

# Experimental proof of the spin of the photon\*

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## 1. Introduction

The hypothesis that the light-quantum possesses an intrinsic spin in addition to energy and momentum has the merit of enabling the corpuscular concept of radiation to become a complete and intelligible picture. The ratio of the energy to the linear momentum of the quantum determines the velocity of light while the ratio of energy to spin determines its frequency, and the sense of the spin determines the polarisation characters. The energy, momentum and spin of the photon have their counterparts in the Maxwellian field theory and indeed may be regarded as a translation into the language of the quantum theory of ideas derived from the classical electrodynamics. The transference of the classical concepts of radiation energy and radiation momentum into the quantum theory was effected by Einstein, and placed on a firm experimental basis by the work of Millikan and of Compton. The spin of the photon, on the other hand, remains at the present time a somewhat nebulous mathematical abstraction without any convincing experimental support. It is the purpose of the present paper to show how experimental studies on the scattering of light by gases enable us to give precision to the idea of photon spin and firmly to establish its existence as a fact of observation.

## 2. The theory of photon spin

In his well-known derivation of the Planck radiation formula from quantum statistics, Prof. S N Bose<sup>1</sup> obtained an expression for the number of cells in phase-space occupied by the radiation, and found himself obliged to multiply it by a

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\* A preliminary note (communicated by cable on 28th September 1931), published in *Nature* of the 3rd October, 1931, on page 576 contained the essential results of this paper.

A further note (communicated to *Nature* by cable on 12th October 1931) contains an account of the results obtained with liquids.

<sup>1</sup>S N Bose, *Z. f. Phys.* (1924) 26, 178.

numerical factor 2 in order to derive from it the correct number of possible arrangements of the quantum in unit volume. The paper as published did not contain a detailed discussion of the necessity for the introduction of this factor, but we understand from a personal communication by Prof. Bose that he envisaged the possibility of the quantum possessing besides energy  $h\nu$  and linear momentum  $h\nu/c$  also an intrinsic spin or angular momentum  $\pm h/2\pi$  round an axis parallel to the direction of its motion. The weight factor 2 thus arises from the possibility of the spin of the quantum being either right-handed or left-handed, corresponding to the two alternative signs of the angular momentum. There is a fundamental difference between this idea, and the well-known result of classical electrodynamics to which attention was drawn by Poynting<sup>2</sup> and more fully developed by Abraham<sup>3</sup> that a beam of light may in certain circumstances possess angular momentum. The detailed calculation for a spherical wave<sup>4</sup> (assumed to have a total energy  $h\nu$ ), on the basis of the classical field theory shows its angular momentum to be  $\pm h/2\pi$  only in the case of circular polarisation, right-handed and left-handed respectively, while for a plane-polarised spherical wave, the angular momentum vanishes, and for elliptically polarised light, it has intermediate values. Thus, according to the classical field theory, the angular momentum associated with a quantum of energy is not uniquely defined, while according to the view we are concerned within the present paper, the photon has always an angular momentum having a definite numerical value of a Bohr unit with one or other of the two possible alternative signs. Following Dirac,<sup>5</sup> we may regard plane polarised light, *not* as a quantum without spin, *but* as a quantum which has an *even* chance of having plus or minus a Bohr unit as its angular momentum. Elliptically polarised light would be similarly regarded as characterised by *unequal* chances of possessing spin with the two alternative signs.

### 3. Selection rules in light-scattering

It will be recalled<sup>6</sup> that the principle of the conservation of angular momentum was the postulate from which the selection rules for the emission or absorption of radiation from atoms were first derived. The fact that the selection rules were also later established by an application of the correspondence principle of Bohr and follow as mathematical deductions from the wave-mechanics of the atom has tended to diminish the significance of their original derivation in relation to

<sup>2</sup>J H Poynting, *Proc. R. Soc.* (1909) **82**, 560.

<sup>3</sup>M Abraham, *Physikal. Z.* (1914) **15**, 914.

<sup>4</sup>A Sommerfeld, *Atomic Structure and Spectral Lines*, English Edition, Appendix 9 (1923).

<sup>5</sup>P A M Dirac, *Quantum Mechanics*, Cambridge University Press (1930) p. 131.

