TIME CORRELATION OF PHOTONS EMITTED BY EXCITED XENON ATOMS

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Time correlation of emitted photons is observed when xenon atoms are excited by electron impact. The correlation is equivalent to the average "simultaneous" (within $\sim 5 \times 10^{-7}$ sec) emission of 2.2 ± 0.3 photons per excited xenon atom. A calculation which takes the intensity of induced cascade transitions into account yields a correlation that is equivalent to the "simultaneous" emission of at most 1.27 photons per excited atom.

At the present time there is growing interest in the details of light emission and absorption processes. This interest has been stimulated by the development of laser technology and by several obscure aspects of certain optical phenomena. Among the problems is the relationship between the continuity of the wave fields of quanta that results from their coherence, and the discrete absorption of the energy carried by the quanta.

Like earlier publications,[1-3] the present paper is concerned with the possibility of a time correlation between photons (or, more exactly, photoelectrons) that could be associated with the coherence of emitted quanta. Several authors (in [4-7] and elsewhere) have investigated correlations resulting from coherent emission. The conditions under which correlation was observed corresponded to a very small effective aperture for light "capture," thus creating special geometric conditions for coherent illumination of light receivers (photomultiplier cathodes). The correlation observed under these conditions has been studied and explained from both classical and quantum points of view (in [8-11] and elsewhere). However, it has not been possible to arrive at a unique conception of the light quantum emission process.[12]

The correlations are here investigated with the largest possible aperture, permitting the explicit registration of individual light quanta representing cascade transitions of excited atoms. Under such conditions, when a system can "see" quanta from cascade transitions, it was of interest to clarify the possibility of correlation between effects produced by the quanta themselves as coherent light sources (see [13-14], for example).

The problem has been stated in the foregoing manner because if the indicated correlation is found to be completely absent certain conclusions will then become possible in a more fundamental form and the feasibility of more basic experiments will be indicated. Indeed, the very possibility of registering a cascade correlation points to the existence of a finite "time" length of the emitted quanta. Hence their locations on a "time" axis can be predetermined, for example, by using optical shutters to cut off possible connections with other portions of the system. By directing a beam of such quanta toward the semitransparent mirror of a dual beam interferometer and registering the absence of correlation between the "channels" it can be demonstrated that only one of the channels contains that portion of a quantum which is responsible for its energy (or is able to eject photoelectrons). On the other hand, if interference between the "channels" is established under the given experimental conditions, it will be shown that "coherent" information is propagated along both "channels" simultaneously. This ability of a quantum to transport "material" energy along only one of the channels and to transmit "immaterial" coherent information along both channels simultaneously would demonstrate that it is possible to construct more than one model of quanta. We would thus again be enabled, for example, to approach the essential nature of interference phenomena. In the course of such research we might at an early stage observe effects that would elucidate in some way the described behavior of light quanta.

For the experiments in question it would clearly be sufficient to utilize radiation that originates in two spectral transitions forming a cascade and permitting the registration of a cascade correlation. The optical discrimination of either component of the cascade would make the experiments possible. In most instances, however, excited atoms emit a considerable number of spectral lines belonging to both single and cascade transitions, and formidable technical difficulties then are encountered in the large-aperture ($\sim 4\pi$) separation of spectral lines. When correlations were observed in this case it was only possible to learn what contribution to the correlation came from cascade transitions. The present work is devoted mainly to this problem; here Xe is investigated instead of Ar.[3]

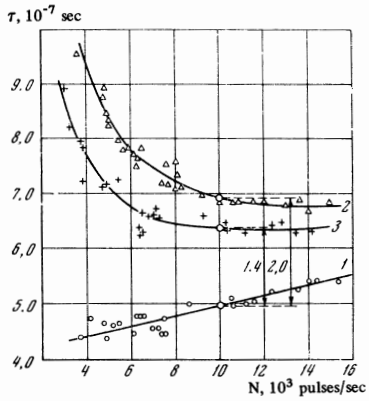
The time correlation was measured using the conventional method of registering coincidences between photoelectronic pulses from two photomultipliers (FÉU-13) that "see" the radiation from the excited gas. Excitation was produced by a continuous beam of ~ 150-eV electrons; the gas pressure was $\sim 3 \times 10^{-3}$ Torr. The length and diameter of the luminous Xe "column" were ~ 13 mm and ~ 1 mm, respectively. Light was focused on the photomultipliers with the aid of conical and cylindrical aluminized mirrors, with nearly 4π geometry.[15] The multipliers exhibited spectral sensitivity in the region 2800–5800 Å.[1] It was most convenient not to measure the correlation directly according to the number of coincidences arising when the counting rates in the channels were $N_1 = N_2 = N$, but from the "effective" resolution time τ of the system, defined by the

formula for random coincidences:

$$\tau = N_c / 2N^2.$$

It is easily seen that for the purpose of determining the resolving time of the system the photomultipliers must be illuminated by independent light sources (two lamps) that yield N_c random coincidences for a given channel counting rate N; the resolving time can then be calculated from the foregoing formula.

