A STUDY OF THE TIME CORRELATION OF PHOTONS FROM SEPARATE GROUPS OF SPECTRAL LINES OF NEON II

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It is shown that on excitation of Ne by electron impact, time correlation of photons from separate groups of closely spaced and cascade-independent NeII lines occurs. The correlation is about 3 times higher than could occur on these spectral lines via cascade transitions from other lines of the spectrum.

 \mathbf{L}_{N} studies of light emission there is considerable interest in questions of the coherence of the emitted radiation^[1-6]</sup>. The concepts of "spatial" or "temporal" coherence are usually defined in terms of correlations of the phase of the radiation at various points in space or time. Many experiments on extremely weak light beams have shown that the coherence persists even when light quanta pass through an optical system independently of each other, one at a time. These experiments show directly that the individual atomic radiators are themselves coherent sources of light (e.g., emission from extremely rarefied gases). In this case the coherence may easily be explained by the "wave model" of the emission of energy by an atom. However as is well known this model (if one makes no additional hypotheses) is in direct contradiction^[7] with the "localized" quantum absorption of the emitted energy that is known to occur experimentally (e.g., in the photoelectric effect).

In this regard as well as in considering the problem of coherence in lasers, this is currently a subject of theoretical and experimental investigations [8-28]. In the experimental papers [12,19] it was asked whether or not photons belonging to two coherent light beams are independent. These experiments showed no correlation; however, similar experiments, with more intense beams and under somewhat different conditions, demonstrated the existence of the correlation effect [20,21,28]. The present author described^[22] an investigation of an analogous question; that is, of the correlation due to the coherence of the radiation. The experimental conditions of that paper were different from those described in the previous references^[12,19-21,28] and were in fact the conditions producing the maximum possible sensitivity to correlations which might be due to cascade transitions. As far as is known to the author, the study of correlations due

to the coherence of the radiation under these conditions had not been carried out previously, although it was pointed out in the paper of Brannon and Ferguson^[19] that the sensitivity of their system was sufficiently great for observation of the "natural" coincidences due to cascade transitions. Nevertheless their experiment was a repitition of the Brown and Twiss experiment, the conditions of which were far from optimum for detection of photon correlations due to cascade transitions.

In the present experiment, as in [22], particular attention was given to the solid geometry of the light collection and to the efficiency of photon detection; by improving these factors we were successful in considerably increasing the sensitivity of the system to cascade transitions. These experiments resulted in the observation of temporal correlations of photons, or more exactly of photoelectrons, from individual atoms of helium, neon, argon and xenon (the results obtained from helium, argon and xenon atoms will be published later). Study of the correlations in the radiation from $neon^{[22]}$ showed that a definite contribution to the correlation was made by photons emitted in cascade transitions from one and the same atom. It may be assumed that another contribution to the correlation might be made by stimulated transitions, whose probability will be strongly dependent both on the radiation density and on the density of the particular excited state of the atoms.

It was shown earlier [22] that the temporal correlation of photons from neon is due to a group of spectral lines in the ultraviolet (3200-3700 Å) emitted by singly ionized NeII atoms. It was also shown that the magnitude of this correlation corresponded to the "simultaneous" radiation of approximately two photons for each collision of an electron with a gas atom, and it was further shown



FIG. 1. Schematic arrangement of the apparatus.

that this "degree" of correlation could not be due to cascade transitions. To make a more detailed investigation of this question, the present work was undertaken to study the correlations in individual regions of the emission spectrum; this makes it possible to a certain extent to separate the correlations due to cascade transitions from the correlations which are not due to cascade processes. Consideration of the emission spectrum of Ne II which gives rise to the correlation shows that many of the spectral lines occur in separate groups (cf. Fig. 7 in [22]) and that these groups occupy only a small spectral interval (about 5-10 Å). These groups may be isolated easily with the help of narrow-band interference filters. The most intense of these groups of spectral lines is the group of lines 6, 7 and 8 (cf. Table I in [22]); these lines comprise about $\frac{1}{4}$ of the total intensity of the emission spectrum. It is natural to expect that, since the overall emission spectrum exhibits two-photon correlations, this group of spectral lines should make a very important contribution to the correlation. Moreover the spectral lines 6, 7 and 8 are not (as may be seen from the energy level diagram in Fig. 11 of [22]) due to cascade transitions and therefore the existence of correlations caused by cascading can arise only via interactions with the remaining part of the spectrum.

