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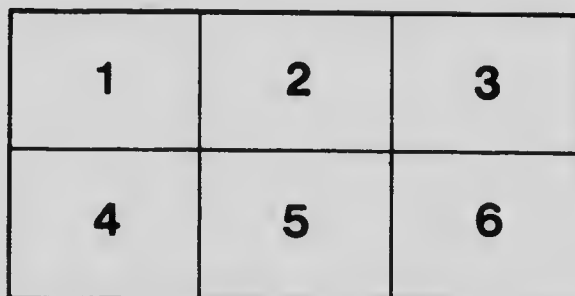
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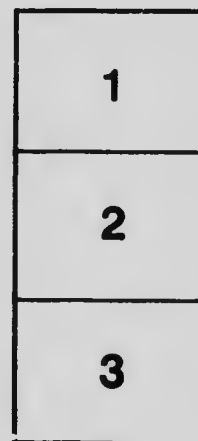
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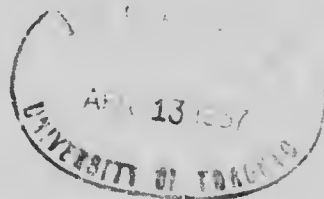


**UNIVERSITY OF TORONTO
STUDIES**

**PAPERS FROM THE CHEMICAL
LABORATORIES**

**No. 112: THE SCATTERING OF LIGHT BY DUST-FREE
LIQUIDS, BY W. H. MARTIN**

(REPRINTED FROM THE JOURNAL OF PHYSICAL CHEMISTRY, VOL. XXIV)



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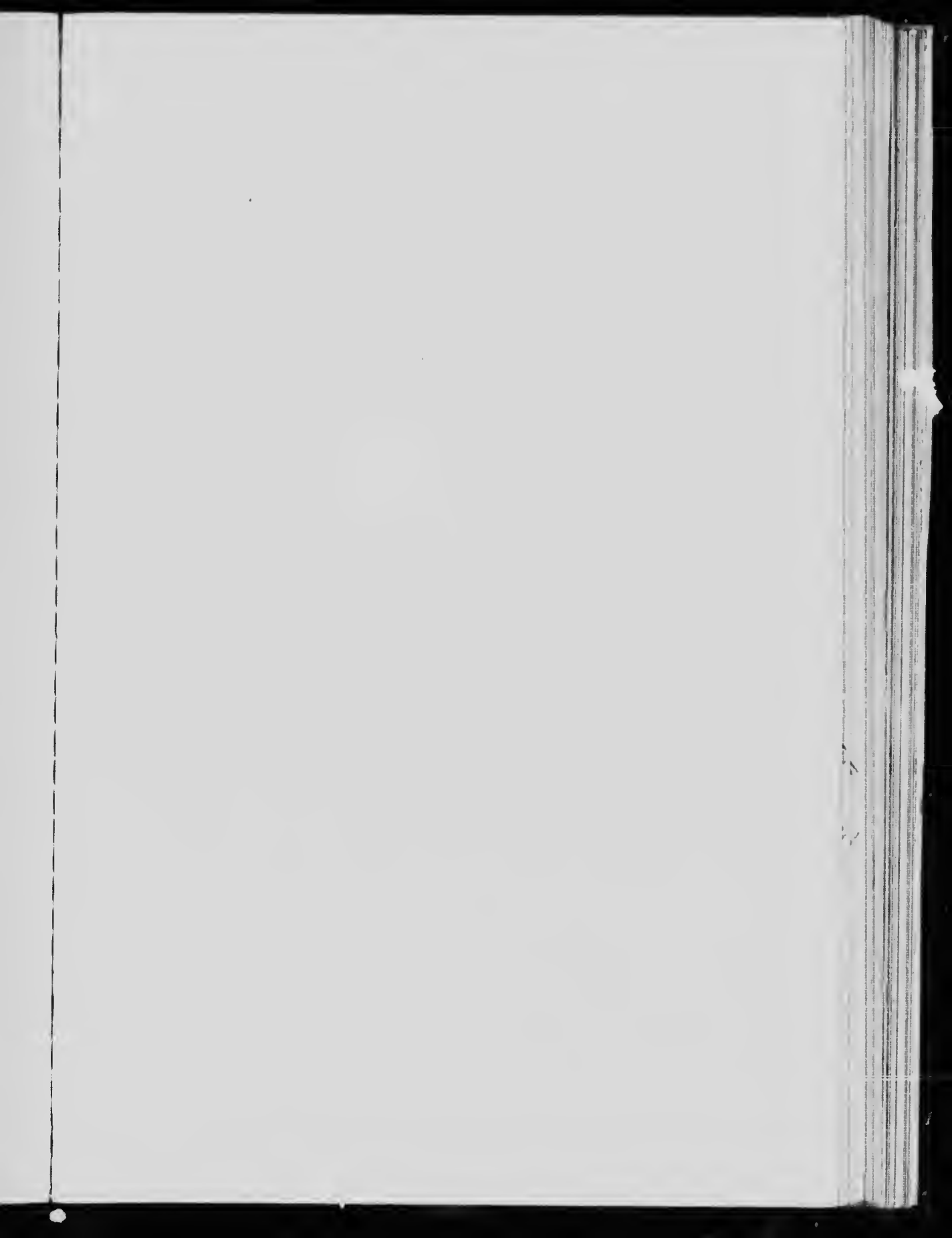
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THE SCATTERING OF LIGHT BY DUST-FREE LIQUIDS.

