

X-ray Excitation of Thermal Spikes

R. J. Weiss

Army Materials and Mechanics Research Center, Watertown, Massachusetts 02172

(Z. Naturforsch. **28a**, 679–681 [1973]; received 16 January 1973)

Dedicated to Professor Dr. G. Borrmann on his 65th birthday

For x-ray scattering from amorphous media at large $\sin \theta/\lambda$ ($> 1.00 \text{ \AA}^{-1}$) momentum and energy is either imparted to a single electron (Compton effect) or to a single atom (Thermal Spike effect). In both cases the impulse approximation provides a convenient method for treating the scattering. Measurements of the ratio of Thermal Spike scattering to Compton scattering were made on paraffin.

Introduction

The discovery of anomalous transmission by Professor Borrmann and the subsequent availability of perfect Silicon and Germanium crystals have combined to provide the most exciting development in x-ray physics since the discovery of the Compton effect. In such perfect crystals essentially all atoms scatter in phase when certain Bragg conditions are satisfied so that the scattering is strictly elastic and the entire crystal takes up the momentum. We should like to point out that under certain diffraction conditions (i. e. $\sin \theta/\lambda$ greater than $\sim 1.0 \text{ \AA}^{-1}$) amorphous solids (and liquids) behave in a diametrically opposite manner i. e. all coherency between atoms is essentially lost and each atom scatters independently so that energy and momentum are taken up by a single atom.

Consider a non-crystalline solid like paraffin scattering Ag $K\alpha$ x-rays ($\lambda = .559 \text{ \AA}$). At low values of $\sin \theta/\lambda$ ($0.1 - 0.3 \text{ \AA}^{-1}$) there is observed a pattern of broad diffraction peaks typical of amorphous solids (and liquids) but for $\sin \theta/\lambda > 1.0 \text{ \AA}^{-1}$ the “elastic” scattering is devoid of structure and the intensity falls off monotonically with increasing angle. It is important that such a measurement be made with a monochromator after the sample to separate the “elastic” component from the inelastic since most of the scattering from paraffin is inelastic (Compton). The quotation marks around the word elastic denote a quasi-elastic process since a small amount of energy ($< 1 \text{ eV}$) is absorbed. If the energy and momentum are absorbed by a single atom, the atom is excited to many times its thermal energy and this can be described as a Thermal Spike. An energy analysis of the beam scattered from paraffin at large $\sin \theta/\lambda$ would reveal both the “elastic” $K\alpha_1 - K\alpha_2$

components plus a broad inelastic (Compton) component whose center of gravity is shifted more than one keV.

Theory

For amorphous media in the limit of large $\sin \theta/\lambda$ the total cross section for the “elastic” and inelastic (Compton) components can be related quite simply to the core one electron wave functions, Ψ_i . The “elastic” component total cross section is the sum of the free atom cross sections, each free atom cross section being given by

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{m c^2} \right)^2 K^2 \left(\sum_{i=1}^N f_{ii} \right)^2, \quad (1)$$

$$f_{ii} = \int \Psi_i^* \exp\{i(\mathbf{k} - \mathbf{k}_0 \cdot \mathbf{r})\} \Psi_i d\tau \quad (2)$$

where the summation is over the N electrons and K is the usual polarization factor. Equation (2) is just the standard expression for the atomic scattering factor. Of course at large $\sin \theta/\lambda$ ($> 1.0 \text{ \AA}^{-1}$) only the core electrons make a significant contribution to Equation (2). For the inelastic component the Waller-Hartree theory plus higher order corrections (Currat, DeCicco and Weiss¹) gives the total cross section as

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{m c^2} \right)^2 K^2 \left\{ Z \left(\frac{\omega_1}{\omega_2} \right)^2 \sum_i (f_{ii})^2 - \sum_{i \neq j} (f_{ij})^2 \right\}, \quad (3)$$

$$\omega_2 = \omega_1 / (1 + 2 \omega_1 \sin^2 \theta / m c^2) \quad (4)$$

where Z is the atomic number and ω_1 the incident x-ray energy. For large $\sin \theta/\lambda$ the ratio of inelastic to “elastic” total cross sections can be evaluated from free atom wave functions since only the unperturbed inner shell electrons give significant contributions to f_{ii} or f_{ij} in Equations (1) and (3).



Dieses Werk wurde im Jahr 2013 vom Verlag Zeitschrift für Naturforschung in Zusammenarbeit mit der Max-Planck-Gesellschaft zur Förderung der Wissenschaften e.V. digitalisiert und unter folgender Lizenz veröffentlicht: Creative Commons Namensnennung-Keine Bearbeitung 3.0 Deutschland Lizenz.

