The Production of New Radiations by Light Scattering.—Part I.

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(PLATES 1, 2.)

1. *Introduction.*

In two preliminary papers* we have recorded the discovery that when monochromatic light is scattered in a transparent medium (be it gas, vapour, liquid, amorphous solid or crystal), the diffused radiation ceases to be monochromatic, and several new lines or sometimes bands (associated in many cases with a continuous spectrum) appear in the spectrograms of the diffused radiation. Further, the new radiations are, in general, strongly polarised. That the phenomenon is entirely distinct from what is usually known as fluorescence is clear from the fact that the effect is observed when both the exciting radiation and the new radiations generated by it are far removed from the characteristic ultra-violet and infra-red frequencies of the medium. As an illustration we may mention the case of transparent crystalline quartz in which the effect is very well shown with the 4358 A.U. line of mercury as the exciting line, the new lines also appearing in the indigo-blue region of the spectrum. Our preliminary studies have proved conclusively that the effect arises in the following way: The incident quantum of radiation is either scattered as a whole, in which case we have the classical scattering, or else is absorbed in part by the molecules of the medium, the remaining part appearing as a scattered quantum. The part absorbed shifts the molecule to a level of energy

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* Raman, 'Ind. J. Phys.,' vol. 2, p. 387 (1928); Raman and Krishnan, 'Ind. J. Phys.,' vol. 2, p. 399 (1928).

different from the initial state. The possibility of a process of this kind, in respect of the electronic state of an atom, was first contemplated by Smekal,* and figures prominently in the theory of dispersion due to Kramers and Heisenberg,† and in the papers of Schrödinger.‡ Our experiments furnish definite proof of the possibility of such processes, and show that they may occur also in such complicated systems as the molecules of a vapour or a liquid or even in a complete crystal. In the series of papers of which this is the first our further studies of the new radiations will be discussed.

2. *Experimental Methods.*

The experimental arrangements were exactly the same as those described in the previous papers. The liquids to be examined were rendered dust-free in the usual manner by repeated slow distillation *in vacuo,* and the final distillate was contained in a bulb of about 500 to 600 c.c. capacity. A 3000-c.p. mercury arc served as the source of monochromatic illumination and was concentrated with an 8-inch condenser, at the centre of the liquid bulb, which was kept immersed in a suitably blackened glass tank containing water. By interposing a strong solution of quinine sulphate and a plate of blue glass between the condenser and the liquid, practically all the lines of the incident mercury spectrum excepting the 4358·3 line and its close companions were cut out. In the case of carbon tetrachloride, where the modified lines were sufficiently close to the exciting line, no filters were used.

The spectrum of the scattered light was taken with a Hilger quartz spectrograph (E_2), using very rapid photographic plates (Ilford Iso-zenith, H. & D. 700). In the case of the single line pictures of benzene and toluene an exposure of about 40 hours was necessary, while for carbon tetrachloride, for which the complete mercury arc was incident, an exposure of only 25 hours was given.

3. *Experimental Results.*

In Plates 1 and 2 are reproduced the spectrograms of the scattering by benzene, toluene and carbon tetrachloride. The plates were measured with a Hilger travelling micrometer and the wave-lengths of the modified lines were calculated, using the mercury lines (feebly transmitted by the filters) as standards for reference. A simplified Hartmann formula was used for interpolation.

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* 'Naturw.,' vol. 11, p. 873 (1923).

† 'Z. Physik,' vol. 31, p. 681 (1925).

‡ 'Abhandlungen zur Wellenmechanik,' Leipzig, p. 112 (1927).

Table I.—Spectrum of Benzene Scattering.

Unmodified Lines.			Modified Lines.			Origin of the Modified Lines.	
Wave-length I.A.	Wave-number (*in vacuo* per cm.)	Intensity.	Wave-length I.A.	Wave-number (*in vacuo* per cm.)	Intensity.	Wave-length of the exciting line.	Difference of wave-numbers.
			4178·0	23928	0	4358·3	− 990
			4246·7	23541	0	4358·3	− 603
4339·2	23039	15					
4347·5	22995	30					
4358·3	22938	200					
			4476·4	22333	1	4358·3	605
			4525·8	22089	0	4358·3	849
			4534·5	22047	0	4339·2	992
			4543·4	22004	0	4347·5	991
			4555·1	21947	10	4358·3	991
			4593·9	21762	2	4358·3	1176
			4683·5	21346	1	4358·3	1592
			5029·6	19877	1 Broad	4358·3	3061

Table II.—Spectrum of Toluene Scattering.

