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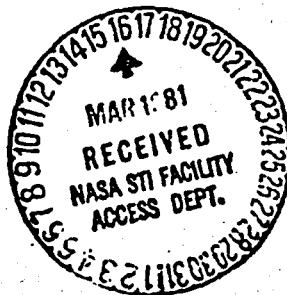
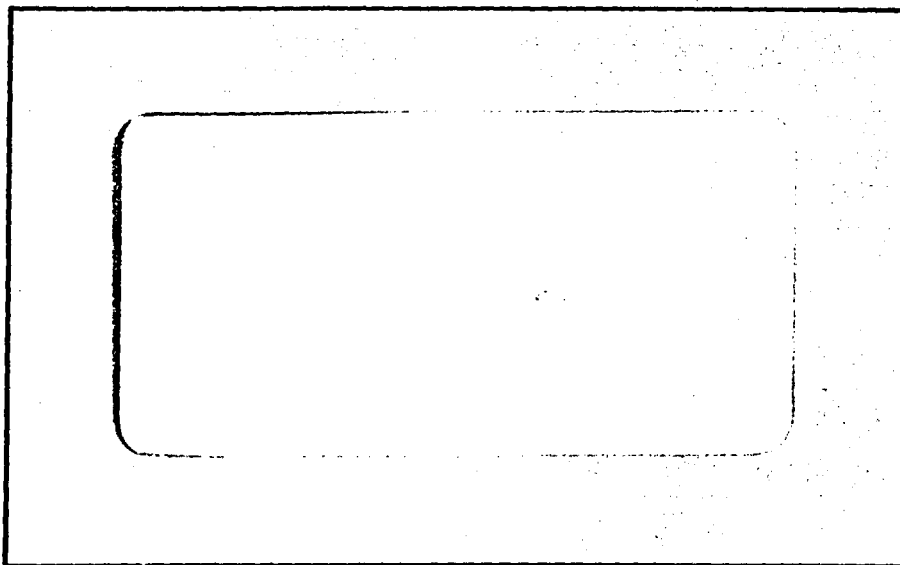
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Department
of
Physics



**Colorado
State University**

Fort Collins, Colorado



FINAL REPORT FOR NASA GRANT NSG-2287
(Grant Period: December 1977 to August 1980)

VELOCITY MEASUREMENTS BY LASER
RESONANCE FLUORESCENCE

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Velocity Measurements by Laser Resonance Fluorescence

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I. Introduction

This is the final report for NASA grant NSG-2287 entitled "Velocity Measurements by Laser Resonance Fluorescence of Individual Atoms". The research was supported from December 1, 1977 to August 31, 1980 by NASA Ames Research Center with grants totaling \$65,000. The experimental research has continued after the termination date of the grant and is still continuing without government support. We have delayed our final report in order to include some of the recent experimental results.

The goals of this research project were: (1) to use the photon-burst correlation method to detect single atoms in a buffer gas, (2) to demonstrate real-time flow velocity measurements with laser induced resonance fluorescence from single or multiple atoms, and (3) to evaluate this method as a tool for wind tunnel flow measurement. These types of experiments have never been attempted before. It is thus not surprising that our research progress has been somewhat slower than we had originally anticipated. Nevertheless, all these goals have now been accomplished. As a side benefit, the demonstration of the ability to probe a single atom has generated considerable interest in the physics community and has opened up new possibilities in basic research. One example is the diffusion coefficient measurements we have made. Other possibilities include a variety of fundamental physics experiments which require single atom sensitivity including the solar neutrino problem, geophysical abundance measurements, and detection of nuclear explosions and radioactive leaks.

The main portion of this report (Section II) is a brief summary of our research progress where the major achievements will be stated in chronological order. We have recently prepared a detailed summary (Appendix I) of our research findings on this project. It is the text of a talk we will present at the SPIE Meeting in Washington, D.C., April, 1981. It will be published by SPIE in a book form, entitled "Laser Spectroscopy for Sensitive Detection". In Section III, we list our publications and presentations related to this project, as well as the Ph.D. theses partially supported by this research project. Reprints of two of the technical publications are included in Appendices II and III.

II. Summary of Research Progress (1978-1981)

During the first six months we set up our experimental apparatus and measured the collection and detection efficiencies. We tried to detect single atoms in one vacuum cell and one flow cell, but were unsuccessful because of the presence of a high level of stray light. During the second half of 1978, we made a detachable flow cell consisting of a center section made of pyrex glass where sodium atoms are seeded into flowing buffer gases and two end sections made of brass mounted with Brewster windows. Baffles and a light trap were used and the interior was blackened to reduce the stray light reaching the detector. The results with this cell were much better. The stray light was reduced to a level comparable to the Rayleigh scattering from the buffer gas used. With the measured total detection efficiency of 0.4%, single sodium atoms in 200 Torr of helium gas were detected and their transit times across a laser beam were measured. The detection of a single sodium atom in a 200 Torr buffer helium gas typically represents a detection sensitivity of one part in 2×10^{18} under our experimental conditions.

Continued experimentation on the measurement of single atom transit-time had led to the conclusion that the apparent single atom motion we measured actually represented single atoms diffusing across the laser beam rather than atoms moving with the average flow speed of the buffer gas. In order to measure the flow speed, a new flow system had to be built to produce an average flow fast enough to make diffusional motion negligible. We also concluded that a higher detection efficiency was needed. While we were designing a fast flow nozzle and ordering an ellipsoid light collector, we decided to use our setup in which diffusion dominates flow to measure diffusion coefficients of Na-He, Na-Ne and Na-Ar under different gas pressures. We also examined the contribution of sodium atoms in excited states to the measured diffusion coefficients. This represents an exciting new area of research which has not yet been fully explored by us. We ended up spending more than a year of time investigating diffusion coefficients. A few experimental problems still remain to be overcome. The details of our diffusion coefficient experiments are given in our SPIE paper (Appendix I of this report).

A faster flowing system was finally put together and an ellipsoidal collector was obtained near the end of July 1980 (one month from the end of the grant period). After testing the system parameters and measuring the laser beam profiles and the detection efficiency, the first unambiguous flow speed measurements of sodium atoms in helium flow were made near the end of 1980. The signal-to-noise for these experiments is borderline for single atom detection. We found that when two laser beams were used in order to make a velocity measurement, correlation functions with good signal-to-noise ($S/N \geq 5$) can be obtained only with multiple atoms. This is because of high background light levels and low total

detection efficiency (about 1.5%) even with our ellipsoidal collector. Figure 1a shows a flow velocity measurement taken in 0.5 sec with a flow speed of 88 m/s. About 60,000 sodium atoms contributed to the observed correlation function. If an average is taken over 10 seconds (Fig. 1b), excellent signal-to-noise can be obtained. We have not tested higher flow speeds due to the limited capacity of our vacuum system. We expect that speeds faster than 88 m/s can be measured with this technique, but the measurement time may need to be increased. Further testing and alignment of our light collection system should improve the total detection efficiency by up to a factor of 10. Then the detection time could be made considerably smaller (at least 10 times faster). We have also been trying to measure velocities using one laser beam. Since the stray light is less in this case, it is possible to measure the flow velocity of a single atom. The velocity determination is not as clear cut in this case. We are currently investigating the accuracy which can be obtained with this method.

