THE REFLECTION OF ATOMS FROM CRYSTALS

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Abstract

Beams of mercury, cadmium, and arsenic incident upon clean cleavage surfaces of rock-salt give rise to specularly reflected beams. Measurement shows that the velocity of the beam of cadmium atoms reflected from rock-salt may be represented by

$$\lambda=\frac{h}{mv}=2d\left(2.26-\frac{\phi}{\frac{1}{2}mv^2}-\cos^2\theta\right)^{1/2}.$$

A beam specularly reflected from a first crystal is specularly reflected at a second if incident upon it at the same angle at which reflection from the first took place. If the angles are nearly equal there is still some specular reflection at the second crystal but the intensity of the specular beam drops off with increasing difference of angles and the incident beam is scattered at random. A beam of cadmium atoms issuing from a boiler at 440°C and striking a rock-salt crystal at an angle of 45° gives rise to a specular beam containing about 17 percent of the incident atoms. The intensity of the beams specularly reflected at 22.5° and 67.5° are in the ratio 1:1.38. These facts indicate that associated with motion of translation of uncharged atoms and molecules there is a wave phenomenon of the type postulated by de Broglie.

THE fact that a beam of atoms incident upon a clean cleavage surface of a crystal may give rise to a well defined reflected beam making the same angle with the normal to the crystal surface as does the incident beam[1] suggests at once the wave-particle dualism exhibited in the Compton effect and the Davisson and Germer experiments. To understand this phenomenon in terms of the hard elastic spheres of Maxwell's kinetic theory or of the planetary atomic systems of Bohr is difficult, at least. On the other hand the wave, or, more properly superposition characteristics of the new quantum theory lead us at once to expect just such phenomena. Not in every case however does the incidence of an atomic beam upon a crystal surface give rise to a specularly reflected beam. We have already reported the existence of specularly reflected beams of mercury and cadmium from sodium chloride crystals and our failure to obtain specular reflection of sodium and atomic hydrogen.[2] We have since observed specular reflection of cadmium and arsenic beams from potassium chloride crystals and also of arsenic from sodium chloride, and have been unable to obtain specular reflection of either of these substances from orthoclase or flourite crystals. Arsenic vapour[3] is tetra-atomic at the temperature (ca. 250°C) at which we worked, so that specular reflection of molecules as well as of atoms is evidently possible.

[1] A. Ellett and H. F. Olson, Phys. Rev. **31**, 643 (1928).

[2] Johnson, Jour. Frank. Inst. **206**, 301 (1928) has since reported specular reflection of hydrogen from rock-salt.

[3] Landolt-Börnstein, Physikalisch-Chemische Tabellen.

If we consider the wave picture of de Broglie and Schroedinger then familiar phenomena of optics and the known behavior of electrons suggest that the specular reflection of atoms is analogous either to the Bragg type of x-ray reflection, or to reflection from a plane mirror, or from a plane grating. The experiments reported in the present paper appear to establish definitely the "wave" nature of the phenomenon, and to rule out the two latter possibilities. In brief we have been able to show; first, that a specularly reflected beam has acquired a property by virtue of which it is entirely specularly reflected from a second crystal only when incident upon that crystal at the same angle at which it was initially reflected, second, that a specularly reflected beam is made up of atoms all having very nearly the same velocity,

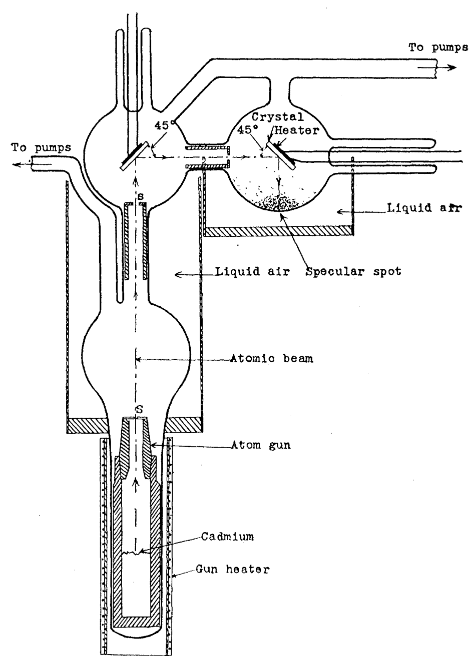

Fig. 1. Type of tube used for successive reflections.

and that this velocity varies with the angle of reflection in a characteristic way, third, that the intensity of the specularly reflected beam varies with the angle of reflection and that the ratio of intensities of the beams reflected at two angles is the ratio of the relative numbers of atoms in the incident beam having wave-lengths appropriate for reflection at these angles. These experiments will be taken up in the above order.

