

Variation of Compton Scattering with Changes in Structure of Quartz (SiO_2)

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The ratio between coherent and incoherent Mo $K\alpha$ radiation, scattered at 90° from amorphous quartz, has been found to be 1.70. This ratio is compared with various theoretical models and found to be in disagreement with them. Measurements have also been made of the Compton spectrum from the (110) plane of quartz, and from the plane perpendicular to l_c axis. The Compton cross section is found to depend on the crystal structure.

I. INTRODUCTION

THIS work was begun in order to obtain experimental data on the ratio of coherent and incoherent scattering from amorphous SiO_2 . This ratio is required in studies of electron density distribution in such amorphous materials. Since this ratio disagreed with theory, the same ratio was then measured for several other samples of amorphous quartz, which all showed the same disagreement.

The measurement of the Compton spectrum of a single crystal of quartz was carried out with the purpose of investigating the influence of the crystal structure of the large-angle Compton cross section. The interesting aspect of the present work is the fact that the Compton cross section depends on the crystal structure in the large-angle region, although only for the small angles is there a theoretical explanation of this effect.

II. THEORY

It is well known that the coherently scattered intensity is proportional to f^2 , where f is the total atomic scattering factor. In the case that the incident radiation has a frequency much higher than that of the absorption edge, f is given by the simple relationship

$$f = f_0 = \sum_{n=1}^Z f_{nn}, \quad (1)$$

where

$$f_{nn} = \int \psi_n^* \exp(i\mathbf{S} \cdot \mathbf{r}) \psi_n d\mathbf{r} \quad (2)$$

is the wave function of the n th electron in the atom, $\mathbf{S}$ is the scattering vector of magnitude $S = (4\pi/\lambda) \sin\theta$, and Z is the atomic number.

In general the total atomic scattering factor f is a complex number given by

$$f = f_0 + \sum_k \int_{\omega_k}^{\infty} \frac{\omega^2 (dg/d\omega)_k}{\omega_i^2 - \omega^2} d\omega + \frac{i}{2} \pi \omega_i \sum_k (dg/d\omega)_{ki}, \quad (3)$$

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where the summation is over all absorption edges, ω_i = incident frequency, ω_k = frequency of absorption edge, $(dg/d\omega)_k$ = density of oscillators of type k at frequency ω , and $(dg/d\omega)_k d\omega$ = the number of virtual oscillators of type k having frequencies between ω and $\omega + d\omega$.¹ The f factor has been calculated by several approaches and is given in various tables.¹⁻³ The incoherent intensity scattered by a free atom in electronic units is given, according to the assumptions of Waller, Breit, and Dirac, by

$$I_{\text{inc}} = R^3 \left(Z - \sum_n |f_{nn}|^2 - \sum_{m \neq n} |f_{mn}|^2 \right), \quad (4)$$

where

$$f_{mn} = \int \psi_m^* \exp(i\mathbf{S} \cdot \mathbf{r}) \psi_n d\mathbf{r}, \quad (5)$$

$R = (\nu'/\nu)$, ν' = frequency of scattering photon, ν = frequency of incident photon. This must certainly be modified for atoms bound in a solid. The Compton intensity has been calculated by several approaches as a function of S for most of the elements.

Although several authors found disagreement between theoretical and experimental results⁴ in the case where the scatterer was a solid, the reason may be that the wave functions which were used to calculate the Compton spectrum were more or less the wave functions for single atoms, and in the case of the solid the wave function must be quite different. It is expected that the Compton intensity will depend on the orientation of the crystal if the recoil energy is about equal to the Fermi energy and the Brillouin zone is not a sphere.⁵ This has already been observed in the case of LiF .⁶

III. EXPERIMENT AND RESULTS

The experiment utilizes a bent-crystal spectrometer with a single crystal of silicon cut to reflect from the (440) plane. The radiation used was Mo $K\alpha$ from a

¹ Don T. Cromer, *Acta Cryst.* **18**, 17 (1965).

² Don T. Cromer and J. T. Waber, *Acta Cryst.* **18**, 104 (1965).

³ *International Tables for X-Ray Crystallography* (The Kymoch Press, Birmingham, England, 1962), Vol. III.

⁴ N. H. March, *Proc. Phys. Soc. (London)* **A67**, 9 (1954).

⁵ H. Curien, *Rev. Mod. Phys.* **30**, 232 (1958).

⁶ N. G. Alexandropoulos, Ph.D. dissertation, Athens University, Greece, 1964 (unpublished).

TABLE I. Experimental results and calculated values for coherent and incoherent scattered intensity.