In short, after considerable efforts, we have demonstrated that single atoms and their real-time diffusional motion in a buffer gas can be measured by the fluorescence photon-burst method. By averaging over many atoms, we have measured flow velocities up to 88 m/s in a time of 0.5 sec. We expect that higher flow speeds can be measured by this method and that the measurement time can be reduced by a factor of 10 or more by careful experimental design. At the present time, this method is clearly not ready for incorporation in high speed wind tunnels. For one thing, we do not know if the stray light level will be higher or lower in a wind tunnel. It may be lower because the walls of the observation region are farther from the detector than in our experimental apparatus. We also do not know what detection efficiency can be obtained

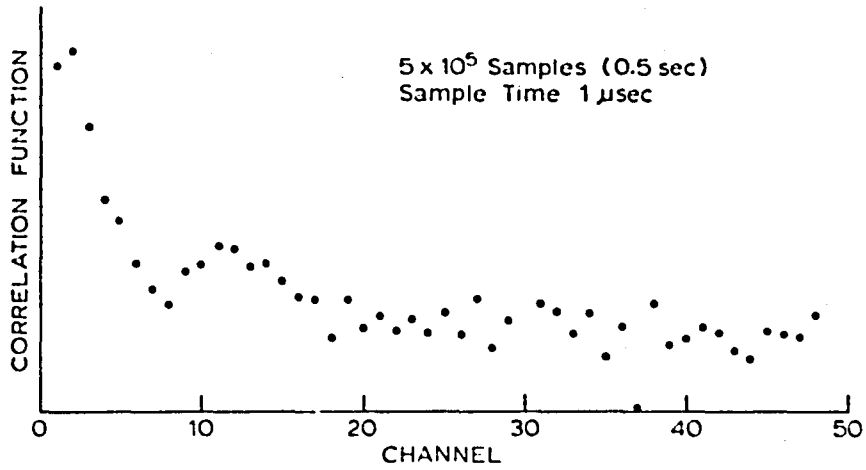


Figure 1a. A two-beam correlation function for a velocity measurement in 0.5 sec. Measured speed = 88 m/s.

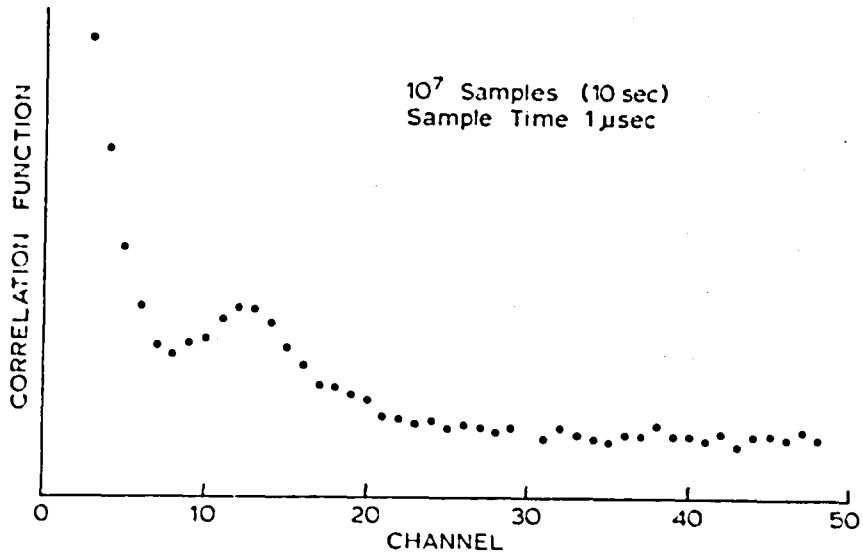


Figure 1b. A two-beam correlation function for a velocity measurement in 10 sec. Measured speed = 88 m/s.

in a wind tunnel situation. Our research indicates that the total detection efficiency must be at least 1% for practical flow speed measurements. Any reduction in stray light would reduce the measurement time from those we have achieved.

III. Publications, Presentations and Thesis Work

Three technical publications, six presentations (three invited) and two Ph.D. theses resulted from this research project. We list them in chronological order as follows:

1. W. M. Fairbank, Jr., C. L. Pan and C. Y. She, "Measuring the Velocity of Single Atoms in Real-Time," presented at the 1978 International Quantum Electronics Conference, May, 1978.
2. C. Y. She, C. L. Pan, J. V. Prodan and W. M. Fairbank, Jr., "Measuring the Velocity of Individual Atoms by Laser Resonance Fluorescence," presented at the 1979 Conference of Laser Engineering and Applications (CLEA), May 1979.
 - * This presentation was selected for a "lay-language" write-up and for discussion at a press conference.
 - * As the result of this presentation, this research project was featured in both Laser Focus (August 1979) and Science News (June 16, 1979).
3. W. M. Fairbank, Jr. and C. Y. She, "Single-Atom Detection with Lasers," Optics News, Spring 1979 (see Appendix III).
4. W. M. Fairbank, Jr., C. L. Pan, J. V. Prodan and C. Y. She, "Direct Measurement of Atomic Diffusion Coefficients by Resonance Fluorescence Correlation Techniques," presented at the Symposium on Atomic Spectroscopy, Tucson, September 1979.
5. C. L. Pan, "Detection and Transit-Time Measurements of Individual Sodium Atoms Diffusing in a Helium Flow by the Laser Resonance Fluorescence Correlation Technique," Ph.D. Thesis, Physics Department, Colorado State University, Winter 1979.
6. W. M. Fairbank, Jr., "Laser Fluorescence Detection of Atomic Motion and Rare Events," Invited talk presented at the American Physical Society Meeting, Chicago, January 1980; Bull. Am. Phys. Soc. 25, 44 (1980).

7. C. Y. She, J. V. Prodan, C. L. Pan and W. M. Fairbank, Jr., "Physiochemical Measurements of Single Atoms in Gases via Fluorescence Correlation," Invited talk presented at the American Chemical Society Meeting, Houston, March 1980.

* This paper was selected for a "lay-language" write-up.