Reflection from two Crystals in Succession[4]

Figure 1 is a sketch of the apparatus used when the beam was reflected from the first crystal at an angle of 45°, and from it the method is evident.

[4] A preliminary report was read at the New York Meeting of the Physical Society, Phys. Rev. **33**, 124 (1929).

The results are presented graphically in Fig. 2, together with the velocities of the beams specularly reflected at the angles in question.

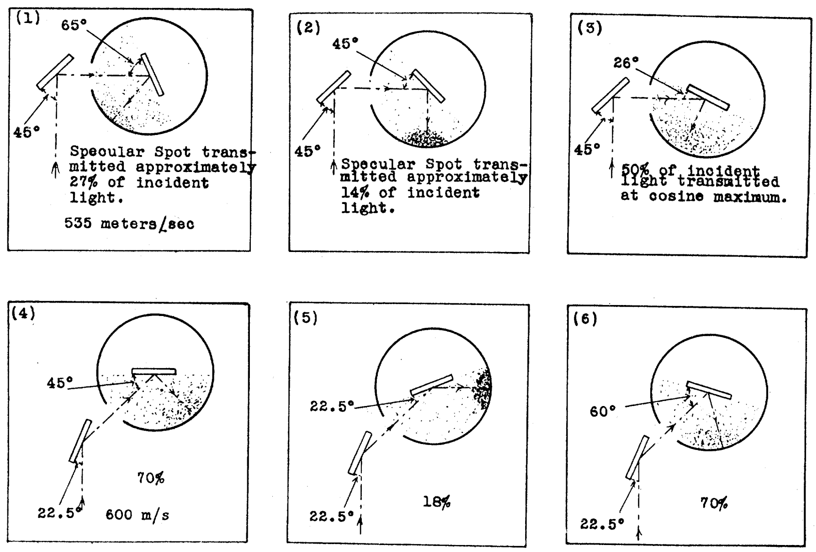

Fig. 2. Results of experiments on successive reflections.

Direct Measurement of the Velocity of Reflected Beams[5]

Figure 3 shows the apparatus, the method being that developed by Eldridge[6] for the experimental verification of the Maxwell distribution law. Atoms issuing from the slit S_1 pass through a second wide slit S_2 and impinge upon a

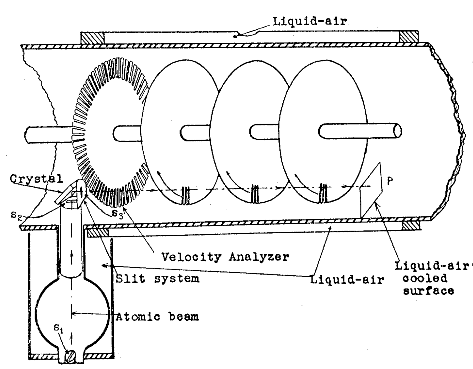

Fig. 3. Apparatus used to measure velocities of reflected atoms.

large area of the reflecting crystal. Part of the specularly reflected beam passes through the narrow slit S_3 and then those atoms which are moving in a direction appropriate to their velocity pass on through the rotating sectored

[5] A preliminary report appeared in Science, **68,** 89 (1928).
[6] J. A. Eldridge, Phys. Rev. **30,** 931 (1927).

disks and form a deposit upon the surface P. By rotating the disks first in one direction and then the other two deposits were formed. The plate P was then run through a microphotometer and the density of the deposits measured.

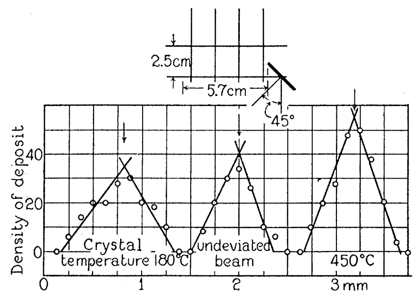

Fig. 4a. Speed of rotor, 72 r.p.s.; velocity of beam, 535m/s.