Quartz samples	Incoherently scattered radiation			Coherently scattered radiation			Ratio
	Intensity at the peak (counts/min)	Integrated intensity (arbitrary units)	Half-width (deg)	Intensity at the peak (counts/min)	Integrated intensity (arbitrary units)	Half-width (deg)	
			Experimental				
Single-crystal scattering surface parallel to (110)	92±1.7	228.75±4.2	1.1 ±0.5	126±2.3	113.24±2.4	0.40±0.5	
Single-crystal scattering surface perpendicular to I_c axis	118±2.2	294.64±5.5	1.1 ±0.5	110±2.0	96.17±1.8	0.45±0.5	
Polycrystalline	116±2.2	303.54±5.5	1.1 ±0.5	204±3.7	178.08±3.2	0.45±0.5	
Amorphous	78±1.15	206.46±3	1.15±0.5	154±2.3	121.0 ±1.8	0.40±0.5	1.70
			Theoretical				
Ionic bond		8.0901			3.719		2.1752
Covalent bond		7.522			3.737		2.0127

Machlett tube, operated with a self-rectified circuit at a stabilized tube current of 17.5 mA and an unstabilized peak tube voltage of 32 kV. The diffracted beam was measured by a scintillation detector. A background of less than 1 count/min was realized by using a small NaI crystal (5 mm×10 mm) on an EMI 9524 A photomultiplier and a pulse-height analyzer.

The spectrum of the totally scattered Mo $K\alpha$ radiation from amorphous quartz is shown in Fig. 1.

In order to have better separation of the coherent and incoherent components of the scattered radiation, the fluorescence spectrum of Mo was obtained. For this purpose a piece of Mo replaced the scatterer, and the spectrum obtained (Fig. 2) is similar to the spectrum of the coherently scattered component. By comparison of the spectra in Figs. 1 and 2, the two components are clearly resolved (dashed line, Fig. 1), according to the separation procedure given in Appendix I. The spectrum of the scattered Mo $K\alpha$ radiation from the single-

crystal quartz cut 5° off the (110) plane is given in Fig. 3, and that of the crystal cut perpendicularly to the I_c axis is given in Fig. 4. Figure 5 shows the spectrum of the polycrystalline quartz (the peak on the small-angle side in all these spectra may come from tube-target impurities). Table I gives the integrated Compton intensity as well as the ratio of the coherent and incoherent intensities calculated from the values given in the *International Tables for X-Ray Crystallography* (Vol. III) and for two different assumptions.

In the first case it is assumed that the bonding in SiO_2 is ionic, and in the second case that it is covalent. Both theoretically calculated ratios differ substantially from the experimental ratio for amorphous SiO_2 , with which they are compared.

IV. CONCLUSIONS

The results show a disagreement between the experimental and the theoretical value of the ratio between

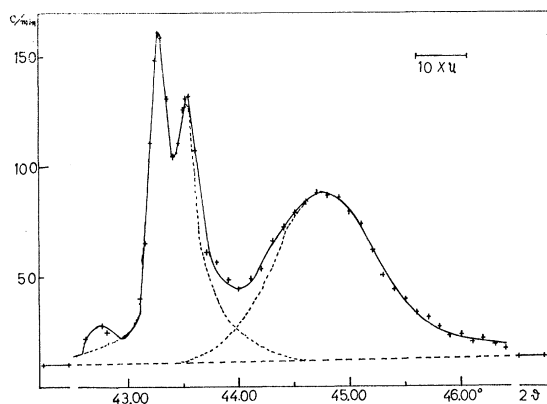


FIG. 1. Solid line—the spectrum of Mo $K\alpha_1$, $K\alpha_2$ radiation scattered on amorphous quartz. The dashed line shows the two spectrum components.

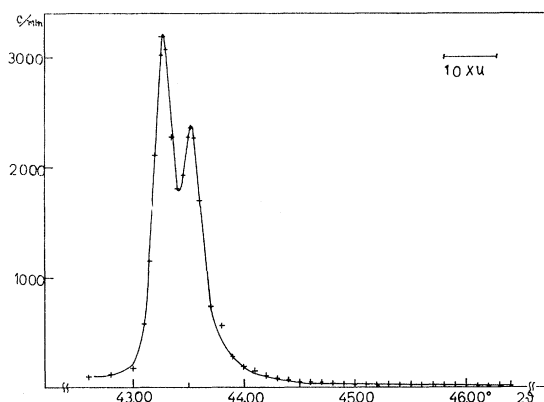


FIG. 2. The spectrum of Mo $K\alpha_1$, $K\alpha_2$ fluorescence radiation.

