Shape of the Modified Compton Line for Hydrogen and Ceylon Graphite Scatterers

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Hydrogen under ten atmospheres pressure was used to scatter almost directly backward the characteristic K radiation of a molybdenum-target x-ray tube. After an exposure of 1914 hours in a focusing curved-quartz-crystal spectrograph the resulting modified line was studied with a microphotometer. The breadth of the composite modified line of the alpha-doublet was 15.2 x.u. This is ten percent greater than the breadth computed by B. Hicks from the electron momentum distribution. That the discrepancy is greater for H_2 than for the previously studied He is probably because the H_2 calculations were not carried to so high a degree of approximation. The unmodified line was completely absent. The shift as determined by fluorescence reference lines is less than that required for free electrons by an amount appropriate to the binding energy of electrons in the H_2 molecule.

A renewed search was made for evidence of any departure of the electron momenta from an isotropic directional distribution in Ceylon graphite, a diamagnetic crystal. This time the diamagnetic axis of the graphite was placed normal to the direction along which the experiment resolves the electron velocities. There was no indication of two lines or prominences symmetrically disposed on either side of the center of the Compton line corresponding to shifts for the directly approaching and receding electrons.

A small error in the previous determination of the shift defect for helium is noted.

I. INTRODUCTION

THE authors have previously shown by a comparison of experiment and theory that the modified line in the Compton effect has a broad structure whose shape reflects directly the velocity distribution of the electrons (their linear momenta) in the atoms of the scattering body; the broadening effect on the modified line is to a classical approximation (which in the present case is a very good one) simply a Doppler broadening caused by the quantum motions of the electrons scattering the radiation. Including the results of the present paper we may say that we have shown the breadth of the line to vary quantitatively as a function of all four of the available variables, scattering angle, primary wave-length, scattering material and physical state (solid or gaseous) in accord with the predictions of the above concept.[1]

Recently the authors[2] succeeded in obtaining for the first time[3] a direct spectrum of modified x-radiation scattered by a gas, helium. For the purpose of comparison with theory Mr. Bruce Hicks[4] of this Institute kindly consented to compute the momentum distribution to be expected in the helium atom[5] and the shape of the Compton "line" which should result from scattering of the radiation by electrons with such a distribution. At the same time Hicks also computed the momentum distribution and resulting line shape for the hydrogen molecule H_2 which latter turned out to be very appreciably narrower than the Compton line for a helium scatterer. Such a marked dependence of Compton line

[1] P. A. Ross, Proc. Nat. Acad. Sci. **9**, 246 (1923). G. E. M. Jauncey, Phys. Rev. **25**, 314 and 723 (1925). DuMond, Phys. Rev. **33**, 643 (1929); DuMond and Kirkpatrick, Phys. Rev. **37**, 136 (1931); **38**, 1094 (1931); DuMond and Hoyt, Phys. Rev. **37**, 1443 (1931); DuMond, Rev. Mod. Phys. **5**, 1 (1933); DuMond and Kirkpatrick, Phys. Rev. **52**, 419 (1937).

[2] DuMond and Kirkpatrick, Phys. Rev. **52**, 419 (1937).

[3] Kappeler, Ann. d. Physik **27**, 129 (1936). This author had studied the shape of the Compton line from a gaseous scatterer (neon) previous to our study of helium, but without obtaining a direct spectrum of the scattered radiation. Kappeler used a refinement of a method first proposed and used by P. A. Ross in which by varying the angle of scattering and hence the shift the modified line is made to cross an absorption discontinuity defined by a filter. The shape of the Compton line can be deduced from the results.

[4] B. Hicks, Phys. Rev. **52**, 436 (1932).

[5] A. L. Hughes, Phys. Rev. **53**, 50 (1938). This author, by the study of inelastically scattered electrons, has developed a very beautiful and precise method of experimentally observing the distribution of electron momenta in the atoms of a gas which furnished in the above reference a very satisfactory check on the theoretical predictions of Hicks as to helium.

breadth on the scattering atom furnishes a valuable check on the validity of our concept of the Doppler broadening of the line, and we have therefore undertaken the present study with a gaseous hydrogen scatterer.