Unmodified Lines.			Modified Lines.			Origin of the Modified Lines.	
Wave-length I.A.	Wave-number (*in vacuo* per cm.)	Intensity.	Wave-length I.A.	Wave-number (*in vacuo* per cm.)	Intensity.	Wave-length of the exciting line.	Difference of wave-numbers.
			4176·1	23939	0	4358·3	− 1001
			4213·7	23725	0	4358·3	− 787
			4262·3	23455	0	4358·3	− 517
			4315·6	23165	1	4358·3	− 227
4339·2	23039	15					
4347·5	22995	30					
4358·3	22938	200					
			4399·5	22723	2	4358·3	215
			4424·9	22593	1	4358·3	345
			4448·4	22474	0	4347·5	521
			4459·3	22419	2	4358·3	519
			4479·8	22316	1	4358·3	622
			4501·7	22208	0	4347·5 4358·3	787 730
			4512·8	22153	4	4358·3	785
			4547·1	21986	0	4347·5	1009
			4558·0	21933	7	4358·3	1005
			4563·0	21909	1	4358·3	1029
			4591·0	21776	1 Broad	4358·3	1162
			4600·8	21729	2	4358·3	1209
			4638·0	21555	1	4358·3	1383
			4648·3	21507	0	4358·3	1431
			4664·0	21435	0	4358·3	1503
			4685·5	21336	2	4358·3	1602
			4994·3	20017	0	4358·3	2921
			5027·3	19886	1	4358·3	3052

Table III.—Spectrum of Carbon Tetrachloride Scattering.

Unmodified Lines.			Modified Lines.			Origin of the Modified Lines.	
Wave-length I.A.	Wave-number (*in vacuo* per cm.)	Intensity.	Wave-length I.A.	Wave-number (*in vacuo* per cm.)	Intensity.	Wave-length of the exciting line.	Difference of wave-numbers.
			4273·5	23393	1	4358·3	– 455
			4299·6	23251	2	4358·3	– 313
			4317·2	23157	2	4358·3	– 219
4339·2	23039	15					
4347·5	22995	30					
4358·3	22938	200					
			4400·3	22719	7	4358·3	219
			4418·5	22626	8	4358·3	312
			4447·3	22479	10	4358·3	459
			4509·3	22170	2	4358·3	768
			4514·0	22147	2	4358·3	791

4. *Infra-Red Spectra of the Molecules.*

As has been shown in our previous papers, the shift in frequency of the modified lines must correspond to a characteristic frequency of the molecule. In the following table are exhibited the shifts in wave-number of the different modified lines and the corresponding infra-red wave-lengths characteristic of the molecule. The infra-red absorption spectra of these liquids have been studied by Coblentz,* Bell† and others, and their values are also reproduced in the table for comparison.

Considering the uncertainties in the direct measurement of infra-red spectra, the agreement between the values of the characteristic wave-lengths calculated from light-scattering and those measured directly should be considered satisfactory; thus confirming the conclusions drawn in our previous papers regarding the origin of the modified lines.

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* Quoted by J. W. Ellis, 'Phys. Rev.,' vol. 27, p. 305 (1926).

† 'J. Am. Chem. Soc.,' vol. 47, p. 2814 (1925).

Table IV.—Infra-Red Spectra of the Molecules.

Benzene.			Toluene.			Carbon Tetrachloride.		
Shift in wave-number.	Infra-red wave-length.		Shift in wave-number.	Infra-red wave-length.		Shift in wave-number.	Infra-red wave-length.	
	Calculated.	Observed (Bell).		Calculated.	Observed (Coblentz)		Calculated.	Observed (Coblentz)
Cm.$^{-1}$	μ	μ	Cm.$^{-1}$	μ	μ	Cm.$^{-1}$	μ	μ
3061	3·27	3·3						
			3052	3·28				
			2921	3·42	3·4			
			1602	6·24	6·2			
1592	6·28	6·2						
			1503	6·65				
			1431	6·99	6·9			
			1383	7·23				
			1209	8·27				
1176	8·57	8·5						
			1162	8·60	8·6			
			1029	9·71				
			1005	9·94	9·9			
991	10·09	9·7 to 10·2						
849	11·78	11·8				791	12·64	13
			786	12·72	12·1			
						768	13·02	
			730	13·69	13·9			
			622	16·07				
605	16·52							
			520	19·23				
						457	21·9	
			345	29·0				
						312	32·0	
						219	45·6	
			215	46·5				

