Nonclassical Radiation of a Single Stored Ion

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The resonance fluorescence of a single atomic ion stored in a radio-frequency trap was investigated. The photon correlation in the fluorescent light shows antibunching, and the probability distribution of the photon number is sub-Poissonian, both of these being properties of a nonclassical radiation field.

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In this paper we demonstrate the application of a radio-frequency trap to study the resonance fluorescence of a single stored atomic ion which was cooled by radiation pressure using laser light.[1,2] The reemitted radiation corresponds to a nonclassical radiation field as manifested by the photon statistics of the fluorescence radiation. To investigate this, the second-order correlation function (intensity correlation) is observed, this being proportional to the probability of a second photon being detected after a first one within a time interval t. For thermal and noncoherent light this probability has a maximum for $t=0$ and decreases for larger t. This behavior is called photon bunching since it indicates that the photons occur in "clusters." Coherent light displays a value for the intensity correlation that is independent of t. Quantized fields show additional dependences: The smallest value can occur at $t=0$; this is termed antibunching. Such a field is produced by, for example, a single stored ion in the following way: After a photon is emitted the trapped ion returns to the ground state; before the next photon can be emitted, the ion has to be excited again. This happens through Rabi nutation in the external laser field. On the average a time of half a Rabi period has to elapse until another photon can be observed. The probability of two photons being emitted a short time after each other is therefore very small.[3]

Previous experiments to investigate antibunching in resonance fluorescence have been performed by means of laser-excited collimated atomic beams. The initial results obtained by Kimble, Dagenais, and Mandel[4] showed for the second-order correlation function $g^{(2)}(t)$ a positive slope characteristic of photon antibunching, but $g^2(0)$ was larger than $g^2(t)$ for $t \to \infty$. This was due to number fluctuations in the atomic beam and to the finite interaction time of the atoms.[5,6] Later the analysis of the experiment was refined by Dagenais and Mandel.[6] Another experiment with a longer interaction time was performed by Rateike and Walther.[7] In the latter experiment the photon correlation was also measured for very low laser intensities.

The fluorescence of a single ion should also display the following property: The probability distribution of the photon number recorded in a finite time interval t is narrower than Poissonian, which means in other words that the variance is smaller than the mean value of the photon number. This is because the single ion can only emit a single photon. Antibunching and sub-Poissonian statistics are often associated. They are, nevertheless, distinct properties and need not necessarily be simultaneously observed,[8] as is the case in the experiment described here. Although there is evidence of antibunching in the atomic-beam experiments,[4,6,7] the photon counts were not sub-Poissonian as a result of fluctuations in the number of atoms. In the experiment by Short and Mandel[8] this effect was excluded by use of a special trigger scheme for the single-atom event. In the setup described in our work these precautions are not necessary since we have no fluctuations in the atomic number.

The centerpiece of the experiment is a Paul radio-frequency trap[1] mounted inside a stainless-steel ultrahigh-vacuum chamber. At a pressure of 5×10^{-11} mb single ions can be stored for periods of about 10 min. The ion trap and detection optics are mounted on a conflat flange (CF35) (Fig. 1). With a ring diameter of 5

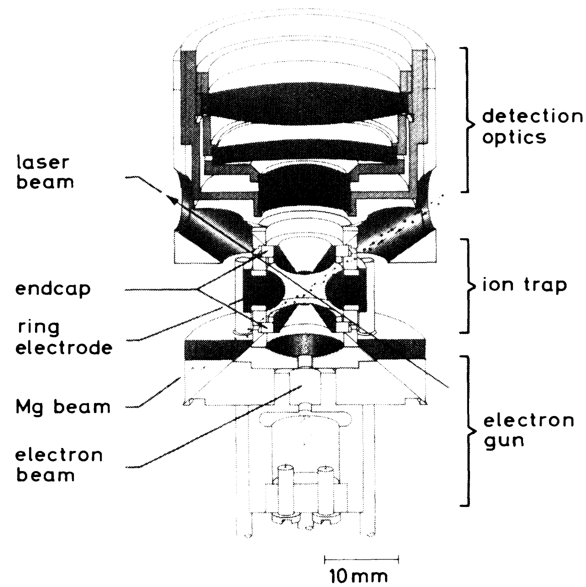

FIG. 1. Radio-frequency trap. The fluorescence light of the stored ion is observed through a bore in the upper pole cap.

mm and a pole cap separation of 3.54 mm this trap is much larger than other single-ion radio-frequency traps.[9-11] Close confinement of a single ion at the center of the trap is achieved by photon-recoil cooling.[12] The size allows a large solid angle for detection of the fluorescence radiation. The resonance fluorescence is transmitted through a molybdenum mesh covering the conical bore in the upper pole cap and then imaged onto a photomultiplier (RCA C31034-A02) by a three-element quartz lens system. The overall efficiency η of the detection is 4×10^{-4}, yielding a high single-ion fluorescence counting rate of 55 kHz.