II. Hydrogen Scattering Experiment

The reader is referred to our previous article[2] on the Compton line with helium gas as scatterer for details of the experiment, apparatus and procedure which were identical in the present work save for the scattering chamber. This latter we moved somewhat with respect to the primary and the scattered beams and we lengthened it from one meter to 1.5 meters. These improvements permitted us to reduce the pressure from 14 to 10 atmospheres with still an appreciable gain in scattered intensity for a given exposure time.

The characteristic K-line radiation from a molybdenum target was scattered almost directly backward by the hydrogen for 1914 hours; the voltage and current supplied to the tube were 60 kv and approximately 20 milliamperes. As before, test films behind the film proper in the focusing curved crystal spectrograph were used to test the progress of the exposure.

The reference spectrum on the edges of our film was accidentally fogged. Fortunately certain fluorescence lines which always appear on our film from the lead backing in the scattering chamber could still be used in conjunction with other spectra previously made in the same instrument to establish the exact position of the unshifted lines.

Thus we were able, in spite of the mishap, to determine the exact shift of the Compton line scattered by hydrogen. This was done in the same manner as we did for the case of helium.[2]

III. Results of Hydrogen Scattering Experiment and Their Interpretation

Computation of Predicted Line Profile

We have used, because of its intensity, the alpha-doublet of the molybdenum K spectrum as the primary radiation. The profile of each of the two shifted lines resulting from the two members of this doublet is so much broader than the doublet separation that the doublet character

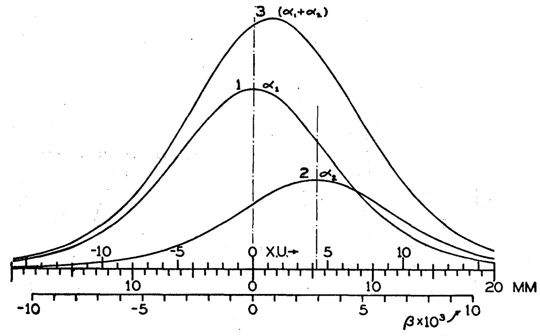

Fig. 1. Hicks' computed curve for the Compton line as broadened by his theoretically computed electron momentum distribution in the hydrogen molecule is here shown as curves 1 and 2 plotted to correct wave-length scale for the cases of Mo $K\alpha_1$ and $K\alpha_2$ as primary radiation and for our scattering angle. Curves 1 and 2 are plotted with relative intensities in the ratio (2 : 1) and with a wave-length separation of 4.27 x.u. (the separation of the alpha-doublet in the Mo K spectrum). Thus curve 3, which is the sum of 1 and 2, gives the correct shape to be compared with our observed shifted line. These curves show that the doublet nature of the primary radiation has but a slight distorting effect on the line shape.

of the primary radiation disturbs the shape of the profile but little even for the case of hydrogen scattering which gives the narrowest shifted line we have yet observed. Fig. 1 shows how we have corrected the theoretical shape for the doublet nature of the source by plotting Hicks theoretical curve to correct wave-length scale in the two positions corresponding to shifted α_1 and shifted α_2 with the appropriate relative intensities (2 : 1) and adding the two resulting curves. This gives us a master theoretical curve for comparison of the profile with our experimental results. This master comparison curve on which appear the positions of the centers of shifted α_1 and α_2 also serves to determine the shift very conveniently. This is done by sliding the master curve horizontally along the wave-length scale until it coincides for best fit with the observed microphotometer trace and then measuring the shift from the position of either member of the unshifted alpha-doublet to the line center of the same member marked on the shifted master curve.

In every case the vertical or ordinate scales of theoretical and experimental curves have been adjusted so as to normalize these curves to the same area.