The present paper treats the relation of these three spectral lines among themselves and with the remainder of the spectrum insofar as it relates to correlations. The experimental measurements were carried out with the apparatus whose essential parts are shown in Fig. 1. An electron beam E (W = 150 eV) formed by a special focusing system into a filament of about 7μ diameter was passed through the near focal line of an aluminum elliptical mirror M (the mirror aperture was about 2π , the diameter at the small (near) focus was 56 mm, the separation between foci was about 686 mm, magnification was about $55 \times$, and the resolution was about 50 lines/mm in the central region of the near focus¹⁾). This beam excited fluorescence from the neon atoms ($P = 10^{-3}-10^{-4}$ mm Hg). The light emitted was collected by the mirror into a beam which was passed through a semi-transparent aluminum mirror S and split into two beams, one of which was incident on the photomultiplier I and the other incident on photomultiplier II. The photomultipliers detected the individual photoelectrons; the resulting pulses were then fed to counting and coincidence systems^[23,24].

A permanently mounted UFS-1 filter transmitted only the ultraviolet spectral lines from 3000-4000 Å and rejected the intense group of yellow and red lines at 5800-6500 Å. This produced a spectral distribution equivalent to that used to measure the correlations in [22], where a UFS-1 filter was also used. The diaphragm D was located at the far focus of the elliptical mirror and served to define a region of the excited gas (at the near focus) of dimensions $100 \times 100 \times 100 \mu$. For measurements on individual groups of spectral lines, narrow-band interference filters could be placed in positions 1 and 2 of Fig. 1; these filters consisted of 25 to 29 layers of PbF_2 and cryolite deposited on quartz, had a transmission of from 30 to 60%, and a passband at half height of from 18 to 30 Å^{2} The light-collection geometry in the present case was some 5 to 7 times less efficient than that used in the author's previous experiments^[22,23]</sup>. This somewhat decreased the sensitivity of the system to correlations and decreased the accuracy of the measurements. To insure greater reliability of measurement there was in addition to the primary coincidence circuitry^[23] an additional parallel system for detecting coincidences in which one of the channels contained a pulse delay of about 200 nsec.

¹⁾The mirror was prepared by the author.

²⁾The filters were prepared by the author according to technical directions given by A. Yu. Klement'ev of the laboratory of F. A. Korolev (MGU).

Both of these coincidence circuits allowed simultaneous determination of number of coincidences due to the time correlations of photons and determination of the number of purely random coincidences. As in ^[22] the temporal correlation of the photons was defined in terms of a quantity $\Delta \tau$ which is determined experimentally as follows:

$$\Delta \tau = \tau - \tau_0 = (N_{\text{tot}} - N_{\text{rand}}) / 2N_1 N_2, \qquad (1)$$

where N_{tot} is the total number of coincidences per second obtained with the light beam and N_{rand} is the number of random coincidences per second obtained with the auxiliary coincidence circuitry, and where N_1 and N_2 are the single channel rate in pulses per sec.

The number of random coincidences N_{rand} was determined for arbitrary rates N_1 and N_2 by measurements either with "monitors" (by illuminating the photomultipliers with independent light sources) or else by using the supplementary coincidence circuitry. The quantity $\Delta \tau$ may be easily calculated from the intensity of the photon source. Under the conditions of the present experiment (reflection of photons from the elliptical mirror and separation of the light beam by the semi-transparent mirror) the detection of photons took place as if there were no angular correlation between the beams.