BY W. H. MARTIN

1. Historical

If a strong beam of light be concentrated in a flask of water and viewed in a direction at right angles to that of the beam against a black background, the path of the light through the liquid is clearly visible. The cause of this "light cone" in carefully distilled liquids has been a subject for controversy at least since 1869, when Lallemand and Soret disagreed as to its cause. Lallemand¹ thought it a property of the liquid itself, while Soret² attributed it wholly to particles suspended in the liquid.

Some years later the problem was attacked and apparently settled by two different experimenters: Spring³ described the preparation of "optically empty" water by two different methods, while Tyndall described the preparation of optically empty air by filtration through cotton-wool. Lobry de Bruyn and Wolff⁴ confirmed Spring's work, preparing dilute solutions of sodium chloride and other salts, which they said scattered no light. They found, however, that solutions of saccharose, raffinose and phosphomolybdic acid could not be made optically empty by the methods used. "Nous croyons pouvoir tirer de nos recherches la conclusion, qu'il faut répondre affirmativement à la question posée comme titre de cette communication, et que par conséquent les solutions 'véritables' de substances à poids moléculaire élevé sont susceptibles de provoquer la diffraction de la lumière. Ce résultat indique donc la continuité entre les solutions, vraies et les pseudo-solutions."

Several years ago, when some experiments on the be-

¹ Comptes rendus, 69, 1294 (1869).

² Ibid., 69, 1192 (1869).

³ Rec. Trav. chim. Pays.-Bas., 18, 153, 233 (189) See also 19, 24, 25, 29.

⁴ Ibid., 23, 155 (1904).

havior of solutions of partially miscible liquids at the saturation point were being carried out in this laboratory by W. J. Fawcett,¹ it was suggested that the reason for the difficulty in supersaturating such solutions was that dust particles served as nuclei for the formation of the second phase. The author of the present paper, in the winter of 1912-13, attempted to remove the dust from liquids by Spring's method, but found it impossible to obtain optically empty liquids either by this or by other methods, and came to the following conclusion:²

"The general result was that the light beam in water and in aqueous alcohol—the two liquids most thoroughly investigated—consists of two parts; a part which is removed by each of the methods of purification and a part which is not removed by any of the methods and which *is constant in intensity irrespective of the method of purification used*. This permanent part is faint and is plane-polarized, and can be seen only if the room is dark, the light beam very intense, and the vessel clean and free from striae."

As far as I am aware, this is the first conclusive evidence for the scattering of light by pure liquids—contrast indeed by solids and gases. This paper was presented at the meeting of the Royal Society of Canada in May, 1913. Almost at the same time Le Blanc and Kangro, in a preliminary communication,³ presented to the Deutsche Bunsen-Gesellschaft in August, 1913, and elaborated later⁴ describe their attempts to repeat Spring's preparation of optically empty liquids. They found they could still see the light cone if all foreign light was excluded; that strong solutions scattered more light than weak; and that solutions of colloids scattered more light than solutions of crystalloids. Finally they described an elaborate method of distillation, which, however, did not

¹ Proc. Roy. Soc. Canada, 7, III, 219 (1913).