8. C. L. Pan, J. V. Prodan, W. M. Fairbank, Jr. and C. Y. She, "Detection of Individual Atoms in Helium Buffer Gas and Observation of their Real-Time Motion," Opt. Letters 5, 459 Nov. 1980 (see Appendix II).
9. W. M. Fairbank, Jr., C. Y. She and J. V. Prodan, "Detection of Single Atoms and Measurements of their Motion by the Photon Burst Method," to be presented at the Society of Photo-Optical Instrumentation Engineers (SPIE) Meeting, April 1981 and published as a book chapter (see Appendix I).
10. J. V. Prodan, a Ph.D. thesis to be written soon.

IV. Appendices

Three technical publications resulted from this research project and are attached as appendices. Appendix I is a technical summary of the research progress to date.

1. W. M. Fairbank, Jr., C. Y. She and J. V. Prodan, "Detection of Single Atoms and Measurements of their Motion by the Photon Burst Method," to be presented at the Society of Photo-Optical Instrumentation Engineers (SPIE) Meeting, April 1981 and published as a book chapter (see Appendix I).
2. C. L. Pan, J. V. Prodan, W. M. Fairbank, Jr. and C. Y. She, "Detection of Individual Atoms in Helium Buffer Gas and Observation of their Real-Time Motion," Opt. Letters 5, 459 Nov. 1980 (see Appendix II).
3. W. M. Fairbank, Jr. and C. Y. She, "Single-Atom Detection with Lasers," Optics News, Spring 1979 (see Appendix III).

Appendix I

Detection of single atoms and measurement of their motion by the photon burst method

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Abstract

Using a digital correlator, we record the burst of photons emitted by a single atom as it passes through a resonant laser beam. A high efficiency light collection system allows us to register many counts from each atom. The transit time of the atom across the laser beam is computed from the displayed autocorrelation function. By averaging over many atoms, we determine diffusion coefficients for ground state sodium in helium, neon and argon. We have also used this technique to measure rapid flow speeds of gases.

Introduction

Resonance fluorescence has long been recognized as a sensitive technique for detection of small numbers of atoms. With flame or electrical excitation, about one nanogram (10^{13} atoms) of an element is detectable.¹ With modern dye laser excitation, single atoms and ions can now be detected.²⁻⁷ In this paper we will first review briefly the conditions required for detection of single atoms by the photon burst method. Then we will describe our most recent experiments in which the temporal characteristics of the photon burst from a single atom are recorded by a real-time digital correlator. In this way we record information about the motion of a single atom. We will also report measurements of the average motion of many atoms by this method, from which diffusion coefficients of ground state and excited state atoms are determined, and gas flow velocities are measured.

Theory

A two-level atom (Fig. 1a) can be excited many times per second if a dye laser is tuned to its resonant frequency, ν_{12} . An atom excited at time $t = 0$ will decay by spontaneous emission of a photon at $t \approx \tau$, the excited state lifetime. It is then ready to absorb and emit a second photon. Thus, approximately $1/\tau = A_{21}$ photons per second can be absorbed and emitted by a single atom in a resonant laser beam. Taking stimulated emission into account, the exact expression for the maximum spontaneous emission rate is

$$R_s = A_{21}g_2/(g_1 + g_2) . \quad (1)$$

where g_1 and g_2 are the degeneracies of the two states. For most atoms, this number is about 10^7 - 10^8 sec^{-1} . Therefore, a large number of photons can be obtained from a single atom, so long as the atom spends a modest amount of time (>1 μsec) in the laser beam.

The laser intensity required to saturate the atomic transition is given by

$$I_s = 4\pi^2hc \Delta\nu / [(1 + g_2/g_1)\lambda^3] . \quad (2)$$

For atoms in vacuum, the linewidth, $\Delta\nu$, of the resonant velocity group is small (about 10 MHz), and saturation intensities are low, on the order of 10 mW/cm^2 . Thus low power CW dye lasers can be used with beam areas as large as 1 cm^2 . For atoms in buffer gases at atmospheric pressure, $\Delta\nu \approx 10$ GHz, and $I_s \approx 10$ W/cm^2 . Beam areas of 1 mm^2 or smaller are usually used in this case.

The total number of photons emitted by an atom in a saturated resonant laser beam is given by

$$F = a R_s T . \quad (3)$$

where T is the time the atom spends in the beam. The constant a is on the order of unity. It takes into account the path of the atom through the beam and the Gaussian variation of the laser intensity across the beam. For example, consider atoms which move in approximately straight line motion with velocity v (e.g., atoms in vacuum or atoms in a rapidly flowing buffer gas). For these atoms, $T = 2w/v$, where w is the laser beam radius. If the

laser intensity in the center of the beam is equal to $2I_s$, then $a = 0.56$ for those atoms which pass through the center of the laser beam.⁸ Atoms in a stationary buffer gas will diffuse randomly through the laser beam. Their transit times will vary greatly. A typical transit time will be $T = w^2/D$, where D is the diffusion coefficient of the atoms in the buffer gas.

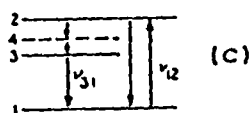
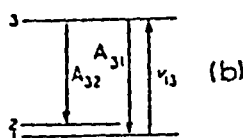
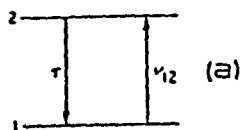


Figure 1. Atomic level schemes: (a) two-level atom, e.g., sodium; (b) multilevel atom, e.g., cesium; (c) atom with intermediate metastable level, e.g., barium.

As an example, we have calculated the parameters for detection of single sodium atoms by the photon burst method in vacuum and in 200 Torr of helium. The results are given in Table 1. In vacuum, 43 photons are emitted as a typical sodium atom passes through the center of the laser beam. A total detection efficiency (light collection efficiency times detector quantum efficiency) of 5% is therefore required in order to get multiple counts from single atoms. The fraction of laser photons reaching the detector by scattering off windows, walls, etc., is typically 3×10^{-11} in our experiments. Thus the average number of stray light photocounts expected during a transit time is only 0.04. Greenless, et al. have shown that if one looks for bursts from single atoms, some discrimination against background stray light can be achieved in spectroscopy experiments.³

For sodium atoms in helium buffer gas, higher powers and smaller laser beam radii are usually used. The long transit time characteristic of diffusion motion makes detection requirements less severe than in vacuum. In the example of Table 1, 4860 photons are emitted. A total detection efficiency of only 0.5% still yields 24 signal counts from a single atom. Due to the longer sample time, higher laser power, and contribution of Rayleigh scattering, background light is significant in this case. One can see from Table 1 that single atom detection is still possible, but the expected signal-to-noise is only 4.3.

Many atoms do not have the simple two-level system that sodium and other alkalis have. For example, in some cases (e.g., cesium) it may be desirable to excite a higher resonant state, as shown in Fig. 1b. For these transitions, R_s is typically an order of magnitude less than for the first resonance, and I_s is an order of magnitude greater.⁹ Thus it will be difficult to use these transitions for single atom detection in vacuum. It may still be possible to detect single atoms in buffer gases using these higher resonances, but stray light will be a difficult problem due to the higher laser powers required.