The resolving time calculated in this manner did not depend very strongly on the channel counting rates, as is shown by the straight line 1 in the figure. For $N = 10^4$ pulses/sec the resolving time was $\sim 4.9 \times 10^{-7}$ sec (represented by the large circle on line 1).



Measurements of $\Delta\tau$ (correlation) in Xe and Ne. 1—resolving time of the coincidence circuit; 2—correlation in Xe, and 3—correlation in Ne under the same geometrical experimental conditions.

The resolving time was governed by the duration of the electric pulses formed as photomultiplier outputs. The resolving time was not much diminished by the fact that the cascade correlation "times" (the lifetimes of levels for allowed transitions) had the same order of magnitude (10^{-7}–10^{-8} sec) as the "times" of the coherent "length" of quanta (the damping time of the classical atomic dipole is 10^{-7}–10^{-8} sec) during which photoelectrons could be ejected. When the relative magnitudes of these times are unknown, a reduction of the resolving time could affect correlations unfavorably with respect to the possibility of observing a "coherent" correlation.

After the resolution of the system was measured, the photomultipliers were both exposed to the light of the luminous Xe "column" and τ^* was recalculated from the number N_c^* of coincidences compared with a counting rate N. Since, in the present instance, for identical N a somewhat greater number N_c^* of coincidences was observed than in the case of illumination by independent light sources, τ^* was also found to be larger, as is shown by curve 2. The excess $\tau^* - \tau_0 = \Delta\tau = \Delta N_c / 2N^2$, which is proportional to the number of extra coincidences $\Delta N_c = N_c^* - N_c$, indicated the occurrence of some time-correlated emission of photons (more exactly, photoelectrons) from the excited gas column. The figure shows that for $N = 10^4$ pulses/sec we obtain $\tau^* = \sim 6.9 \times 10^{-7}$ sec and $\Delta\tau \approx 2 \times 10^{-7}$ sec.

Curve 3 of the figure represents τ^* for Ne as measured under the same experimental conditions.

Without performing any additional measurements of the aperture, we can use this same curve as a calibration curve for computing the number of correlated photons emitted by Xe from the number of correlated photons emitted by Ne under earlier simpler experimental conditions.[1] In this earlier work for $N = 10^4$ pulses/sec from Ne the correlation $\Delta\tau \approx 1.4 \times 10^{-7}$ sec was considered to indicate the average "simultaneous" emission of two photons from each emitting atom. A simple calculation shows that if Xe atoms emit n_{Xe} independent photons "simultaneously" the ratio of the correlations is

$$\frac{\Delta\tau_{Xe}}{\Delta\tau_{Ne}} = (n_{Xe} - 1)\frac{\eta_{Xe}}{\eta_{Ne}},$$

where η_{Xe} is the quantum efficiency of the photomultipliers for the xenon emission spectrum, and η_{Ne} is the same for neon. Inserting here the experimental values of $\Delta\tau_{Xe}$ and $\Delta\tau_{Ne}$ that are obtained from the figure at $N = 10^4$ pulses/sec and also the efficiency ratio $\eta_{Xe}/\eta_{Ne} \approx 1.2$ (since the Ne and Xe spectra are somewhat separated), we obtain the following average number of photons emitted "simultaneously" by xenon atoms:

$$n_{Xe} \approx 2.2 \pm 0.3 \text{ photons/event}.$$

More precisely, this equation gives the average number of photoelectrons impinging "simultaneously" on the photocathodes of the photomultipliers. The word "simultaneous" is here being used to designate time intervals between photons that do not exceed the resolving time of the coincidence circuit ($\sim 5 \times 10^{-7}$ sec).

The foregoing correlation was observed for Xe emission from both excited neutral atoms (XeI) and the excited ions XeII and XeIII, because of the sufficiently high energy ($\sim$150 eV) possessed by the exciting electrons. Most of the (registered) spectral intensity appeared in the interval from $\sim$2900 to $\sim$5500 Å. The wavelengths and relative intensities of the emitted spectral lines were measured with Q-12 and ISP-28 instruments. The total number (123) of lines represented allowed transitions of the neutral atom (XeI), singly ionized atom (XeII), and doubly ionized atom (XeIII). Wavelengths were determined by comparing the spectrograms with Fe emission spectrograms; intensities were determined from the photographic densities of the negatives (Panchrome 10N-1000 aerial film). The relative distribution of line intensities was determined from the photographic densities, which are known to depend on the spectral sensitivity of the photographic emulsion and on the contrast factor γ for given spectral regions.

The foregoing factors definitely affected the calculated contribution of cascade transitions to the total correlation. Characteristic (photographic density versus light intensity) curves of the negatives were plotted (using the 9-step attenuator of a DFS-8 instrument to obtain an intensity scale) at 3000, 4000, and 5000 Å. The somewhat variable spectral sensitivity of the emulsion for Xe lines was also determined. By examining carefully each group of observed cascade transitions with respect to an intensity effect of the involved spectral lines because of the aforementioned factors, it was

found that the contribution of the cascade correlation was sometimes enhanced slightly (~5–10%) and sometimes diminished. The combined effect of these factors (the contrast and spectral sensitivity) was compensated on the average, within the experimental error limits, because of the large number of participating spectral lines and their location in a relatively small interval of the spectrum (the most intense group of lines was in the range 4000–4600 Å). The conversion from photographic density to relative intensity was performed in accordance with the characteristic curve at $\lambda = 4000$ Å.