² The Tyndall effect in liquids. Proc. Roy. Soc. Canada, 7, III, 219 (1913); Chem. Abs., 8, 3739 (1914).

³ Zeit. Elektrochemie, 19, 794 (1913).

⁴ Zeit. phys. Chem., 87, 257 (1914).

give optically empty water. The authors were nevertheless unwilling to grant that dust-free liquids might scatter light. "To attribute the scattering to the dissolved salt molecules is a belief for which no chemist or physicist is ready. It is much less far-fetched and more plausible to ascribe the scattering to fine suspended particles * * * *"

In 1915 Cabannes¹ observed the scattering of light in dust-free air, and a short time later R. J. Strutt² carried out some experiments—the results of which are discussed later in this paper—in which he measured the relative intensity and the polarization of the light scattered by a number of gases.

The conception that dust-free liquids and gases scatter light is one which seems to have been gradually forced on the experimenters more or less against their will, largely because Tyndall and Spring, by their classical researches on the removal of dust particles, had left such a strong impression that the scattered light was entirely due to this dust.

The very noticeable light scattered by many organic liquids (see p. 487) was observed by Lobry de Bruyn, who, however, called it fluorescence,³ apparently because he noted that a picric acid filter cut off the light. In this conclusion he appears to have disregarded the fact, experimentally shown by Tyndall, that very small particles scatter to an appreciable extent only the short wave lengths of light. Spring,⁴ too, observed that the cyclic organic compounds showed this faint blue "fluorescence" to a much greater extent than did aliphatic compounds. There can be no doubt that these two men, and probably many others, have observed the scattering of light; but, lacking any strict definition of fluorescence, they failed to distinguish between the two phenomena.

The work begun in 1912-13 by the author, was interrupted

¹ Comptes rendus, **160**, 62 (1915); see also **168**, 340 (1919).

² Proc. Roy. Soc., **94A**, 453 (1918); **95A**, 155, 476 (1919); Nature, **104**, 412 (1919).

³ Rec. Trav. chim. Pays.-Bas, **23**, 163 (1894).

⁴ Ibid., **16**, 1 (1897).

at that time to be taken up again recently. In view of the increased interest attached to the early results by reason of the later developments in the study of light-scattering, it is thought best to include in the present paper a more complete account of some of the results then obtained.

2. Preparation of Dust-Free Liquids

Three methods for removing the suspended motes were used: distillation, envelopment, and cataphoresis.

A. *Distillation of Water.*—The method of distillation in vacuum with the special form of apparatus (Fig. 1) was found

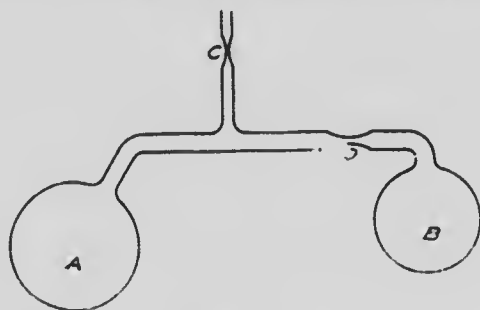


Fig. 1

to be much more convenient and more generally applicable than the other methods, and later was used entirely for the removal of motes from liquids of various kinds. Ordinary distillation of water in air or even in vacuum with boiling removes very few of the motes. With the apparatus shown the distillation can be conducted at a temperature far below the boiling point of the liquid and *without ebullition*.

Sufficient water is put into the bulbs to more than half fill each of them. By heating or by the use of an air pump the liquid in the bulbs is boiled until about one-third has been evaporated, after which the apparatus is sealed off at C, care being taken that no air is left in the bulbs. By keeping the bulb A 10° warmer than B a distillation without boiling is effected in a few hours. The first distillates are shaken back from B to A so as to wash back the dust. In most cases

the motes were all removed after four distillations. The tube T serves as a reflux condenser in which there is continuous condensation of the liquid. After distillation is completed the bulb B is sealed off at D.