In many atoms, metastable states or ground state fine structure or hyperfine structure states exist between the ground state and resonant excited state (e.g., state 3 in Fig. 1c). If excited atoms emit a photon at frequency ν_{31} and drop into state 3, they become transparent to the laser beam at ν_{12} . Thus the laser induced fluorescence process may be

Table 1. Comparison of Parameters for Single-Atom Detection in Vacuum and in a Buffer Gas at 300 K

Parameter	Vacuum	200 Torr of He
Beam diameter, $d=2w$	1 mm	0.4 mm
Saturation intensity, I_s	16 mW/cm ²	5.4 W/cm ²
Laser power ($I_0/I_s=1.9$)	0.12 mW	6.5 mW
Rms velocity	460 m/sec	-
Diffusion coefficient	-	1.9 cm ² /sec
Transit time, T	2.2 μ sec	2.2 μ sec
Saturated fluorescence rate, R_s	3.6×10^7 sec ⁻¹	4.1×10^7 sec ⁻¹
Photons emitted, $F(I_0/I_s=1.9)$	43	4860
Detection efficiency, ϵ	5%	0.5%
Background count, $B(\text{time } T)$		
due to stray light	4×10^{-2}	21
($B=3 \times 10^{-11}$)		
due to Rayleigh scattering	0	10
One-atom signal count, S	2.2	24
Signal-to-noise ratio, S/\sqrt{B}	11	4.3

prematurely terminated. For the alkaline earth atoms Ba, Ca, and Sr, this occurs after about 25-100 photons.⁹ We may expect that single alkaline earth atoms can only be detected by the photon burst method if a detection efficiency of at least 5% is used. One alternative, however, is to use two resonant lasers, one at ν_{12} and one at ν_{32} . Neuhauser et al. have shown that this solves the bottlenecking problem in level 3, and allows a continuing fluorescence cycle.⁶

Experimental apparatus

Two types of light collection systems which we have used in our experiments are shown in Fig. 2. The light collection efficiency of the f/l lens and mirror system of Fig. 2 was measured to be 4%. This could have been improved by a factor of 2 if antireflection

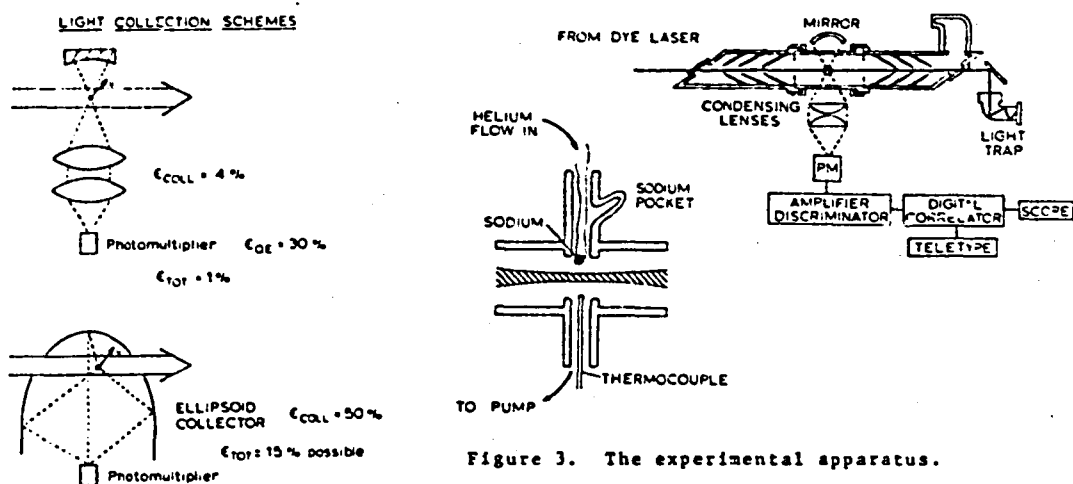


Figure 3. The experimental apparatus.

Figure 2. Light collection systems used in our experiments.

coatings were used on the lenses and the walls of the sample cell. The quantum efficiency of our C31034A photomultiplier tube was measured to be 30%. Thus the total detection efficiency of this system was about 1%. This is more than adequate for detection of single atoms in slowly moving buffer gases (e.g., column 2, Table 1), but it is not sufficient for detection of single atoms in vacuum or supersonic flows (e.g., column 1, Table 1). The ellipsoidal collector in Fig. 2 is a polished ellipsoid available from Helles Griot Corporation. The atoms and the laser beam cross at one focus of the ellipsoid. The detector is placed at the other focus. We measured a light collection efficiency of 50% with the ellipsoid and a total detection efficiency of 15%. This system was used in our experiments with fast helium flows.

The complete experimental apparatus is shown in Fig. 3. The laser beam passes down the center of a long cell with three baffles at each end designed to block stray light which is scattered from the cell windows. Sodium atoms are seeded into the buffer gas flow by heating with resistance wire a small pellet of metallic sodium which is hanging in the inlet tube. In our diffusion experiments a very slow flow of buffer gas carries the sodium atoms into the vicinity of the laser beam, where they diffuse in and out of the beam. In our gas flow measurements, we substituted a glass nozzle for the inlet tube, so that high flow speeds could be obtained. Bursts of photocounts from single atoms are processed by an amplifier-discriminator and fed into a Malvern single-clipped digital correlator (Model 7023). Correlation functions were displayed in real time on an oscilloscope and recorded on a teletype when a permanent record was desired.

Single atom correlation functions

Usually the sodium seeding level was adjusted so that there was much less than one sodium atom in the laser beam at a time, as determined from the total resonance fluorescence count rate. The measurement time was set for 100 digital samples, corresponding to a few milliseconds in a typical experimental run. For most of our runs no photon correlations other than noise fluctuations were observed. For as many as 20% of the

measurements (depending on the sodium concentration used), the correlation functions displayed discernable photon bursts. One such correlation function is shown in Fig. 4. This burst extends over about 12.5 channels or 250 μsec . The actual atomic transit time is $\sqrt{2}$ times larger than this, or $T = 350 \mu\text{sec}$. The total number of photons in the burst can be read from the ordinate intercept,¹⁰ if the average noise level is subtracted. For this burst we read about 16 counts above noise. The conditions of this experiment were fairly close to those tabulated in column 2 of Table 1. The actual prediction for this case is 21 counts. Thus the observations agree well with theory. We believe this to be a burst from a single atom moving through the laser beam. When the laser was tuned off resonance we saw no correlations approaching this quality, just small random noise fluctuations.

