{r}_m - \mathbf{r}_n)} \\ &\times \exp \left(-4\pi^2 kT \sum_{u=1}^3 \sum_{v=1}^3 S_\lambda^u S_\lambda^v [G^{uv}(0) \right. \\ &\quad \left. - G^{uv}(\mathbf{r}_m - \mathbf{r}_n)] \right). \quad (12) \end{aligned}$$

Letting

$$2M = 4\pi^2 kT \sum_{u=1}^3 \sum_{v=1}^3 S_\lambda^u S_\lambda^v G^{uv}(0), \quad (13)$$

Eq. (12) can be rewritten as

$$\begin{aligned} I &= Nf^2 e^{-2M} \sum_m \sum_n e^{2\pi i S_\lambda \cdot (\mathbf{r}_m - \mathbf{r}_n)} \\ &\times \exp \left(4\pi^2 kT \sum_{u=1}^3 \sum_{v=1}^3 S_\lambda^u S_\lambda^v G^{uv}(\mathbf{r}_m - \mathbf{r}_n) \right). \quad (14) \end{aligned}$$

The last exponential function is commonly expanded into a power series with the first term giving the Bragg peak attenuated by e^{-2M} , while the second term yields the first order TDS,

$$I_1 = Nf^2 e^{-2M} 4\pi^2 kT \sum_{u=1}^3 \sum_{v=1}^3 S_\lambda^u S_\lambda^v \tilde{G}^{uv}(S_\lambda). \quad (15)$$

Subsequent terms of this series give second, third,..., order TDS, and involve multiple convolutions by summation of $\tilde{G}^{uv}(g)$. Normally their contribution to the total TDS is significantly smaller than that from the first order TDS. The Green's function is found to peak sharply at the Bragg points in reciprocal space so that it can be approximated by a Dirac δ -function at the zone center beyond the first order. This is equivalent to the method suggested by Warren¹ to take into account higher order TDS. This effectively assumes that the shape of the higher order TDS has the same shape as the first order which is a good approximation if $2M$ is small. With this approximation, the total TDS for a single crystal is given by

$$I_{\text{TDS}}(S_\lambda) = Nf^2(1 - e^{-2M}) 4\pi^2 kT \sum_{u=1}^3 \sum_{v=1}^3 S_\lambda^u S_\lambda^v \tilde{G}^{uv}(S_\lambda), \quad (16)$$

which requires the Green's function and not the eigenvalues and eigenvectors of it. This expression, strictly speaking, can be applied to materials with only one kind of atom. If the material under investigation is a random solid solution of more than one kind of atom, and the interatomic force constants are obtained by fitting a model of "average" atoms, with this set of force constants and an average atomic scattering factor.

III. TDS FOR POLYCRYSTALLINE MATERIALS WITH TEXTURE

In an actual diffraction experiment with polycrystalline materials, the diffraction vector determined by the instrument, S_λ , is localized to a small volume in reciprocal space for each diffraction angle, and the reciprocal lattices of the crystallites in the sample take on all possible orientations. Each crystallite has a set of reciprocal axes (b_1 , b_2 , and b_3) with position in reciprocal space given by

$$S'_\lambda = h_1 b_1 + h_2 b_2 + h_3 b_3. \quad (17)$$

The rotation of each crystallite and therefore its reciprocal axes is relatable to the texture found in the sample. This causes the same S_λ to be decomposed into different (h_1, h_2, h_3) , all located on the surface of a sphere, for each crystallite. For brevity, S'_λ is used in the following discussions. The measured TDS from such a polycrystalline sample is therefore the average of $I_{\text{TDS}}(S'_\lambda)$ over all orientations weighted by the volume fraction of the sample that scattered at a position in reciprocal space located by the condition $S_\lambda = S'_\lambda$. Thus S'_λ takes on all orientations while keeping the magnitude constant.

