Detection of individual atoms in helium buffer gas and observation of their real-time motion

C. L. Pan, J. V. Prodan, W. M. Fairbank, Jr., and C. Y. She
Department of Physics, Colorado State University, Fort Collins, Colorado 80523

Received July 14, 1980

Single atoms are detected and their motion measured for the first time to our knowledge by the fluorescence photon-burst method in the presence of large quantities of buffer gas. A single-clipped digital correlator records the photon burst in real time and displays the atom's transit time across the laser beam. A comparison is made of the special requirements for single-atom detection in vacuum and in a buffer gas. Finally, the probability distribution of the bursts from many atoms is measured. It further proves that the bursts observed on resonance are due to single atoms and not simply to noise fluctuations.

Chiefly because of the availability of broadly tunable lasers, it has become possible in the last few years to detect a single atom and to measure its motion. In fluorescence techniques, the atom is detected through the photons it emits when it is excited by a resonant laser beam. In ionization techniques, the atom is ionized by the resonant lasers, and the resulting electron and/or ion is detected. Only two of the previous experiments have been able to record the presence of one atom at the time it was in the laser beam. Hurst et al. detected single atoms created by photodissociation or radioactive decay in coincidence with the creation event with nearly 100% efficiency. Greenlees et al. recorded bursts of six or more fluorescence photons in a microsecond when a single sodium or barium atom in vacuum passed through their cw laser beam. We present in this Letter a third demonstration of single-atom detection in which we record the temporal characteristics of the photon burst from a single atom using a real-time digital correlator. In this way, we not only detect single atoms but we also measure their motion.

An atom that moves in a straight line through the center of a saturated resonant laser beam of Gaussian spatial profile will emit a total number of photons in real time and displays the atom's transit time across the laser beam. A comparison is made of the special requirements for single-atom detection in vacuum and in a buffer gas. Finally, the probability distribution of the bursts from many atoms is measured. It further proves that the bursts observed on resonance are due to single atoms and not simply to noise fluctuations.

$F = aR_s T,$

where $R_s$ is the saturated fluorescence rate and $T$ is the atomic transit time across the laser beam of diameter $d = 2w$. For the optimum case, the intensity in the beam center is 1.91 times the saturation intensity, and the constant $a = 0.55$. Typical experimental parameters for detecting single sodium atoms at room temperature in vacuum and in 200 Torr of helium are presented in Table 1. For detection in vacuum, it is important that the total detection efficiency (the product of optical collection efficiency and detector quantum efficiency) be high, on the order of 10%. This efficiency is not achievable with simple lenses, mirrors, and available photomultipliers. Greenlees et al. used an ellipsoidal light collector to get 5% total detection efficiency. In column 1 of Table 1 we predict that about two photocounts per atom will be detected for typical experimental conditions. This is consistent with the observations of Greenlees et al. Because of the short transit time of an atom in vacuum, background count can be made negligible. A larger laser beam is useful in this case because it increases the transit time, $T$, and hence the fluorescence yield, $F$, according to Eq. (1). In a buffer gas, the atomic transit time is much longer, so smaller laser beams and lower collection efficiencies can be tolerated. A simple lens detection system is sufficient. Because of pressure broadening, higher powers are needed to saturate the atomic transition. In this case, competition from stray light and Rayleigh scattering is quite important. Single-atom detection in helium is still relatively easy. In gases with high Rayleigh-scattering cross sections, single-atom detection may be difficult, although the burst method has the

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Vacuum</th>
<th>200 Torr of He</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beam diameter, $d = 2w$</td>
<td>1 mm</td>
<td>0.4 mm</td>
</tr>
<tr>
<td>Saturation intensity</td>
<td>16 mW/cm²</td>
<td>5.4 W/cm²</td>
</tr>
<tr>
<td>Optimum laser power</td>
<td>0.12 mW</td>
<td>6.5 mW</td>
</tr>
<tr>
<td>Rms velocity</td>
<td>460 m/sec</td>
<td>~</td>
</tr>
<tr>
<td>Diffusion coefficient</td>
<td>~</td>
<td>1.9 cm²/sec</td>
</tr>
<tr>
<td>Transit time, $T$</td>
<td>2.2 μsec</td>
<td>212 μsec</td>
</tr>
<tr>
<td>Saturated fluorescence rate, $R_s$</td>
<td>$3.6 \times 10^7$ sec⁻¹</td>
<td>$4.1 \times 10^7$ sec⁻¹</td>
</tr>
<tr>
<td>Photons emitted, $F$ (optimum)</td>
<td>43</td>
<td>4860</td>
</tr>
<tr>
<td>Background count, $B$ (time $T$)</td>
<td>$4 \times 10^{-2}$</td>
<td>21</td>
</tr>
<tr>
<td>due to stray light ($\beta = 3 \times 10^{-11}$)</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>due to Rayleigh scattering</td>
<td>$2.2$</td>
<td>$24$</td>
</tr>
<tr>
<td>One-atom signal count, $S$</td>
<td>$11$</td>
<td>$4.3$</td>
</tr>
<tr>
<td>Signal-to-noise ratio, $S/\sqrt{B}$</td>
<td>~</td>
<td>~</td>
</tr>
</tbody>
</table>
advantage that only the fluctuations of the constant stray-light signal compete with the single-atom bursts.