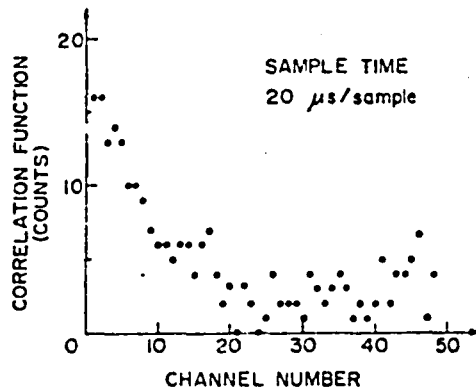


Figure 4. Correlation function of the fluorescence photon burst emitted by one sodium atom crossing one laser beam. The experimental parameters are: laser power 3.8 mW, beam radius $w = 212 \mu\text{m}$, measurement time, 100 samples, or 2 msec, measured detection efficiency 0.37%.

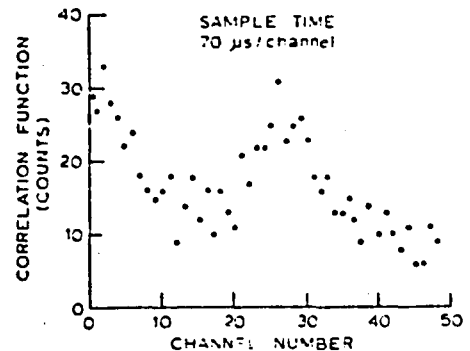


Figure 5. Correlation function obtained when two laser beams are used.

One of the original goals of our research was to use the photon burst method as a means of noninvasive measurement of gas flow velocities. Current Laser Doppler Velocimetry (LDV) techniques used in wind tunnels, for example, rely on scattering from dust particles, which are so large that they probably lag behind the actual flow at sonic speeds. We proposed to use seeded atoms as scatterers and detect the resonance fluorescence photons emitted by the atoms.⁹ Rather than LDV, we chose to use the simpler Laser Time-of-Flight Velocimetry (LTV) method developed by Bartlett and She.¹¹ In the LTV method, two parallel laser beams are used, and two photon bursts are seen as the particle passes through the two laser beams. The correlation function for the two beam case has a second hump due to the cross correlation between the two photon bursts. The transit time between the beams, t , is read from the location of the second hump on the correlation function. The particle velocity is computed from $v = d/t$, where d is the measured distance between the two laser beams.

When we directed two parallel laser beams into the cell, we saw some single hump correlations similar to Fig. 4 and some two-humped correlations such as Fig. 5. Unfortunately, the apparent flow velocities recorded in this manner were quite variable. This was puzzling to us. When we finally estimated the rate of diffusion for these experimental conditions, we discovered that diffusion motion was in fact much faster than the flow of the gas. Thus the correlation function in Fig. 5 may be due to two separate atoms diffusing in and out of the laser beams, or a variety of other possibilities.

Diffusion coefficient measurements

Since the correlation function in Fig. 4 does represent a measurement of the diffusion time (350 μsec) for a particular atom in a single laser beam, it is possible to determine the diffusion coefficient for sodium in helium by averaging correlation functions for many atoms. To do this, we let the correlator accumulate a correlation function for about 10^7 samples, in which time many atoms pass through the laser beam. The resulting correlation function is shown in Fig. 6. Theory predicts that the average correlation function for diffusing atoms is given by¹⁰

$$R(\tau) = A + \frac{B}{\sqrt{(1 + 4D\tau/w_x^2)(1 + 4D\tau/w_y^2)}} \quad (4)$$

In Eq. (4) any ellipticity in the laser beam cross section is taken into account by incorporating semi-major and semi-minor axes, w_x and w_y , into the theory. The constant A in Eq. (4) represents the uncorrelated background noise in the experiment. The solid line in Fig. 6 is a three-parameter (A,B,D) fit to the experimental correlation function. The measured laser beam radii were used for the parameters w_x and w_y .

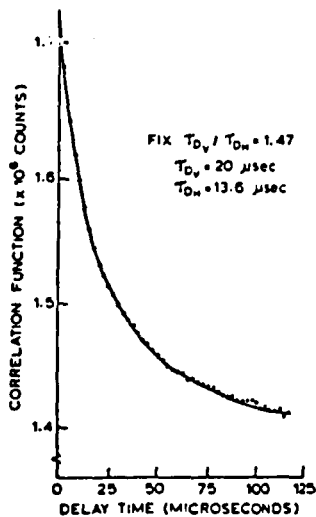


Figure 6. Correlation function averaged over many atoms. The solid line is a least-squares fit to Eq. (4).

Using this method, we have made diffusion coefficient measurements for sodium in helium, neon and argon buffer gases at pressures from 100 Torr to 400 Torr. For all three gases, the product Dp , diffusion coefficient times pressure, is a constant, as it should be. For example, the results of one set of runs in helium is shown in Fig. 7. The error estimates for each point are standard deviations generated by the computer fit to the averaged correlation functions. The average diffusion coefficient shown, $Dp = 360 \pm 16$ Torr cm^2/sec represents a weighted average with a three standard deviation of the mean statistical error limit. The results for several sets of runs in the various inert gases are given in Table 2. The error limits listed in parentheses are purely statistical.

We had originally expected that this method should give diffusion coefficients accurate to better than 10%. We have discovered, however, that our method of radius measurement (finding the $1/e^2$ points of the laser intensity) in these runs introduced significant systematic errors. This was checked by comparing radii measured by the $1/e^2$ method with radii determined by computer fitting measurements of the complete Gaussian profile. The results of these two methods differed by up to 6 μ (up to 12%). Since the radii w_x and w_y appear in Eq. (4) as w_x^2 and w_y^2 , the resulting uncertainties in our diffusion coefficients are 20-25%. The final error limits listed in Table 2 take these possible radii errors into account. We are now in the process of repeating all our diffusion coefficient measurements. In our latest runs, we measure the complete laser beam profile in two dimensions. This gives us a much better determination of the parameters w_x and w_y . We expect that our new diffusion coefficient measurements will have less than 10% uncertainty.

It should be noted that Eq. (4) is correct only if the intensity in the center of the laser beam, I_0 , is well below the saturation intensity, I_s . For all the diffusion coefficient runs described above, the laser power was attenuated until $I_0 \ll 0.1 I_s$. It is also a property of the single-clipped correlation function that the average number of counts should be less than 0.1 per sample time in order for this function to accurately reproduce the actual correlation function. This condition was also satisfied in our diffusion experiments.

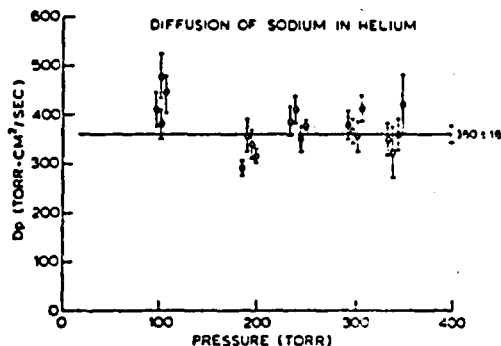


Figure 7. Values of Dp measured in one set of experimental runs. The error limits are purely statistical.