Figure 2 shows a few sample microphotometer traces, nine of which were averaged by reading

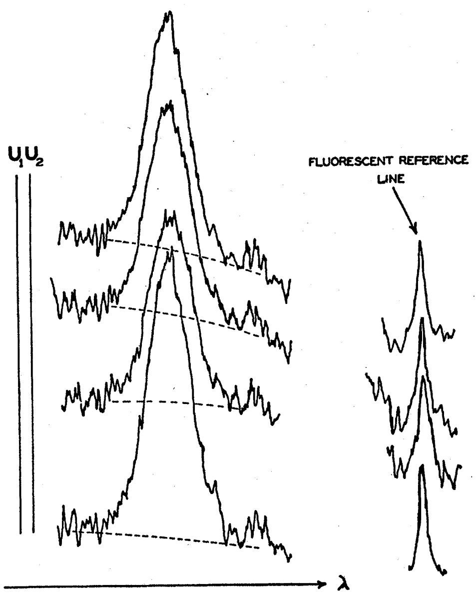

FIG. 2. Samples of single microphotometer traces of the Compton line with gaseous hydrogen as the scatterer. The unshifted wave-length positions are marked as U_1 and U_2. Nine such traces were numerically averaged to obtain the final results for comparison with the theoretical curves.

some eighty ordinates (every millimeter), taking the numerical average of these readings, and replotting these averaged results. As usual the background of the spectrum is found to have a slight slope and we are obliged to determine the average slope for all our microphotometer traces and subtract from the final averaged trace a straight line background having this slope.

The abscissa scale of Hicks' computed profiles is given by him in terms of the variable $\beta = v/c$, where v is the electron velocity responsible for the broadening of the Compton line. This is transformed into wave-length units by the formula

$$l = 2\beta\lambda^*, \quad (1)$$

where l is the abscissa of the shifted line measured from its center in wave-length units and λ^* is given by

$$\lambda^* = \tfrac{1}{2}(\lambda_1^2 + \lambda_c^2 - 2\lambda_1\lambda_c \cos\theta)^{\frac{1}{2}}, \quad (2)$$

where θ is the scattering angle, λ_1 is the primary wave-length, $\lambda_c = \lambda_1 + (h/mc)(1-\cos\theta)$ is the shifted wave-length for initially stationary electrons.

The abscissa scale of the computed theoretical profile must next be transformed into centimeters on the microphotometer plate to facilitate comparison with the microphotometer curves. This involves a knowledge of the dispersion which the geometry of the transmission-type focusing curved-quartz-crystal spectrograph imposes on the original film and also the ratio of abscissa magnification imposed by the recording microphotometer. This latter has been determined as 10.0024 ± 0.001 uniform over its entire range to the indicated precision which is far higher than necessary for our present purposes. As for the dispersion of the spectrograph the method of its determination and the errors involved have been treated at length in our paper on helium scattering to which the reader is referred.[2]

In Fig. 3 the full line gives the theoretical Compton line profile derived from Hicks' computations while the dots are the observed values obtained by averaging the microphotometer traces. Experiment thus indicates that the Compton line is here about ten percent broader than Hicks' theory predicts. Our helium results supported Hicks' predictions with much better agreement than this. Hicks is of the opinion that his results for molecular hydrogen might well be in error by this amount as the approximation to which these somewhat more difficult calculations were carried out was not as high as in the case of his helium calculations. Also in the hydrogen case it was difficult for him to estimate the error committed at a given stage of approximation. Hicks points out that for his helium calculations at one of his earlier stages of approximation which gave an error of 2 percent in the energy levels he found an error of 10 percent in the momentum (breadth of the Compton line at half-maximum height). Hicks' calculations for H_2 were only carried to a stage of approximation which yielded about 2 percent error in the energy levels (owing to the rapidly increasing difficulties for this molecule) and it is interesting to note that our experimental results seem to indicate the error in momentum here to be about 10 percent again as in the case of helium.

A. L. Hughes[6] has also observed this slight excess (11 percent) for the actual momentum breadth in the H_2 distribution (as compared to

[6] A. L. Hughes, Phys. Rev. 54, 189 (1938).