Zum 01.01.2015 ist eine Anpassung der Lizenzbedingungen (Entfall der Creative Commons Lizenzbedingung „Keine Bearbeitung“) beabsichtigt, um eine Nachnutzung auch im Rahmen zukünftiger wissenschaftlicher Nutzungsformen zu ermöglichen.

This work has been digitalized and published in 2013 by Verlag Zeitschrift für Naturforschung in cooperation with the Max Planck Society for the Advancement of Science under a Creative Commons Attribution-NoDerivs 3.0 Germany License.

On 01.01.2015 it is planned to change the License Conditions (the removal of the Creative Commons License condition “no derivative works”). This is to allow reuse in the area of future scientific usage.

Experiment

The ratio of the inelastic to "elastic" components scattered from paraffin (or polyethylene both giving identical results) was measured at a scattering angle $2\theta = 159^\circ$ for both Ag K α ($\lambda = .559 \text{ \AA}$) and Ag K β ($\lambda = .497 \text{ \AA}$). Energy analysis of the scattered x-rays was made with an LiF (400) crystal coupled to two sets of Soller slits (0.1° divergence). Detection was with a NaI scintillation counter. In a separate measurement the ratio of K α to K β was made by replacing the sample with a Ag foil that was caused to fluoresce by the continuum radiation. In Table 1 are given the observed and calculated [Eqs. (1) and (3)] ratios of the cross sections showing good agreement between them.

Table 1. The observed ratio of the total inelastic (Compton) scattering to the total "elastic" scattering of paraffin (or polyethylene) for the Ag K α and K β components. The calculated ratios are from Eqs. (1) and (3) using Hartree-Fock core scattering factors. Included also are the observed and calculated ratios of the K β to K α components corrected for the relative intensities of K β to K α components in the primary beam.

$\sin \theta/\lambda$	inelastic/elastic ratio		elastic K β /elastic K α ratio	
	calculated	observed	calculated	observed
AgK $\alpha_{1,2}$				
1.77 \AA^{-1}	28.7 ± 0.3	27.7 ± 1.0		
AgK $\beta_{1,2,3,4}$			0.61 ± 0.01	0.57 ± 0.06
1.98 \AA^{-1}	47.0 ± 0.5	48.3 ± 2.0		

Discussion

We can now consider the qualitative aspects of the "elastic" and "inelastic" interactions. For the inelastic (Compton) component Currat, DeCicco and Weiss¹, Eisenberger and Platzman², and Phillips and Weiss³, have shown the validity of the so called impulse approximation. The impulse approximation can be applied to the experimental conditions under which the ejected electron is given an energy large compared to the potential energy it experiences in its ground state. Thus the electron is very rapidly moved out of its potential field and the x-ray and electron exchange energy and momentum under conditions of essentially constant potential. Of course after the electron is ejected the system relaxes but the x-ray has already entered the detector before this occurs. Thus the problem reduces to a consideration of conservation of kinetic energy and momentum during the collision. Because one need only consider the kinetic energy of the electron be-

fore and after the collision the differential cross section for the process can be described in terms of the ground state momentum wave functions of the electrons (since kinetic energy is the square of the momentum) Phillips and Weiss 1968.

For the "elastic" component scattered by an amorphous substance we have a direct analogy with the inelastic (Compton) scattering in that the energy given to the atom as a whole ($\sim 0.2 \text{ eV}$) is large compared to the potentials coupling the atoms together and producing the phonon spectrum. Thus the atom is caused to recoil rapidly compared to the motion of its neighbours so that energy and momentum are exchanged between x-ray and atom in essentially a constant potential. We wish to emphasize that it is not necessary to expel the atom from its lattice position but merely to give it an energy large compared to its thermal energy. As a result of the collision a thermal spike is created. (In crystals this process is more complex since the reciprocal lattice can absorb most of the momentum and only normal phonon excitation occurs.) In the impulse approximation thermal spike scattering can be viewed as an atomic "Compton" effect so that the Compton wave length shift $\Delta\lambda$ need only be modified by the ratio of the electron mass to the nuclear mass.

$$\Delta\lambda = \frac{2h}{mc} \sin^2 \theta \rightarrow \frac{2h}{Mc} \sin^2 \theta \quad (5)$$

(Compton) (thermal spike)

where M is the nuclear mass and m the electron mass. The picture then in paraffin for large $\sin \theta/\lambda$ is that the $1s^2$ core electrons on the carbon atoms interact with the x-rays through the scattering factor and these electrons drag the carbon nucleus along in the collision process. It is this reasoning that leads to Eq. (1) while the experimental results of Table 1 provide confirmation.