5. *Light Scattering and Infra-Red Spectroscopy.*

Attention was drawn in our previous papers to the usefulness of light scattering as a convenient and accurate method in infra-red spectroscopy. In a single spectrogram taken in the visible region we get all the infra-red frequencies of the molecule simultaneously photographed, and they could be measured much more accurately than with an infra-red spectrometer. For example, in the case of sharp, bright lines, as some of those appearing in the spectrograms are, we can, with an instrument of larger dispersion and using suitable comparison standards, measure them correct to a hundredth of an Ångström unit, which corresponds in the near infra-red, say, 3 μ, to an accuracy of about 1 part in 60,000, and in the extreme infra-red, say, 20 μ, to 1 part in 10,000. In fact, the accuracy is limited only by the width of the modified line.

The appearance in the scattered spectra of toluene and carbon tetrachloride of modified lines corresponding to several hitherto unknown frequencies of the molecules, in the extreme infra-red region as far as 46 μ,* emphasises the ready applicability of the method especially to regions not accessible to the ordinary infra-red spectrometer.

6. *Enhancement of Frequency in Light Scattering.*

While most of the modified lines are of smaller frequency than the exciting line there appear in all the three spectrograms (figs. 2, 3 and 5) some relatively feeble lines whose frequencies *exceed* the frequency of the exciting line by an infra-red frequency of the molecule. These lines are particularly conspicuous in the case of carbon tetrachloride, where corresponding to each of the three prominent lines on the longer wave-length side of the exciting line we have a weaker line on the shorter wave-length side. The appearance of these lines of enhanced frequency proves in the first place the existence in the liquid at ordinary temperatures of some molecules at a level of energy higher than the normal by that corresponding to an infra-red frequency of the molecule, and secondly, that the incident light induces a return of these molecules to the normal state. That is to say, while most of the molecules taking part in modified scattering are in the normal state and absorb a part of the incident light quantum, thus giving rise to a scattered radiation of smaller frequency, there is also a small number in the liquid, already in the higher level of energy, which under the influence of the incident radiation can be induced to part with their energy by a return to the normal state, thus giving rise to a scattered radiation of correspondingly higher frequency. In the existence of these lines of enhanced frequency we have for the first time a direct experimental proof of *induced emission* of radiation by molecules.

It may be of interest to recall in this place that the idea of an induced emission of radiation or what has sometimes been described as a "negative absorption of radiation," was first put forward by Einstein in his celebrated paper† on the derivation of Planck's radiation formula, and forms an essential feature of his theory. The idea also figures prominently in the theory of dispersion developed by Kramers and Heisenberg (*loc. cit.*).

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* In fact in the case of toluene there is a line corresponding to even longer wave-lengths which could not, however, be measured in the particular negative.

† 'Phys. Z.,' vol. 18, p. 121 (1917).

The relative intensities of the positive and negative lines corresponding to a given molecular frequency ν can be easily calculated. The proportion of molecules in the higher level of energy is obviously given by the relation

$$f = e^{-\frac{h\nu}{k\mathrm{T}}},$$

where h is the Planck constant, k is the Boltzmann constant per molecule, and T is the absolute temperature. If we assume, as we might reasonably, that the transitions of the molecule to and from the higher level are equally probable, then the above ratio, viz., $e^{-\frac{h\nu}{k\mathrm{T}}}$ also represents the ratio of the intensities of the corresponding negative and positive lines.

Taking for example the case of carbon tetrachloride, corresponding to the shifts in wave number 219, 312 and 457 (cm.$^{-1}$) the calculated values for the ratio of intensities of the negative and positive lines are 1 : 2·8, 1 : 4·4 and 1 : 8·8 respectively, at the temperature of the experiment, viz., 30° C. Accurate measurements of the actual intensities of the lines have not been made. A rough estimate, however, of the intensities from the negative, using for comparison a series of graded exposures taken on the same negative and in the same region of the spectrum, gave the values 1 : 3, 1 : 5, and 1 : 10 respectively, in good agreement with the calculated values.