Hicks' predictions) by his method of inelastically scattered electrons.

In order to make quite sure that the excess observed over Hicks' calculated breadth might not be an instrumental effect, we narrowed the exploring slit of our recording microphotometer until it admitted less than half the intensity used in making our traces of the line profiles and we ran another profile with this new slit. The breadth of this curve was identical with that obtained before and we conclude therefore that this cannot explain the excess breadth.

This shifted line for a hydrogen scatterer is the narrowest so far observed at this scattering angle and primary wave-length in accord with the lower electron velocities to be expected for hydrogen. Our observed breadth at half-maximum height for the shifted composite $\alpha_1+\alpha_2$-line is 15.2 x.u. which corresponds to a breadth at half-maximum for a strictly monochromatic primary line of 14 x.u. We arrive at this correction on the assumption that the ratio of breadth of the composite shifted line to breadth of the pure shifted line will be the same in the actual case as in the case of the Hicks' theoretical curves which approximate the observed curves. This ratio is 1.09.

For comparison we list below the breadths at half-maximum of Compton lines for two gases and one solid used as x-ray scatterer, all corrected as above to give the breadth for a strictly monochromatic primary line of wave-length 710 x.u. at scattering angles approximately 180°.

H_2, 14 x.u.; He, 18.6 x.u.; Graphite, 21.4 x.u.

Determination of the Shift

Our exposure shows no unmodified line so we have determined the shift directly and separately from each of our microphotometer records as follows: The positions on the microphotometer recording plate of the center of shifted α_1 and α_2 were found by fitting to the actual microphotometer curve of the Compton line a plotted composite curve (like the one in Fig. 1) on which the center lines of the shifted α_1- and α_2-bands appear. These positions were next measured with reference to a fluorescence line which appears on our spectra whose position is accurately known relative to the unshifted α_1- and α_2-lines from previous work.

The average shift so measured turns out to be 59.12 mm on our microphotometer plates or 5.912 mm on the original film and the deviations of the nine individual measurements on the nine plates from the mean indicate the probable error of the mean as ± 0.08 mm on the microphotometer plate or ± 0.008 mm on the original film (± 0.13 percent). From the average dispersion of the spectrometer (in Siegbahn wave-length units) for the range of wave-lengths between Mo $K\alpha_1$ and shifted Mo $K\alpha_1$, which is 80.622 x.u./cm, we compute the shift to be 47.66 x.u. and correcting this slightly to express it in terms of absolute or grating wave-lengths we obtain finally for the observed shift 47.75 ± 0.06 x.u. where the error is based only on the internal consistency of the nine observations.

The Shift of Initially Stationary Free Electrons

We have recomputed the mean effective value of $(1-\cos\theta)$ averaged over the scattering volume with appropriate weighting for distance from source and from the spectrometer (see our article on helium scattering[2]) and we obtain

$$(1-\cos\theta)=1.9933.$$

Taking a value of 24.15×10^{-11} cm for h/mc we obtain 48.13×10^{-11} cm for the shift to be expected at our mean effective scattering angle for free initially stationary electrons on the simple Compton theory.

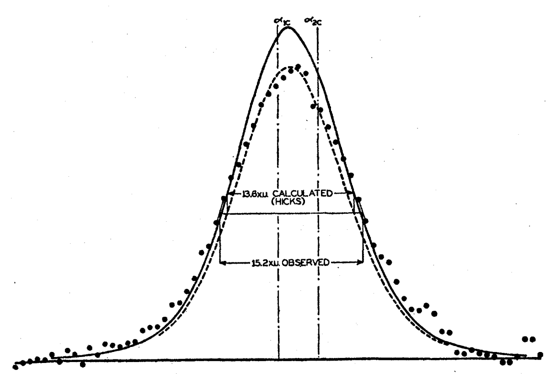

FIG. 3. Comparison of calculated and observed Compton line profiles for a gaseous hydrogen scatterer. The dots are the averages of measurements made on nine microphotometer traces. The full and dashed lines are each Hicks' calculated curve corrected for the doublet nature of the primary radiation. The vertical scale of the full line curve has been normalized to the same area as the observed curve while the dashed curve has been adjusted to have the same maximum ordinate as the observed curve.