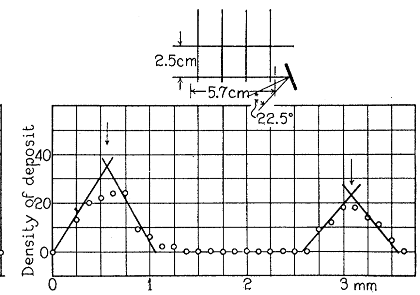

Fig. 4b. Speed of rotor, 73.5 r.p.s.; velocity of atom beam, 500m/s.

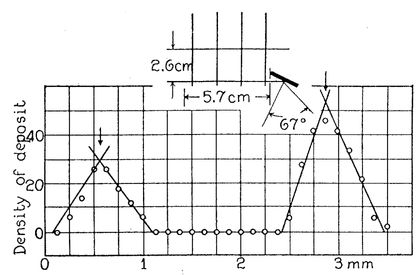

Fig. 4c. Speed of rotor, 76 r.p.s.; velocity of atom beam, 600m/s.

Fig. 4. Density curves obtained with apparatus shown in Fig. 3.

Densities were translated into relative numbers of cadmium atoms per unit area by the method described by Eldridge. Since the slit S_3 and the slits in

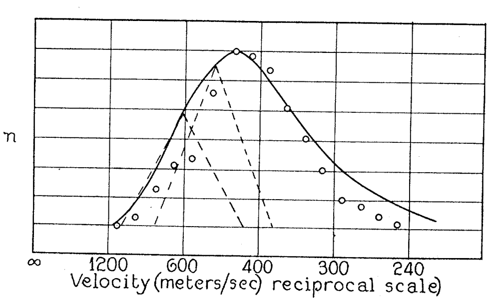

Fig. 5. Comparison of velocity distribution of cadmium atoms before and after reflection.

the sectored disks were the same width a beam of atoms having all the same velocity will form a deposit three times as wide as the slits with density varying linearly from zero at the edges to a maximum at the center.

The density curves obtained are shown in Fig. 4 (a, b, c), together with the rate of rotation of the sectored disks, the distance of the slit S_3 from the plate P, and the velocity corresponding to the observed separation of the deposits. It is evident from the inspection of these curves that the reflected beam of atoms contains only a narrow range of velocities, otherwise the deposits would not approximate so closely to the type to be expected for a purely monochromatic (single velocity) beam. Fig. 5 shows the deposit formed when the direct beam from the gun is incident upon the slit S_3. The heavy curve shows the distribution to be expected for a Maxwell distribution of velocities at the temperature of the gun, due allowance being made for the finite slit width of the velocity analyzer. The dotted triangles are the types of deposit obtained by reflection from rock-salt at 22.5° and 67.5°, as in Fig. 4, but reduced to the scale of the present drawing for purposes of comparison. Fig. 4 (a) shows the displaced deposits obtained from the specular beam reflected at 45° from a crystal which was kept at 180°C, when the disks were rotated clockwise and at 450°C when they were rotated counter clockwise. The central undisplaced deposit was obtained by rotating the disks very slowly (about 15 or 20 r.p.m.). Since the outer deposits are the same distance from the undisplaced deposit it is evident that the velocity of the specularly reflected beam does not depend critically upon the temperature of the crystal. It likewise is significant that the form and breadth of the displaced deposits do not differ greatly from that of undisplaced deposit, for the latter, obtained with the disks rotating very slowly, must have just the shape to be expected for a single velocity beam at high speeds of rotation.

The Fraction of the Incident Beam Going into the Specular Beam

To determine what part of the incident beam goes into the specular deposit and what part is diffusely reflected it is necessary to measure the relative intensities of incident and specular beams. This is readily done by weighing the deposits formed when the beams are intercepted by liquid air cooled surfaces. In the apparatus, Fig. 6, a and b are thin pieces of platinum upon which the cadmium was deposited. To eliminate errors due to variation in the rate of emission from the boiler the collecting surfaces were exposed alternately for brief periods. As soon as the apparatus had warmed to room temperature the platinum receivers and deposits were removed, placed in a dessicator, and weighed upon a micro-balance. Repetition of the weighings after two and three days showed that the deposits did not evaporate. Similar deposits on glass evaporate quite appreciably in this length of time.