Since study of the emission due to cascade transitions in Ne II^[22] indicated that cascades leading to the simultaneous emission of three or more photons were practically nonexistent, and since there is no other information on this matter, one may limit oneself in calculating the correlation to collisions which give rise to the simultaneous emission of no more than two photons; this is in agreement with the results of the previous paper^[22].

€.10⁻⁸, sec



Taking this into account and neglecting the insignificant variation of photomultiplier sensitivity over the spectral range of investigation, we find for $\Delta \tau$:

$$\Delta \tau = \frac{N_2}{N_{\rm ph} N_{\rm ph}} \frac{1}{(1 - e^{-\tau_0/T})},\tag{2}$$

where $N_{ph} = N_1 + 2N_2$ is the total intensity of the photon source (number of photons per sec), N_1 is the number of electron-atom collisions per second leading to the emission of a single photon, N_2 is the number of electron-atom collisions per second giving rise to the simultaneous emission of two photons, τ_0 is the coincidence resolving time of the apparatus, and T is the total effective lifetime for the levels taking part in the optical transitions of the spectral lines under investigation.

Since (as will be published shortly) for the present group of lines $T < \tau_0 = 3 \times 10^{-8}$ sec, the exponential term may be neglected in Eq. (2). It is clear that the ratio N_2/N_{ph} is independent of the emission intensity N_{ph} . When $N_2/N_{ph} = \frac{1}{2}$, Eq. (2) describes the case in which each electronatom collision leads to the simultaneous emission of two photons. It is further clear from Eq. (2) that the magnitude of the photon correlation time $\Delta \tau (N_{ph})$ determined in this way for the above experimental conditions is independent of the light collection geometry as well as independent of the photomultiplier gain and is determined only by the intensity of the photon source.

By changing the electron beam current (at constant gas pressure and for given positions of the interference filters) one can vary the intensity of the photon source and thus make measurements of $\tau(N_{\rm ph})$ and $\tau_0(N_{\rm ph})$. The results of these measurements are shown in Fig. 2. The lower curve

FIG. 2. Measurements of the correlations $\tau(N_{ph})$ and $\tau_0(N_{ph})$ in neon: • – for the monitors; 0 – for points obtained without any interference filters; Δ – points obtained with an interference filter in channel 2 isolating spectral lines 6, 7 and 8. No interference filter in channel 1; + – points obtained with interference filters in both channels isolating spectral lines 6, 7, and 8.

gives the values of τ_0 obtained both from the monitor measurements (the photomultipliers being illuminated by independent light sources) and from measurements made on the beam using the auxiliary coincidence scheme. The figure shows that the resolving time of the coincidence circuitry is (within wide limits) essentially independent of the single channel rates and is $\tau_0 = 3 \times 10^{-8}$ sec. The dotted line is calculated from Eq. (2) for N₂/N_{ph} = $\frac{1}{2}$ and corresponds to the values $\tau(N_{ph}) = \Delta \tau(N_{ph}) + \tau_0$.

The experimental points corresponding to values of $\tau(N_{\text{ph}})$ were calculated from Eq. (1) for the following combinations of filters:

1. No interference filters in the light beams (open circles in Fig. 2).

2. An interference filter isolating lines 6, 7, and 8 was placed in one of the paths (position 2 in Fig. 1).

3. Interference filters isolating lines 6, 7, and 8 were placed in both light beams (position 1 in Fig. 1; the crosses in Fig. 2).

4. A neutral density filter (from $3200-3700 \text{ \AA}$) with a transmission of 12-15% was placed in one of the channels (position 2 in Fig. 1); there was no filter in the other light beam (open circles in Fig. 2).

5. Neutral density filters of the same kind were placed in both beams (position 1 in Fig. 1) (open circles in Fig. 2).

The interference filter used to isolate spectral lines 6, 7, and 8 had a peak transmission of about 50% and a half-width of about 20 Å. The integrated transmission of this filter for a given spectral line was about 12-15%. It is clear from Fig. 2 that the experimental points corresponding to filter combinations 1-5 above follow the dashed curve within the limits of error of the measurement.