Technically, Eq. (1) is not applicable to a sodium atom in 200 Torr of helium because the diffusion motion does not follow a straight line. Nevertheless, we can use Eq. (1) to obtain a rough estimate for the photon yield if we set $T$ equal to a typical diffusion time across the laser beam, $d^2/4D$, where $D$ is the diffusion coefficient of sodium in 200 Torr of helium, 1.9 cm$^2$/sec. With this approximation, we expect to obtain 24 counts in the photon burst from a typical sodium atom. During this time, the constant background signal will be 31 counts, with a typical fluctuation of $\sqrt{31} = 5.6$ counts. Thus the expected signal-to-noise ratio in single-atom photon-burst detection is about 5:1. This is consistent with what we observed in our experiments.

The apparatus we used for single-atom detection is shown in Fig. 1. It consists of a cell in which buffer gas at a given pressure (typically 200 Torr) is flowing slowly (typically 1 cm/sec) across one beam from a Coherent 599 single-frequency dye laser. Three baffles are used on each side, and a light trap at the end reduces the stray light reaching the detector to about 3 X $10^{-11}$ of the incident laser light. The fluorescence photons emitted by a sodium atom crossing the laser beams are collected at right angles to both the laser beam and the helium flow by a pair of f/1 condenser lenses with a matching spherical mirror. The detector is an RCA 31034A photomultiplier cooled to $-30^\circ$C. The photocounts are processed by a Malvern single-clipped digital correlator (Model 7023) operating either in the autocorrelation or the probability density mode. A few sodium atoms are seeded into the helium flow by heating a small piece of sodium metal in the inlet tube. The average number of sodium atoms in the viewing volume, $N$, is monitored by measuring the total fluorescence count rate, $C_F$, at low laser powers, $P$. It is given by

$$N = \pi w^2 C_F h\nu / P e \sigma. \tag{2}$$

The peak-absorption cross section, $\sigma$, of the pressure-broadened transition is determined by measuring the line profile and using the standard formulas for integrated cross section. At 200 Torr of He, the measured linewidth is 3.8 GHz, and $\sigma = 2.6 \times 10^{-12}$ cm$^2$. The detection efficiency, $\epsilon$, is determined in two ways: (1) by measuring the unsaturated resonance fluorescence from a sealed-off sodium cell in vacuum and using the known equilibrium density and absorption cross section at room temperature in Eq. (2) and (2) by measuring the Rayleigh scattering that is due to 200 Torr of He and using the known Rayleigh cross section$^{12}$ in Eq. (2). Both methods give the same answer, which ranged from 0.3% to 1.3% in our experiments.

After the sodium-seeding level was adjusted so that there was much less than one atom in the beam, the correlator was turned on and off repetitively. The measurement time was set for 100 digital channels, corresponding to a few milliseconds in a typical experimental run. For many runs, no photon correlations other than small noise fluctuations were observed. When the laser was tuned on resonance with the $D_2$ line, as many as 20% of the observations (depending on sodium concentration used) yielded correlation functions displaying discernable photon bursts. These were the bursts produced by single atoms traversing the laser beam. One such correlation function is shown in Fig. 2. A single photon burst extending over 12.5 channels can be clearly seen. The atomic transit time can be determined by $\sqrt{2}$ times the number of channels times the sample time, or $T \approx 350$ usec. The total number of photons in the burst can be read from the ordinate intercept$^{11}$ if the average background noise level is subtracted. We read about 16 counts in this burst. The measured laser power, $P$, beam-waist radius, $w$, and detection efficiency, $\epsilon$, for this experiment were 3.8 mW, 212 $\mu$m, and 0.37%, respectively. These conditions are not far from those tabulated in column 2 of Table 1. The exact prediction for this experiment using the measured transit time, $T = 350$ usec, is 21 counts. Thus the experiment and theory agree quite well, considering the approximations made.