The receptor a of course receives a portion of the diffusely scattered atoms as well as the specular beam. This diffuse scattering appears to be entirely random and was corrected for on the assumption that this is true. That is, it was assumed that the amount of diffuse scattering into a cone of solid angle $d\omega$ making an angle θ with the normal to the crystal surface was proportional to $d\omega \cos \theta$. In Table I are given the actual weights of specular and direct deposits, and the corrected weight of the specular deposit. From this it appears that 16 to 17 percent of the incident beam is specularly reflected at 45°.

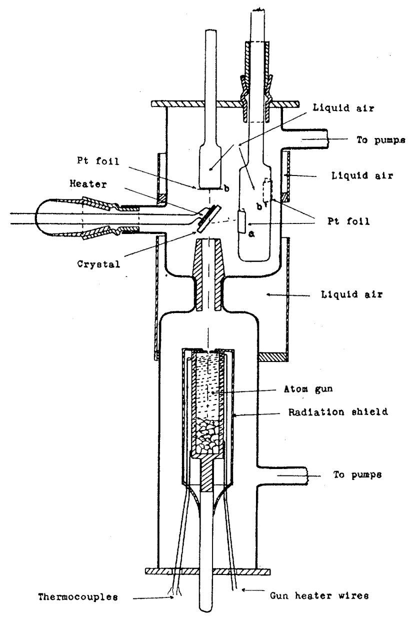

Fig. 6. Type of tube used to obtain quantitative reflection data.

To determine whether the intensity of specular reflection varied with the angle of incidence essentially the same apparatus was used, save that the crystal was mounted to rotate about an axis lying in the plane of the crystal

TABLE I. *Weights of specular and direct deposits.* (*45°*)

Specularly reflected cadmium Uncorrected	Specularly reflected cadmium Corrected	Direct beam	Percent specularly reflected
(1) 0.283 mg	0.190	1.060	17.9
(2) 0.197	0.128	0.800	16.0

surface passing through the center of the incident beam. This was done to eliminate the possibility of changes of intensity due to changes in the reflecting surface. Specularly reflected beams were received upon the receptors

TABLE II. *Weights of deposits and corrections for diffuse scattering.*

Angle		Weight of cadmium				Ratio
		Uncorrected		Corrected		
θ_1	θ_2	θ_1	θ_2	θ_1	θ_2	θ_2/θ_1
22.5	67.5	0.265	0.384	Negligible	0.357	1.35
20.5	66.0	.057	.094	correction	.088	1.54
22.0	66.0	.042	.056		.052	1.24
				Average		1.38

a and b^1, placed on opposite sides of the rotatable liquid air container. This avoided the accumulation of diffusely scattered atoms upon one receptor while the other was being deposited. Variation due to change in rate of emission from the boiler was avoided just as before. Table II gives the weights of deposits and corrections for diffuse scattering.

Discussion

The experiments on reflection from two crystals in succession show that the probability of specular reflection depends upon the velocity and the angle of incidence, for there is evidently a selective action at the first reflection which determines whether the beam when incident upon the second crystal shall be specularly or diffusely reflected. It is not reasonable to suppose that this selection is with respect to anything except velocity. Direct measurements of the reflected velocities confirm this. It is too early to say definitely that the wave-length associated with the translatory motion of an atom is h/mv, as quantum mechanics would have it. Bethe[7] has shown that in the wave picture of the Davisson and Germer experiments there is a quantity analogous to the refractive index in optics, so that the dependence of velocity or wave-length upon the angle of refraction gives an equation

$$\lambda = h/mv = 2d(\mu^2 - \cos^2\theta)^{1/2} \tag{1}$$

for the space grating type of reflection. Here

$$\mu = (1 - \phi/\tfrac{1}{2}mv^2)^{1/2}$$

where ϕ is the average potential energy of the particle when inside the crystal and v its initial velocity. Using this value of μ in Eq. (1) we get

$$\frac{h^2 + 8d^2m\phi}{4d^2m^2v^2} = 1 - \cos^2\theta .$$

So that $1/v^2$ plotted against $\sin^2\theta$ should give a straight line through the origin. So far as our present data go the relation is linear, but the curve does not pass through the origin, see Fig. 7. In fact the data agree within the errors of observation with the equation

$$\lambda' = h/m_Hv = 2d(2.26 - \cos^2\theta)^{1/2}$$

Where λ' is the wave-length characteristic of a proton and the refractive index is constant and equal to 1.5 nearly. However it is hard to justify the use of this wave-length[8] and the agreement may be accidental. If the mass of cadmium atom is used we may secure the same agreement by making

[7] Bethe, Naturwiss. **15**, 787 (1927).