Combinations 4 and 5, which were used as controls, indicate that attenuating the light with the neutral density filters is equivalent to decreasing the photon detection sensitivity; their insertion did not [in agreement with Eq. (2)] cause a change in the measured value of $\Delta \tau(N_{ph})$. Combinations 2 and 3 indicate the existence of correlations not only between spectral lines 6, 7, and 8 and the remaining portion of the spectrum but also of correlations arising from lines 6, 7, and 8, which are not related to each other by cascade transitions. If one assumes the correlations detected in experiment 3 above are due to emissions involving the simultaneous radiation of two photons, then the data may be used to calculate that fraction of the photons (compared to the total number) which participate in such emissions [that is, one may

find the value of $2N_2/N_{ph}$ in Eq. (2)]. To do this one needs the experimentally determined values of the correlations as a function of the intensity of photon source N^{*}_{bh}. The interference filters used in these experiments, which had a maximum transmission of about 50% and a spectral pass band of 12-15%, are equivalent in effect to two successive filters, one of which is neutral and has 50% transmission and the other of which is complementary to the original filter (hence its transmission at the peak must be about 100% and its transmission band must be $(12-15\%) \times 2 = 24-30\%$. It is clear that the measured value of the correlation $\Delta \tau (N_{\rm ph})$ is not affected by the action of the neutral filter (experiment 5) and that the complementary filter isolates 24-30% of the photons from the total number, i.e., $N_{ph}^* = (0.24 - 0.30)N_{ph}$.

Putting this photon intensity in Eq. (2) and using the results of Fig. 2 we find $2N_2/N_{ph} = 0.24 - 0.30$ ~ 24-30%. On the other hand (in a fashion similar to the calculation made in [22]) experiment 3 may be used to calculate the maximum possible correlation which could be produced in this case by cascade transitions alone. To do this it is necessary to make use of the relative intensities of the spectral lines shown in the table (the first four columns are taken from Table I in [22]; these data were obtained by passing the radiation through the interference filter. It is clear from the values in the table that the overall radiation intensity going through the filter concentrated in lines 5, 6, 7, 8, 10, and 11 along with other spectral lines not related by cascades make up 95-96% of the total intensity (taken as 100%). It follows from the energy level diagrams (Fig. 11 in [22]) that these spectral lines are not related by cascades. Thus only the 4-5% remaining intensity may form cascades with these transitions and give rise to a total number of multiple photons 2N₂/N_{ph} $= 2(0.04 - 0.05) = 0.08 - 0.10 \sim 8 - 10\%$. Thus the excess of the correlation obtained in experiment over the correlation calculated for cascade transitions is a factor $3[(0.24-0.30)/(0.08-0.10) \sim 3]$. This indicates that the correlations are probably due to other processes. The relative number of photons corresponding to non-cascade type correlations may be easily seen to be

$$2N_2^+ / N_{\rm ph} = [(0.24 - 0.3) - (0.08 - 0.10)] = 0.16 - 0.20 \sim 16 - 20\%.$$

As mentioned previously correlations in isolated spectral lines were studied in [12,19-21,28]. Brown and Twiss [20] observed and measured correlations which were explained by Purcell [25] to be the result of the "anomalous" increase in the