The Defect in the Shift

As Ross and Kirkpatrick have shown both theoretically and experimentally[7] x-ray scattering by bound electrons should take place with slightly less shift than for free electrons. The recoiling electron ejected from the atom by the Compton effect may be thought of as slightly "loaded" with a small fraction of the mass of its parent atom through the agency of the binding forces and the greater effective m in the expression h/mc calls for a smaller shift. Our present results seem to support this defect in the shift, our value for the defect for hydrogen being $48.13-47.75=0.38$ x.u. This should be compared with a defect we obtained for the case of helium scattering of 0.53 x.u. The lower binding energy of hydrogen as compared with helium leads one to expect a lower value for the defect in the shift and our results seem consistent with this. We do not wish however to attach undue faith to our shift-defect measurements which may well be in error by considerably more than the "probable" value ± 0.06 x.u. indicated by the application of the theory of errors to our shift measurements.

Correction of a Slight Error in our Shift Defect for Helium

In the present computations we have discovered a small error which we committed in computing the mean effective value of $(1-\cos\theta)$ for our paper on the Compton effect with a helium scatterer.[2] We find that the mean value of $(1-\cos\theta)$ should have been, for that work, 1.994 instead of 1.997.

This has no effect whatever on any of the conclusions of that paper except the computed value of the shift to be expected for free stationary electrons and the resultant shift defect. Those should be corrected to 48.16 x.u. and 0.53 x.u., respectively.

Internal Evidence That Our Shifted Line Was Scattered by Hydrogen Only

The absence of unmodified scattering for weakly bound electrons is well understood. When the energy imparted to the electron by the radiation (recoil energy) is large compared to the binding energy there is a high probability that the electron will be ejected from the atom and the energy balance then requires a change of wave-length. For hydrogen even more so than for helium the unmodified scattering is utterly negligible and no unshifted lines whatever could be observed on our film. Every other substance which could conceivably be present in our scattering chamber would have an atomic number equal to or greater than carbon which for our primary radiation and scattering angle gives unmodified lines of the same height approximately as the modified line. The absence of unmodified lines is thus conclusive evidence that our Compton line was scattered by the hydrogen alone. The lead lining and back wall of our chamber can easily be shown to scatter negligibly in comparison to the hydrogen because its fluorescence absorption so greatly limits the thickness of the layer from which scattering can occur.

[7] Ross and Kirkpatrick, Phys. Rev. **45**, 223 (1934); **46**, 668 (1934).

IV. Experiment with Ceylon Graphite Scatterer

Reason for the Experiment

Five conditions influence the breadth and shape of the Compton modified line, and of these we believe we have experimentally verified the effects of four. The five are: the scattering angle, the primary wave-length, the material of the scatterer, the physical state (solid, liquid or gaseous) of the scatterer, and finally an effect which might be expected if the electrons in the scatterer do not on the average have their linear momenta isotropically directed in the crystalline lattice of the scatterer. The fifth condition would make the shape of the Compton line dependent on the orientation of the crystalline structure of the scatterer with reference to the incident and scattered beams of radiation. So far we have been unable to reveal the fifth effect.

For radiation observed under a given scattering angle θ we have shown[8] that there exists a natural reference axis in space (which is very nearly the bisector of the supplement of the

[8] This "natural reference axis" is "the vector change in light momentum for scattering at angle θ by an initially stationary electron." See DuMond and Kirkpatrick, Phys. Rev. **37**, 142, 143 (1931); **38**, 1094, 1100 (1931); DuMond, Kirkpatrick and Alden, Phys. Rev. **40**, 168, 170 (1932); DuMond, Rev. Mod. Phys. **5**, 1 (1933).

scattering angle and lies in its plane) such that it is the component electron momentum resolved along this axis which is responsible for the Doppler broadening of the Compton line. As Jauncey[9] has later pointed out, the shape of the Compton line is a picture of the component electron momentum distribution. It is along this natural axis that the momenta are to be resolved.

