INVESTIGATION OF THE TIME CORRELATION OF PHOTONS EMITTED BY EXCITED ARGON ATOMS

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When argon atoms are excited by electron impact, a time correlation between the photons of the spectral lines Ar I and Ar II appears and corresponds in magnitude to "simultaneous" emission of an average of ~2.75 ± 0.3 photons in each act of gas-atom excitation. A calculation in which the intensity of the spectral lines in cascade transitions is taken into account yields a maximum of ~1.4—1.5 photons per impact.

THE question dealt with in this paper, just as in the preceding ones [1,2], is essentially the possibility of occurrence, under certain conditions, of a time correlation between photons as a result of the coherent properties of the light emission [3-9]. The experiments were described in [1,2,10] and consisted essentially of the following. Pulses from two photomultipliers (FÉU-I and FÉU-II, Fig. 1), operating in a mode in which individual photoelectrons are recorded [11], were analyzed by a coincidence circuit.

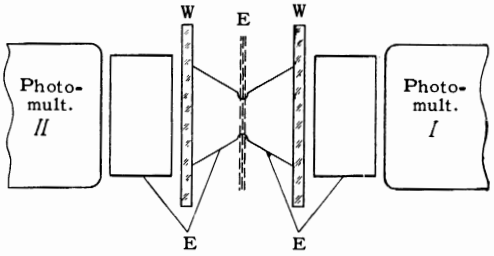

FIG. 1. Schematic diagram of experiment.

For calibration purposes, the photomultipliers were first illuminated by independent light sources and the number of random coincidences N_c (counts/sec) was determined for a given counting rate N (counts/sec). The resolution time of the system, $\tau_0 = N_c/2N^2$, was determined from these quantities by using the formula for random coincidences.

The photomultipliers were then illuminated from a common source (a "radiating" beam of electrons E in argon), the light from which was gathered unto photomultipliers by means of mirrors M, and the number of coincidences was again determined. In this case, at the same counting rates N as before, the number of coincidences turned out to be somewhat larger, $N_c + \delta N_c$. Consequently, the same formula yielded also the larger value $\tau = (N_c + \delta N_c)/2N^2$.

Under the experimental conditions, this excess was interpreted as a result of a certain time-correlated photon emission in the argon-atom excitation acts. It was assumed that such a photon correlation could be due to cascade transitions. However, an estimate of their magnitude, which is essentially the subject of the present paper, has shown that cascade transitions alone are not sufficient to explain the total correlation. This raised the following question: Can the resultant excess correlation be ascribed to a "coherent" origin, as was indeed assumed initially? It is clear that such a step would be justified, of course, only if there are no other possible ways for the excess correlation to arise.

The experiment was carried out under the following conditions:

1. The density of the excited argon corresponded to a gas pressure $\sim 10^{-3}$ mm Hg, at which the mean free path time of the atoms was considerably larger than the lifetime of the levels ($\sim 10^{-8}$ sec) for allowed transitions.

2. The excitation was produced with a monochromatic beam of electrons (150 eV), making it possible to vary the number of excited argon atoms monotonically. The intensity was set at such a value that the photons could be emitted by the atoms independently of one another, with an average interval considerably longer than their natural temporal coherence length. The part of the glowing string of excited argon atoms from which the light was gathered by the conical mirrors (Fig. 1) was 13 mm long and 1 mm in diameter. When the counting rate of each channel was

$\sim 10^4$ counts/sec, the beam current was $\sim 3 \times 10^{-10}$ A.

3. The light-gathering geometry (aperture) reached $\sim 4\pi$. This made it possible, in the presence of a finite photomultiplier dark current ($\sim$100 counts/sec) to raise to the utmost the ratio of the number of true coincidences due to the correlation to the number of random coincidences due to the finite time resolution of the system. The region of the spectral sensitivity of the system was determined essentially by the spectral sensitivity of the photomultipliers, which extended from $\sim$3000 to 6000 Å, inasmuch as the aluminum-coated mirrors M and the quartz windows W (Fig. 1) did not change the spectral composition of the radiation.

If the total correlation (connected with the excess over the random coincidences) is characterized by a quantity $\Delta\tau = \tau - \tau_0$ and is interpreted as the result of "simultaneous" emission of n independent photons in each argon-atom excitation act, then we can readily obtain for its magnitude

$$\Delta\tau = (n-1)\frac{\eta}{4N}\frac{\Omega}{4\pi}, \qquad (1)$$

where η is the quantum efficiency of the photomultipliers, N the counting rate in each channel (counts/sec), and $\Omega/4\pi$ is the light-gathering solid angle.

To identify the correlation observed in argon with the number of photons emitted in each act of argon-atom excitation, we compared the quantity $\Delta\tau(N)$ measured for argon with analogous quantity for neon, measured under the same geometrical conditions. The correlation for the neon was measured earlier [1] and corresponded to "simultaneous" emission of an average of about two photons in each gas-atom excitation act.

