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Photon antibunching and sub-Poissonian statistics from quantum jumps in one and two atoms

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Antibunching and sub-Poissonian statistics of the 11-μm photon emission from the $5d^{10}6p\,^2P_{1/2}$ to $5d^96s^2\,^2D_{3/2}$ transition in one and two trapped Hg^+ ions have been inferred from the quantum jumps of the laser-induced fluorescence of the first resonance transition at 194 nm. Each downward step in fluorescence was assumed to mark the emission of an 11-μm photon. Signals from two ions were consistent with the assumption that quantum jumps in the two ions occurred independently. Mandel's Q parameter was determined to be approximately -0.25 for one and two ions.

Studies of collections of a few atoms or of individual atoms reveal properties which are not easily observed with samples of many atoms. One such property is antibunching of the emitted photons. A closely related, though distinct, phenomenon is sub-Poissonian statistics in the number of photons emitted in a given time interval. Photon antibunching has been observed from atoms in a dilute atomic beam by Kimble, Dagenais, and Mandel[1] and by Rateike, Leuchs, and Walther,[2] and from atomic ions in a Paul trap by Diedrich and Walther.[3] Sub-Poissonian photon statistics have been observed from single atoms by Short and Mandel[4] and by Diedrich and Walther.[3] Sub-Poissonian photon statistics have also been observed in Franck-Hertz light,[5] in parametric down conversion,[6] and from squeezed-light generators.[7]

Another effect that might be observed in collections of a few atoms is the onset of cooperative atom-field processes, which are known to occur with large numbers of atoms. Sauter, Blatt, Neuhauser, and Toschek have reported observing simultaneous quantum jumps of two and more Ba^+ ions at a rate higher than that expected from random coincidences.[8] They attributed these multiple quantum jumps to a collective interaction of the atoms and the radiation field. According to a calculation by Lewenstein and Javanainen,[9] collective effects occur only if the atoms are separated by much less than the transition wavelength.

We have previously reported an indirect observation of photon antibunching.[10] Transitions from the $^2S_{1/2}$ ground state to the $^2D_{5/2}$ metastable level of a single Hg^+ ion, induced by a 282-nm radiation source, were observed by noting the sudden cessation of the 194 nm laser-induced fluorescence of the $^2S_{1/2}$ to $^2P_{1/2}$ transition (see Fig. 1). When the fluorescence suddenly reappeared, we assumed that the ion had returned to the ground state by emitting a 282 nm photon. The correlations between the times of these assumed emissions showed antibunching.

In the work reported here, the statistical properties of 11 μm photons emitted from one and two Hg^+ ions were studied. These photon emissions were inferred from the state changes of the Hg^+ ion; these state changes were indicated by the abrupt changes in the 194 nm ion fluorescence. This is similar to the way in which the state of a Rydberg atom has been used to infer properties of the photon field in a cavity.[11] This indirect method of detecting the photon emissions was the same as in Ref. 10, but a different transition was involved. Some advantages of this detection method are high efficiency (about 95%) and low background. Collisions, which can either cause or mimic state changes of the ions,[10] were previously shown to be negligible.[12] Moreover, such independent random events would tend to decrease the antibunching and sub-Poissonian statistics observed here.

The four lowest energy levels of Hg^+ are shown in Fig. 1. The radiative decay rates and branching ratios have been measured previously.[12–15] In our experiment, the ion is subjected to coherent 194 nm radiation, which drives it from the ground $5d^{10}6s\,^2S_{1/2}$ level to the $5d^{10}6p\,^2P_{1/2}$ level. Most of the time, it decays directly back to the ground state, at a rate $\gamma_4 \approx 4\times10^8\ s^{-1}$. However, about once in 10^7 times, it decays instead to the metastable $5d^96s^2\,^2D_{3/2}$ level and emits an 11 μm photon. The rate of this transition is $\gamma_3 = 52\pm16\ s^{-1}$. The $^2D_{3/2}$ level decays at a rate $\gamma_1 = 109\pm5\ s^{-1}$. The probability for the $^2D_{3/2}$ level to decay directly to the ground state is