After the fourth distillation of water there remain no motes, but still there is visible a light cone or opalescence. It is a bluish haze, faint but very definite, and repeated distillations up to ten times do not lessen its intensity. Distillations with the bulb A at 100° and B at 90° , and again with A at 20° and B at 0° give distillates with the same opalescence.¹

Fractional Distillation

By using an apparatus consisting of four bulbs of decreasing size all connected and sealed off with vacuum, and by properly adjusting the temperatures of the various bulbs some water was fractionated three times, but no difference whatever between any of the fractions could be observed.

Distillation in Quartz

Since glass is measurably soluble in water, it was thought possible that the light was scattered by some siliceous suspension from the walls of the vessels. If so it seemed unlikely that quartz and soft glass would give rise to the same amount of light scattering. When a pair of quartz bulbs (Fig. 1) was used, the distillate showed the same amount of light-scattering as in the glass bulbs. The apparatus afterwards constructed to measure quantitatively the light-scattering showed this to be true within the limits of experimental error. The error was, however, larger here than for other measurements, since the quartz had air inclusions which reflected a good deal of light and made it difficult to get consistent results.

¹ In these experiments as in all those described in my former paper, the intensity of the light-scattering by different liquids was judged by passing the same beam of light alternately through the two bulbs held side by side. The quantitative method used in Section 3 of this paper had at that time not been developed.

After the measurements had been taken the 40 cc of water in the quartz bulb B was distilled back again to A so that any dissolved quartz might collect where the last drop evaporated. It proved impossible to find any residue even with a microscope, although one drop of distilled water evaporated on glass leaves a residue visible to the naked eye. The only evidence that there was any residue in the quartz bulb was the fact that, when condensation first occurred on the dry bulb, a ring formation of droplets was seen at the place where the last drop of water had evaporated. The solubility of quartz must then be negligible compared to the solubility of glass, and it seems, therefore, certain that the scattered light can not be attributed to any suspended siliceous material from the glass.

B. Envelopment.—Using Spring's method, gelatinous precipitates of aluminium, cadmium, and zinc hydroxides were precipitated from dilute solutions of their sulphates by addition of slight excess of potash. Solutions were prepared of

Aluminium sulphate,	7 g. per litre
Zinc sulphate,	20 g. per litre
Cadmium sulphate,	5 g. per litre

These were precipitated in each case in corked Erlenmeyer flasks by slight excess of potash, shaken and left to settle for a week. The resulting liquids in a condensed beam of light were found to be free from motes, but in every case the blue opalescence remained equal to that of pure water.

C. Cataphoresis of Water.—The apparatus consisted of a glass bulb with two side tubes containing platinum electrodes separated from the bulb by parchment partitions. The whole was filled with distilled water and a potential of 110 volts was maintained over the terminals for several weeks.

Experiments showed that in five days the motes were all removed and that the opalescence was then the same as that of the best water obtained by distillation in vacuum. Two months additional cataphoresis made no further reduction

in the opalescence. That the dust had not settled from long standing alone was shown by comparing with a similar bulb of water left standing for the same time undisturbed.

It seemed, therefore, certain that the light-scattering observed in water and other distilled liquids was not caused by foreign particles, but was a property of the liquids themselves, and quantitative measurements of its intensity, were therefore, undertaken.

3. Measurement of the Scattered Light

A. *Preparation of the Liquids.*—All the liquids investigated, ranging in boiling points from -10° (sulphur dioxide) to 250° (monochloronaphthalene) were freed from motes by the method of distillation in vacuum described above. No special precautions were taken to free the liquids of dissolved volatile impurities, as is proved later (see page 487) that in general liquids with high light-scattering power, with the possible exception of carbon bisulphide, have high formula weights and proportionately high boiling points. Consequently these could not be present in large amounts in the distillate unless the latter were itself a high-boiling liquid, and in this case its light-scattering power would be but slightly affected by the presence of impurities.

Kahlbaum's chemicals were used in all cases except methyl alcohol and benzene, which were Baker's C. P., chloronaphthalene which was Eastman's, and sulphur dioxide which was the commercial article from a steel cylinder.

