Photon Antibunching and Non-Poissonian Fluorescence of a Single Three-Level Ion

M. Schubert, I. Siemers, R. Blatt, W. Neuhauser, and P. E. Toschek
Institut für Laser-Physik, Universität Hamburg, D-2000 Hamburg 36, Germany
(Received 3 September 1991)

The photon pair correlation in the laser-excited fluorescence of a single trapped and cooled Ba^+ ion shows antibunching and, in addition, novel nonclassical phenomena absent in the fluorescence of two-level atoms. They include excessive transient values of the correlation caused by optical pumping, and temporally extended sub-Poissonian photon emission probability which arises from the transient excitation of nonabsorbing Raman coherence. The fluorescence also displays sub-Poissonian photon statistics.

PACS numbers: 42.50.Dv, 32.50.+d

Although light is known to carry information on its source encoded in the correlation functions of all orders, it is the second-order or intensity correlation which provides the main information on intensity fluctuations [1]. Its detection requires two measurements of the light flux. On the microscopic level, the intensity correlation is represented by the correlation of photoelectrons recorded in two events of detection [2], from which the characteristics of photon statistics have been inferred [3]. Atomic resonance fluorescence is a case in point: Measurements of its intensity correlation by comparing the time-separated photon counting signals in one or two channels of detection have revealed nonclassical properties of the light for which the atomic interaction with the excitation light and the vacuum field is responsible. In particular, sub-Poissonian photon statistics [4] have been observed [5,6], and also the rise of the correlation of the two detected photons upon the increase of their time separation τ close to zero ("antibunching" [6,7]). So far, the observations have included dilute atomic beams [5,7] or a single ion in a rf trap [6]. The involved atomic particles could be well approximated as two-level systems with the monochromatic laser light cyclically exciting the resonance line.

We have, for the first time, recorded the intensity correlation of the resonance fluorescence of a single ion which *cannot* be modeled as a two-level system. The observed intensity correlation reveals novel features that are not seen in the resonance fluorescence of two-level atoms. These features include a maximum photon correlation which is much larger than what is possible with two-level atoms, and also photon antibunching with much larger time constants of the initial photon anticorrelation.

A single Ba^+ ion, stored in a 1-mm rf trap of 25-MHz drive frequency [8], was laser cooled [9] to less than 3 mK, and its laser-excited resonance fluorescence was recorded by two photon counting channels placed in opposite directions. The relevant levels of Ba^+ and the wavelengths of the two dye-laser-generated light fields are shown in Fig. 1. The $6^2P_{1/2}$ resonant level decays with 8-ns lifetime to the ground state $6^2S_{1/2}$ and also to the metastable level $5^2D_{3/2}$, with branching ratio 2.85 in favor of the ground state. Since the metastable level lives for 17 s [10], the two light fields are required for the elimination of optical pumping and the generation of a continuous flux of fluorescence. A 0.35-mT magnetic field perpendicular to both propagation and polarization of the light—and parallel to the direction of observation—serves to avoid optical pumping in the $D_{3/2}$ sublevel manifold. Spurious surface charge on the trap electrodes and the mean light pressure tend to shift the dc center of the trap from its ac center. This effect was compensated by four electrodes placed in the ring plane of the trap suitably biased such that no modulation by the drive frequency shows up in phase-synchronized counting of the fluorescence. Since the driven motion grows with the ion's excursion from the trap center by the secular oscillation, this detection technique allows us to estimate the ion

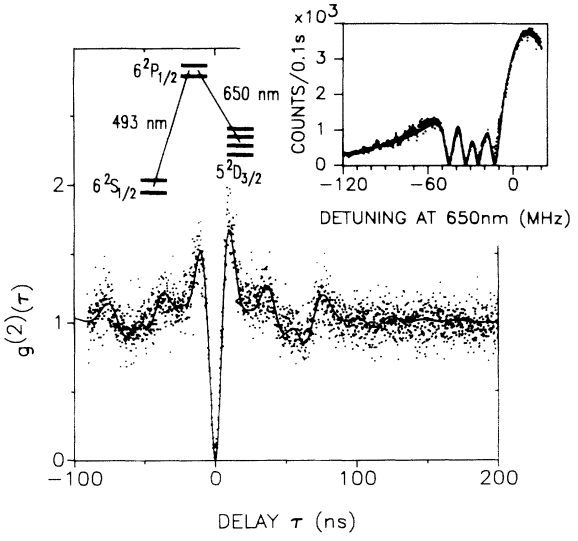

FIG. 1. Intensity correlation $g^{(2)}(\tau)$, green-green ($\tau > 0$) and all-all ($\tau < 0$), of resonance fluorescence, and partial level scheme of single Ba^+. $\Delta_r = 1.5$ MHz. Solid line: Calculation. Inset: Excitation spectrum of green and red Ba^+ fluorescence. Solid line: Fit with the parameters $I_r = 0.29$ W/cm^2, $I_g = 0.38$ W/cm^2, $\Delta_g = -29$ MHz, $B = 0.5$ mT, $\gamma_{sd}/2\pi = 20$ kHz which were used in the calculation of $g^{(2)}$. Dark lines at $\Delta_r = \Delta_g \pm (\frac{11}{5}, \frac{3}{5})\mu_B B/h$.