[8] Evidence from specific heats (D. M. Dennison Proc. Roy. Soc. **A115**, 483 (1927); Heisenberg, Zeits. f. Physik **41**, 239 (1926)) makes it appear rather certain that protons, like electrons, are governed by the Fermi-Dirac statistics. For alpha-particles however this is probably not the case so that it is not impossible that the wave accompanying the translatory motion of a cadmium atom may have the wave-length which the de Broglie equation would attribute to a helium atom.

$$\mu=(2.26-\phi/\tfrac{1}{2}mv^2)^{1/2}$$

in Eq. (1), and we get a refractive index less than unity.

If diffraction patterns of the plane grating type can be obtained they will serve to fix the wave-length uniquely. With space grating diffraction involving the same power of the velocity on both sides of the equation this

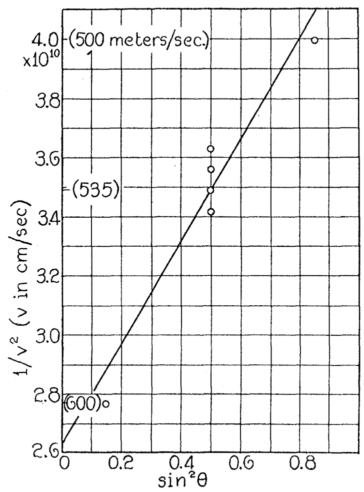

Fig. 7. Experimental curve plotting $\sin^2\theta$ against $1/v^2$.

cannot be done, unless an independent method of measuring ϕ is available. Where the atom reflected is a constitutent of the crystal, ϕ might be computed from heats of formation, and in other cases data on the stability of monomolecular surface layers might yield significant results.

Intensity of Reflected Beams and Resolving Power of Crystals

The intensity of a reflected beam of wave-length λ may be expected to depend upon the number of atoms in the incident beam having wave-lengths between λ and $\lambda+\Delta\lambda$, and upon the resolving power of the crystal. In reflection of x-rays the resolving power $R=\lambda/\Delta\lambda$ will be constant provided the number of crystal planes reflecting is constant. So in the present somewhat analogous situation it may not be unreasonable to suppose that the range of wave-lengths $\Delta\lambda$ over which reflection occurs will vary inversely as λ. The number of atoms in the incident beam having wave-lengths between λ and $\lambda+\Delta\lambda$ is

$$dN=N\left(\frac{m}{2\pi kT}\right)^{3/2}\frac{h^4}{m^4}e^{-h^2/2kTm^2\lambda^2}\frac{1}{\lambda^5}d\lambda.$$

If the probability of reflection drops off regularly decreasing to zero in a range $\Delta\lambda=R/\lambda$ then the ratio of the numbers reflected by crystals set to reflect velocites λ_1 and λ_2 will be

$$\frac{n_1}{n_2}=\left\{\exp\left[-\frac{h^2}{2kTm^2}\left(\frac{1}{\lambda_1^2}-\frac{1}{\lambda_2^2}\right)\right]\right\}\left(\frac{\lambda_2}{\lambda_1}\right)^4.$$

Taking λ_1 and λ_2 to correspond to velocities of 500 to 600 meters per second, the values observed for angles of 22.5 and 67.5°, we find this ratio to be 1.35. The mean of the observed values for angles near these two, given in Table II is 1.38. Obviously in order that 16 percent of the incident beam may be specularly reflected, as at 45° incidence, requires reflection to occur over a considerable range. To form a rough idea of what this range may be it may be well to note that 16 percent of the atoms emerging from the gun have velocities in the range 535 ± 45 meters per second. This represents merely a lower limit, as complete reflection over such a range, dropping at once to zero outside is not to be thought of. On the other hand the range over which appreciable reflection occurs probably does not greatly exceed ± 45 meters per second, as otherwise there would be noticeable widening of the deposits obtained with the velocity analyzer. The same conclusion may be drawn from the experiments on reflection from two crystals in succession. When the difference in velocities for the two crystals is as much as 45 meters per second very little specular reflection occurs at the second crystal, most of the beam incident upon it being scattered at random.