Figure 2 shows the measured values of $\tau(N)$ and $\tau_0(N)$ in argon and neon under identical geometrical conditions. Inasmuch as the correlation in neon, $\Delta\tau = 1.45 \times 10^{-7}$ sec at $N = 10^4$ counts/sec, corresponds to "simultaneous" emission of two photons from atom, i.e., n = 2, the value $\Delta\tau = (2.83 \pm 0.3) \times 10^{-7}$ at the same counting rate for argon, according to (1), corresponds to an average emission $n = 2.95 \pm 0.3$ photons. Allowing in such a calculation for a certain difference in the efficiencies of the photomultipliers ($\eta_{Ne}/\eta_{Ar} \approx 0.9$) with respect to the somewhat shifted spectral regions of the emission from argon and neon, we obtain for the average number of emitted photons in argon a value $n = (2.95 \pm 0.3) \times 0.9 \approx 2.75 \pm 0.3$ photons per act.

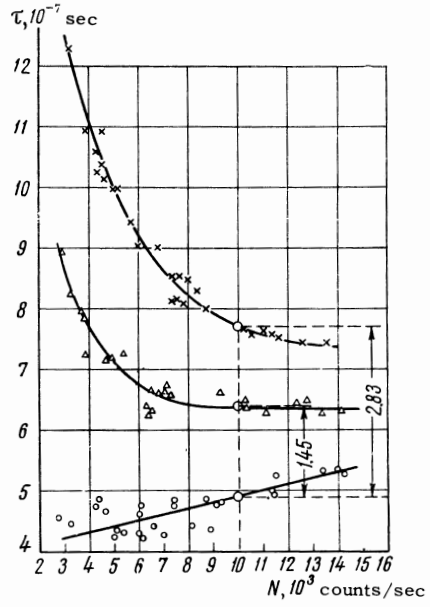

FIG. 2. Measurements of the correlation in argon and in neon, $\tau(N)$ and $\tau_0(N)$: ○—monitor measurements, △—measurements in neon, ×—measurements in argon.

The correlation observed in the argon is the result of emission of the appreciable number of spectral lines excited by electrons with energy $\sim$150 eV. Investigations of the spectral composition of emission (using ISP-28 and Q-12 instruments), under conditions close to those at which the correlation was measured ($P \sim 10^{-3}$ mm Hg, W = 140 eV), have shown that, in the spectral region of the multipliers (see Fig. 2 of [1]), the spectral lines predominantly excited are those of Ar II. Excitation of a large number of spectral lines could lead to the formation of cascade transitions, which could make a definite contribution to the measured correlation.

To estimate this possible contribution, we measured the wavelengths and of the line intensities, and identified the spectral lines, using known data [12-13], with the corresponding transitions. Altogether 137 lines were measured in the spectral region 3000—5000 Å. The majority of the most intense lines are located in the maximum of the photomultiplier spectral sensitivity. In identifying the spectral lines with the corresponding transitions, we used both the term tables [14] and the results of Minnhagen [12], who systematized the Ar II emission in detail. He also took into account, in classifying the terms, the possible existence of different couplings between the spin and orbital momenta of the electrons. The actual positions of many terms were refined and determined anew, and the observed transitions were accordingly identified.

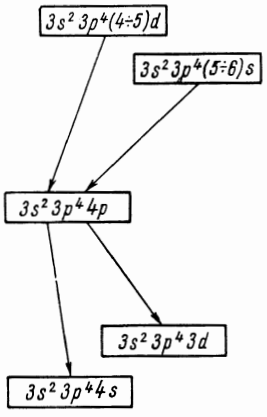

FIG. 3. Cascade transitions between Ar II lines. The rectangles show the electron configurations of levels.

Since the accuracy with which the line positions were determined was ~0.5 Å in the short-wave part of the spectrum and ~4 Å in the long-wave region, we took into account all the closely-located (within these limits) tabulated argon lines, regardless of the line actually excited. The intensity distribution among such spectral lines (for which we knew only the summary intensity) was effected in such a way as to obtain the maximum intensity of the "cascades" when the entire emission spectrum is taken into account.

There are no "cascades" among the Ar I lines in this region of the spectrum, and all the cascade transitions between the Ar II lines are shown in the scheme of Fig. 3. In this scheme, the electron configuration $3s^2 3p^4 4p$ ("intermediate") denotes a system of 19 levels, resulting from the L-S coupling between the momenta. The levels in the "upper" ($3s^2 3p^4(5—6)s$, $3s^2 3p^4(4—5)d$) and the "lower" ($3s^2 3p^4 4s$, $3s^2 3p^4 3d$) electronic configurations are also the result of the L-S coupling.