<sup>6</sup>A Sommerfeld, *Atombau und Spektrallinien*, 4th German Edn. (1924) p. 317.

radiation theory. More recently, however, Oppenheimer<sup>7</sup> has stressed the close connection which exists between the question of photon spin, the selection rules in spectroscopy and the conservation of angular momentum in radiation processes. If we assume that the spin of the photon is a whole Bohr unit (plus or minus), it follows at once in agreement with observation that the *total* angular momentum of the atom may not remain zero during the process of radiation but must increase or decrease by one unit. The simplicity of this derivation is very appealing, and we may therefore extend its principle to the problem of light-scattering. It is an empirically established result that in the scattering of light by various simple diatomic molecules as H<sub>2</sub>, O<sub>2</sub>, N<sub>2</sub>, etc. the rotational quantum number *K* of the molecule obeys the rule  $\Delta K = 0, \pm 2$ . The zero value for the change in quantum number gives the QQ branch or the undisplaced Rayleigh line in the spectrum of scattering by the gas, and the value 2 with the alternative signs gives the RR and PP branches of the rotational scattering, which appear as a close group of lines respectively on the low and high frequency sides of the undisplaced line. The selection rule stated was derived by Rasetti, Dieke and others from the conception of the third intermediate level in light-scattering. As pointed out in recent notes in *Nature*,<sup>8</sup> they also follow directly from the principle of conservation of angular momentum. We assume that the spin of the photon has the value  $\pm h/2\pi$  and for simplicity confine our attention to the case in which the quantum is scattered without any change in its direction of motion and the molecule has its axis of rotation fixed in the same direction. If the photon has a spin equal to  $+h/2\pi$ , it might either retain the same value or change to  $-h/2\pi$ , as the result of the scattering process. If it has initially the value  $-h/2\pi$ , it might either retain this value or change to  $+h/2\pi$ . The change in spin of the photon is thus 0 or  $\pm 2$  Bohr units. The corresponding change in the rotational quantum number of the molecule would be equal but opposite in sign, and would therefore be 0 or  $\pm 2$ . The PP and RR branches of the scattering in the forward direction would on this view consist of photons which have experienced a reversal of spin. In other words, if the incident light be circularly polarised, the Rayleigh or undisplaced scattering would be circularly polarised in the same sense, and the rotational scattering would be *reversely* circularly polarised. The fact that this inference from theory is supported by the observations of Bar and of Bhagavantam may be regarded as furnishing strong support for the view that the photon has an intrinsic spin. It must be confessed, however, that as all the results stated can also be derived from the classical radiation theory, either with or without the help of the quantum mechanics, the argument can scarcely be regarded as a conclusive proof of the existence of photon spin.

<sup>7</sup>J R Oppenheimer, *Phys. Rev.* (1931) 38, 726.

<sup>8</sup>C V Raman and S Bhagavantam, *Nature* (1931) 128, 114; C V Raman *Nature* (1931) 128, 545.

#### 4. Scattering of spinning photons

A careful consideration shows that a difference between the results of the classical field theory and the idea of spinning photons is to be expected in respect of the spectral distribution of the light scattered by the molecules of a gas. If we regard radiation as a particle possessing an intrinsic spin and impinging on a rotating molecule, the *sense* of the spin which the molecule possesses in relation to that of the photon should obviously enter into the problem in a fundamental way. If the molecule is spinning in a right-handed sense, it is obvious that if it gains energy of spin from the encounter, it must also gain angular momentum from it in a right-handed sense; this obviously it cannot if the photon before the encounter has a left-handed spin, since the latter can take up but not give up right-handed spin. Similarly a molecule with left-handed spin cannot gain both energy and spin from a right-handed photon. We may in fact make out the following scheme of forbidden and allowed transitions:

Molecule spin	Photon spin	PP $K \rightarrow K - 2$	QQ $K \rightarrow K$	RR $K \rightarrow K + 2$
Right	Left	Allowed	Allowed	Forbidden
Right	Right	Forbidden	Allowed	Allowed
Left	Right	Allowed	Allowed	Forbidden
Left	Left	Forbidden	Allowed	Allowed

It will be seen that while the transitions which give the QQ branch are always allowed, those which could contribute to the intensity of the PP and RR branches are forbidden in half the total number of possible cases and allowed only in the other half. On the other hand, in the treatment of the problem of light-scattering on the Maxwellian field theory, the function of the radiation is assumed to be merely that of furnishing a periodic electric force at the position of the molecule which would polarise it and cause to emit secondary radiation as a rotating Hertzian dipole. The question of the sense in which the molecule rotates does not arise, and has no influence on the final results. In other words, it is implicitly assumed that in the collision between a photon and a molecule, either a gain or a loss of energy by the latter is always allowed. As we have seen, this is not the case if the existence of an intrinsic photon spin and the conservation of angular momentum are taken into account.

