FEBRUARY 15, 1930 PHYSICAL REVIEW VOLUME 35

THE REFLECTION OF BEAMS OF THE ALKALI METALS FROM CRYSTALS

By John B. Taylor*
General Electric Company, Schenectady, N. Y.

(Received January 9, 1930)

Abstract

Beams of lithium, potassium, and caesium were reflected from crystals of sodium chloride and lithium fluoride, as a means of studying the wave nature of these atoms. Incident angles from 2° to 60° were investigated. Although one one-hundredth percent of specular reflection could have been detected, no trace of such a reflection or of diffraction was found. The measured angular distribution of reflected atoms followed closely the cosine law. Apparatus, and the detecting device depending on positive ion emission by which this sensitivity of measurement was obtained, are described.

The reflection of a beam of atoms from a crystal or ruled grating has for some time been recognized as a means of investigating the wave nature of the atoms.[1] Recently the author has described a method of detection and intensity measurement of molecular beams.[2] This method has proved so reliable and sensitive that it gave opportunity for a very detailed study of the reflection of the alkali metals from crystals. This has been done, using the detector for measuring the angular distribution of the atoms reflected when a beam of alkali metal atoms is allowed to fall upon a crystal.

Reflection experiments have already been carried out for a few atoms. Knauer and Stern[3] have obtained specular reflection of helium and hydrogen molecules from sodium chloride and potassium chloride crystals. In addition they have measured distinct surface diffraction beams arising from the crossed grating. These occur, within the present limits of the experiment, at the angle calculated from the surface grating formula using the de Broglie wave-length, $\lambda = h/mv$, obtained from the velocity and mass of the helium atom. Johnson[4] has shown that hydrogen atoms are specularly reflected from sodium chloride. Kirschbaum[5] also reports experiments with hydrogen atoms. No evidence of diffraction was found in either case. Ellett, Olson, and Zahl[6] have obtained specular reflection of cadmium, mercury, arsenic, and antimony from sodium chloride crystals. Certain features of their results not in

---

* National Research Fellow, University of California, Berkeley, California.

[1] O. Stern, Zeits. f. Physik **39**, 762 (1926).

[2] J. B. Taylor, Zeits. f. Physik **57**, 242 (1929).

[3] F. Knauer and O. Stern, Zeits. f. Physik **53**, 779 (1929); O. Stern, Naturwiss. **21**, 391 (1929).

[4] T. H. Johnson, J. Frank. Inst. **206**, 301 (1928).

[5] H. Kirschbaum, Ann. d. Physik **2**, 213 (1929).

[6] A. Ellett and H. F. Olson, Phys. Rev. **31**, 643 (1928); Ellett and Zahl, Phys. Rev. **31**, 1122 (1928).

agreement with a surface grating action are yet to be explained. They also report a trial of reflection of sodium from sodium chloride. No regular reflection was observed.

In the present work, beams of lithium, potassium and caesium have been reflected from crystals of sodium chloride and lithium fluoride. The most probable de Broglie wave-lengths in question, at furnace temperatures of 900°, 500°, and 450°K respectively, vary from about 0.2×10^{-8} cm for lithium to 0.07×10^{-8} cm for caesium. No trace of specular reflection or diffraction has been found. On the contrary the intensity of reflection measured at various angles with the surface follows almost exactly the cosine law of Knudsen; i.e. all of the atoms are reflected diffusely from the point of incidence. One-tenth of one percent specular reflection could easily have been observed in the present experiments. In some cases one one-hundredth of a percent could have been detected. In the other experiments mentioned the specular reflection has varied from almost one hundred percent[7] in the case of helium, to 10 to 20 percent in the case of cadmium, and one or two percent for atomic hydrogen.

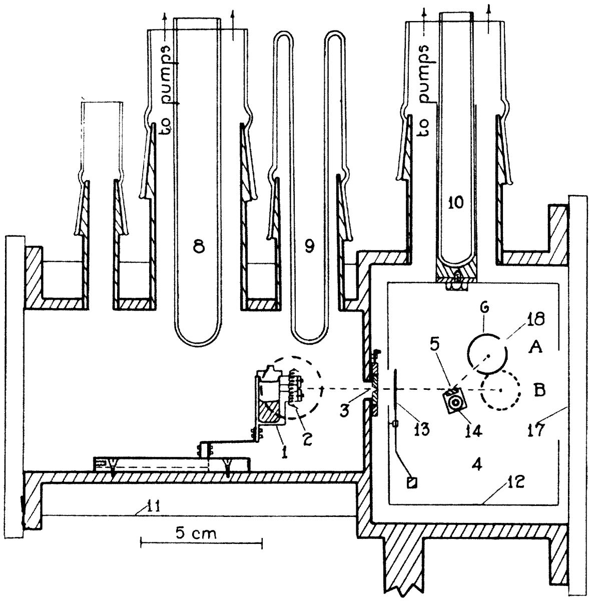

Fig. 1. Section of apparatus showing path of beam to crystal and detector.