Our first attempt to reveal the existence of anisotropic electron momenta was made with Ceylon graphite crystals[10] which exhibit marked diamagnetic anisotropy and which occur in small thin hexagonal flakes the diamagnetic axis being normal to the flakes. If the diamagnetism could be attributed to a fairly numerous class of electrons in the crystal lattice executing thin, flat, approximately circular orbits these we thought might have no component momentum normal to the plane of the orbit. We therefore tried the experiment with the axis of diamagnetic anisotropy of the crystals directed along the above mentioned "natural reference axis" for our incident and scattered beams of radiation. Scattering from a sufficiently populous class of electrons having no component momentum along this natural reference axis should contribute a sharp shifted line in the center of the broad band from the remaining electrons. Nothing of the sort was observed. This might have been either because such a class of electrons existed but was insufficiently populous or because the electrons responsible for the diamagnetism did not behave as we pictured them. Oppenheimer pointed out that electrons spatially restricted in the axial direction in thin flat orbits would by the uncertainty principle be required to have a wide spread in momentum in the corresponding (axial) direction.

The scattering blocks we used, and which we still have, are built up out of the tiny crystal flakes orientated by the combined action of settling in a fluid and of a superposed magnetic field. The flakes are held together by a very tiny quantity of gum binder.[11] Very recently it has occurred to us to try placing these blocks with

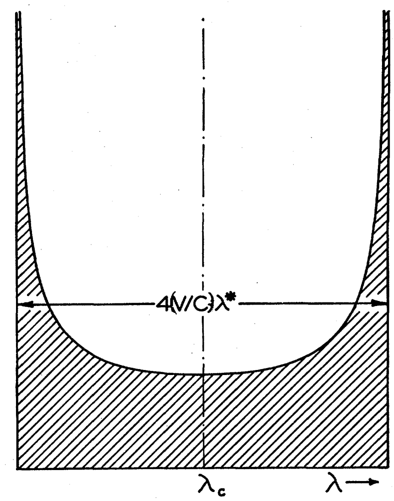

FIG. 4. Ideal modified spectrum to be expected from a class of electrons revolving at speed, v, in circular orbits with their axes normal to the natural reference axis for the scattering experiment.

the diamagnetic axis *normal* to the natural reference axis of the scattering experiment. In this new position if there exists a class of electrons responsible for the diamagnetic anisotropy executing approximately circular orbits at roughly one constant tangential velocity one should expect the radiation scattered by such a class to exhibit two peaks symmetrically disposed on either side the center of the Compton line and corresponding to the shifts for the electrons approaching and receding parallel to the natural reference axis. Fig. 4 is a sketch of the intensity distribution to be expected ideally for a class of circular orbits with uniform tangential speed. Some rough approach to this might perhaps be anticipated in the form of two bumps or a mere flattening at the top of the shifted lines.

Experimental Results and Interpretation

Figure 5 shows a microphotometer record from our scattered spectrum with the orientated Ceylon graphite scatterer. The diamagnetic axis of the graphite was normal to the natural reference axis of the scattering experiment and the plane of the graphite flakes was parallel to the plane of the scattering angle. The effective value of the scattering angle was approximately 137° with a range of inhomogeneity of about ±4°. The exposure of 54 hours with the molyb-

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[9] G. E. M. Jauncey, Phys. Rev. **46**, 667 (1934).

[10] DuMond, Kirkpatrick and Alden, Phys. Rev. **40**, 165 (1932).

[11] We are much indebted to Messrs. A. Goetz, A. B. Focke and A. Faessler (see Phys. Rev. **39**, 168 (1932)) for purifying the graphite and preparing the orientated blocks.