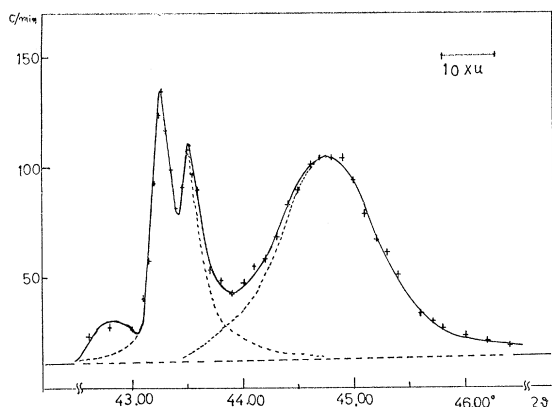


FIG. 3. Spectrum of Mo $K\alpha_1$, $K\alpha_2$ scattered on single-crystal quartz cut 5° off the plane (110).

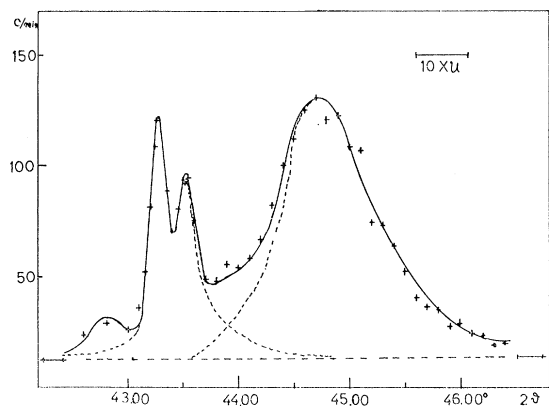


FIG. 4. The spectrum of Mo $K\alpha_1$, $K\alpha_2$ scattered on single-crystal quartz cut perpendicular to l_0 axis.

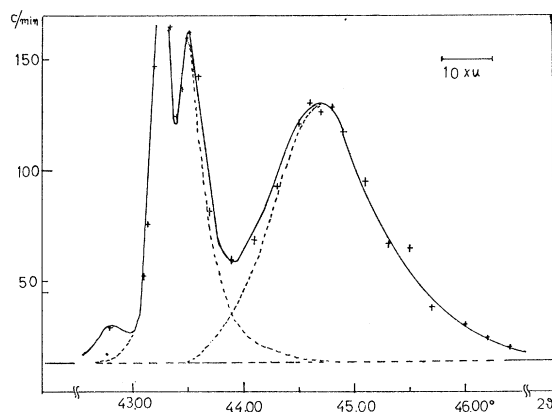


FIG. 5. Spectrum of Mo $K\alpha_1$, $K\alpha_2$ scattered on polycrystalline quartz.

coherent and incoherent scattered radiation; they also indicate that the Compton intensity depends on the structure of the scatterer. The last observation would not be difficult to explain in the case in which the recoil energy is a few eV⁷; in the present case, however, the recoil energy is some hundred eV.

⁷ H. Curien, Rev. Mod. Phys. **30**, 232 (1958).

A possible explanation may be that the assumption that the wave functions for atomic Si and O are also good for describing the crystalline SiO₂ is incorrect, and that in fact the wave function is much more dependent on the orientation of **S** with respect to the crystal axis.

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Thanks are also due to Dr. R. C. Buschert for his initial suggestion that the ratio between coherent and incoherent scattered radiation be measured.

APPENDIX: SEPARATION OF THE COHERENT AND INCOHERENT COMPONENTS IN THE SPECTRUM OF SCATTERED X RADIATION

A technique for separating these two components is given in this Appendix. This is useful in all cases but particularly so in the case of small-angle scattering in which the two components overlap.

Let $g(E)$ represent the pure line profile of the Compton band due to strictly monochromatic radiation, and $\Phi(E)$ the pure line profile of the Compton band due to the primary line $\varphi(E)$. Then⁶

$$\Phi(E) = \int_{-\infty}^{+\infty} \varphi(E_0) g(E - E_0) dE_0. \quad (A1)$$

Let the Compton line profile, as generated by the spectrometer, be $F(E)$; this is the convolution of the pure line profile $\Phi(E)$ and the over-all weight function $\sigma(E)$ of the apparatus (apparatus profile). $F(E)$ and $\Phi(E)$ are given by^{8,9}

$$F(E) = \int_{-\infty}^{+\infty} \sigma(E_0) \Phi(E - E_0) dE_0, \quad (A2)$$

$$f(E) = \int_{-\infty}^{+\infty} \sigma(E_0) \varphi(E - E_0) dE_0. \quad (A3)$$