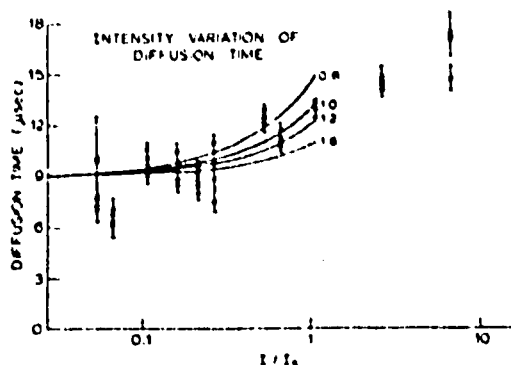


Figure 8. Variation of apparent diffusion time with laser intensity. The numbers on the theoretical curves represent the ratio of the excited state diffusion coefficient to the ground state diffusion coefficient.

When the laser intensity is increased above I_0 , two factors affect the apparent diffusion time calculated from the computer fits to observed correlation functions. One is the fact that the atom spends a significant portion of the time in the excited state. In the excited state the atom will experience a different rate of collisions with buffer gas atoms, and it should have a different diffusion coefficient. The observed diffusion should represent a weighted average of excited and ground state diffusion coefficients. The second factor is that the portion of the laser beam over which the atom is saturated grows larger as the laser power is increased. This leads to longer apparent diffusion times. We have made diffusion runs at a constant pressure (200 Torr) of helium using laser intensities from $I_0/I_0 = 0.05$ to 7. The diffusion times calculated from the observed correlation functions are plotted in Fig. 8. An exact solution for the correlation function is difficult to derive in this case. We have made approximate solutions which are good for intensities $I/I_0 \leq 1$. Theoretical curves are displayed in Fig. 8 for various excited state diffusion coefficients. The error bars displayed in Fig. 8 represent one standard deviation, as determined by the computer fit to the correlation functions. Since the data displayed was taken on two different days, additional systematic errors of about 10% should be added to these error bars to take into account possible variations in the laser beam size and shape. Because of the large experimental uncertainties at low I/I_0 , and the lack of theoretical calculations for $I/I_0 > 1.0$, it is difficult to draw a conclusion about the excited state diffusion coefficient from Fig. 8. However, it does appear that the excited state diffusion coefficient is larger than the ground state diffusion coefficient. From a simple hard sphere model one might expect just the opposite, because the orbit radius of the outer electron in the excited atom is larger than in the ground state atom. On the other hand, calculations using recent Na-He potential curves¹² yield 300 Torr-cm²/sec and 380 Torr-cm²/sec for the ground- and excited-state diffusion coefficients. This ratio of 1.27 is consistent with the results in Fig. 8. It is clear that more accurate experimental data and theoretical curves for $I/I_0 > 1$ are needed in order to get a good determination of the excited state diffusion coefficient. Work is in progress in these areas.

Very recently, Gil'mukhanov and Shalagin in the USSR, have proposed that the difference between ground state and excited state diffusion coefficients could be exploited to separate isotopes.¹³ They report observation of high sodium concentrations at one end of a sodium cell due to this effect.¹⁴ Measurements of the diffusion coefficient of excited state atoms by methods such as ours may prove important in the future in evaluating isotope separation schemes of this type.

Gas flow velocity measurements

Recently we have installed the glass nozzle in the ellipsoidal light collector and have attempted to measure the speed of sodium atoms in a fast helium flow. We have been unsuccessful in observing good single atom correlation functions because the total detection efficiency has been low, about 1.4%. We were recording only about one count per atom in this case. One problem is that when two beams straddle the ellipsoid focus, the

Table 2. Measured values for the diffusion coefficient of ground-state sodium atoms in buffer gases at 315 K. Each measurement represents the weighted average of 7-24 runs taken on a single day at pressures from 100 Torr to 400 Torr.

Buffer Gas	D_p	Statistical Error Limit
Helium	429: 96	(±33)
	360: 91	(±16)
90% Neon, 10% He	323: 65	(±10)
	428: 103	(±31)
Argon	196: 40	(±10)
	250: 61	(±21)

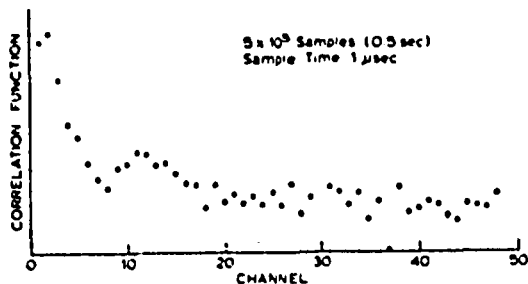


Figure 9. A two-beam correlation function averaged over 5×10^5 samples, or 0.5 sec. A second hump is observed at about channel 12, indicating a transit time of 12 μ sec between beams.

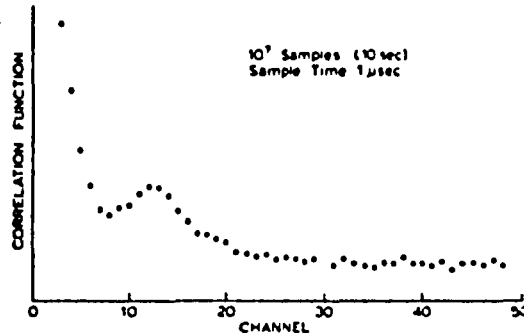


Figure 10. A two-beam correlation function averaged over 10^7 samples, or 10 sec. A more accurate determination of the second peak position can be made in this case than in Fig. 9.

collection efficiency from each beam is less than optimum. We have, however, observed averaged two-humped correlation functions from multiple atoms. Figures 9 and 10 show averages over 5×10^5 samples (0.5 sec, 60,000 atoms) and 10^7 samples (10 sec, 1.2×10^6 atoms). One can see that moderately accurate velocity measurements can be made in 0.5 sec, and quite accurate measurements in 10 sec. From the measured beam separation, $d = 1.056$ mm, the flow velocity is determined to be $v = 88$ m/sec. With improvements in detection efficiency of a factor of 10, the measurement time could be 10 times smaller. So far we have not yet measured speeds as high as the speed of sound in room air, 340 m/sec. But our results do show that fairly fast flows can be measured in a reasonably short time (<1 sec) by the photon burst method.

Conclusions

In summary, we have shown that the temporal nature of the photon burst from a single atom can be recorded in real time with moderate signal-to-noise. This can be used to characterize the motion of that atom. By averaging over many atoms, quite accurate values for diffusion coefficients or fast gas flow velocities can be determined in a short period of time.