Before sealing off the distillation apparatus about one-third of the liquid was allowed to boil away, and after the distillation about one-third of the liquid remained in the large bulb; so that the distillation was a fractional one in which the middle fraction alone was used.

B. *Apparatus for Measuring the Intensity of the Light-scattering.*—A parallel beam of light from a carbon arc (about 60 V, 12 A) passed through a narrow horizontal slit S and was focused by a lens L in the middle of the bulb B of the

liquid to be studied; then, after traversing a cell C of ammoniacal copper sulphate solution to make its color comparable with that of the scattered light, it fell on a piece of white paper at the back of the box. A quinine sulphate filter Q was used to prevent any possible fluorescence, although it made no noticeable difference. A small section (about 1 cm) of the beam of light passing through the liquid was observed

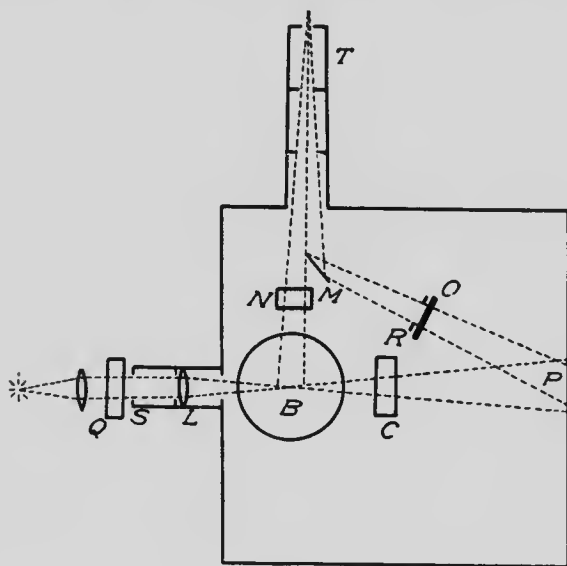


Fig. 2

through T, and appeared as an oblong of light about 1.5 mm wide. It was found that the lens L made this oblong brighter and its edges more definite. The image of the slit R, illuminated by light from the paper P, was reflected in the mirror M and was seen in focus beside the oblong of light. The whole box and its contents except the bulb of liquid were painted dead black, and the tube T was stopped down with several black diaphragms to prevent reflection from its sides.

By means of an optical wedge O of transmission 1 to 1/10 calibrated with an accuracy of about one percent, and supplemented by a number of uniformly fogged photographic plates of a transmission measured photometrically to the same accuracy, the light from the back of the box was matched with the scattered light from the bulb. In this way a comparison of the intensity of the light scattered by the various liquids was obtained. For measuring the extent of the polarization a Nicol was placed at N and readings of the intensity were taken with the Nicol in two positions at right angles to one another. A very little light was reflected from the glass bulbs containing the liquids, but not enough to seriously interfere with the measurements.

C. The Relative Intensity of the Scattered Light.—The following results are calculated from the averages of a great many readings, generally 18 or 24, made in sets of 6, each set made on a different occasion and with new adjustments. Very few results calculated from individual measurements differed by more than 10 percent from the average result thus obtained. A typical set of scale readings, in which a decrease of 1 in the scale reading corresponds to an increase in the light-scattering power by a factor 1.176, is as follows:

Heptane—13.3, 12.5, 13.8, 13.6, 13.4, 13.3. Average, 13.3.

The relative intensities of the scattered light (toluene = 1) calculated from three such sets of readings as the above are as follows:

Heptane—0.390, 0.384, 0.348.¹ Average, 0.374.

Table of the relative intensities of the light scattered by different liquids:

¹ The agreement between the individual measurements would warrant the expectation of a much better agreement between the results calculated from the averages of the sets of 6 measurements. The variation in these results was found to be largely attributable to the variation in the illuminated area at the back of the box, caused by the slight difference in size and shape of the bulbs. It would probably be better to use parallel light and vessels with plane sides.