Equation (A3) is the relation between $f(E)$, (the primary line profile generated by the spectrometer), $\sigma(E)$, and $\varphi(E)$. Assuming that $R(E)$ represents the spectrum scattered by the specimen (the solid line of Figs. 1, 3, 4, 5), then between $R(E)$, $f(E)$, $F(E)$, and the background α_1 , the following relation is true:

$$R(E) = \beta f(E) + F(E) + \alpha_1. \quad (A4)$$

Here, β is a constant that can be determined by comparison of the spectrum $R(E)$ and the spectrum of the primary radiation (Fig. 2), which is the primary line profile as generated by the spectrometer plus a constant

⁸ L. Alexander, J. Appl. Phys. **25**, 155 (1954).

⁹ F. W. Jones, Proc. Roy. Soc. (London) **A166**, 16 (1938).

background α_2 ;

$$r(E) = f(E) + \alpha_2. \quad (\text{A5})$$

$r(E)$ is obtained by putting in place of the scatterer a piece of the same material as the x-ray-tube target, and exciting it to fluorescence. From (A4) and (A5) one obtains

$$\frac{R(E) - \alpha_1}{r(E) - \alpha_2} = \beta + \frac{F(E)}{f(E)}, \quad (\text{A6})$$

and from (A6),

$$\frac{R(E) - \alpha_1}{r(E) - \alpha_2} = \beta, \quad (\text{A7})$$

for the intervals of values of E in which $F(E) = 0$. Therefore, from (A7) and the observed spectra $R(E)$

and $r(E)$, $F(E)$ is determined as well as $f(E)$ by (A4) and (A5) (see dashed line in Figs. 3, 4, 5), even in the case in which the scattering angle is only a few degrees and the components overlap.

From (A1), (A2), and (A3) one finds

$$F(E) = \int_{-\infty}^{+\infty} f(E_0)g(E-E_0)dE_0. \quad (\text{A8})$$

This result is of considerable importance because $f(E)$ and $F(E)$ are known functions, obtained from direct measurements. From the solution of (A8) for $g(E)$ one gets the ideal Compton spectrum without knowing $\sigma(E)$ and $\varphi(E)$, the apparatus profile and the true profile of the primary line, respectively.

Oscillatory Photoconductivity in InSb†

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The oscillations in the low-temperature photoconductive spectral response of p -type InSb are found to be strongly dependent on electric field strength in both intrinsic and extrinsic photoconductivity. The oscillatory structure gradually disappears at electric fields higher than ~ 20 V/cm. As a function of increasing temperature, the position of the minima of the oscillations shifts towards lower energies and the size of the oscillations decreases. A qualitative interpretation, consistent with all observations, shows that the primary reasons for oscillatory photoconductivity are: (a) the preferential momentum losses of carriers due to longitudinal optic phonon emission in the direction of the electrical potential gradient; (b) the fact that the steady-state distribution of carriers generated by photon absorption is drastically different from equilibrium.

I. INTRODUCTION

OSCILLATIONS in photoconductive spectral response have recently been observed in many semiconductors.¹ In this paper we report the main results of an extensive investigation of both the intrinsic and extrinsic oscillatory photoconductivity in InSb. In particular the dependence of the shape of the oscillations on electric field strength and on temperature have not been reported elsewhere. Partly because of these experiments, we present an interpretation of the phenomenon of oscillatory photoconductivity which differs in important respects from those given earlier. Our qualitative interpretation is borne out by a theo-

retical calculation of the photoconductive response with the help of the Boltzmann equation.²

II. EXPERIMENTAL

The preparation of InSb single crystals used in this work has been described elsewhere.^{3,4} Our samples generally contained 10^{13} to 10^{15} cm⁻³ excess acceptor impurities.

The photoconductive spectral response was measured with a Perkin-Elmer model 13 monochromator whose light beam was chopped at ~ 200 cps. A dc voltage was applied across the sample in series with a load resistor. The ac voltage across the load resistor was amplified and recorded. In this way, only the light-induced change in current is measured, i.e., the dc "dark current" is suppressed. For measurements of the extrinsic photo-

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¹ Table I summarizes the materials and types of optical transitions in which oscillatory photoconductivity has been observed.

² H. J. Stocker and H. Kaplan, following paper, Phys. Rev. **150**, 619 (1966); hereafter referred to as II.

³ We acknowledge the use of one high-purity single crystal of InSb supplied by Texas Instruments.

⁴ W. Engeler, H. Levinstein, and C. R. Stannard, Jr., J. Phys. Chem. Solids **22**, 249 (1961).