Acknowledgements

The authors would like to thank Robert L. McKenzie and Ci-Ling Pan for helpful discussions. This work was supported in part by a grant from NASA/Ames Research Center.

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

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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,¹⁻⁴ the atom is detected through the photons it emits when it is excited by a resonant laser beam. In ionization techniques,⁵⁻⁸ 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.*^{5,7} detected single atoms created by photodissociation or radioactive decay in coincidence with the creation event with nearly 100% efficiency. Greenlees *et al.*⁴ recorded bursts of six or more fluorescence photons in about a microsecond when a single sodium or barium atom in vacuum passed through their cw laser beam. We present in this Letter a third demonstration⁹ of true 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 photons¹⁰

$$F = \alpha R_s T, \quad (1)$$

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,¹⁰ the intensity in the beam center is 1.91 times the saturation intensity, and the constant $\alpha = 0.55$. 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 1. Comparison of Parameters for Single-Atom Detection in Vacuum and in a Buffer Gas at 300 K

Parameter	Vacuum	200 Torr of He
Beam diameter, $d = 2w$	1 mm	0.4 mm
Saturation intensity	16 mW/cm ²	5.4 W/cm ²
Optimum laser power	0.12 mW	6.5 mW
Rms velocity	460 m/sec	-
Diffusion coefficient	-	1.9 cm ² /sec
Transit time, T	2.2 μ sec	212 μ sec
Saturated fluorescence rate, R_s	3.6×10^7 sec ⁻¹	4.1×10^7 sec ⁻¹
Photons emitted, F (optimum)	43	4860
Detection efficiency, ϵ	5%	0.5%
Background count, B (time T)		
due to stray light ($\beta = 3 \times 10^{-11}$)	4×10^{-2}	21
due to Rayleigh scattering	0	10
One-atom signal count, S	2.2	24
Signal-to-noise ratio, S/\sqrt{B}	11	4.3

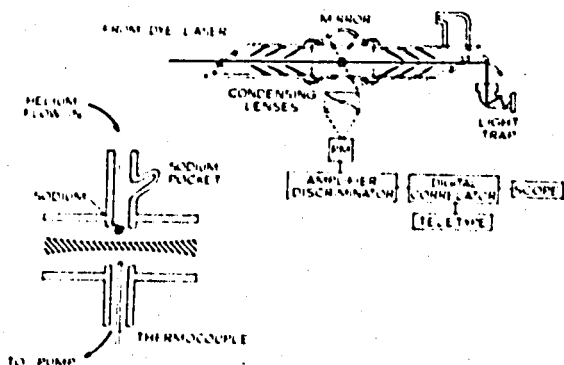


Fig. 1. The experimental apparatus.

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, $1.9 \text{ cm}^2/\text{sec}$. 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 3×10^{-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°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_T , at low laser powers, P . It is given by¹¹

$$N = \pi w^2 C_T h\nu / P \sigma \quad (2)$$

The peak-absorption cross section, σ , 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 $\sigma = 2.6 \times 10^{-12} \text{ cm}^2$. The

detection efficiency, ϵ , 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 section¹² 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 $T \approx 350 \mu\text{sec}$. The total number of photons in the burst can be read from the ordinate intercept¹¹ 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, ϵ , for this experiment were 3.8 mW, $212 \mu\text{m}$, 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, $T = 350 \mu\text{sec}$, 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

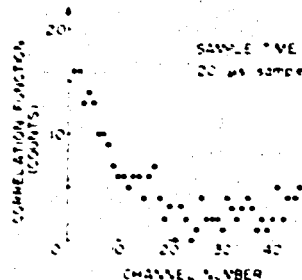


Fig. 2. Single-clipped correlation function of the fluorescence photon burst emitted by one sodium atom crossing the resonant dye-laser beam in 200 Torr of helium. The experimental parameters are: laser power $P = 3.8 \text{ mW}$; beam-waist radius $w = 212 \mu\text{m}$; measurement time, 100 samples or 2 msec; measured detection efficiency, 0.37%.

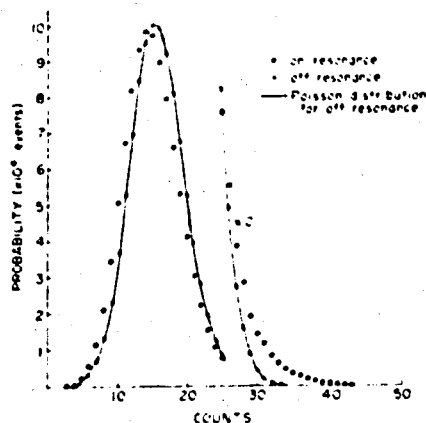


Fig. 3. Probability distributions of detected photon counts on and off resonance compared with a Poisson distribution (solid line). The sample time was set at 250 μ sec. Each run represents 10^6 total samples.

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 D_2 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 14.2 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,¹⁰ 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.¹³ In our initial flow cell, shown in Fig. 1, we could perform experiments only at low flow speeds¹¹ (<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.¹¹

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.

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Single-Atom Detection

with **Lasers**

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W. M. FAIRBANK, JR., and C. Y. SHE



Primarily because of the development of broadly tunable dye lasers, it has become possible to detect a single atom and also to measure its velocity. In the last five years, a number of different single-atom detection schemes have been proposed and demonstrated. In some cases, the detected atoms were in a vacuum; in other experiments, detection was possible even in the midst of 10^{19} atoms/cm³ of a different atomic or molecular species.

In this article we will review the different methods of single-atom detection. As an example, we will concentrate on our own single-atom velocity measurements by the method of laser-resonance fluorescence. Our emphasis on the resonance-fluorescence

method should not be construed as an endorsement of this technique as the best of the different schemes. In fact, the exact conditions of each experiment and the types of dye lasers available to the experimenter will dictate which single-atom detection method is the best choice.

All the single-atom detection methods that have been demonstrated to date involve the same first step: excitation of the atom to a low-lying excited state by a dye-laser beam tuned to the resonant wavelength of the atom. This is represented by the large arrow in Fig. 1. In most cases, this step can be performed quite easily. A few milliwatts of dye-laser power is usually sufficient to saturate the transition, creating a high probability that the atom is in the upper level. The excited atom then is detected in one of the six different ways shown in Fig. 1.