Liquid	Formula weight	Relative intensity of scattered light (toluene = 1)	R liquid ¹ R toluene
<i>Inorganic Compounds:</i>			
Water	18	0.060	0.074
Sulphur dioxide	64	0.100	0.276
<i>Aliphatic Compounds:</i>			
Methyl alcohol	32	0.160	0.162
Ethyl alcohol	46	0.180	0.279
Ether	74	0.236	0.467
Ethyl acetate	88	0.210	0.488
Isobutyl chloride	93	0.375	
Heptane (mixture of heptanes)	100	0.374	
Isobutyl butyrate	144	0.320	
<i>Aromatic Compounds:</i>			
Benzene	78	0.91	0.864
Toluene	92	1.00	1.00
Chlorobenzene	112	1.52	1.09
Ethyl benzoate	150	1.55	1.39
Chloronaphthalene (alpha)	102	4.30	2.19

It is evident from the above results that there is a relation between the formula weight and class of substance on the one hand, and the intensity of the scattered light on the other hand:

(1) Aliphatic compounds scatter relatively little light; Benzene and its derivatives scatter decidedly more light; Chloronaphthalene, a compound with two benzene rings in its formula, scatters light to a still greater extent.

(2) The intensity of the scattered light with certain exceptions increases with increasing formula weight for the same class of compounds.

A great many physical properties, of course, bear some relation to the formula weight and chemical formula of the compound. For example the refractive index is found to be considerably greater for benzene and its derivatives than for aliphatic compounds, and greater still for naphthalene and its derivatives.

¹ See page 488 for meaning of R liquid.

Lord Rayleigh, before any experiments on light-scattering by dust-free gases and liquids had been made, formulated a relation between the index of refraction of the medium and the light-scattering caused by particles in the medium. According to his equation¹ the intensity of the scattered light for different media and for any given wave length of light is proportional to $\frac{(\mu - 1)^2}{n}$, where μ is the index of refraction and n the number of scattering particles per unit volume. This relation was deduced to apply to the light-scattering by any suspension of spherical particles of dimensions small compared to the wave-length of light. Its application to dust-free media would rest on the assumptions that the media are "discontinuous" and that there are present in the media secondary and spherical sources of light.

R. J. Strutt² has shown that, within the limits of his experimental error, the relation given above, where

$$n = \frac{\text{Density}}{\text{Formula weight}}$$

held true for the gases investigated.

In the case of liquids, by giving n^3 the same meaning, the relative values of (R liquid =) $\frac{(\mu - 1)^2}{n}$ have been listed in the above table for the various liquids, the value R toluene being taken as unity. It appears from the table that, although the relation does not hold so well for liquids as for gases, there is, nevertheless, some agreement between the observed and calculated values.

D. *The Polarization of the Scattered Light.*—Readings of the intensity were taken with the Nicol in two positions at right angles to one another. It was found that in every case the vibrations of the scattered light were transverse to the direction of the exciting beam. The results are given in

¹ Phil. Mag., 47, 375 (1899); Collected Works, Vol. 4, 397, Equation 14.

² Proc. Roy. Soc., 95A, 155 (1919).

³ In assigning a value to n the ordinary formula weight derived from the formula has been used. There is no generally accepted way of defining the formula weight of a pure liquid.

Column I of the following table. Column II gives the values obtained by R. J. Strutt for the corresponding gases:

Liquid	I Intensity in plane of pol'n $\times 100$ Intensity in plane at right angles	II Results for the gaseous phase (R. J. Strutt)
<i>Inorganic Compounds:</i>		
Water	6.7	
Sulphur dioxide	22.0	
<i>Aliphatic Compounds:</i>		
Methyl alcohol	7.0	
Ethyl alcohol	7.1	
Ether	9.3	1.7
Ethyl acetate	22.8	
Isobutyl chloride	18.0	
Heptane	13.4	
Isobutyl butyrate	17.3	
<i>Aromatic Compounds:</i>		
Benzene	54	6.0
Toluene	54	
Chlorobenzene	52	
Ethyl benzoate	55	
Chloronaphthalene (alpha)	78	
Carbon bisulphide ¹	70	12.0
Colloidal silica solution	0.25	

It appears from the above results:

(1) That for those substances which scatter very little light, the polarization is fairly complete; and that the polarization decreases as the light-scattering power increases.