To understand what happens in the case of a forbidden transition, we may adopt a hint given by the classical treatment. From the latter, it is clear that the whole of the scattering would appear in the QQ branch if the molecule were optically isotropic, and that the existence of the PP and RR branches in scattering is due entirely to the fact that the molecule is optically anisotropic. It is possible to

separate the scattering by an anisotropic molecule into two parts, namely the isotropic part which is completely polarised in a transverse direction, and which appears solely in the QQ branch, and the anisotropic part which consists mainly of unpolarised light, and is divided between the PP, QQ and RR branches. If the anisotropic molecules instead of being allowed to rotate were imagined to be held fixed in randomly distributed orientations, the classical theory tells us that the PP and RR branches would disappear, their contribution being merely transferred to the QQ branch, so that the intensity and state of polarisation of the aggregate scattering would remain unaffected. We may therefore make the assumption that when a  $K \rightarrow K - 2$  transition, or a  $K \rightarrow K + 2$  transition is forbidden by reason of photon spin, we have simply a  $K \rightarrow K$  transition: in other words the photon is scattered without change of energy. Adopting this view we conclude that the effect of photon spin on the scattering of light by a rotating molecule is to diminish the intensity of the PP and RR branches to one half the value given by the classical theory, the intensity of the QQ branch being correspondingly increased, so that the intensity and state of polarisation of the total scattering remain unaffected.

### 5. Depolarisation of scattering

The foregoing conclusions have a further important consequence. It is clear from what has been said above that the QQ branch of the scattering consists mainly of polarised light, while the PP and RR branches consist mainly of unpolarised light. Hence the defect in polarisation of the QQ branch, when spectroscopically separated from the PP and RR branches would be *smaller* than that of the total unresolved scattering consisting of all the three branches. The extent of the difference would be very considerable if we are to believe the classical theory according to which most of the anisotropic scattering appears in the PP and RR branches. According to the theory of photon spin, on the other hand the intensity of these branches is diminished to one half the value given by the classical theory with a corresponding increase in the intensity of the QQ branch. We should therefore expect the defect of polarisation of the QQ branch to be much nearer that of the total scattering than is indicated by the classical theory. This fact furnishes us with a convenient *experimentum crucis* for the existence of photon spin.

The magnitude of the effects to be expected is readily calculated. As has been explained by Bhagavantam in a recent paper,<sup>9</sup> in the case of simple linear molecules such as  $N_2$ ,  $O_2$ ,  $CO_2$ ,  $N_2O$ ,  $CS_2$ , etc. the anisotropic scattering according to the classical theory, finds a place in the QQ branch only to the extent  $\frac{1}{4}$ th of the whole. That this result is characteristic of the Maxwellian concepts of

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<sup>9</sup>S Bhagavantam, *Indian J. Phys.* (1931) 6, 331.

radiation is shown by the fact that Manneback's theory<sup>10</sup> of scattering by diatomic molecules based on the wave-mechanics and Bhagavantam's treatment based on the classical or Maxwell-Boltzmann statistics of molecular rotation give precisely the same results. For symmetric top molecules such as  $\text{NH}_3$ ,  $\text{C}_6\text{H}_6$ , etc. the ratio should be  $\frac{1}{3}$  instead of  $\frac{1}{4}$ . Following the theory of photon spin, if we transfer one-half of the aggregate intensities of the PP and RR branches to the QQ branch, the anisotropic scattering would be found in the latter to the extent of  $\frac{2}{3}$  of the whole in the case of linear molecules and  $\frac{2}{3}$  of the whole for symmetric top molecules.

The defect of polarisation in the transversely scattered light is known for the case of various molecules from the earlier investigations in which spectroscopic resolution was not used, and represents the depolarisation of the PP, QQ and RR branches taken together. The scattering with large frequency shifts due to the vibration of molecules is of comparatively negligible intensity. Taking the best accepted values and using the relations stated above, we obtain the following table showing the depolarisation of the QQ branch to be expected according to the classical field theory and the theory of photon spin.