Can we measure the wave length shift in thermal spike scattering? For Ag K α ($\lambda = .559 \text{ \AA}$) scattered through an angle of 160° by paraffin the wave length shift Eq. (5) is $\sim 4.3 \times 10^{-6} \text{ \AA}$. With a silicon crystal (12, 12, 0) as an analyzer we expect a shift in Bragg angle of 2.8 seconds while for germanium (14, 14, 0) we expect 7.4 seconds. Even though the natural widths of the Ag K α lines are many times the shift it should still be possible to measure it.

Conclusion

In conclusion it would appear desirable to measure the wave length shift associated with thermal spike scattering and corroborate the application of the impulse approximation to high angle scattering from amorphous media. Such high angle scattering could also be used to measure the scattering factors for core electrons of heavy atoms (for example Hg as a liquid) where relativistic effects are important. In addition thermal spike scattering can be used to standardize Compton scattering since the "elastic"

component can be accurately calculated from Hartree-Fock free atom core scattering factors. Lastly such thermal spike scattering may be followed by emission of a photon and this might provide a convenient method to study these localized excitations. By employing W K α instead of Ag K α the thermal spike energy and the angular shift in the analyzer are increased by almost a factor of 10. Under these conditions one might even measure an atomic "Compton" profile and determine the atomic momentum distributions.

¹ R. Currat, P. D. DeCicco, and R. J. Weiss, Phys. Rev. **4**, 4256 [1971].

² P. Eisenberger and P. Platzmann, Phys. Rev. **A 2**, 415 [1970].

³ W. C. Phillips and R. J. Weiss, Phys. Rev. **111**, 790 [1968].

Elastic Scattering of Bloch Waves in Absorbing Crystals Containing Lattice Defects

M. Wilkens, K.-H. Katerbau, and M. Rühle

Max-Planck-Institut für Metallforschung, Stuttgart, Germany

(Z. Naturforsch. **28 a**, 681–690 [1973] ; received 22 January 1973)

Dedicated to Professor Dr. G. Borrmann on his 65th birthday

The differential equations of Bloch wave type which are used in transmission electron microscopy for the calculation of the diffraction contrast from lattice defects are discussed with special reference to crystals with anomalous absorption (i. e. with a complex crystal potential). Anomalous absorption is included, the eigenvalue equation by which the Bloch waves are defined in the perfect reference lattice, becomes non-Hermitian. It is shown that for some particular contrast effects not only the eigenvalues but also the eigenvectors must be corrected for the imaginary part of the crystal potential. As a consequence the orthogonality of the Bloch waves is violated. The correction of the eigenvectors may result in long-range contrast tails of the images of lattice defects (intra-band scattering). If, however, only the peak positions of the contrast profiles are considered, it suffices in practical cases to correct only the eigenvalues as is usually assumed for the diffraction in perfect crystals.

1. Introduction

The elastic scattering of fast electrons in crystals can be described conveniently assuming a complex crystal potential. In transmission electron microscopy of crystalline specimens this was first demonstrated by Hashimoto, Howie and Whelan¹. While the real part of the potential is responsible for the elastic diffraction of the electrons into the Bragg-reflexions the imaginary part accounts for inelastic scattering processes which lead to an attenuation of the wave functions of the elastically scattered electrons by absorption. One of the most striking phenomena which was successfully explained by such a complex crystal potential is the anomalous absorp-

tion first discovered by Borrmann^{2,3} during his studies of the X-ray diffraction in perfect crystals. (For the quantum mechanical justification of the imaginary part of the crystal potential see Yoshioka⁴ or, e. g., the review article by Kambe and Molière⁵.)

If the wave function of the elastically scattered fast electrons inside a perfect crystal is expressed in terms of Bloch waves the solution of the Schrödinger equation can be reduced to the solution of an eigenvalue equation which is Hermitian for crystals without absorption and which becomes non-Hermitian when absorption is taken into account by an appropriate imaginary part of the crystal potential. Since, in general, the imaginary part of the potential is small compared with the real part, it is usually assumed that the non-Hermitian eigen-

Reprint requests to Dr. M. Wilkens, Max-Planck-Institut für Metallforschung, D-7000 Stuttgart, Azenbergstr. 12.