	Wave length	Degree of excita- tion	Intensity before filter (arb. units)	Intensity, past the filter for groups of lines, arb. units							
Line				Lines 6, 7 and 8		Lines 10 and 11		Lines 12 and 13		Lines 44, 45, 46 and 47	
				×10-2	nor- mal- ized	×10-2	nor- mal- ized	×10-2	nor- mal- ized	×10-2	nor- mal- ized
1 2 2	3229.50 3230.16		3.12	3.12	0.2	1,25	0,156	0.94	0.165	61	8,85
5 4 5	3232.38 3297.74 3309.78		0.24	2.04	0.13	0.43	0.054	$1.44 \\ 0.67$	$0.254 \\ 0.118$	1 ,49	0.216
ĕ	3319.75	ΠÌ	7.0	205	25.0	2,01	0.57	7 0	4 30	31 6	4.6
7	3320.29	IJĴ	7,9	395	20.2	51,4	6.43	1.9	1.55	31.0	4.0
8 9	3323.75 3334.87	II II	$17.24 \\ 0.96$	862 25	$55 \\ 1.59$	$\begin{array}{c} 120\\ 16.8 \end{array}$	15 2,1	$17.24 \\ 1.44$	$3.04 \\ 0,254$	69 3.17	10.0 0,46
10 11	3344.43 3345.49	$\begin{bmatrix} \Pi \\ \Pi \end{bmatrix}$	13.9	188	12.0	530	66.3	3 2	5.63	42	6.1
12 13	3377,23 3378,28		11.72	29 .3	1.87	42.2	5.27	445	78.5	23.4	3,4
14 15	3390.56 3392.78		3.6	6,5	0,41	7.6	0.95	41.4	7,3	6.5	0.94
16 17	3416.87 3417.71		0.84	0.84	0.054	0.67	0.084	2.35	0.415	1.51	0.22
18 19	3418.007 3447,703	IJ	1.2	0,6	0,041	0.6	0.075	0,72	0.126	2.16	0.31
20 21	3453.10	$\left[\begin{array}{c} II\\ I \end{array} \right]$	1.56	0.8	0,051	0.78	0.097	0.78	0.137	2.8	0.4
22 23	3464.339		0,48	0.19	0.012	0.24	0.03	0,192	0.034	0.87	0,12
24	3472.571	1'	0.12	0.05	0.0032	0.048	0.06	0.048	0.0085	0.216	0.031
25 26	3480.75	$\prod_{\mathbf{H}}$	4,31	1.72	0.11	1.72	0.215	1.29	0,228	7,8	1.1
20 27	3501.215		0.12	0.04	0.0025	0.048	0.06	0.036	6 0063	0.216	0.031
28 29	3503,61	11)	1/ 12	4.94	0.27	5.64	0 705	4 23	0.75	25 4	37
30	3542.28	ี่ม่า	14,12	4.24	0,41	0.04	0.105	4,20	0,10	20,4	0.1
31 32	3542,90	II }	0.12	0.04	0,0025	0.048	0.06	0,036	0.0063	0,24	0.035
33	3568.53		4.8	1.9	0.12	1.96	0.245	0.96	0,169	11.5	1.67
35 35	3571.20 3574.64	II	3.35	1.34	0.085	1.34	0.168	0.67	0,118	8,7	1.26
36 37	$3593.526 \\ 3593.640$	$\begin{bmatrix} I\\I\\I \end{bmatrix}$	0.96	0.58	0.037	0,48	0.6	0.192	0,034	3,64	0.53
38 39	3594.18 3632.75		0.36	0.54	0 034	0.22	0.027	0.072	0.0127	3.06	0.44
40 41	3633,665 3643,89		4.2	2.04	0.001	0.94	0.405	0.26	0.062	12 6	4 92
42	3644.86	II∫	1.2	2.04	0.15	0.64	0,105	0.30	0.003	12.0	1.05
43 44	3664.112 3694.197	II II	1.08	0.96	0.061	0.53	0.066	0,192	0.034 0.151	8.2 54	7.8
45 46	3709,64	\prod_{Π}	3.6	12.6	0.8	7.2	0.9	3,6	0.63	240	34.8
4 7	3727.08	ii'	1,2	5.64	0.36	3.0	0.37	1.8	0.32	58	8.4
48	3766.29	II }	0.36	2.9	0.185	1.8	0.225	1.26	0.22	6.1	0.88
49 50	3769.449 3777.16		0,24	2.2	0.14	1.44	0.18	1.1	0,194	3,5	0.5
Tota1	Total intensity of the lines, %			15.7	100	8	100	5.69	100	6.93	100

photon fluctuations when a photomultiplier cathode is illuminated by coherent light. Although the conditions of the present experiment differed considerably from those in which this correlation effect was observed, it in nonetheless important to estimate its possible magnitude under the present experimental circumstances and to compare it with the results obtained.