Apparatus

The beam forming arrangement, furnace, slits, etc. are practically the same as have been described previously.[8] Figure 1 gives a schematic view

---

[7] Recent experiments of O. Stern about to be published, and as described to the author in private communication.

[8] J. B. Taylor, Zeits. f. Physik **52**, 846 (1929); **57**, 242 (1929).

of the situation. Atoms of the alkali metal evaporate through a fine slit in the steel oven (1) which is heated by radiation or by electron bombardment from the tungsten spiral (2). The beam is then formed by a second slit (3) and passes into the reflection chamber (4). Here it impinges on the crystal surface (5). The reflected atoms are received on the filament of the detector (6) which can be moved to intercept and measure the number of atoms leaving the crystal at any angle.

In the sketch the detector in position (*A*) observes the specular beam. The detector was a platinum cylinder eighteen×fifty mm with a coaxial pure tungsten filament 0.05 mm in diameter. The cylinder was held ten volts negative to the filament and at a temperature of about 1500°K. Every alkali metal atom striking the filament leaves as a positive ion.[9] Thus the positive ion current is at once a measure of the intensity of the beam or of the reflected atoms at the point. The detector is carried on the ground joint (7) Fig. 2, by which it may be moved to cover various angles of reflection. For lithium the detector filament was oxygen on tungsten.

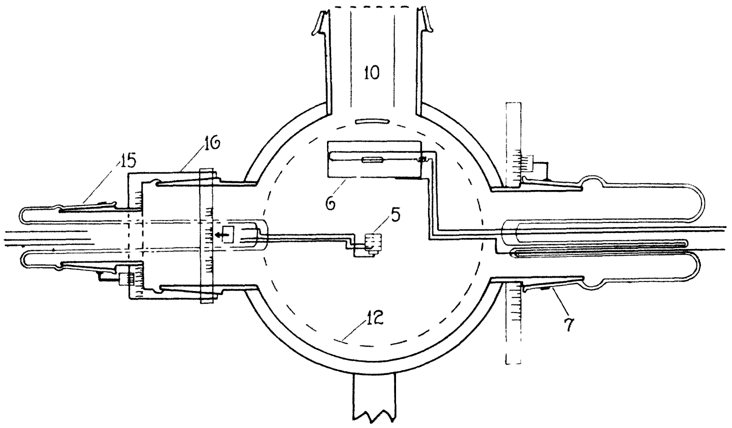

Fig. 2. End view of detecting chamber showing mounting and motions of crystal and detector.

The oven and crystal chambers were evacuated separately by two large Langmuir pumps. Liquid air traps (8), (9), (10), served to protect the chambers from grease vapors from the joints. The walls of the apparatus were held below room temperature by the water jacket (11). Added protection is given the detector and crystal by a sheet copper shield (12) cooled by liquid air from the trap (10). (13) is a shutter operated by action of a small magnet on an iron lever. It allowed interuption of the beam at any instant.

The crystal (5) is carried in the holder and heater (14). A tungsten spiral running through the body of the holder allows heating the crystal by radiation to at least 500°C. The holder is mounted on the ground joints (15) and (16) Fig. 2. (15) is eccentric in (16) so that by turning (16) the crystal can be

---

[9] I. Langmuir and K. H. Kingdon, Proc. Roy. Soc. **21**, 380 (1923).

raised into or lowered out of the path of the beam. By turning (15) the crystal is rotated on an axis through its surface and coaxial with the motion of the detector, to set the crystal at any incident angle with the beam.

Since the detector motion is coaxial with the crystal motion, the filament remains equidistant from the crystal at all angles and the positive ion currents require no correction for distance.

Graduations on the various joints permitted accurate readings of the angular settings.

Crystals used. Natural crystals of sodium chloride were used in a few experiments. Later to secure more perfect crystals, they were grown from the melt by the excellent method of Kyropolous.[10] Single crystals 3 to 5 cm in diameter were easily made in a few hours. Lithium fluoride was first purified by this method from an impure melt, and then crystalized from a melt of the pure product. The crystals were held at 2 to 300°K until ready for use. After cleaving they were immediately inserted in the apparatus and evacuation and heating begun at once.

During the reflection experiments the crystals were held at temperatures varying from room temperature to about 500°C. The elevated temperatures were to prevent if possible the momentary condensation or adsorption of alkali metal atoms on the crystal surface, and to insure a surface free from adsorbed gases.

The crystals used were 5 to 10 mm$\times$6 mm.

PROCEDURE

The oven was loaded and the experiment begun as described[8] previously, with the addition of the crystal introduction. After five to eight hours pumping and heating of the crystal the oven was brought gradually to a temperature giving the desired beam intensity. This changing oven pressure could be readily followed by moving the detector into the path of the beam, (position B in Fig. 1), with the crystal lowered. In addition when the desired intensity was reached the detector filament was moved by 0.01 to 0.05 mm steps through the beam to measure its intensity from point to point. Thus in size and intensity the incident beam was accurately known. Fig. 3 gives examples of such measurements. The small displacements of the filament during this measurement were controlled by use of a traveling microscope with eyepiece micrometer. Through the glass window (17) and the detector opening (18), the filament could be seen as a bright line against the scale.