Conversely, we can take the observed intensity relationships between the different positive and the corresponding negative lines as an experimental verification of the assumption made in the earlier paragraph, that the forward and backward transitions of the molecule are equally probable. The probabilities of transitions of the molecule from the normal state to *different* energy levels are, of course, different, and the differences in the intensities of the different modified lines of degraded frequency are obviously due to this cause.

The rapid fall in the value of the ratio as ν increases offers a ready explanation why we do not get negative lines corresponding to large shifts of frequency. Also as the temperature is increased, since the proportion of molecules in a higher energy level also increases, we should expect the negative lines to brighten up relatively to the corresponding positive lines. Experiments are in progress to test this point.

7. *Continuous Spectrum accompanying the Scattered Lines.*

The molecular frequencies in the near and extreme infra-red, determining the shifts of the various modified lines discussed in the previous sections, are presumably vibrational frequencies. The question naturally arises whether we should not expect also shifts corresponding to the pure rotational frequencies of the molecules. For example, in the case of benzene, taking the rotational frequency of the molecule to be of the order of 100 μ the corresponding shifts for the 4358 line will be slightly less than 20 Å.U., and should be capable of being detected. A visual examination with a direct vision spectroscope, of benzene, toluene and some other liquids, showed a nebulosity or continuous spectrum accompanying the prominent lines in the scattered spectrum. The actual spectrograms reproduced here and in our earlier papers also show a general broadening of the lines. However, in view of the presence of a general photographic halation accompanying the bright lines—the plates used were not backed—we thought it desirable to take fresh pictures with a larger dispersion instrument, and they are reproduced here. Fig. 7 (Plate 2) shows the nebulosity accompanying the 4358 line scattered by liquid benzene, fig. 6 being the direct mercury spectrum diffused by a plate of ground glass, taken with the same instrument.* Fig. 8 shows the scattering by carbon tetrachloride where, however, the nebulosity is not so conspicuous as in benzene.

From the fact that in the case of benzene the extension of the nebulosity from the exciting line is of the same order of magnitude as the rotational frequency of the molecule, one is tempted, by analogy with the explanation of the origin of the modified lines, to attribute the nebulosity to a combination of the rotational frequencies of the molecule with the frequency of the incident radiation. Owing to the continual impedance to rotation which must be present in a dense medium, it is not difficult to understand why we get a continuous spectrum instead of discrete lines. In connection with this explanation it may be pointed out that the nebulosity extends unsymmetrically on the two sides of the exciting line, being more conspicuous on the longer wave-length side.

Whether the comparatively feeble continuous spectrum in carbon tetrachloride scattering is due to a smaller rotational frequency of the molecule or is in any way connected with its symmetrical nature, is more than we can venture to answer at present. Further observations, especially in the vapour state, or in the liquid at different temperatures, are obviously necessary before we can fully understand the origin of this nebulosity.

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* In this connection see an interesting paper by Cabannes and Daure, ' Comptes Rendus, June 4, 1928.

8. *Polarisation of Scattered Radiations.*

In view of the imperfection in polarisation of the classical light-scattering at 90°, it becomes of importance to ascertain the nature of the polarisation of the modified radiations. For this purpose two spectrograms were taken with equal exposures and under identical conditions, side by side on the same plate, using a nicol in front of the slit of the spectrograph. During the first exposure the shorter axis of the nicol was perpendicular to the track in the liquid, and during the second, parallel to the track.

Fig. 9 gives the direct spectrum of the mercury arc, incident in the liquid, fig. 10 the scattered spectra of benzene, fig. 11 of carbon tetrachloride, and fig. 12 of amyl alcohol.

The following are some of the interesting results which emerge from a study of these spectrograms :—

(1) All the unmodified lines (*i.e.*, classical scattering) are polarised to practically the same extent.

(2) For a given shift of frequency, the modified lines excited by the different incident lines are polarised to the same degree, but differ in polarisation from the unmodified lines.

(3) The modified lines corresponding to different frequency shifts are polarised to different extents, the intensity of the weaker component varying from almost nothing to about 40 or 50 per cent. of that of the stronger line. For example, in the case of carbon tetrachloride, the most prominent modified line (which corresponds to a shift of 457 cm.^{-1}) is more or less completely polarised, while the other two prominent lines show a large imperfection in polarisation.