In the case of an ideally random polycrystalline sample, all the reciprocal lattice directions oriented in the direction of S_λ have the same probability. The TDS for such a sample is just the integral of $I_{\text{TDS}}(S'_\lambda)$ over the surface of a sphere, divided by the total solid angle 4π . However, in a polycrystalline material with a texture, the relative volume fractions of crystallites that have their reciprocal space direction S'_λ oriented in a particular sample direction (defined by S_λ) is given by the inverse pole density function (or inverse pole figure), $R(S'_\lambda)$, of that sample direction.³ This can deviate greatly from the random case. The measured TDS from a polycrystalline material with a texture can be expressed as

$$I_{\text{TDS}}^P(S_\lambda) = Nf^2(1 - e^{-2M}) 4\pi^2 kT \int_{\text{all orientations}} \frac{1}{4\pi} \times R(S'_\lambda) \sum_{u=1}^3 \sum_{v=1}^3 S'^u_\lambda S'^v_\lambda \tilde{G}^{uv}(S'_\lambda) d\Omega_{S'_\lambda}, \quad (18)$$

in electron units, where $d\Omega_{S'_\lambda}$ is the differential solid angle of S'_λ .

Experimentally, the inverse pole density function has been traditionally derived from the measurement of several complete pole figures which are used to obtain the orientation distribution function. This in turn is integrated to obtain the inverse pole density function.³ He, Rao, and Houska⁵ recently developed a simplified method to obtain the orientation distribution function for a polycrystalline material with a true fiber texture or a fiber texture artificially produced by spinning a sample having a more complex texture. This simplified method applied to a cubic system can be more generally applied to obtain the inverse pole density function for any texture in any crystal system. For the special case of a fiber texture, the inverse pole density function for the fiber axis is equal to the orientation distribution function.³ In this method, each crystallographic direction, S'_λ , is characterized by a set of spherical polar angles (Ω, ψ) . The inverse pole density function is expanded in terms of spherical harmonics according to

$$R(\Omega, \psi) = \sum_{n=0}^{\infty} A_{n0} \bar{P}_n^0(\cos \Omega) + 2 \sum_{n=1}^{\infty} \sum_{m=0}^n [A_{nm} \bar{P}_n^m(\cos \Omega) \cos m\psi + B_{nm} \bar{P}_n^m(\cos \Omega) \sin m\psi], \quad (19)$$

where $\bar{P}_n^m(\cos \Omega)$ is the normalized associated Legendre polynomial.⁶ If the series is truncated to a finite number of terms related to the finite sharpness of the texture distribution, the coefficients A_{nm} and B_{nm} can be determined from the values of $R(\Omega, \psi)$ at a finite number of reciprocal space points corresponding to the Bragg reflections, with the direction of the diffraction vector, S_λ , coinciding with the direction for which the inverse pole density function is being constructed. The value of $R(\Omega, \psi)$ at these points can be calculated from the ratio of the experimental to the theoretical integrated intensities for an ideal powder.⁵ After the co-

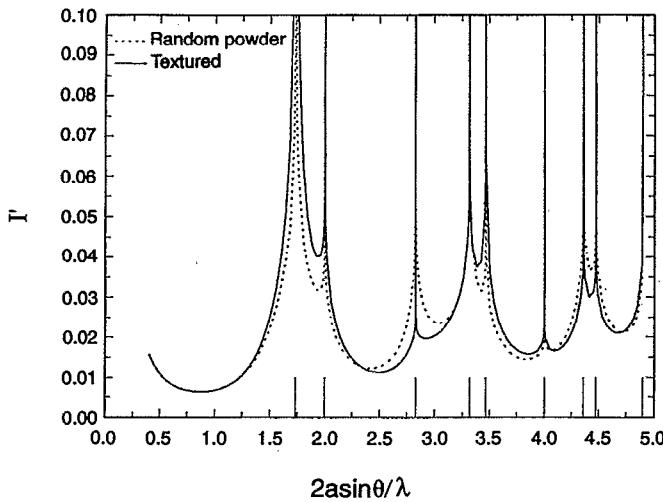


FIG. 1. TDS from polycrystalline materials with and without texture. The plotted normalized intensity I' is defined as $I_{\text{TDS}}^P/Nf^2(1-e^{-2M}) \times 16\pi^2kT(\sin\theta/\lambda)^2$. The attenuation factor $2M$ is evaluated to be $0.00523T(\sin\theta/\lambda)^2$, where T is in K and λ is in Å.

efficients are determined, the values of the inverse pole density function for all other orientations of S'_λ can be evaluated from Eq. (19). If the texture is not too severe, Eq. (19) may be truncated to a small number of terms and only a few number of Bragg peaks are required.