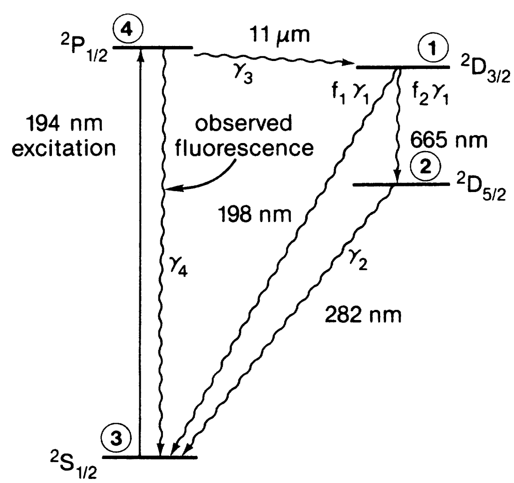

FIG. 1. Diagram of the lowest four energy levels of Hg^+. Only the 194-nm fluorescence was observed directly.

$f_1 = 0.491 \pm 0.015$. The probability for it to decay to the metastable $5d^9 6s^2\,{}^2D_{5/2}$ level is $f_2 = 1 - f_1$. The ${}^2D_{5/2}$ level decays to the ground state at a rate $\gamma_2 = 11.6 \pm 0.4$ s^{-1}.

The density-matrix equations for this system are

$$d\rho_{11}/dt = -\gamma_1\rho_{11} + \gamma_3\rho_{44}\,, \tag{1a}$$

$$d\rho_{22}/dt = f_2\gamma_1\rho_{11} - \gamma_2\rho_{22}\,, \tag{1b}$$

$$d\rho_{33}/dt = f_1\gamma_1\rho_{11} + \gamma_2\rho_{22} + \gamma_4\rho_{44} - i(\rho_{34} - \rho_{43})\mu E(t)/\hbar\,, \tag{1c}$$

$$d\rho_{44}/dt = -(\gamma_3 + \gamma_4)\rho_{44} + i(\rho_{34} - \rho_{43})\mu E(t)/\hbar\,, \tag{1d}$$

$$d\rho_{34}/dt = [i\omega_{43} - \tfrac{1}{2}(\gamma_3 + \gamma_4)]\rho_{34} + i(\rho_{44} - \rho_{33})\mu E(t)/\hbar\,. \tag{1e}$$

The state labels 1, 2, 3, and 4 refer to the ${}^2D_{3/2}$ level, the ${}^2D_{5/2}$ level, the ${}^2S_{1/2}$ level, and the ${}^2P_{1/2}$ level, respectively. Here, ω_{43} is the ${}^2S_{1/2}$ to ${}^2P_{1/2}$ transition frequency, μ is the electric dipole matrix element for that transition, and the applied electric field is $E(t) = E\cos(\omega t)$. The rates γ_1, γ_2, and γ_3 are all much less than γ_4. Therefore, the ${}^2S_{1/2}$ and ${}^2P_{1/2}$ populations quickly reach a steady state with respect to each other. For times much greater than the lifetime of the ${}^2P_{1/2}$ level, the system is effectively a three-level system, described by rate equations for the population variables $P_1 \equiv \rho_{11}$, $P_2 \equiv \rho_{22}$, and $P_0 \equiv \rho_{33} + \rho_{44}$. We derive these equations similarly to the way Kimble, Cook, and Wells[16] reduced the density-matrix equation for a three-level system to rate equations for a two-level system. The three-level equations are

$$dP_1/dt = -\gamma_1 P_1 + \gamma_0 P_0\,, \tag{2a}$$

$$dP_2/dt = f_2\gamma_1 P_1 - \gamma_2 P_2\,, \tag{2b}$$

$$dP_0/dt = f_1\gamma_1 P_1 + \gamma_2 P_2 - \gamma_0 P_0\,, \tag{2c}$$

where

$$\gamma_0 \equiv \gamma_3\Omega^2/(4\Delta^2 + \gamma_4^2 + 2\Omega^2)\,, \tag{3a}$$

$$\Omega \equiv \mu E/\hbar\,, \tag{3b}$$

$$\Delta \equiv \omega - \omega_{43}\,. \tag{3c}$$

Let $g^{(2)}(\tau)$ be the intensity correlation function of the 11-μm field generated by the radiative decay from the ${}^2P_{1/2}$ level to the ${}^2D_{3/2}$ level, normalized such that $g^{(2)}(\infty) = 1$. Photon antibunching occurs if $g^{(2)}(0) < 1$. The function $g^{(2)}(\tau)$ is proportional to $P_0(\tau)$, where $P_0(\tau)$ is obtained from the solution of Eqs. (2a)-(2c), subject to the initial conditions $P_0(0) = 0$, $P_1(0) = 1$, and $P_2(0) = 0$. This is because the average rate at which ${}^2P_{1/2}$ to ${}^2D_{3/2}$ transitions occur is the product of γ_3 and the probability that the ion is in the ${}^2P_{1/2}$ level (which for given Ω and Δ is proportional to P_0). The initial conditions signify that at time $\tau = 0$, the ion has just decayed to the ${}^2D_{3/2}$ level. The solution of the equations yields