(4) The negative lines (*i.e.*, of enhanced frequency) are polarised to the same extent as the corresponding positive lines.

(5) The strong modified lines are usually more polarised than the feeble ones.

(6) The general continuous spectrum appearing in the scattering by amyl alcohol is also strikingly polarised.

A tentative explanation of these results regarding the polarisation of the modified lines may be suggested by analogy with the ideas put forward by the late Lord Rayleigh and by Born to explain the observed imperfection of polarisation of the classical scattering. In the classical theory the molecules of the medium behave under the influence of the incident light-vector, like oscillating doublets, which, as a consequence radiate out energy. If the moment induced in the molecule is in the same direction as the inducing force, the transverse scattering would obviously be completely polarised. The imperfection in polarisation experimentally observed therefore suggests that there are three principal directions in the molecule mutually perpendicular to one another, only along which moments can be induced, and that to varying extents. If the incident force lies along any one of these directions the induced moment is along the same direction. In every other case we have to resolve the incident force along the principal axes of the molecule and then determine the corresponding moments induced in these directions. Incidentally, we may point out that on this basis, assuming the molecules to be independent scattering centres, as is very nearly the case in a gas, the ratio of the intensities of the two principal polarised components of the scattered light can never exceed 50 per cent., the latter limit being reached when the molecule is polarisable only along one axis.

Carrying now the analogy to the case of modified scattering let us tentatively assume that for a given energy transition of the molecule, there are three principal directions in the molecule, the Einstein coefficients of probability of transition under the influence of an external force incident respectively along these three directions being different. As regards the actual mechanism which leads to such an anisotropy we shall for the present leave it an open question. The imperfection in polarisation of the modified lines follows then as a necessary consequence. If, further, we take these principal axes, as well as the corresponding principal Einstein coefficients, to be different for different energy transitions of the molecule, the differences in the intensity and the degree of polarisation of the different modified lines are also readily explained. That for a given frequency shift the polarisation is independent of the inducing line is also obvious. The identity in the degree of polarisation of the positive and negative lines corresponding to a given frequency shift will then merely be a consequence of the equal probability of transitions in the positive and negative directions; that is to say, for the same reasons for which, as we pointed out in sec. 6, the intensities of the negative and positive lines are in the ratio of the number of molecules in a correspondingly higher level of energy, to the number in the normal state.

It is significant in connection with the above explanation that there is no modified line in the spectrograms for which the ratio of the intensities of the principal polarised components is greater than 50 per cent. Physically interpreted, this limiting value of the polarisation signifies that the vibration to which the frequency under consideration corresponds, can take place only along a definite axis in the molecule, which is *a priori* not improbable.

It remains now to discuss the relation between the polarisation of the unmodified radiations and that of the different modified ones. The modified radiations are presumably incoherent, *i.e.*, the radiations at any given instant from neighbouring molecules have no definite phase relationship, so that the polarisation of the modified radiation would be characteristic of the molecule and independent of its state of aggregation. On the other hand, the unmodified radiations from neighbouring molecules are definitely correlated in phase, and their polarisation will be characteristic of the molecule only in the vapour state. Actual calculation shows the polarisation in the liquid state to be considerably smaller. Thus for a fair comparison we should take the polarisation of the modified lines as given by the spectrograms with that of ordinary classical scattering not in the liquid but in the vapour. The vapour values for the ratio of components of classical scattering are benzene 4·5 per cent., amyl alcohol, 1·2 per cent., and carbon tetrachloride 0·5 per cent.

On making the actual comparison we find that most of the modified lines are much less polarised than the unmodified ones. If we can take the degree of anisotropy of the transitions corresponding to the infra-red frequencies as evidenced by the imperfection of polarisation of the corresponding modified lines, as being typical of the anisotropy of all other kinds of energy transitions of the molecule which determine its refractivity, especially the electronic transitions of ultra-violet frequencies, then the smaller polarisation of the modified lines is easily understandable; because the anisotropy of the molecule as a whole, which determines the polarisation of classical scattering, is merely a resultant of the anisotropies of all the individual transitions (suitably weighted) of the molecule, and must, therefore, necessarily correspond to a greater spherical symmetry.