Table I

Molecule	Total depolarisation	Depolarisation of QQ branch (classical)	Depolarisation of QQ branch (spin theory)
$\text{N}_2$	3.6%	3.9%	2.3%
$\text{O}_2$	6.4	1.7	4.1
$\text{CO}_2$	9.8	2.7	6.4
$\text{N}_2\text{O}$	12.5	3.5	8.3
$\text{CS}_2$	11.5	3.2	7.6
$\text{C}_6\text{H}_6$	4.2	1.4	2.8

## 6. Experimental method and results

The earliest indication of the failure of the classical field theory was furnished by measurements of the intensity of the lines in the spectrum of the light scattered by hydrogen gas.<sup>11</sup> An attempt to reconcile the data obtained with Manneback's theory and the known depolarisation of the total scattering proved unsuccessful; the observed intensity of the rotational lines was found to be just half that to be expected on the basis of Manneback's theory. It was this discrepancy in fact which suggested the ideas outlined in the preceding paragraphs. Except in the

<sup>10</sup>C Manneback, *Z. f. Phys.* (1930) 62, 224.

<sup>11</sup>S Bhagavantam, *loc. cit.*

case of hydrogen, it is not easy to determine the intensity of the lines in the PP and RR branches as they are very close to each other and to the QQ branch and are not completely separable from the photographic halation accompanying the latter. The alternative method of the study of the depolarisation of scattering with and without spectroscopic resolution is simpler and also of a more general application, as it can be applied to the case of molecules having high moments of inertia which give unresolved wings instead of discrete lines in the PP and RR branches.

The general plan of the experiments is very simple. The light of the mercury lamp transversely scattered by the gas passes through a square-ended nicol and is focussed on the slit of a high-illumination spectrograph. Two photographs are taken of the spectrum of the scattered light, respectively with the nicol oriented in two perpendicular positions, and the state of polarisation is determined by varying the relative exposures until the horizontal and vertical components are obtained with equal intensity. The depolarisation of the QQ branch is determined by narrowing the slit of the spectrograph so that with the exposures given, only the undisplaced Rayleigh lines are recorded. A second set of measurements made with the slit open as wide as possible gives the depolarisation of the unresolved scattering the comparison of which with the accepted value acts as a check on the perfection of the experimental arrangements.

Numerous experimental difficulties present themselves in the exact determination of such small depolarisations as those observed with gases. The experience gathered from the long series of investigations carried out at Calcutta by I Ramakrishna Rao<sup>12</sup> of the unresolved scattering and the more recent work of Bhagavantam<sup>13</sup> using spectroscopic methods has proved very helpful in dealing with these difficulties. Amongst the major pitfalls which present themselves may be mentioned the following:

(a) False light derived from stray illumination of the walls of the container, and specially of the background. This consists mainly of unpolarised light, and if not completely eliminated would result in enormous errors.

(b) Errors due to lack of transversality of the illuminating rays to the direction of observation. When using extended sources of light such as a quartz mercury arc, the errors are very serious.

(c) Polarisation of the rays occurring within the spectrograph due to oblique refraction at the surfaces of the prisms.

After much preliminary work devoted to elimination of these sources of error, the following arrangements were adopted for the actual measurements. CO<sub>2</sub> and N<sub>2</sub>O gases were chosen as the working substances, as they give an intense scattering and large depolarisation: the errors due to false light were therefore far

<sup>12</sup>I R Rao, *Indian J. Phys.* (1927) 2, 61.