To do this one makes use of the quantity ρ_{c} which is the designation used by Brown and Twiss for the relative number of photons giving rise to nonrandom "intrinsic" coincidences. As may be seen from their formulas this number of photons is proportional to the photomultiplier counting rate

 $N_0.$ According to these formulas, in the paper by Brannon and Ferguson [19] for $N_0=5\times 10^4~{\rm pulses}/$ sec, the photon fraction should have been $\rho_{\rm C} \leq 1.7$ $imes 10^{-8}$; in the paper of Adam, Janossy and Varga^[12] where N_0 was 100 pulses/sec, ρ_C should have been less than 10^{-9} ; in the paper of Rebka and Pound^[21], for $N_0 = 4 \times 10^4$ pulses/sec, ρ_C should have been $\sim 8 \times 10^{-5}$; in the paper of Brown and Twiss^[20] where $N_0 = 3 \times 10^9$ pulses/sec, ρ_C was 2.5×10^{-4} ; and in the paper of Janossy, Naray and Varga^[28] for $N_0 = 4 \times 10^4$ pulses/sec (if one assumes that the quantum efficiency of their photomultiplier was 10^{-2}) $\rho_{\rm C}$ was of the order of 2.5×10^{-4} .

Considering the agreement between the experi-

ments of Fig. 2 and Eq. (2) the relative number of correlated photons in the present work is determined as follows: $\rho_c = 2N_2^+/N_{bh}$ and hence depends neither on the source intensity nor on the photomultiplier counting rate N_0 . For the spectral lines 6, 7 and 8 it has the value $\rho_{\rm C} \sim 1.6 - 2 \times 10^{-1}$ which is three orders of magnitude larger than the largest of the previously mentioned values. If one uses the equations of Brown and Twiss to calculate the correlation applicable to the conditions of the present experiment then the fraction of the correlated photons has a value $5 \times 10^{-4} - 5 \times 10^{-3}$ %. In making this calculation we used the following values of the parameters which occur in Eq. (7) in the paper of Brown and Twiss^[20]: the photomultiplier counting rate $N_0 = 10^3$ pulses/sec; spectral linewidth $B = 10^8$ cps (which is approximately the natural linewidth); the parameter $\sigma = 1$; and the degree of partial coherence $\Phi(\theta D/\lambda_0) = 1$ is taken to be the maximum, for which the relative number of photons $\rho_{\rm C}$ has its greatest value. The photomultiplier efficiency was taken to be 10^{-2} . Thus an estimate of the magnitude of the correlation which could be due to the fluctuation effect under the present experimental conditions and a comparison of this with the results obtained shows that the correlations in spectral lines 6, 7, and 8 are due to some other mechanism.

Experiments similar to experiments 2 and 3 above were carried out with other groups of spectral lines (10 and 11; 12 and 13; 44, 45, 46, and 47; and others), which were isolated by interference filters (the relative intensities of the lines after passing through the filters are shown in the table). The results of these measurements are tentative and simply indicate that the correlation obtained (on the scale of Fig. 2) is equal to or does not exceed by 1.5-2 times the value corresponding to the dashed curve. The results of the present experiment provide satisfactory evidence that the primary part of the time correlation of the photons exhibited in isolated groups of spectral lines is not due to cascade or bunching processes. They also indicate that in order to construct hypotheses to explain their origin it will be necessary to carry out investigations with still greater spectral resolution in order to decide the existence or absence of correlations in a single spectral line.

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