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LETTER TO THE EDITOR

The effect of atomic number fluctuations on photon antibunching in resonance fluorescence

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Abstract. It is suggested that the correct processing of data in a recent experiment of Kimble, Dagenais and Mandel implies that the short-time value of the correlation function of photons emitted by a single atom in resonance fluorescence is zero. This result is found by firstly correcting an apparent error in the treatment of the background radiation and secondly by taking account of Poisson fluctuations in the number of radiating atoms.

In a recent experiment the phenomenon of antibunching of photoelectric counts has been reported (Kimble *et al* 1977). The joint probability density $P_2(t, t+\tau)$ of photoelectric pulse pairs $n(\tau)$ was measured in the resonance fluorescence off a dilute atomic beam. Values of $n(\tau)$ were recorded for time delays τ in intervals of 2 ns using a start–stop arrangement with two separate detectors. The observed pulse pair data were corrected theoretically to take account of the considerable background of elastically scattered laser light, and after normalisation the photocount correlation function $1+\lambda(\tau)$ was determined. Two unusual features were then noted: (i) the value of $\lambda(0)$ was approximately $-0{\cdot}6$; and (ii) the curve rose with positive slope to a peak of $\lambda(25\text{ ns})\approx 1{\cdot}3$. Since $\lambda(\tau)\leqslant\lambda(0)$ and $\lambda(\infty)=0$ for any classical ergodic process, the reduced data, if correct, point to photon antibunching not describable in classical terms (Stoler 1974).

Without theoretical correction, however, the data of Kimble *et al* show a value of $\lambda(0)=0$ though still a positive slope. This may be seen by considering the random content of any channel, equal to $N_s\Delta\tau R_2$ (N_s = number of start pulses, $\Delta\tau$ = channel width, R_2 = stop rate). Evaluation of this gives a random content per channel of 299 to be compared to the extrapolation of $n(0)$ of about 295 from figure 2 of Kimble *et al*. To deduce a negative $\lambda(0)$ Kimble *et al* invoke substantial coherent beating between background and fluorescent light in the third term on the right-hand side of their equation (7). In our experience, however, full heterodyne efficiency is very difficult to achieve in practice even under the best conditions of controllable light beams and careful wavefront matching. In the experimental arrangement used by Kimble *et al* of large-angle collection via a microscope objective it would seem that wavefront matching should not be expected between a small radiating atom and a background spread over a 100 μm aperture. Thus the third term should have a heterodyne efficiency factor inserted, probably of the order of 10^{-2}, and thus becomes negligible. With this supposition $\lambda(\tau)$ has the same general form with a peak $\lambda(25\text{ ns})\approx 1{\cdot}4$, but most importantly, $\lambda(0)$ is now equal to 0, within the experimental error indicated, rather than −0·6. The negative value of $\lambda(0)$ claimed is thus, if the above argument is accepted, not correct.

Additional considerations, however, can be applied to rescue the situation and retain the hypothesis of $\lambda(0) = -1$ for a single atom. Kimble *et al* estimate that the mean number of contributing atoms in the atomic beam is around 1 within a factor of 2 or 3. They comment that the field produced by two radiating atoms located at random positions would give an expected value of $\lambda(0) = -0{\cdot}5$. Such an experimental result would obtain if there were only two and always two atoms in the beam. In fact the actual number of atoms in the probe volume will fluctuate and it seems reasonable to assume the distribution to be Poisson. The expected result for a fluctuating number may be deduced as follows (for somewhat similar discussion see Pusey 1977).

We assume, as in our earlier discussion of the background contribution, that the fields from different atoms add incoherently. It is then permissible to add photon numbers. We write

$$n_1(t) = \sum_{i=1}^{N(t)} a_{1i}(t) + b_1(t) \tag{1}$$

where $n_1(t)$ is the total photon count at detector 1 in interval $\Delta\tau$ at time t, $a_{1i}(t)$ is the photon count at detector 1 due to the ith atom, $b_1(t)$ is the background count (assumed incoherent) and $N(t)$ is the number of atoms in the probe volume at time t. The mean photon number is then given by

$$\langle\overline{n_1}\rangle = \bar{N}\langle a_1\rangle + \langle b_1\rangle \tag{2}$$