<sup>13</sup>S Bhagavantam, *Indian J. Phys.* (1931) 6, 319.

smaller than for other gases such as  $N_2$  and  $O_2$ . The gases were taken from commercial cylinders under pressure, and were found to be free from dust. They were transferred under pressure into a containing tube of fused quartz blackened outside, armoured in steel and fitted with dark background, diaphragms, etc. as already described and illustrated in a previous paper by Bhagavantam in this Journal. So long as the working pressure was less than 20 atmospheres, the changes in depolarisation due to the deviation from Boyle's law was negligible. The source of illumination was a quartz mercury lamp, the entire luminous area of which was covered up by a metal plate containing an aperture 2 cm in diameter through which the light emerged, and an image of which was focussed by a large glass condenser at the centre of the quartz tube containing the gas. The distance from the centre of the condenser to the illuminated aperture on the one side, and the centre of the gas tube on the other was about 20 cm. The restriction of the illuminating area of the lamp had the effect of greatly diminishing the importance of false light from the back of the observation tube, as was shown by actual trial exposures with a gas evacuated. It also reduced the error due to lack of transversality of the illuminating rays. As was shown by I Ramakrishna Rao,<sup>14</sup> the use of a condenser for focussing a source of light of small angular aperture introduces no error under this head, so long as observations are made of the scattered light at the exact point of focus. The polarisation of light occurring within the spectrograph was carefully determined, and allowed for in making the photographic time exposures.

The experimental results are given in table II.

Table II. Depolarisation in %

Gas	Observed (wide slit)	Observed (narrow slit)	Observed diminution	Calculated diminution (Classical)	Calculated diminution (spin)
$CO_2$	11.0	7.0	4.0	7.1	3.4
$N_2O$	12.0	7.0	5.0	9.0	4.2

The measurements are clearly decisive in favour of the spin theory.

## 7. Observations with liquids

It is well known that the light scattered by liquids at ordinary temperatures exhibits a notable defect in polarisation which is larger and much easier to

<sup>14</sup>I R Rao *loc. cit.*



observe and measure than the corresponding effect in gases. Further when, spectroscopically analysed, the scattered light exhibits very marked wings on either side of the undisplaced line due to the exciting radiation. These wings represent the PP and RR branches of the scattering by molecules of the liquid, and their existence together with their observed depolarisation is *prima facie* evidence that the molecules rotate while scattering radiation. The close packing of the molecules in a liquid (as compared with a gas) has a great influence on the intensity of the coherent part of the scattering which is determined by the compressibility of the fluid and not by the molecular density. This constitutes the 'isotropic' scattering which is fully polarised and appears solely in the QQ branch. The intensity of the anisotropic scattering which is incoherent and therefore in a truer sense molecular in origin, is determined by the density of the liquid, but cannot be assumed to be strictly proportional to it owing to the influence of a complicating factor, namely the local polarisation field acting on the molecules within the liquid. We are however concerned here not with the total intensity of the anisotropic scattering (which may be taken to be that actually observed under the given conditions), but with the manner in which it is shared between the PP, QQ and RR branches. There is reason to suppose that the close packing in a liquid might influence the statistical distribution of the rotational energy of the molecules, making it depart very considerably from that obtaining in the case of a gas. If such were the case, the distribution of intensities *within* the PP and RR branches would be affected. So long, however, as we are free to assume that the material particles which scatter the light (whatever they may be) are free to take up spin from the photons and rotate with appreciable frequencies, there must be a separation of the anisotropic scattering into the PP, QQ and RR branches in a manner consistent with the principles of conservation of energy and angular momentum.

The considerations set out above make it clear that except in the case of highly viscous liquids where large molecular groups may be assumed to exist, we shall probably be justified in regarding the general principles discussed in the earlier part of the paper with reference to gases to be applicable with full force to the case of liquids as well. Especially in the case of non-associated liquids such as  $\text{CS}_2$ ,  $\text{C}_6\text{H}_6$ , etc. we should be justified in determining the part of the total anisotropic scattering which appears in the QQ branch, as a fraction of the whole, assuming the individual chemical molecule to be the scattering particle. The total anisotropic scattering may be determined from the observed depolarisation of the aggregate scattering. In this way the depolarisation of the QQ branch to be expected may be readily calculated. The results along with the observed values<sup>15</sup> for two simple liquids  $\text{CS}_2$  and  $\text{C}_6\text{H}_6$  are given in Table III.

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<sup>15</sup>Taken from the recent unpublished work of S Venkateswaran on a large number of liquids.

Table III. Depolarisation in %

Unresolved	Observed (wide slit)	Observed (narrow slit)	QQ branch (spin theory)	QQ branch (classical)
CS <sub>2</sub> 64.0	64.0	56.0	55.5	36.4
C <sub>6</sub> H <sub>6</sub> 42.0	41.5	34.0	33.5	20.8

The measurements are clearly decisive in favour of the spin theory.