Thus, the "cascades" are the result of a transition of an electron from some level of the "upper" electron configuration to some level of the "intermediate" electron configuration, and then to a level of the "lower" electron configuration. Since in this case the transitions are allowed ($\Delta L = \pm 1$), the lifetimes of the "intermediate" level system is $\sim 10^{-8}$ sec. Inasmuch as the resolution time of the coincidence circuit was ~50 times larger, the emission of two photons in a cascade after $\sim 10^{-8}$ sec was registered as a "simultaneous" two-photon transition. A calculation of the fraction of such two-photon transitions was carried out in accordance with the following principle: The lowest of the obtained intensities of the upper and lower transitions (of any chosen cascade) was ascribed to two-photon transitions, and the difference between the intensities of these two transitions was ascribed to single transitions. This was followed by summation of the intensities of the two-photon and single transitions in all the obtained 18 cascades.

Naturally, to obtain a correlation corresponding to the emission of an average of more than two photons in one act, it is necessary that the emission spectrum contain a sufficient number of "triple" cascades, i.e., cascades with "simultaneous" emission of three photons. An analysis has shown that in this region of the spectrum there are only three "triple" cascades, with a total intensity of merely 1.57% of the total intensity of the emission spectrum.

Calculation of the intensity of the cascade transitions and of the intensity of the remaining spectral lines leads to the following distribution of the relative intensities: The intensity of the cascade transitions in Ar II amounts to 52.128%; the intensity of the transitions which do not enter in the "cascades" when the "cascades" are counted in accordance with the scheme of Fig. 3, is 25.208%; the intensity of single transitions of Ar II is 11.688%; the intensity of Ar I transitions is 9.56%; the intensity of unidentified Ar I and Ar II transitions is 1.416%.

In addition, an examination of the single transitions with total intensity 8.136%, the lower levels of which are the terms $(^1S_0)\,4s^2S$, $(^1D_2)\,3d^2D$, $(^1D_2)\,3d^2P$, and $(^1S_0)\,3d^2D$ (Figs. 1a and b of Minnhagen's paper [12]), reveals the possibility of formation of cascade transitions with transitions which do not enter in the "cascades" when counted in accordance with the scheme of Fig. 3. Such a possibility can be the result of the formation of "red" and "infrared" transitions of the type $4s \rightarrow 4p$ and $3d \rightarrow 4p$, which are not registered by the photomultipliers, from the indicated levels to the intermediate system of levels. If in such a case we include also this group of spectral lines (8.136%) in the "cascade" formation, and also the group of unidentified spectral lines (1.416%), then we obtain for the upper limit of the intensity of the cascade transitions

$$(52.128 + 2 \cdot 8.136 + 2 \cdot 1.416)\,\% \approx 72\%.$$

The remaining radiation intensity, i.e., ~28%, goes to the remaining non-cascade transitions. It is easy to see that such an intensity distribution among the cascade (72%) and single (28%) lines should lead to a correlation corresponding to an average emission of at most $1 + (0.72/2)/ \times$ $\times [0.72/2 + 0.28] = 1.57$ photons per gas-atom excitation act in contrast with the experimentally obtained value $\sim 2.75 \pm 0.3$.

Thus, an excessive ("supercascade") correlation, corresponding to emission of $1 + (2.75 \pm 0.3$

– 1.57) = 2.18 ± 0.3 photons in each gas-atom excitation act, arises in the radiation of argon atoms. If we characterize the magnitude of this correlation by a relative number of "double" (time-correlated) photons $\rho_c = N_2/N_{ph}$, where N_2 is the flux of "double" photons and N_{ph} is the total photon flux, then in this case this number will not depend on the counting rate of the multipliers and, as can be readily seen, has a value ~1.

However, in the well known paper of Brown and Twiss [6], the analogous number ρ_c is proportional to the counting rate of the multipliers and, for example, at a count $N = 10^4$ counts/sec (average counting rate in our case) it amounts to only $\sim 10^{-3}$—10^{-4}. This indicates that the "excessive" correlation occurring in this case cannot be due entirely to the correlation-occurrence mechanism described in the cited paper.

Inasmuch as the given excessive correlation is calculated as the difference between the "total" and "cascade" correlations, its existence, naturally, depends on the reliability with which their absolute magnitudes were determined. It is easy to see that the obtained absolute value of the "total" correlation is based on the reliability of the method with which the absolute quantum sensitivity of the recording system is measured [11]. Indeed, it is sufficient to increase by merely a factor of three times the quantum sensitivity of the system (during the course of its measurements), in order that the magnitude of the "total" correlation turn out to be equal to the "cascade" correlation, thereby reducing the "excess" correlation to nothing. In the present paper this quantity was measured with an accuracy ± 20%, which greatly exceeded the indicated possible error.

The probability that the "cascade" correlation has been underestimated as a result of incomplete identification of the spectral lines can be seen, from a description of the method employed, as quite low, since we have assumed throughout such a distribution of the spectral lines and such a transition configuration, which would yield the maximum possible contribution to the "cascade" correlation, thereby overestimating its magnitude.

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79