9. *Dependence on Wave-Length.*

It is well known that the classical scattering falls off rapidly with increasing wave-length, following Rayleigh's λ^{-4} law, and it will, therefore, be of interest to find how the intensity of the modified radiations depend on the wave-length of the exciting line. No quantitative measurements have yet been made which would enable us to decide this question. But we may draw attention here to the spectrogram of carbon tetrachloride scattering, which throws some light on the point. Comparing the intensities of the green λ 5460·7 line and the ultra-violet λ 3906·5 line of the mercury arc, as they appear in the spectrum of the scattered light the former line is much brighter, whereas comparing the modified lines excited by them, those excited by the green line are distinctly feebler than the ones excited by the ultra-violet line. It would thus appear that the intensity of the modified line changes with wave-length even more rapidly than is indicated by Rayleigh's inverse fourth-power law.

This is further confirmed by visual observations on the spectrum of scattered light with a direct vision spectroscope. Though as seen through the spectroscope the scattered green mercury line is far more intense than the indigo or violet lines, the modified lines excited by the green line could not be detected, whereas those due to the violet and indigo lines are very conspicuous.

10. *Summary.*

In two preliminary papers the authors have shown that when any transparent medium, be it gas, vapour, liquid, amorphous solid or crystal, is irradiated by monochromatic light the radiations scattered by the molecules contain several spectral lines of modified frequencies, the difference between the incident and scattered frequencies corresponding to a characteristic infra-red frequency of the molecule. The present paper describes further studies on these radiations.

(*a*) Using the λ 4358 group of lines of the mercury arc as the exciting radiations, the scattered spectra of benzene, toluene and carbon tetrachloride have been photographed and measured.

(*b*) The characteristic infra-red frequencies of the molecules are calculated from the frequencies of the modified lines and are compared with the values obtained from direct measurements of infra-red absorption. The calculation gives several molecular frequencies hitherto unknown.

(*c*) The usefulness of light-scattering as a powerful, convenient and accurate method of exploring molecular spectra, is pointed out.

(*d*) While most of the modified lines are of smaller frequency than the exciting line, there are some relatively feeble lines whose frequencies exceed the frequency of the exciting line by an infra-red frequency of the molecule. In the appearance of these lines we have for the first time a direct experimental proof of *induced emission* (or negative absorption) of radiation by molecules.

(*e*) The scattered lines are sometimes accompanied by a nebulosity or continuous spectrum, extending unsymmetrically on the two sides. Its origin is discussed.

(*f*) The modified radiations scattered at 90° exhibit striking polarisation, the degree of polarisation being different for lines corresponding to different frequency shifts. A tentative explanation is suggested.

(*g*) Some preliminary remarks are made regarding the dependence of the intensities of the modified lines on the wave-length of the exciting line.

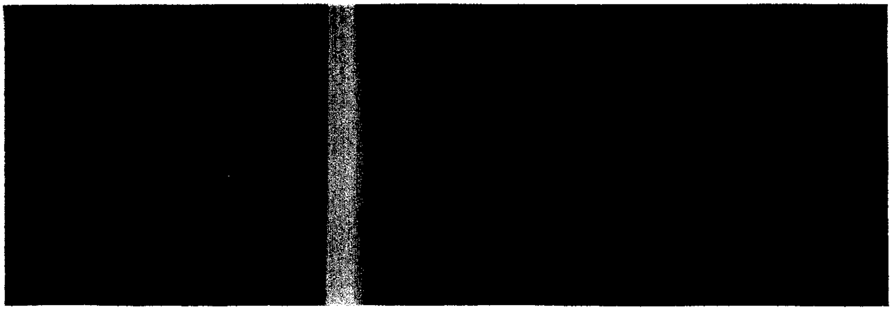

Fig. 1.—
Incident
Spectrum.

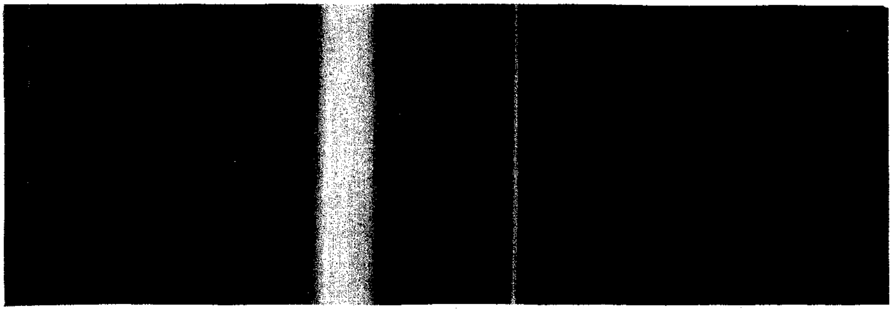

Fig. 2.—
Benzene
Scattering.