$$g^{(2)}(\tau) = 1 - C_+\exp(-\gamma_+\tau) - C_-\exp(-\gamma_-\tau)\,, \tag{4}$$

where

$$\gamma_\pm \equiv \tfrac{1}{2}\{(\gamma_0 + \gamma_1 + \gamma_2) \pm [(\gamma_0 + \gamma_1 + \gamma_2)^2 - 4f_2\gamma_0\gamma_1 - 4\gamma_1\gamma_2 - 4\gamma_0\gamma_2]^{1/2}\}\,, \tag{5a}$$

$$C_\pm \equiv \pm\gamma_\mp(f_1\gamma_\pm - \gamma_2)/[\gamma_2(\gamma_+ - \gamma_-)]\,. \tag{5b}$$

All of the constants required to calculate $g^{(2)}(\tau)$ are fixed by the radiative decay rates of the Hg^+ ion, except for the transition rate γ_0 from the fluorescence-on state (cycling between the ${}^2S_{1/2}$ and ${}^2P_{1/2}$ levels) to the fluorescence-off state (either the ${}^2D_{3/2}$ or the ${}^2D_{5/2}$ level). This rate was determined by dividing the number of downward quantum jumps in fluorescence by the time that the ion was in the fluorescence-on state. Since $g^{(2)}(0) = 0$, this system exhibits photon antibunching.

The apparatus has been described previously.[10,14] The Hg^+ ions were confined in a radio-frequency (Paul) trap under high-vacuum conditions. Ions were created inside the trap volume by electron impact ionization of neutral Hg atoms. A 5-μW beam of narrow-band, cw 194-nm radiation was focused to a waist w_0 of approximately 5 μm at the position of the ions. The frequency of the 194 nm radiation was tuned below the atomic resonance, so the ions were laser cooled. The 194 nm photons emitted by the ions were collected by a lens system and focused onto a photomultiplier tube. The net detection efficiency was approximately 5×10^{-4}. Peak signals were approximately 50000 counts/s per ion. The data consisted of recordings of the numbers of photons detected in a series of as many as 100000 consecutive 1-ms time intervals. An example of the data is shown in Fig. 2 of Ref. 12. Each one-ion data set was analyzed by considering the ion to be in the fluorescence-on state if the number of detected photons was above a discriminator level and in the fluorescence-off state if it was not. This discriminator level was set to minimize the number of false transitions due to noise. Each transition from the fluorescence-on state to the fluorescence-off state was assumed to indicate the emis-

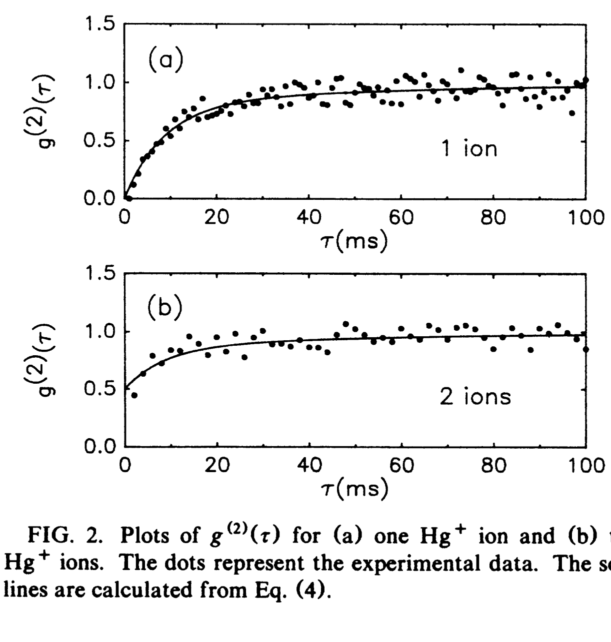

FIG. 2. Plots of $g^{(2)}(\tau)$ for (a) one Hg^+ ion and (b) two Hg^+ ions. The dots represent the experimental data. The solid lines are calculated from Eq. (4).

sion of an 11 μm photon. Analysis of simulated data, which included noise, indicated that about 5% of the transitions would have escaped detection, because they were preceded by or followed by a quantum jump in the other direction within a time interval of about 1 ms. The transitions from the fluorescence-off to the fluorescence-on state were not studied in detail, since it was not possible to tell from the 194 nm fluorescence whether a particular transition resulted from the emission of a 198 or 282 nm photon.