Typical trapping parameters are an ac voltage amplitude of 570 V, a dc voltage of 10 V, and a radio-frequency of 11 MHz, resulting in radial and axial secular frequencies of 0.73 and 1.1 MHz, respectively. The laser system is a ring dye laser system (CR 699-21) pumped by an Ar^+ laser (Innova CR 20). The spectral bandwidth of the dye laser, being 1 MHz, is small compared with the natural linewidth of the $^{24}Mg^+$ $3^2S_{1/2}$-$3^2P_{3/2}$ transition used in our experiments. The 560-nm radiation is frequency doubled by use of a 30-mm potassium dihydrogen phosphate crystal. To improve the low potassium dihydrogen phosphate conversion efficiency, the crystal is placed inside an external ring resonator.[13]

In order to investigate the intensity correlation in the fluorescence, an ordinary Hanbury-Brown–Twiss setup with two photomultipliers and a beam splitter was used. The output pulses of the two photomultipliers are amplified and discriminated by an EG&G model 584 constant-fraction discriminator. The time delay between the photomultiplier pulses is converted by a time-to-amplitude converter (TAC) into a voltage amplitude. A delay of 63 ns in one of the channels permits us to measure $g^{(2)}(t)$ for negative t and to check the symmetry of the correlation function. The output pulses of the TAC are stored in a multichannel analyzer running in pulse-height-analyzing mode.

The photon correlation signals show the nonclassical antibunching effect connected with Rabi oscillations, which are damped out during the excited-state lifetime, and an undamped modulation (Fig. 2). This modulation cannot be accounted for by a geometrical effect, as in earlier observations,[14] because the oscillation amplitude of the stored ions is much smaller than the beam waist of the laser in the trap (this can be inferred from the Doppler width of the excitation spectrum). However, even in a region with homogeneous intensity the resonance fluorescence of an oscillating ion is modulated, because the laser frequency and thus the excitation probability are modulated in the ion rest frame. The mean frequency is tuned to the long-wavelength edge of the resonance line for photon-recoil cooling. This results in a dependence of the fluorescence intensity on the atomic velocity. The harmonic oscillation of the ion therefore results in an intensity being harmonically modulated with the same frequency. Larger oscillation amplitudes

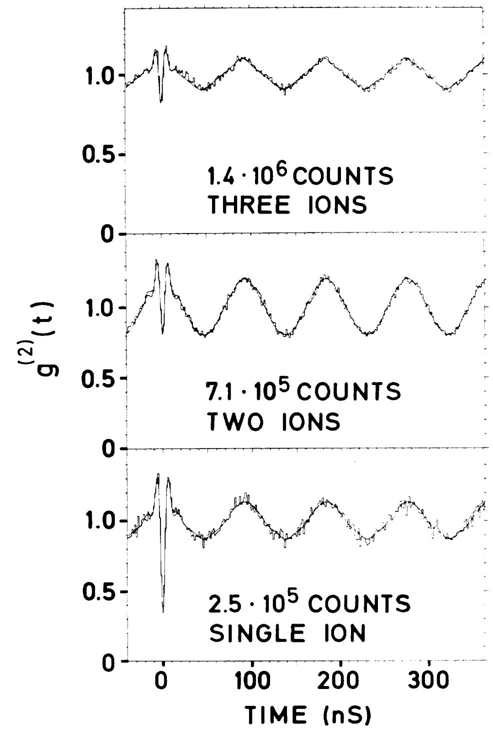

FIG. 2. Intensity correlation for one, two, and three ions. The antibunching signal occurs around $t=0$ and decreases with increasing ion number. The periodic signal at larger t is a result of the micromotion of the ion. The triangular shape obtained for three ions is caused by the increased Coulomb repulsion. The deviation from zero at $t=0$ is caused by accidental coincidences due to stray light. The number of ions is discriminated by discrete steps in the fluorescence radiation.