temperature as less than 3 mK, in agreement with the absence of motional sidebands in the spectral measurements. The frequency fluctuations of the lasers relative to their controlling reference cavities amount to less than 1 Hz (green) and 100 Hz (red) [10^{-4} < (sampling time) < 1 s] [11]. The drift of the cavity resonances is less than 270 Hz/s as measured with an I_2-stabilized He-Ne laser. The emission bandwidths of the lasers —essentially caused by residual fluctuations of the control cavities—are on the order of 6 kHz.

The green light of the 493-nm laser was tuned down from the Ba^+ resonance by a few MHz in order to provide cooling for the ion. At first, the red laser was scanned, by an acousto-optical modulator, across the $D_{3/2}$-$P_{1/2}$ line, and the total fluorescence was recorded. The excitation spectrum shows a "dark line" [12–15] when the red detuning matches the green one, and the laser difference frequency is resonant with the dipole-forbidden line $S_{1/2}$-$D_{3/2}$. This dark line represents the formation of the antisymmetric superposition of $S_{1/2}$ and $D_{3/2}$ states which decouples from the light. The dark line is Zeeman split by the magnetic field [16] into four components (Fig. 1, inset).

Optical Bloch equations for a three-level, eight-sublevel system have been solved, and their steady-state solution for the P-level population versus red laser detuning—i.e., the excitation spectrum—has been fitted to the observed spectra. The light intensities I_i ($i=r,g$) and the strength of the magnetic field B at the location of the ion were measured with best accuracy when using the ion as the probe. Thus, they were unequivocally derived, for each specific measurement, from a fit together with the detuning of the green light Δ_g—which directly shows up in the spectrum—and the effective decay rate of the sd coherence γ_{sd} which includes the bandwidths of the laser radiations.

As the next step, the intensity correlation (or photon pair correlation) of the fluorescence was recorded, with 0.6-ns temporal resolution, by making the signal of detection channel 1 trigger the "start" input of a time-to-digital converter (TDC), and the signal of detection channel 2, with matched delay, the "stop" input. The overall detection efficiency was 2×10^{-3}, and the TDC output versus delay τ represents the conditional probability of detecting a photon at time τ given the detection of a photon at time zero. Accidental counts from residual stray light and the dark counting rate of the photomultipliers, which total about 6%, have been subtracted according to a standard procedure [17]. The Poisson level of the correlation—the delayed coincidence rate of statistically independent photons—has been simultaneously derived from the mean counting rates in a manner that leaves this level unaffected by a possible drift of the counting rates, and the data have been normalized by this value. In the start channel, only green light was admitted, whereas the stop channel registered both radiations. In a measurement of the photon pair correlation of a single ion the start signal derives from a photon that *prepares* the internal ion state in a particular way when the photon is green, and differently when it is red. Thus, the conditional probability for the detection of the second photon depends on the color of the *first* detected photon, since the reexcitation rate of the ion for the emission of the second photon depends on the wavelength. On the other hand, that normalized probability does not depend on the color of the *second* photon which induces the stop signal since the probability equals the normalized value of the $P_{1/2}$ population which is measured by both kinds of photons alike.

When the delay τ is negative (Fig. 1), the first photon is actually detected in the stop channel, and start and stop signals are interchanged. Thus, with $\tau>0$ the pair correlation for *green light only* is recorded, whereas at $\tau<0$ the correlation is for the fluorescence *irrespective* of its color.

The normalized intensity correlation function [3]

$$g^{(2)}(\tau)=\langle E^-(t)E^-(t+\tau)E^+(t+\tau)E^+(t)\rangle \times\langle E^-(t)E^+(t)\rangle^{-2}$$

