Compton Scattering of X-Rays in Crystals

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INTRODUCTION

In his report, Professor Laval summarizes the French work on the thermal agitation of atoms in crystals. We propose to speak of the study of Compton effect in crystals, which forms a necessary complement, and is in addition a fruitful study.

I. FOUR TYPES OF BEHAVIOR OF X-RAY PHOTONS IN CRYSTALS

(I.1)

An x-ray photon falling on a crystal may be scattered without changing its wavelength (Bragg scattering).

(I.2)

It may be scattered with a very small relative change of wavelength.

This is the scattering by thermal agitation (see J. Laval[1–3]). The photon exchanges energy and momentum with one or more phonons. Since the energy of the phonons is of the order of kT (0.025 ev at ordinary temperature), and that of the x-ray photons is of the order of 10 kev, the relative change of wavelength λ of the photon is of the order of 10^{-6}. On the other hand, the momenta of the phonons are of the same order of magnitude as the momenta of the photons (the wavelengths λ of the photons and Λ of the phonons are of the order of magnitude of 1 A). Figure 1 shows the scattering angle ϕ resulting from the momentum exchange. The quantities $1/\Lambda$ are not defined except to a translation of the reciprocal lattice.

The behavior (I.1) is merely a special case of (I.2), in which a photon absorbs and reemits the same phonon: there is no change of wavelength of the photon, but there is a change of direction. The point X (Fig. 1) coincides therefore with a lattice point of the reciprocal lattice.

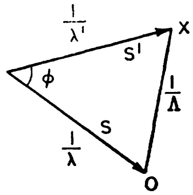


FIG. 1.

[1] J. Laval, Bull. soc. franç. minéral. **52**, 1 (1939).
[2] J. Laval, Bull. soc. franç. minéral. **64**, 1 (1943).
[3] J. Laval, *Rayons X et structures atomiques* (Collège de France, Paris, 1948).

(I.3)

The collision of photon and electron is described by the equations of Compton, which lead to the change of wavelength:

$$\Delta\lambda = \frac{h}{mc}(1-\cos\phi); \quad \frac{h}{mc} = 0.024 \text{ A}.$$

This quantum theory seems to eliminate the process of scattering described in (I.1) and (I.2). As a matter of fact, the scattering types (I.1) and (I.2), called coherent scattering, results from the interaction of the photon with a more complex system than the isolated electron; that is, the atom or crystal considered as a whole. The relation between the coherent scattering (I.1) and (I.2) and the incoherent scattering (I.3) is given by the theory of Waller, which leads to the following ratio of intensities:

$$\frac{I_{\text{incoh}}}{I_{\text{coh}}} = \frac{R[Z - \sum f_{kk}^2 - \sum\sum_{k\neq l} f_{kl}^2]}{(\sum f_{kk})^2},$$

with

$$f_{kl} = \int \psi_k \psi_l^* e^{-2\pi i \mathbf{x}\cdot\mathbf{r}} dv,$$

$R = (\nu'/\nu)^3$,
ν' = frequency of scattered photon,
ν = frequency of incident photon.

This formula is applicable only to the free atom. It must certainly be modified for atoms bound in a crystal in a way that is discussed in Sec. II.

(I.4)

A last type of behavior corresponds to the complete absorption of the photon, leading to the photoelectric effect which is responsible for the larger part of the absorption of x-rays.

As in the Compton effect one has the ejection of an electron, but not the reemission of a photon. This last type of behavior (I.4) is more closely analogous to (I.3) than to (I.1) and (I.2). The quantum field theory developed by Heitler allows one to determine the relative importance of (I.3) and (I.4). The photoelectric effect is considered to be a first-order process leading to a direct transition from one quantum state to another. The Compton effect on the other hand is of the second order, involving an intermediate state between the initial and final states: this is the state where the incident photon has been absorbed and the emergent photon has not yet been re-emitted. It is possible in principle to calculate the matrix elements corresponding to these transitions, and therefore to determine the cross sections for the various processes.