¹ Experiments on carbon bisulphide and solutions of carbon bisulphide and methyl alcohol of various strengths gave results which seemed unreliable on account of the evident discoloration of the carbon bisulphide in the light. They seemed to indicate, however, that carbon bisulphide, a liquid with an extremely high refractive index, scatters a great deal of light which is but slightly polarized. On the other hand, alcoholic solutions of carbon bisulphide scattered about the amount of light one would expect from their dilution, but the scattered light was, contrary to expectation, very largely polarized and the amount of polarization increased with the dilution.

(2) That the scattered light is much less completely polarized by the liquid than by the corresponding gas. (It is noteworthy, however, that where data for both liquid and gaseous form are at hand—namely, for benzene, ether and carbon bisulphide—the order of these three substances when arranged according to their polarization is the same for both phases.)

(3) That the light scattered by any of the liquids studied is very much less completely polarized than that scattered by a colloidal silica solution.

E. *May the Phenomenon be Called Fluorescence?*—The light scattered by all liquids is sky-blue in color when the light of the carbon arc is the exciting source. This means that the shorter wave lengths of light are scattered to a much greater extent than the long (according to Lord Rayleigh's relation¹ the light scattering varies inversely as the fourth power of the wave length).

The scattered light was too faint to observe visually with a spectroscope, but by the help of some Wratten monochromatic light filters, which excluded all the exciting light except a narrow band of the spectrum, the following results were obtained for all the liquids tried:

(1) Exciting light of any color in the visible spectrum, from faintest violet to the beginning of the yellow, was scattered; with exciting light of wave length longer than yellow no scattering could be observed.

(2) The scattered light always had the same color as the exciting light whatever the color of the latter.

(3) A quinine sulphate cell in the path of the exciting light did not noticeably change the intensity nor the polarization of the scattered light.

Previous work by R. J. Strutt showed that for gases and for the one liquid investigated—ether containing dissolved water—the scattered light has the same spectrum as the exciting light.

¹ Phil. Mag., 47, 375 (1899); Collected Works, Vol. 4, 397.

From the information at hand it would appear, therefore, that taking the generally accepted criterion of fluorescence as being alteration in wave length, the light emitted by the liquids studied—which has been sometimes looked upon as fluorescent—is really scattered light. The author contemplates the spectroscopic examination of the emitted light to obtain further data for the various liquids.¹

Many esters and other high-boiling compounds showed before purification a distinct blue-green fluorescence. This was removed by vacuum distillation in the case of ethyl benzoate, the only one studied, and must, therefore, have been due to some impurity.

F. *The Effect of Change of Temperature.*—Measurements of the light scattered by water and toluene were made at 20° and again at 60°, but the variation in the intensity of the scattered light, if any, was within the experimental error of the readings.²

Summary.

(1) A method of preparing dust-free liquids by simple distillation in a vacuum is described.

(2) Liquids were found to scatter light; the short wave lengths being scattered to a much greater extent than the long.

(3) A relation, formulated by Lord Rayleigh and verified in the case of gases by R. J. Strutt, connecting the index

¹ Since this paper was sent in a photograph of the spectrum of the light scattered by chloronaphthalene has been taken and shows only the line spectrum of the glass mercury lamp used as the source of light. There is no sign of continuous spectrum. Since moreover chloronaphthalene is the liquid which one would most suspect from its formula of showing fluorescence there can be no doubt that the phenomenon observed is true scattering.

² Since the intensity of the light-scattering was found to vary approximately as $\frac{(\mu - 1)^2}{n}$, any considerable change in light-scattering with temperature would necessitate a change in n and, therefore, in the formula weight. Ramsay and Shields' surface tension measurements on a great many liquids (Zeit. phys. Chem., 12, 433 (1893)) led them to postulate for many liquids a formula weight which was independent of temperature, and for a few liquids—one of which was water—a formula weight which varied greatly with temperature.

of refraction of the medium and its light-scattering power, showed some agreement in the case of the liquids investigated.

(4) The scattered light is largely plane-polarized in the case of liquids which scatter very little light; and the polarization in different liquids decreases as the relative intensity of the scattered light increases. The polarization is much less complete for liquids than for gases.

(5) Evidence is given that the phenomena observed are due to scattering and not to fluorescence.

This research was begun at the suggestion of Professor F. B. Kenrick, and has been carried out under his direction.

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April, 1920*