has been calculated, from the solutions of the optical Bloch equations which are determined by the parameters of the corresponding excitation spectrum, with the help of the quantum regression theorem [18] with *no extra fit* being required (see solid line in Fig. 1). The observed and calculated correlation functions show a null at zero delay and increase with $\pm\tau$, i.e., they indicate antibunching of the fluorescence photons, since a second excitation and fluorescence emission of the ion needs some time. At small τ, the oscillations which result from the ion's damped optical nutation reveal more than one Rabi frequency—unlike with two active levels—which characterize the various transitions among sublevels with different coupling and frequency detuning. As expected of the different spectral sensitivity of the detection channels, the correlation function shows substantial asymmetry with $\tau>0$ or $\tau<0$. The correlation function of a two-level atom is known to be symmetric and to show, close to $\tau=0$, its maximum value 2 on resonance [4], and 4 on far-off resonance. In contrast, the correlation of the Ba^+ ion shows much larger values, e.g., a value of 12 with the red light field detuned far-off resonance (Fig. 2). This is so because the function is normalized to the steady-state light flux which is small as a consequence of optical pumping among the low-lying levels. Since the ion was reduced to the $S_{1/2}$ state by the emission of the green start photon ($\tau>0$) and finally resides in the $D_{3/2}$ level, light is scattered mostly in the transient regime, and its quanta are highly correlated. This correlation periodically varies as the ion's nutational flopping. With $\tau<0$, the ion has some small chance of being reduced to the $D_{3/2}$ state, and the maximum correlation is smaller. More-

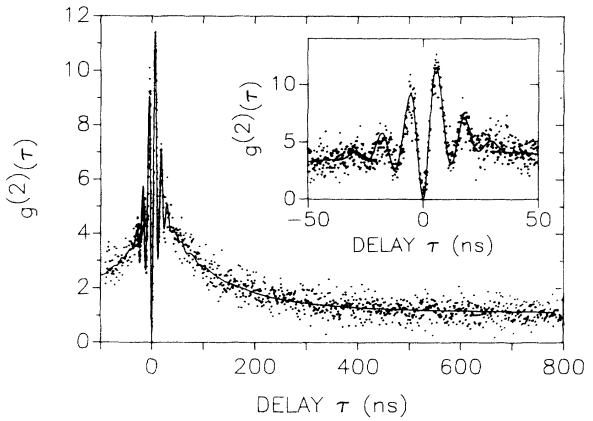

FIG. 2. Correlation $g^{(2)}(\tau)$, as in Fig. 1; red light detuned far-off resonance. Parameters: $I_g=2.2$ W/cm^2, $I_r=660$ mW/cm^2, $\Delta_g=-40$ MHz, $\Delta_r=-110$ MHz, and otherwise as in Fig. 1. Inset: Detail near $\tau=0$.

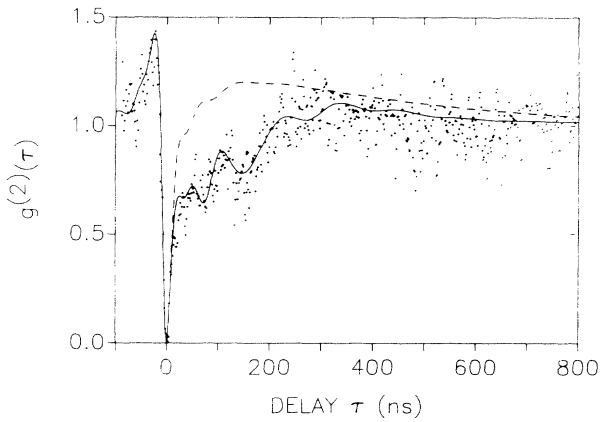

FIG. 3. Correlation $g^{(2)}(\tau)$, as in Fig. 1, with high red and low green laser intensity. Parameters: $I_g=9.6$ mW/cm^2, $I_r=0.1$ mW/cm^2, $\Delta_g=-10.5$ MHz, $\Delta_r=-5.4$ MHz, $B=0.37$ mT, $\gamma_{sd}/2\pi=20$ kHz. Exact calculations including Raman coherence (solid line), and omitting Raman coherence (dashed line).

over, the long time constants of the pumping process, which may exceed the 8-ns decay time of the $P_{1/2}$ level by 2 orders of magnitude, show up in the approach of the intensity correlation to the steady state. In contrast with a two-level system, the longest possible time constant is twice the lifetime of the upper level.

When both light fields are detuned from their resonances by about the same amount, Raman coherence of the ground and $D_{3/2}$ states is excited, which is almost decoupled from the light (the "trapped state" [15]). The photon correlation function when the intensity of the red light is *much higher* than that of the green light is shown in Fig. 3. It displays an extremely extended range of sub-Poissonian emission probability. Starting with the initial antibunching, the correlation function does not develop the signature of the ion's nutational flopping to the resonance level, but rather continued and slowly decaying anticorrelation. After being reduced to the $S_{1/2}$ state, the ion is *not* optically pumped, since the red light is strong. Rather, Raman coherence emerges for a period of time which corresponds to the smallest detuning from one of the Zeeman-shifted two-photon resonances. This time interval is a large fraction of a microsecond as seen in Fig. 3. The solid line in the figure results from a calculation that fully accounts for the *sd* coherence, which was eliminated with the calculation of the dashed line.