lead to higher harmonics in the spectrum, which permits the kinetic energy of the stored ion to be calculated from the Fourier spectrum of the correlation function in the classical region. The correlation spectra of one, two, and three stored ions are shown in Fig. 2. The period of the classical modulation of 91 ns corresponds to the trapping frequency of 11 MHz. No influence of the radial and axial secular motions with frequencies of 0.73 and 1.1 MHz can be seen, although in a quadrupole field the secular and rf-field oscillations should have comparable velocity amplitudes.[15] A deviation from quadrupole symmetry can be caused by surface potentials, which shift even a single ion into a region of stronger radio-frequency field; in addition, the Coulomb repulsion between several stored ions is of influence. A deviation from harmonic modulation can be seen in the three-ion spectrum of Fig. 2, where the relative strength of the third harmonic is 10% compared with 2% in the other spectra shown in the figure.

The size of the antibunching signal around $t=0$ shows a strong dependence on the ion number. A single atom gives $g^{(2)}(0)=0$, whereas photons emitted by independent atoms are not correlated, thus leading to a more and more classical correlation function as the number of photons in the field increases. If there are more than one ion stored in the trap, the Coulomb repulsion keeps them at a distance of several micrometers and the fluctuation in their position is of the same order of magnitude. Therefore, coherent beating of the light emitted by different ions is negligible. Also the background radiation gives no coherent contribution since several coherence zones are observed. When the stray-light counting rates are subtracted the experimental values for $g_N^{(2)}(0)$ (for $N=2$ and 3 ions) agree very well with theory. The results are $[1-g_2^{(2)}(0)]/g_1^{(2)}(0)=0.43$ (theory 0.5) and $[1-g_3^{(2)}(0)]/g_1^{(2)}(0)=0.32$ (theory 0.33).

For a single ion the intensity correlation function is given by

$$g^{(2)}(t)=1-e^{-3\gamma t/4}[\cos\Omega t+(3\gamma/4\Omega)\sin\Omega t],$$

where $\Omega^2=\Omega_R^2+\Delta^2-(\gamma/4)^2$, Ω_R being the Rabi frequency at resonance, γ the natural linewidth, and Δ the detuning.[16] In order to check the intensity dependence of Ω^2, a least-squares fit to the data points was calculated. In the classical region, for 50 ns $<t<$ 360 ns, the data are fitted by

$$f_c(t)=\sum_{i=0}^{5} a_i\cos[i\Omega_{rf}(t-\delta)],$$

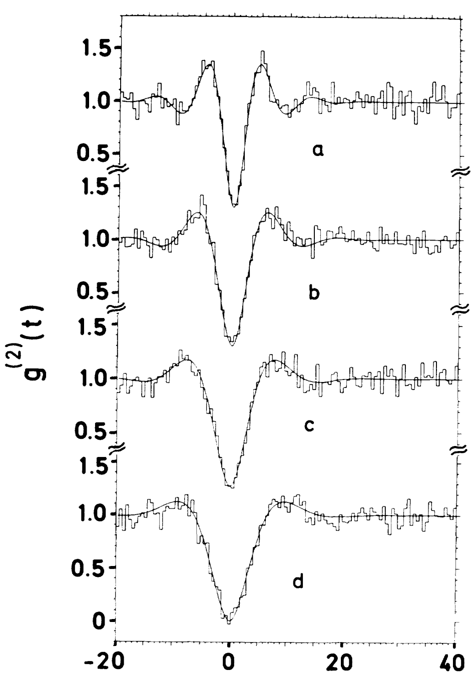

FIG. 3. Antibunching signal of a single ion for different laser intensities. The data have been corrected for the micromotion of the ion; in addition, the background due to accidental coincidences was subtracted.

a_i, Ω_{rf}, and δ being fit parameters. The result for Ω_{rf} is 69 MHz, this being in very good agreement with the trapping frequency of $2\pi\times 11$ MHz. In the region -20 ns $<t<20$ ns, which includes the antibunching signal, the data are fitted by $f_c(t)f_q(t)$, where $f_q(t)=1-ae^{-bt}[\cos(ct)+(b/c)\sin(ct)]$. This follows from the result for $g^{(2)}(t)$ given above, but the factor a is introduced to take care of the stray-light background. Averaging the fit parameter b of 11 independent measurements yields $\tau=3.46\pm0.29$ ns for the lifetime of the $3^2P_{3/2}$ state of ^{24}Mg. This is in good agreement with a Hanle-effect measurement giving $\tau=3.67\pm0.18$ ns.[17] The results for $g^{(2)}(t)$ measured with four different intensities are shown in Fig. 3 together with the corresponding fits. The linear dependence of Ω^2 upon intensity following from these results is shown in Fig. 4.