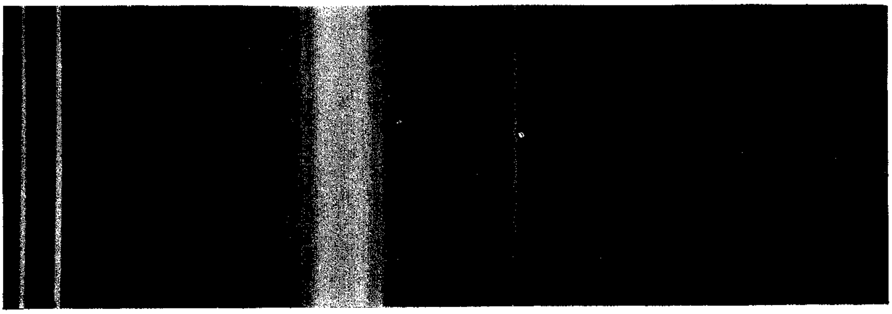

Fig. 3.—
Toluene
Scattering.

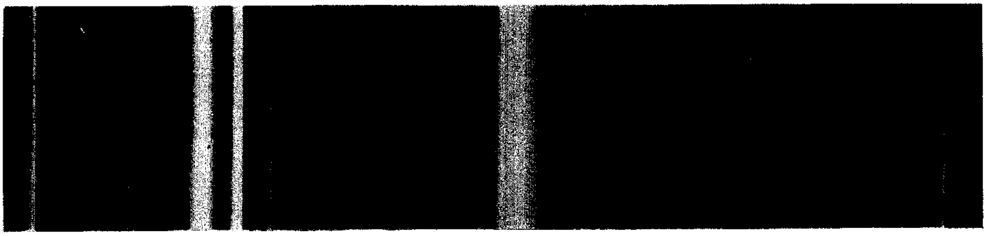

Fig. 4.—
Incident
Spectrum.

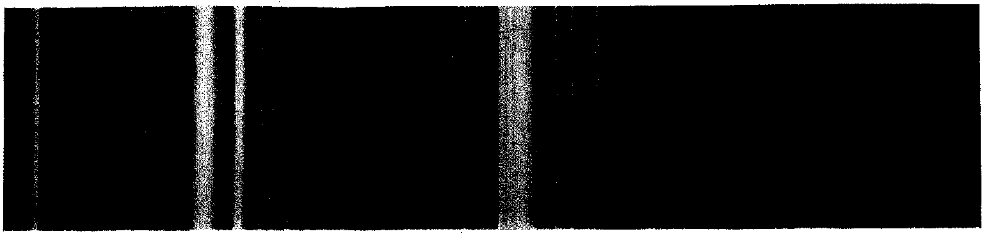

Fig. 5.—
Carbon
Tetrachloride
Scattering.

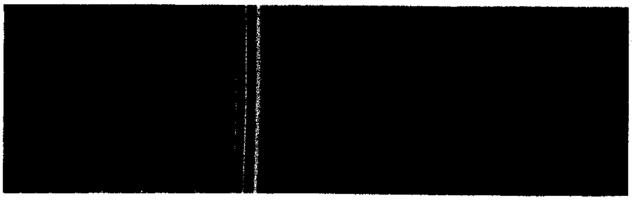

Fig. 6.—
Diffused by
Ground Glass.

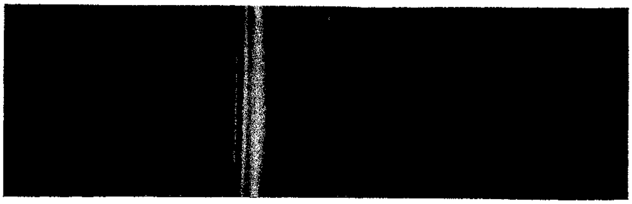

Fig. 7.—
Scattered
by Benzene.