II. MODIFICATIONS OF THE COMPTON EFFECT IN CRYSTALS

(II.1)

It would be natural to think that Compton scattering, being incoherent, and not leading to any interference, would be essentially an atomic phenomenon, independent of the location of the different atoms with respect to each other. But experiment has shown (Laval[1–3]) that, in certain cases, and particularly at small scattering angles, the scattered intensities are much weaker than would be predicted by the theory of Waller for the free atom.

What theoretical reasons can one invoke for this weakening?

(a) The interatomic bonds modify the wave functions of the external electrons. The calculation of I_{incoh} should therefore be made with the wave functions of the crystal and not of the isolated atom.

(b) The electrons ejected from the atom by the Compton effect must travel through the crystal and not in free space. For these recoil electrons, there are therefore forbidden ranges of energy; it follows that the total intensity of Compton scattering in a crystal be less than that corresponding to an assembly of free atoms.

Waller (not published) proposes a general quantitative interpretation of the interaction photon crystal taking into account the fundamental and excited states of the crystal.

(II.2)

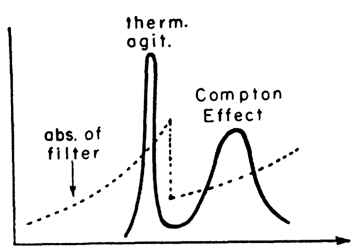
Experimentally one has tried to bring evidence for these various modifications.

(II.2a)

Total diminution of the ratio $I_{\mathrm{incoh}}/I_{\mathrm{coh}}$.

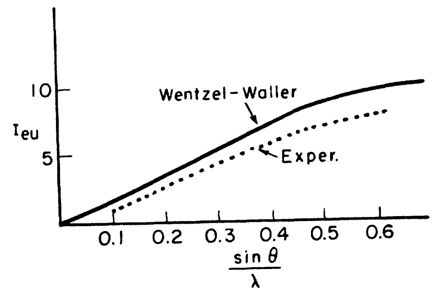
The intensities of scattered x-rays are very weak, and it is difficult to get good experimental conditions for a precise spectrographic analysis of the scattered radiation. The beautiful experiments of DuMond[4]

FIG. 2.



[4] J. W. M. DuMond, Revs. Modern Phys. 5, 1 (1933).

FIG. 3.



and Kirkpatrick, Ross, and Ritland[5] have nevertheless made it possible to study the modification of the form of the Compton modified lines and in some cases the ratios of intensities.

The following experimental devices aid greatly in the study:

(a) One can practically eliminate thermal agitation by studying the scattering for a crystal at very low temperature (see Walker[6]).

(b) One can interpose in the path of the scattered rays a filter having a K- or L-absorption edge at a wavelength very slightly greater than the wavelength λ of emission of the x-ray tube, but less than the wavelength of the Compton modified line (Fig. 2). Using measurements with and without this filter, one determines the two parts of the coherent and incoherent radiation.[7] This method has been applied to aluminum by Curien and Deroche.[8] The results have been verified with good agreement by Walker.[6] The Compton intensity is significantly below the values found for the free atom (Fig. 3).

(II.2b) Energy Threshold for Compton Effect in Insulators

In order that the electron may be emitted from the atom, the energy delivered by the photon must be enough that the electron can jump cross the first forbidden gap. Therefore there should exist a threshold for Compton scattering. Attempts have been made to show this threshold by two types of experiments.

(a) By spectrographic analysis of a beam scattered by a crystal in one direction. The Compton modified line should start suddenly in the direction of short wavelengths, at a distance from the coherent ray corresponding to the energy difference between the top of the last totally occupied band and the bottom of the first empty band. In diamond this jump is $\Delta E = 7$ ev. The experiment has been attempted by Alexopoulos and Brogren.[9] It is difficult to maintain that the results are really conclusive.