With our setup we can easily demonstrate that the fluorescence photons show sub-Poissonian statistics. The setup used for this purpose is the same as that used for the correlation measurements. The discriminators in the two signal channels of the photomultipliers were adjusted so that each of them showed the same average number of trigger pulses. The pulses of one channel start and those of the other stop the TAC, and a pulse-height analysis is performed. In this way the number of events ΔN in which two photons arrive within the time interval ΔT is determined. Because of the low pulse rate the probability that two start signals arrive during the 400-ns dead time of the TAC is very low and can be neglected. The number of events in the interval ΔT is calculated to be $\Delta N=N_{start}P_{stop}(\Delta T)$, $N_{start}=N/2$ being the total number of start pulses and $P_{stop}(\Delta T)$ the probability of detecting a stop pulse in a time interval ΔT centered at zero. Assuming Poissonian statistics for $P_{stop}(\Delta T)$, one has

$$\Delta N=\frac{N}{2}\sum_{i=1}^{\infty}e^{-\langle n\rangle}\frac{\langle n\rangle^i}{i!},$$

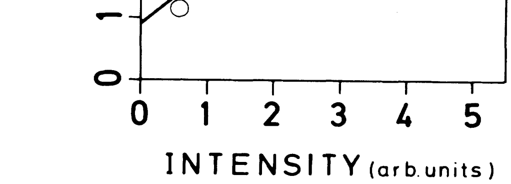

FIG. 4. Square of the modulation frequency Ω vs laser intensity for the results shown in Fig. 3.

where $\langle n\rangle=(N/2)\Delta T/T$ is the average number of counts detected during ΔT at one detector. During a running time of $T=1252$ s a total number of 5.9×10^7 photons were detected. For $\Delta T=4.607$ ns this yields $\Delta N=3.2\times 10^3$ events. Instead we found 1583 events counted by the correlator, a result that differs by 28 standard deviations from Poissonian statistics. This number has not been corrected for background radiation and corresponds directly to the signal recorded by the detector. Because independent detectors were used to measure ΔN, the dead time of the photomultiplier is of no influence.

To compare this result to previous work the normally ordered variance $:Q:$ was calculated with use of the formula[8]

$$:Q:=R\left\{(2/\Delta t)\int_0^{\Delta T}dt'\int_0^{t'}d\tau[g^2(\tau)-1]\right\}.$$

With $R=N/2T$, $g^{(2)}(\tau)=G^{(2)}(\tau)/R^2$ and $G^{(2)}(\tau)=c(j(\tau))/\delta TT$, where $c(j(t))$ is the number of coincidences stored in channel j at time τ; δT is the sampling time per channel. This yields $:Q:=-7\times 10^{-5}$. This value is smaller than that achieved in Ref. 8 because of the smaller overall detection efficiency in the present experiment.

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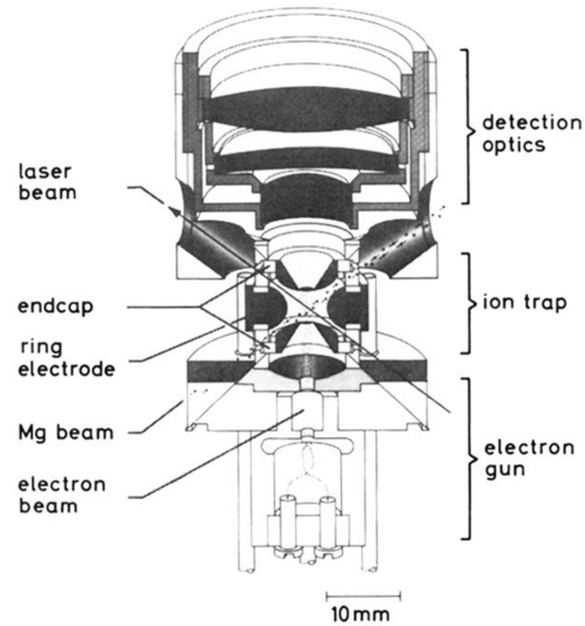

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