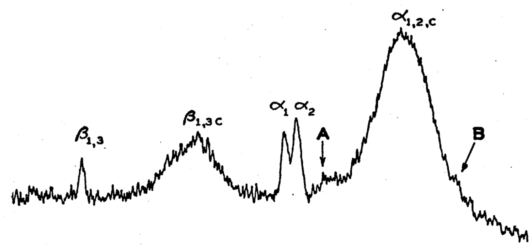

FIG. 5. Microphotometer curve of Mo K radiation scattered by orientated Ceylon graphite with its diamagnetic axis normal to the natural reference axis of the scattering experiment.

denum target tube running at 60 kv and 20 ma served to give a very clear scattered spectrum.

Examination of Fig. 5 shows nothing which we feel inclined to interpret as an effect of anisotropically directed electron momenta. Near the unshifted α-lines to their right there is a shelf, A, with a region of less intense blackening separating it from the α-lines themselves. This less intense region is distinctly visible on the original negative as a white line. There is the bare possibility of interpreting B as a companion shelf to A. However the fact that A and B are not symmetrically disposed around the center of the Compton line argues conclusively against these prominences as maxima corresponding to the velocities of approach and recession of a class of electrons in circular orbits responsible for the diamagnetic anisotropy. The Doppler spread between A and B (if we assume momentarily that these *are* approach and recession maxima) indicates an orbital velocity referred to light velocity of $\beta = v/c = 0.0178$ corresponding to a kinetic energy of 80 volts. This is too low for the K electrons in graphite and too high either for L electrons or structure electrons.

The shelves A and B may correspond to double or multiple scattering inside our scatterer which was rather large and of more or less the same dimensions in all directions. This configuration would be such as to favor multiple scattering whose importance relative to single scattering increases with the size of the scatterer.[12] The spread between A and B is somewhat too broad for the band due to multiple scattering at a primary scattering angle of 137°, however.

[12] For a theoretical discussion of the effects of multiple scattering see DuMond, Phys. Rev. **36**, 1685 (1930).

The shelf at A might be an effect predicted by a number of authors in which one expects to find the continuous modified Compton band sharply limited on its short wave-length side by a discontinuity corresponding to a loss in quantum energy (relative to the primary frequency) equal to the binding energy of the scattering electrons.[13] The position of A calls for a binding energy of over 200 volts which may represent the binding energy of the K electrons in carbon.

The absence of the effect sought may again mean either or both of two things. The class of electrons responsible for the diamagnetic anisotropy in graphite may not have a sufficiently sharply defined momentum distribution approximating the kind we have postulated (circular orbits with uniform tangential velocity) or the class may be insufficiently populous to scatter an observable amount of shifted radiation.

The faintness of the unshifted α_1-line on this exposure is its most curious feature. The intensity of unshifted α_2 relative to the modified doublet corresponds well with our previous experience for graphite scatterers but the unshifted α_1-line should be more than twice as intense as it is. It is difficult to explain this as a vagary of the photographic negative because the line has this faintness over its entire length. We are totally at a loss to understand this weak α_1-line.

ACKNOWLEDGMENTS

We are indebted to Dr. A. Goetz of this Institute for furnishing the purified hydrogen which he pumped under pressure into our scattering chamber.

It is a pleasure to express our gratitude for the financial assistance which supported this research received through the generosity of Dr. Leon L. Watters of New York City.

[13] P. A. Ross, Proc. Nat. Acad. Sci. **9**, 246 (1923). G. E. M. Jauncey, Phys. Rev. **24**, 204 (1924); **25**, 314 (1925); **25**, 723 (1925). Jauncey assumes that an electron must receive in the scattering process enough energy from the radiation to be set free from the atom if the scattering is to be of the modified type. Thus the continuous distribution of shifted radiation which constitutes the Compton modified band must be limited on its short wave-length side at a shift for which the quantum energy lost by the radiation equals the electron binding energy. See also Compton, *X-Rays and Electrons*, page 286, 287; Wentzel, Zeits. f. Physik **43**, 1 (1927); **43**, 779 (1927); **58**, 348 (1929) and Sommerfeld, Ann. d. Physik [5] **29**, 715 (1937).