When the spectrograms were being recorded the Xe pressure and the energy of the exciting electrons were maintained close to the conditions for measuring the correlation. The image of the electronic "filament" was projected on the spectrograph slit (on the middle of the "swallowtail" diaphragm) in such a way that the direction of the electronic "filament" coincided with the optical axis of the lens and the electron current was directed toward the quartz lens (of ~18-mm diameter and ~3.6-cm focal length). The iron arc spectrum was registered along the edges of the Xe spectrogram, for the purpose of interpretation. Under these conditions for registering spectrograms the wavelengths and relative positions of the spectral lines were identical with the measurements obtained in the course of the correlation measurements. The transitions occurring in the excited atoms and ions were then determined from the wavelengths and from tables in [16-18]. Knowledge of these transitions and of their relative intensities made it possible to estimate the upper limit of the cascade-transition contribution to the observed correlation.

Since the level lifetimes for allowed transitions were of the order $\sim 10^{-8}$ sec while the resolving time of the coincidence scheme was considerably longer ($\sim 5 \times 10^{-7}$ sec), all the cascade transitions could be registered as "simultaneous" emissions of two photons.

The spectral lines of the neutral atom (XeI) represented transitions from the electronic configurations

$$5s^25p^5\begin{cases}6p', & 7p, & 9p, & 10p, & 11p\\ 8p, & 4f, & 6f, & 7f, & 8f\end{cases} \qquad (4)$$

to the electronic configuration $5s^2\,5p^5\,6s$, and included no observed cascade transitions in the registered portion of the spectrum. These transitions comprised 7.01% of the total radiation intensity.

Spectral lines of the singly ionized atom (XeII) comprised a considerable fraction of the intensity. These lines represented transitions of the ion from the electronic configurations

$$5s^25p^47s', \quad 5s^25p^47s, \quad 5s^25p^46d', \quad 5s^25p^46d \qquad (5)$$

through a system of intermediate levels (the configuration $5s^2\,5p^4\,6p$) to levels of the electronic configurations $5s5p^6$, $5s^25p^46s$, and $5s^25p^45d$. Since the lower branch of the "cascades" possessed considerably greater intensity than the upper branch, not all of this radiation could be attributed to "two-photon" emission. Only 17.94% of the intensity (8.97% in the upper branch and 8.97% in the lower branch) could be attributed to emission in the form of "coupled" photons, while 27.89% represented "single" photons. A similar analysis of transitions in the doubly ionized atoms (XeIII) yielded 3.38% of the total intensity as coming from "cascades" producing "coupled" photons, and 3.95% as comprising single "uncoupled" photons.

The registered spectrum also included 31.28% of entirely "single" XeII and XeIII transitions, as well as 8.55% of unidentified lines. Since the unidentified lines could in some (unspecified) way form "cascades" with the free single transitions in XeII and XeIII, these lines were classified accordingly in the computations.

The total intensity of observed and assumed "cascade" transitions assigned to XeII, XeIII, and unidentified lines therefore comprised 17.94% + 3.38% + 2 × 8.55% = 38.42%. This intensity of the cascades could yield average "simultaneous" emission not exceeding

$$n = 1 + \frac{38.42/2}{38.42/2 + 61.48} = 1 + \frac{19.21}{19.21 + 61.48} \approx 1.27 \qquad (6)$$

photons per excited Xe atom. The foregoing discussion shows that this last result is smaller by $2.2 \pm 0.3 - 1.27 \approx 0.9 \pm 0.3$ photons per excited atom than the experimental value for the total correlation. Therefore the extra "non-cascade" correlation leads to an average of $1 + 0.9 \pm 0.3 = 1.9 \pm 0.3$ photons per excited xenon atom.

As already mentioned, in the original statement of the present problem it was proposed to treat this extra "ultracascade" correlation as the result of coherent emission. This hypothesis obviously requires us to ascertain the reliability with which the extra correlation can be determined and also to demonstrate the absence of other possible causes thereof. Since this extra correlation represents the difference between the observed and cascade correlations, the reliability of these correlations naturally determines the reliability of the extra correlation. The total correlation was determined by the procedure developed in [15, 19], and the "cascade" correlation was determined by the foregoing method of computing cascade transition intensities. It appears that the actual experimental conditions should not allow the other possible causes (multiple excitation, stimulated emission, Brown and Twiss correlation) for the registration of extra correlation. Nevertheless, the present author has assumed a very cautious attitude with regard to the proposed interpretation.

On the other hand, an experimental solution of the correlation problem as here formulated could, in the author's opinion, yield very useful information about the character of light quantum emission and about the mechanism whereby the quanta are absorbed.

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