where the angular brackets denote averages over the photons and the bars indicate averages over the atomic number fluctuations. Similar equations can be written for the photon numbers at the second detector. Consider now the cross-correlation function $\langle\overline{n_1(0)n_2(\tau)}\rangle$ for delay times τ short compared to the transit time of the atoms through the probe volume so that N remains virtually constant during time τ:

$$\langle\overline{n_1 n_2}\rangle = \overline{\sum_{i=1}^{N}\sum_{j=1}^{N}\langle a_{1i}a_{2j}\rangle} + \bar{N}(\langle a_1\rangle\langle b_2\rangle + \langle a_2\rangle\langle b_1\rangle) + \langle b_1\rangle\langle b_2\rangle \tag{3}$$

where, for simplicity, the τ dependence is understood. Since it is assumed that a_{1i} and a_{2j} are uncorrelated for $i \neq j$, equation (3) becomes

$$\langle\overline{n_1 n_2}\rangle = \bar{N}\langle a_1 a_2\rangle + \overline{N(N-1)}\langle a_1\rangle\langle a_2\rangle + \bar{N}(\langle a_1\rangle\langle b_2\rangle + \langle a_2\rangle\langle b_1\rangle) + \langle b_1\rangle\langle b_2\rangle. \tag{4}$$

For a Poisson distribution, $\overline{N(N-1)} = \bar{N}^2$ so that

$$\langle\overline{n_1 n_2}\rangle = \bar{N}\langle a_1 a_2\rangle + (\bar{N}\langle a_1\rangle + \langle b_1\rangle)(\bar{N}\langle a_2\rangle + \langle b_2\rangle); \tag{5}$$

normalisation by $\langle\overline{n_1}\rangle\langle\overline{n_2}\rangle$ gives

$$\frac{\langle\overline{n_1 n_2}\rangle}{\langle\overline{n_1}\rangle\langle\overline{n_2}\rangle} = \frac{\langle a_1 a_2\rangle/\langle a_1\rangle\langle a_2\rangle}{\bar{N}[1 + (\langle b_1\rangle/\bar{N}\langle a_1\rangle)][1 + (\langle b_2\rangle/\bar{N}\langle a_2\rangle)]} + 1$$

or

$$\lambda(\tau) = \frac{1 + \lambda_{\mathrm{A}}(\tau)}{\bar{N}[1 + (\langle b_1\rangle/\bar{N}\langle a_1\rangle)][1 + (\langle b_2\rangle/\bar{N}\langle a_2\rangle)]}. \tag{6}$$

We see then that, if $\lambda_{\mathrm{A}}(0) = -1$ for a single radiating atom, the measured value $\lambda(0)$ for a Poisson ensemble of atoms should be zero, as found experimentally by Kimble *et al* if we disregard their heterodyne background correction. This result is independent of the strength $\bar{N}$ of the atomic beam and of the signal-to-background ratios $\bar{N}\langle a\rangle/\langle b\rangle$.

It is worth pointing out that, as noted by Kimble *et al* (1977, footnote 7), a negative $\lambda(\tau)$ with positive slope can be produced for $\tau \neq 0$ by a purely classical source, for example, one emitting 'antibunched' pulses of light, each one of which could give rise to a (Poisson) number of photodetections. At zero delay time the correlation function of this signal will have its largest value. However, for delay times long compared to the duration of the pulses but short compared to their separation a result $\lambda(\tau) = -1$ in the first observed channel can be achieved and in consequence an initial positive slope. While this is possible in principle, after the experiments of Clauser (1974) it seems a purely didactic point in the present context. However, the semi-classical hypothesis can only be categorically excluded by measurements at $\tau = 0$, in the present case for instance, by introducing a delay in the stop channel of the apparatus. Such a measurement does not appear to be included in the report of Kimble *et al.*

In summary, we have questioned the validity of the background correction of Kimble *et al* and have suggested that atomic number fluctuations must be considered for the correct interpretation of the published experimental result. We then find that the intercept of $\lambda(\tau)$ at $\tau = 0$ is zero for a Poisson atomic beam of any strength. Only when multiple occupancy of the probe volume is excluded can a result of $\lambda(0) = -1$ be attained experimentally, and the full antibunched character of the radiation be observed.

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