Data were obtained also with two Hg^+ ions, separated by approximately 3 μm. The 194 nm fluorescence switched among three distinct levels, depending on whether 0, 1, or 2 ions were in a metastable level. Discriminator levels were set to distinguish among the three different states. The time resolution was 2 ms. Each downward quantum jump was assumed to indicate the emission of an 11 μm photon from one of the ions. With an imaging photon-detector,[17] one could determine which ion had emitted the photon.

Figure 2 shows observed and calculated plots of $g^{(2)}(\tau)$ for (a) one ion and (b) two ions. The two-ion calculation was based on the assumption that photon emissions from one ion were not correlated with those from the other one. In that case, $g_2^{(2)}(\tau)=\frac{1}{2}[1+g_1^{(2)}(\tau)]$, where the subscript indicates the number of ions. The agreement of the experimental and calculated curves is an indication that the ions were independent, for time scales greater than about 2 ms. The values of γ_0 were about 15 s^{-1} for the one-ion data and about 12 s^{-1} for the two-ion data.

If the ions are independent and are subjected to the same 194 nm radiation field, then certain relationships hold among the probabilities p_0, p_1, and p_2 for zero, one, or two ions to be in the fluorescence-on state. Let p_{on} and $p_{\mathrm{off}}=1-p_{\mathrm{on}}$ be the probabilities for a single ion to be in the fluorescence-on or the fluorescence-off state. Then, $p_0=(p_{\mathrm{off}})^2$ $p_1=2p_{\mathrm{on}}p_{\mathrm{off}}$, and $p_2=(p_{\mathrm{on}})^2$ If the transitions of the ions were correlated in some way, then deviations from these relationships might occur. For a particular 100 s run, the measured values were $p_0=0.1836$, $p_1=0.4887$, and $p_2=0.3277$. For a value of $p_{\mathrm{on}}=0.57224$, obtained by least-squares adjustment, the calculated values are $p_0=0.1830$, $p_1=0.4896$, and $p_2=0.3275$. Similarly good agreement was obtained for the other runs.

Another test of the independence of the two ions was a comparison of the number of apparent double quantum jumps to the number that would be expected from random coincidences, given the finite time resolution of the apparatus. In 400 s of two-ion data, 11 double quantum jumps and 5649 single quantum jumps downward in fluorescence were noted. Data generated by a Monte Carlo simulation, in which the two ions were independent, were analyzed in the same way. The simulated data had 15 double quantum jumps and 5430 single quantum jumps. Thus, the rate of double quantum jumps was consistent with independence of the two ions.

The experimental probability $P_n(\Delta t)$ of detecting n 11 μm photons in a period Δt can be derived from the quantum-jump data, where Δt is any integer multiple of the experimental time resolution. Mandel's Q parameter,[18] which measures the deviation of the distribution fiom Poisson statistics, is

$$Q=(\sigma^2-\langle n\rangle)/\langle n\rangle\,, \tag{6}$$

where σ^2 and $\langle n\rangle$ are the variance and the mean of the distribution. Negative values of Q indicate sub-Poissonian statistics. The theoretical value of Q is[18]

$$Q=\frac{2N}{T\Delta t}\int_0^{\Delta t}dt'\int_0^{t'}d\tau[g^{(2)}(\tau)-1]\,, \tag{7}$$

where N and T are the total number of photons emitted and the total time. Figure 3(a) shows $P_n(\Delta t)$ for one ion, with $\Delta t=200$ ms and $T=400$ s. The Poisson distribution for the same mean ($\langle n\rangle=1.45$) is also shown. For this data set, the measured value of Q, calculated from Eq. (6), is -0.253 ± 0.025; the theoretical value obtained by inserting $g^{(2)}(\tau)$ into Eq. (7) is -0.242. Figure 3(b) shows $P_n(\Delta t)$ for two ions, with $\Delta t=200$ ms, and $T=400$ s. The measured value of Q is -0.242 ± 0.025; the theoretical value is -0.243. The Poisson distribution for the same mean ($\langle n\rangle=2.84$) is also shown. Previous measurements of Q in single atoms have yielded the values -2.52×10^{-3} (Ref. 4) and -7×10^{-5} (Ref. 3). The value observed here is larger because the detection efficiency is greater.