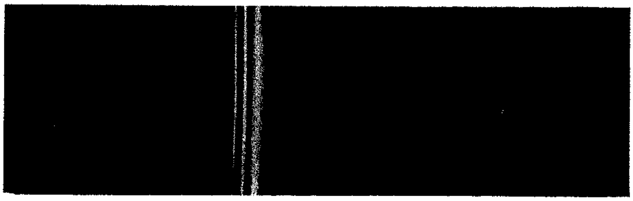

Fig. 8.—
Scattered
by Carbon
Tetrachloride.

Continuous Spectrum accompanying the Scattered Lines.

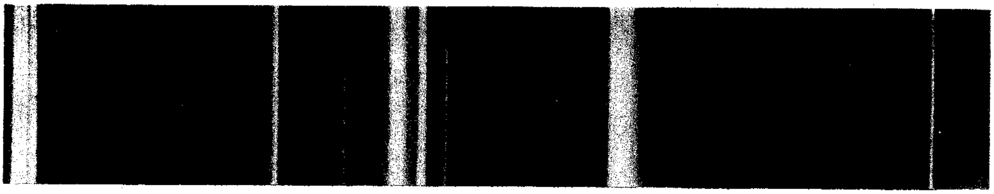

Fig. 9.—
Incident
Spectrum.

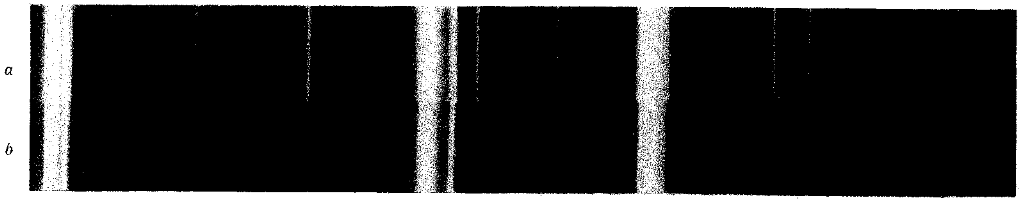

Fig. 10.—
Benzene.

Fig. 11.—
Carbon
Tetrachloride.

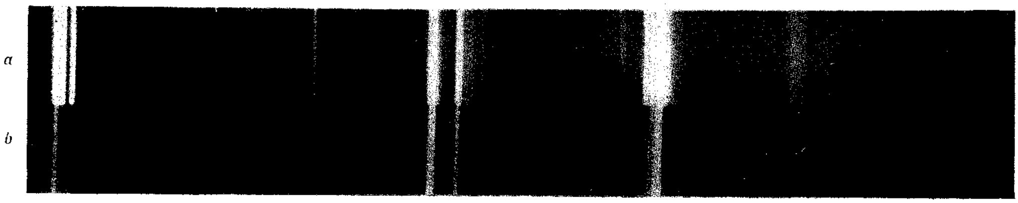

Fig. 12.—
Amyl
Alcohol.

Polarisation of Scattered Radiations.

DESCRIPTION OF PLATES.

PLATE 1.

FIG. 1 gives the spectrum of the mercury arc from which practically all the lines excepting the λ 4358 group have been cut out by interposing a solution of quinine sulphate and a blue glass plate.

FIG. 2 is the spectrogram of the radiations shown in fig. 1 when scattered by liquid benzene.

FIG. 3 is a similar spectrogram of the radiations scattered by toluene.

FIG. 4 is the direct spectrum of the mercury arc which was incident on carbon tetrachloride.

FIG. 5 being the corresponding scattered spectrum. Note the appearance of relatively feeble lines of enhanced frequency.

PLATE 2.

FIG. 6.—Spectrum of the λ 4358 group of lines diffused by a plate of ground glass.

FIG. 7.—Spectrum of the same as scattered by benzene. Note the nebulosity accompanying the scattered line, which is not present in fig. 6.

FIG. 8 shows the same line as scattered by carbon tetrachloride. The nebulosity is not so prominent as in benzene.

FIG. 9 gives the complete spectrum of the mercury arc used for polarisation measurements (for comparison with figs. 10 to 12).

FIG. 10.—Spectrum of the mercury arc scattered by benzene, taken with a nicol in front of the slit of the spectrograph. In (*a*) the shorter axis of the nicol was perpendicular to the track, while in (*b*) it was parallel to the track.

FIG. 11 shows similarly the polarisation of the scattered radiations from carbon tetrachloride.

FIG. 12 is a similar spectrogram of the radiations scattered by amyl alcohol. Note the continuous radiations are also partly polarised.