Figure 1 shows the calculated total TDS [normalized by $Nf^2(1-e^{-2M})16\pi^2kT(\sin\theta/\lambda)^2$] for a fiber texture obtained from a copper 11.6 at. % beryllium alloy. The sample was water quenched after annealing at 780 °C for 15 min. The direction of the instrumental diffraction vector coincides with the fiber axis of the texture so the orientation distribution function given by He *et al.*⁵ may be used in place of the inverse pole density function. Interatomic force constants were determined from neutron scattering experiments by Koo *et al.*⁷ Only the coefficients up to the third nearest neighbors are used. TDS for an ideally random polycrystalline material but with all other conditions identical is also computed and plotted in Fig. 1 for a comparison. The latter curve is essentially the same as the result obtained by Warren² who used an approximate method that assumed a linear dispersion relation between ω and $|g|$. An examination of the maxima and minima in Fig. 2 shows a preferred orientation in the $\langle 111 \rangle$ and $\langle 100 \rangle$ directions and is nonpreferred in the $\langle 110 \rangle$ direction. As a result, the TDS for the sample with texture has higher values near the (111) and (200) Bragg reflections and smaller values near the (220) reflection when compared with the scattering from an ideal powder sample.

IV. OTHER DIFFUSE SCATTERING FROM POLYCRYSTALLINE MATERIALS WITH TEXTURE

The discussion at the beginning of the last section applies not only to the TDS, but also to other forms of diffuse scattering such as short range order scattering. In general, if

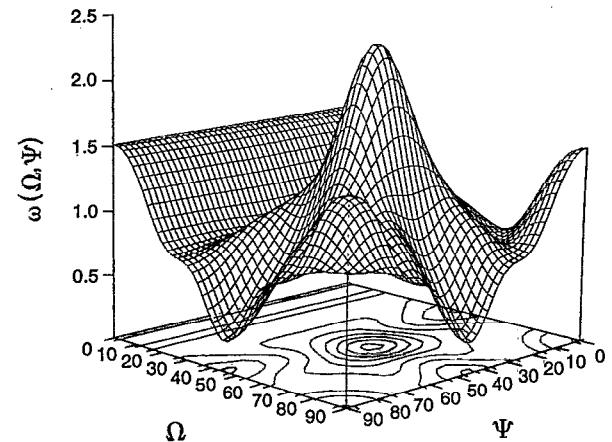


FIG. 2. Orientation distribution function of the texture used to calculate TDS (see Ref. 5). The base plane shows the two dimensional contours of the function. A plane at $\omega=1$ would represent an ideal random powder.

the diffuse scattering for a single crystal is $I^S(S'_\lambda)$, the diffuse scattering from the same material in polycrystalline form with a texture is given by

$$I^P(S_\lambda) = \int_{\text{all orientations}} \frac{1}{4\pi} R(S'_\lambda) I^S(S'_\lambda) d\Omega_{S'_\lambda}, \quad (20)$$

where $R(S'_\lambda)$ is the inverse pole density function, of the texture, for the sample direction that coincides with the diffraction vector, S'_λ . If the single crystal diffuse scattering depends strongly on the direction of the diffraction vector with a fixed magnitude, like the TDS which peaks sharply in the region close to the Bragg reflections, the intensity from the polycrystalline sample with a texture will differ considerably from an ideally random powder. This is shown in Fig. 1 in regions close to the Bragg reflections. On the contrary, if the diffuse scattering is not a strong function of scattering direction like short range order scattering, the resulting intensity from materials with a texture will be similar to that from an ideal powder. Of course, this must be treated with some caution. If the texture of the material is very strong, even a small variation of the scattering with respect to the diffraction direction can lead to a very different diffraction pattern compared with the ideally random one. The example can be considered as having an intermediate texture.

V. CONCLUSION

In this paper, a simplified method for calculating the TDS from single crystals using interatomic force constants is described. This method is based upon the lattice Green's function and does not require the extensive computing required to solve the eigen-equations from lattice dynamics. Only the expressions of the lattice dynamics matrix and its inverse, the Green's function, are needed. The treatment should only be applied with sample temperatures that are

comparable or higher than the Debye characteristic temperature so that equipartition of the thermal energy for the individual normal modes is applicable.

The diffuse scattering from polycrystalline materials with a texture can be calculated from the diffuse intensities from single crystals and the inverse pole density function. A TDS calculation for a copper beryllium solid solution with an intermediate fiber texture has shown a significant difference when compared with that from an ideal powder. In general, the difference between the diffuse scattering from a textured sample and those from an ideal powder depends upon both the severeness of the texture and the directional dependency of the diffuse scattering. The discrepancy will normally become more evident when the texture is strong and when the directional dependency of the scattering is high.

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