[5] Kirkpatrick, Ross, and Ritland, Phys. Rev. 50, 928 (1936).
[6] C. B. Walker, Phys. Rev. 103, 547, 558 (1956).
[7] H. Curien, J. phys. radium 16, 494 (1956).
[8] H. Curien and C. Deroche, Bull. soc. franç. minéral. 79, 102 (1956).
[9] K. Alexopoulos and G. Brogren, Arkiv. Fysik 6, 22 (1953).

(b) One can also make measurements, as J. Blin (unpublished) has done, of scattering at small scattering angles ϕ. At very small angles the Compton effect should be impossible, the energy delivered to the recoil electron being smaller than the required jump ΔE. When with increasing ϕ the energy delivered to the electron becomes equal to ΔE, the Compton effect should appear suddenly, leading to a discontinuity of the total scattered intensity as function of ϕ, this discontinuity varying with the orientation of the crystal with respect to S and S′ (Fig. 1) (the Brillouin zone is not spherical). One observes, in fact, a discontinuity but at a smaller angle than one would expect. The explanation of this disagreement may perhaps be found in the Doppler effect at the moment of collision between photon and electron (which also explains, as DuMond[4] has shown, the width of the Compton modified line). One would therefore have an additional method for studying the distribution of momenta in the electrons of the crystal.

CONCLUSION

Study of Compton scattering is a necessary complement to the study of scattering by thermal agitation of the atoms of the crystal. The experiments carried out up to the present are promising. They cannot be interpreted except by a more careful study of the behavior of electrons in crystals.

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Lattice Dynamics and Thermal Diffuse Scattering of X-Rays

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IT is possible to study in a detailed way the thermal vibration of atoms in a crystalline solid by analyzing the angular distribution of scattering of monochromatic x-rays. A perfectly static crystal lattice would produce very sharp Bragg reflections, and in addition, a certain amount of continuous background between the Bragg reflections because of Compton scattering. In contrast to this static situation, the presence of thermal motion reduces the intensity of the crystalline or Bragg reflections, as well as substantially increasing the intensity of the background scattering. This latter effect is useful in analyzing the thermal motion.

Assuming linear restoring forces between atoms, and that these forces depend only on the nuclear coordinates, we may completely describe the thermal vibrations as a linear superposition of traveling elastic waves whose frequencies are those of the lattice normal mode oscillators. These frequencies, numbering three times the number of atoms in the crystal, extend from zero to approximately 10^{13} cycles per second, with the respective wavelengths ranging from crystalline dimensions down to atomic spacings.

In the first approximation, the background scattering due to thermal motion is related to the elastic waves in the following way. Each elastic wave produces a pair of intensity peaks centered about each crystalline reflection, the spacing between the crystalline and thermal reflections being inversely proportional to the reciprocal of the wavelength of the elastic wave. In other words, the x-ray undergoes additional Bragg reflections due to the periodicity of the elastic wave being superimpoesd on that of the lattice.* The intensity of each such thermal reflection is proportional to the energy of a harmonic oscillator whose frequency is that of the particular elastic wave in question. Assuming equilibrium at a given temperature, the oscillators of lowest frequency will possess the largest amplitudes, which means that elastic waves of long wavelength will produce maximum diffuse intensity. The high-frequency waves will scatter less since these waves are in reduced states of excitation at the same temperature. Owing to the quasi-continuum of lattice vibration frequencies, the diffuse scattering will appear continuous, starting at a maximum near the crystalline reflections and falling to a minimum halfway between, a prediction well in accord with experiment. The process outlined above is a single phonon scattering process, i.e., the x-ray quantum is scattered from one elastic wave. The single phonon scattering is usually dominant, but there are significant contributions to the total diffuse scattering arising from multiple phonon processes wherein the x-ray quantum is scattered successively from two or more elastic waves. These

* The x-ray and phonon of course exchange energy and momentum according to the conservation laws. With x-rays, this energy shift is beyond observation since the x-ray photon energy is a million times that of a phonon. A shift is observable in the cases of slow neutron diffraction and light scattering from transparent solids and liquids (Brillouin scattering).