The width of the beams striking the crystal was changed in different trials from 0.05 mm to 1 mm. The maximum intensity in the undeflected beam at B was varied from 30 cm full galvanometer sensitivity to about 60 meters equivalent deflection. These latter were measured with galvanometer shunted to one-thirtieth of full sensitivity. The galvanometer had a full sensitivity of 4×10^{-11} amps. per mm.

Then by means of the joint (16) the crystal was raised into and parallel to the beam. This could be done very accurately by noting the decrease in gal-

---

[10] S. Kryopolous. Zeits. f. Anorg. Chemie **154**, 308 (1926).

vanometer deflection as the crystal intercepted half of the beam. Next with the shutter (13) closed the crystal was turned to whatever incident angle desired, the shutter opened and observations begun. Incident angles from 2° to 60° were investigated. By joint (7) the detector was moved around the crystal, the intensity of the reflected atoms at any angle being recorded immediately by a certain steady galvanometer deflection. In the neighborhood of the expected specular reflection the steps could be made in fractions of a degree and the whole region explored a number of times to remove any question as to the character of the reflection. Readings could also be made by setting the detector at a given angle and then rotating the crystal to investigate any preferred angles of reflection.

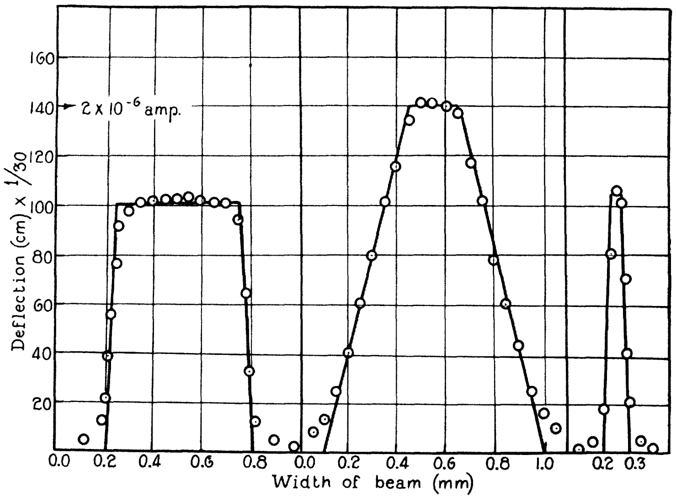

Fig. 3. Examples of intensity measurements of beams used. Solid lines are the geometrical beams constructed from the slit arrangements.

At any angle of reflection, if the shutter was closed the galvanometer deflection fell at once to zero.

At any time the crystal could be lowered and a check made on the constancy of the incident beam.

At the larger beam intensities one-tenth percent of specular reflection would have given 30 to 60 mm galvanometer deflection. However in no case was there the slightest indication of specular reflection.

Fig. 4 shows a reflection measurement with potassium from sodium chloride at an incident angle of 15°. The reflection from lithium fluoride and at other intensities of the main beam gave similar data.

An interesting critical temperature phenomenon was noted in the course of the experiments. If the crystal was allowed to cool, a temperature range was reached where the reflection at any angle fell very quickly practically to zero. This temperature where almost all the incident atoms were condensing on the crystal varied, as to be expected, with the incident beam intensity. It is apparent that the combination of beam and detector offers a precise method of studying surface condensation and evaporation phenomena.

The experiments made so far on the reflections of atoms from crystals have led to no explanation of the efficiency of reflection. The great ease with which the alkali metal atoms are adsorbed on surfaces is believed to explain at least in part their failure to be specularly reflected or diffracted. To avoid this, the possibility is suggested of employing crystals which may be heated to still higher temperatures than in these experiments, or crystals on which the adsorbing forces are much smaller. However unless unusual conditions as to reflecting surface can be secured, it may not be possible to detect an appreciable specular reflection of the alkali metal atoms.

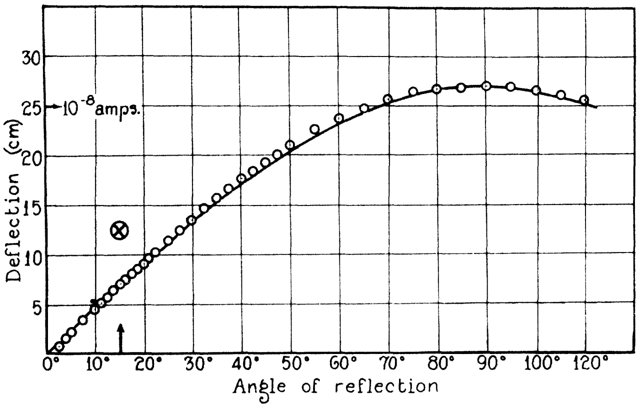

Fig. 4. Showing measured angular distribution of reflection (points) as compared to a cosine distribution (solid curve); the incident beam making an angle of 15° with crystal. Cross indicates intensity which would have been measured at 15° if one-tenth percent of the incident atoms had been specularly reflected.

The writer wishes to express his appreciation of support from the National Research Council and to Professor G. N. Lewis for the facilities so kindly made available at the University of California.