It should be pointed out that the 11 μm photons have not been observed directly. Rather, their existence has been inferred from observations of the 194 nm fluorescence and the density-matrix equations. The 194 nm field does not itself display nonclassical properties, except on time scales of the order of the $^2P_{1/2}$ radiative lifetime, which we do not observe. Other measurements of photon antibunching[1-3] and sub-Poissonian photon statistics[3-7] require fewer assumptions about the nature of the emitter.

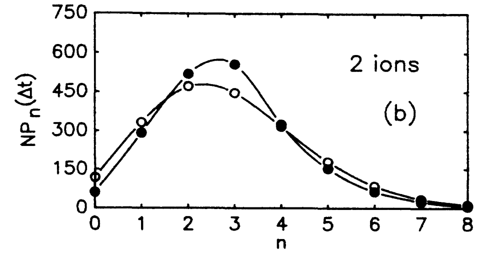

FIG. 3. Plots of the distributions of the numbers of 11-μm photons emitted in 200-ms time intervals from (a) one Hg^+ ion and (b) two Hg^+ ions. The filled circles represent experimental data. The open circles represent Poissonian distributions with the same means as the experimental distributions.

The basic elements of the density-matrix equations have, however, been confirmed by the agreement between the calculated radiative decay rates and the rates deduced from the quantum-jump data,[12] and we are confident in applying them to the present experiment.

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[1]H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977); M. Dagenais and L. Mandel, Phys. Rev. A **18**, 2217 (1978).

[2]F.-M. Rateike, G. Leuchs, and H. Walther, results cited by J. D. Cresser, J. Häger, G. Leuchs, F.-M. Rateike, and H. Walther, in *Dissipative Systems in Quantum Optics,* edited by R. Bonifacio, Topics in Current Physics, Vol. 27 (Springer, Berlin, 1982), p. 21.

[3]F. Diedrich and H. Walther, Phys. Rev. Lett. **58**, 203 (1987).

[4]R. Short and L. Mandel, Phys. Rev. Lett. **51**, 384 (1983).

[5]M. C. Teich and B. E. A. Saleh, J. Opt. Soc. Am. B **2**, 275 (1985).

[6]J. G. Rarity, P. R. Tapster, and E. Jakeman, Opt. Commun. **62**, 201 (1987).

[7]*Squeezed States of the Electromagnetic Field,* edited by H. J. Kimble and D. F. Walls [J. Opt. Soc. Am. B **4**, No. 10 (1987)].

[8]Th. Sauter, R. Blatt, W. Neuhauser, and P. E. Toschek, Opt. Commun. **60**, 287 (1986).

[9]M. Lewenstein and J. Javanainen, Phys. Rev. Lett. **59**, 1289 (1987).

[10]J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).

[11]See, for example, D. Meschede, H. Walther, and G. Müller, Phys. Rev. Lett. **54**, 551 (1985); R. G. Hulet, E. S. Hilfer, and D. Kleppner, *ibid.* **55**, 2137 (1985); W. Jhe, A. Anderson, E. A. Hinds, D. Meschede, L. Moi, and S. Haroche, *ibid.* **58**, 666 (1987).

[12]W. M. Itano, J. C. Bergquist, R. G. Hulet, and D. J. Wineland, Phys. Rev. Lett. **59**, 2732 (1987).

[13]P. Eriksen and O. Poulsen, J. Quant. Spectrosc. Radiat. Transfer **23**, 599 (1980).

[14]J. C. Bergquist, D. J. Wineland, W. M. Itano, H. Hemmati, H.-U. Daniel, and G. Leuchs, Phys. Rev. Lett. **55**, 1567 (1985).

[15]C. E. Johnson, Bull. Am. Phys. Soc. **31**, 957 (1986).

[16]H. J. Kimble, R. J. Cook, and A. L. Wells, Phys. Rev. A **34**, 3190 (1986).

[17]D. J. Wineland, J. C. Bergquist, W. M. Itano, J. J. Bollinger, and C. H. Manney, Phys. Rev. Lett. **59**, 2935 (1987).

[18]L. Mandel, Opt. Lett. **4**, 205 (1979).