Sub-Poissonian emission probability, as demonstrated in the correlation function of Fig. 3, is accompanied by sub-Poissonian statistics of photon counting, as described by negative values of the $Q(T)$ function [4]. These values are not attainable by classical light fields. Figure 4 shows $Q(T)$ calculated from the data of Fig. 3, with $\tau>0$, where T is the sampling time. The error is related to the uncertainty of the determination of the Poisson counting level, and the discrepancy between experiment and theory is consistent with this error. The observed structure of this function remarkably emphasizes the nonclassical nature of the single-ion fluorescence.

In summary, we have observed the intensity correlation function of the resonance fluorescence of a single Ba^+ ion excited by bichromatic light. It has been analyzed by comparison with correlation functions calculated, with no fit, from parameters supplied by independent measurements of excitation spectra. The photon pair correlation of this three-level, eight-sublevel ion shows antibunching and, unlike fluorescence from a two-level atom, vast extra deviations from both Poissonian photon emission probability and statistics which include excess maximum values and protracted anticorrelation. The correlation function reveals the *transient* internal dynamics of the ion, which is characterized by optical pumping and the excitation of Raman coherence, unlike with a two-level atom. The ion's internal dynamics gives rise to substantial sub-

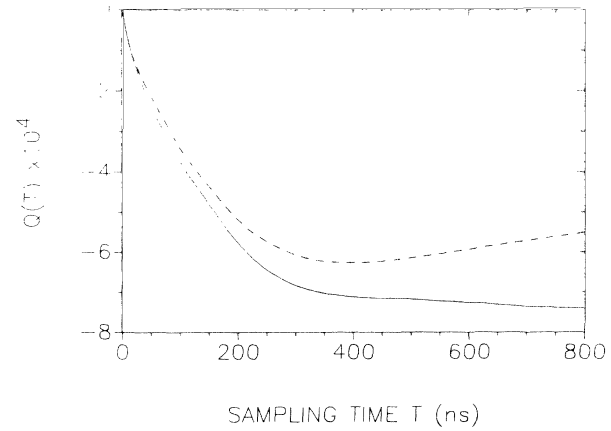

FIG. 4. Mandel's Q function calculated from recorded data (solid line) and from exact full result of Fig. 3 (dashed line).

Poissonian statistics of the photon counting of its fluorescence. Finally the ion approaches the steady state, which is specified by an excitation spectrum, and which is also quite different from what happens with a closed two-level system.

We appreciate discussions with W. Vogel on the theory of photodetection. M.S. and I.S. acknowledge grants by the Claussen-Stiftung, Essen, Germany. This work was supported by the Deutsche Forschungsgemeinschaft.

[1] R. Hanbury Brown and R. Q. Twiss, Nature (London) **177**, 27 (1956).
[2] B. L. Morgan and L. Mandel, Phys. Rev. Lett. **16**, 1012 (1966); D. B. Scarl, *ibid.* **17**, 663 (1966); D. T. Phillips, H. Kleinman, and S. P. Davis, Phys. Rev. **153**, 113 (1967).
[3] R. J. Glauber, Phys. Rev. **130**, 2529 (1963); **131**, 2766 (1963); P. L. Kelley and W. H. Kleiner, *ibid.* **136**, 316 (1964).
[4] L. Mandel, Opt. Lett. **4**, 204 (1979).
[5] R. Short and L. Mandel, Phys. Rev. Lett. **51**, 384 (1983).
[6] F. Diedrich and H. Walther, Phys. Rev. Lett. **58**, 203 (1987).
[7] H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977).
[8] W. Neuhauser, M. Hohenstatt, P. E. Toschek, and H. Dehmelt, Phys. Rev. A **22**, 1137 (1980).
[9] W. Neuhauser, M. Hohenstatt, P. E. Toschek, and H. G. Dehmelt, Phys. Rev. Lett. **41**, 233 (1978).
[10] R. Schneider and G. Werth, Z. Phys. A **293**, 103 (1979).
[11] I. Steiner, V. Enders, F. Elsner, W. Neuhauser, P. E. Toschek, R. Blatt, and J. Helmcke, Appl. Phys. B **49**, 251 (1989).
[12] Th. Hänsch, R. Keil, A. Schabert, Ch. Schmelzer, and P. E. Toschek, Z. Phys. **226**, 293 (1969).
[13] Th. Hänsch and P. E. Toschek, Z. Phys. **236**, 213 (1970).
[14] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento B **36**, 5 (1976).
[15] G. Orriols, Nuovo Cimento B **53**, 1 (1979).
[16] G. Janik, W. Nagourney, and H. Dehmelt, J. Opt. Soc. Am. B **2**, 1251 (1985).
[17] H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. A **18**, 201 (1978).
[18] M. Lax, Phys. Rev. **129**, 2342 (1963).