The average number of sodium atoms in the detection volume was determined from Eq. (2) to be 0.002 in this experiment. Thus it is unlikely that a second sodium atom was present during the recording of the correlation function in Fig. 2. When the sodium-atom flux was increased by turning up the heat to the sodium pellet, we began to see two or more humps in the correlation.

Fig. 1. The experimental apparatus.

Fig. 2. Single-clipped correlation function of the fluorescence photon burst emitted by one sodium atom crossing the resonant dye-laser beam in 200 Torr of helium. The experimental parameters are: laser power $P = 3.8$ mW; beam-waist radius $w = 212$ $\mu$m; measurement time, 100 samples or 2 usec; measured detection efficiency, 0.37%.
functions, representing photon bursts from more than one sodium atom. When the laser was tuned off resonance, we did not see any correlation functions of the quality of those in Fig. 2. We saw only occasional correlations of poor quality, which presumably were due to statistical background fluctuations.

In order to make sure that the observed photon bursts were due to single atoms rather than to noise fluctuations, we did a probability analysis of fluorescence photocounts. The probability density mode of the correlator was used for this study. The sample time was set several times longer than typical atomic transit times so that a fluorescence burst would fall completely within one sample period. Runs were made with the laser tuned on and off the sodium D2 line. The laser power was adjusted such that the total average counts per sample was approximately the same for both runs. The results are shown in Fig. 3. The fluorescence count rate on resonance was 3800 counts/sec, or 0.95 count per sample. This corresponded to about 0.05 atom on the average in the observation volume. The background scattering that was due to stray light and Rayleigh scattering contributed 14.3 and 15.8 counts per sample on the average for the on-resonance and off-resonance runs, respectively.

If the detected photons were all due to random noise and contained no fluorescence bursts, a Poisson distribution would be expected. This is indeed what we observed when the laser was tuned off the sodium resonance. The solid line in Fig. 3 is a Poisson distribution; it fits the off-resonance data quite well. This confirms that the marginal correlations seen off resonance are just small noise fluctuations. In the on-resonance case, the tail of the distribution extends about 5–10 counts beyond the off-resonance distribution. Since the total count rates were adjusted to be about equal, this cannot be explained by any random process. The fluorescence photons must be arriving in bursts on the order of 5–10 counts. Note that in this case we used a smaller beam diameter than in the experiment of Fig. 2; the burst size predicted from Eq. (1), with correction for less than optimum power, is four counts. Thus this experiment is also in agreement with theory.

In our previous paper, we had proposed to measure flow speeds in gases using the photon-burst correlation technique on single atoms as an alternative to laser Doppler velocimetry and laser time-of-flight velocimetry. In our initial flow cell, shown in Fig. 1, we could perform experiments only at low flow speeds (<1 m/sec) with small laser beams (diameter < 1 mm). Under these conditions, the time for a sodium atom to diffuse across the laser beam is in fact shorter than the transit time that is due to the mean flow. Thus it is impossible to measure flow speeds with this apparatus.

On the other hand, the observed transit time in Fig. 2 does represent a direct measurement of the diffusion time of that atom in the laser beam. In that sense, we have actually made a real-time determination of the atom's motion. By averaging the diffusion times of many atoms, we are able to make accurate determinations of the diffusion coefficient of sodium in helium and other buffer gases.

In summary, we have presented experimental data indicating the real-time detection of single sodium atoms in a slow helium flow at 200 Torr. From the width of the correlation function of the detected single-atom fluorescence burst, the transit time for the diffusing atom can be estimated. In this sense, the motion of a single atom has been observed directly. In order to verify that we are indeed detecting single-atom events, we measured the distribution of fluorescence bursts resulting from individual atoms' crossing a laser beam. As expected, the on-resonance photon-burst probability distribution differs considerably from the corresponding Poisson distribution for random noise. All measurements of the burst photon counts agree with the rough predictions of theory.

This research was supported in part by a grant from NASA/Ames Research Center.

References

9. Single ions have also been trapped electrostatically and detected recently using fluorescence methods by W. Neuhausser et al., in Laser Spectroscopy IV, H. Walther and K. W. Rothe, eds. (Springer-Verlag, Heidelberg, 1979), p. 73.