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FLUORESCENCE METHODS

The first four pathways of Fig. 1 are variations on a general method called laser-resonance fluorescence or laser-induced fluorescence. Detection occurs through the spontaneous emission that the atom radiates in all directions when it decays from the excited state back to the ground state via one of the four pathways. The delays before emission are usually short (10^{-7} to 10^{-8} sec), and reexcitation can occur soon after the atom returns to the ground state. This means that up to 10^7 to 10^8 excitations and decays can occur in an atom each second, resulting in 10^7 to 10^8 photons/sec of spontaneous emission. In a typical experimental apparatus the light is collected by lenses and/or mirrors in a direction at right angles to the laser beam. Diaphragms are often carefully placed so that little laser light bounces off surfaces and reaches the detector and photon-counting apparatus. Even with a moderate collection efficiency, on the order of 10^5 counts/sec of signal can be recorded each time a single atom passes into the laser beam. Pro-

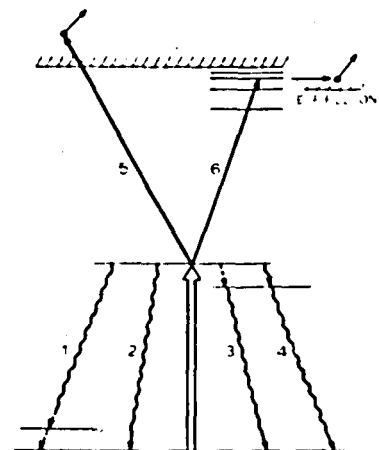


Fig. 1. Different schemes for detecting single atoms with lasers. After an atom is excited by a laser (the large arrow), it may be detected by observing its resonance fluorescence (path 2) or fluorescence at a different frequency (paths 1 and 3), by the method of SONRES (path 4), by photoionization (path 5), and by field ionization (path 6). The nonradiative thermal energy for paths 1 and 3 may also be detected by photoacoustic spectroscopy.

vided that the stray laser light reaching the detector is small, this single-atom fluorescence signal is certainly detectable.

The first experiments in detection at the single-atom level were done by Fairbank *et al.* at Stanford University¹ in 1974. The purpose of their experiments was to measure the vapor density of sodium in an evacuated cell. In order to ensure accurate measurements, the laser intensity had to be kept well below saturation in these experiments. Hence a large laser-beam diameter (1 mm) and low power (3 μ W) were used. Nevertheless, sodium densities as low as 100 atoms/cm³ were measured, representing an improvement of 6 orders of magnitude compared with the best results of conventional methods. In these experiments there were approximately one or two atoms *on the average* in the volume from which fluorescence is collected (note that this volume is not well defined, since a laser beam has no sharp edge and the length of the laser beam seen by the lenses is ill defined because of imperfect focusing). Thus, although individual atoms were not detected in these experiments (the signal was an average over many atoms, one or two at a time), very sensitive detection of matter and the prospects for future single-atom detection were clearly demonstrated.

In the Stanford experiments, the emission followed path 2 in Fig. 1. The signal photons had approximately the same wavelength as the laser, except for a small Doppler shift. Thus it was not possible to distinguish signal and stray-light photons with an interference filter or monochromator. The sensitivity could have been better if the stray light had been lower. In 1977, Gelbwachs *et al.*² of the Aerospace Corporation demonstrated that the stray-light problem could be solved by using path 3 in Fig. 1. They called their method SONRES (saturated optical nonresonant emission spectroscopy). In this scheme, a buffer gas at a moderate pressure is used. The excited atoms collide frequently with buffer-gas atoms. Often the collisions transfer the atom to a nearby excited state. The experimental demonstration of SONRES was done with sodium atoms

in an argon buffer gas at atmospheric pressure. In this case the emitted photon in path 3 is shifted by 6 Å from the laser light. Thus they could use a monochromator to reject all stray laser light and pass only the desired spontaneous emission. With this apparatus, Gelbwachs *et al.* confirmed the sodium-vapor density measurements of Fairbank *et al.* down to 180 atoms/cm³. Since the effective volume was small, the signal at the lowest density came from 0.2 atoms *on the average*.

Greenlees and his co-workers³ at the University of Minnesota proposed a different scheme to beat the stray-light problem of path 2 in Fig. 1 in 1977. They suggested that the signal photons should come in bursts as individual atoms cross the laser beam, whereas the stray-light photons should be distributed randomly in time. By looking for bursts of photons, one should be able to distinguish signal photons from the stray-light noise. Their goal was to do high-resolution spectroscopy on exotic isotopes, for which only a few atoms/sec can be produced.

The problem in this case is that atoms moving with thermal velocities spend only about 10⁻⁶ sec in the laser beam. Thus a detected count rate of 10⁵ counts/sec is not sufficient, for it would yield only 0.1 count per atom, on the average. They solved the problem by using an ellipsoidal mirror to collect the spontaneous emission and achieved a collection efficiency of 46%. The average number of counts recorded per atom was increased to about 1. Because of the statistical nature of the detection process, however, bursts of 2, 3, 4, 5, and 6 counts appeared fairly often from single sodium and barium atoms. Thus, although not every atom was detected as it passed through the laser beam, some individual atoms were detected through the burst of counts they produced. By recording only bursts of 6 counts or more, they found new spectral features that were invisible in the stray-light noise when all counts were registered (Fig. 2). They estimated from Fig. 2 that the burst method increased the sensitivity by a factor of 400.

Similar experiments on single-atom

detection using the resonance-fluorescence technique were carried out independently by Balykin *et al.*⁴ of the Institute of Spectroscopy in Moscow, also in 1977. They used two detectors and the method-of-coincidence counting rather than the photon-burst method to discriminate against stray-light noise. Most of the coincidences in their experiments represented the passage of single atoms through the laser beam.

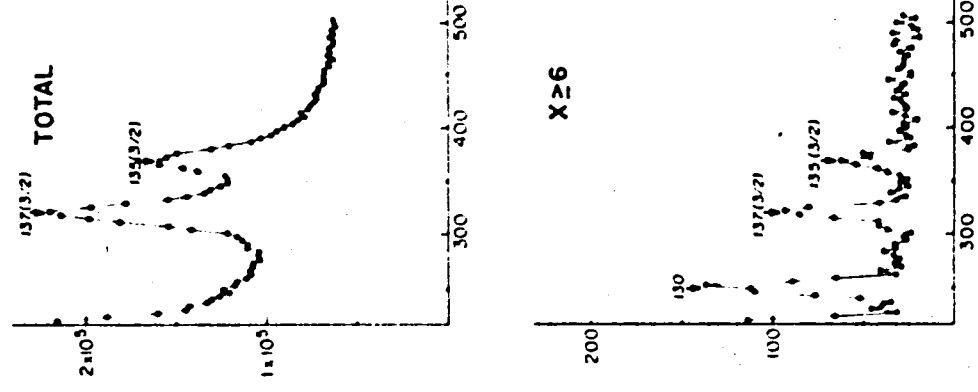


Fig. 2. Frequency sweeps over the 5S35-A transition of a barium atomic beam (taken from Ref. 3). The total counts (upper graph) and frequency of bursts exceeding six photons (lower graph) are plotted as a function of laser detuning (in MHz). The sensitivity of photon-burst spectroscopy is demonstrated very nicely as the peak due to ¹³⁹Ba is clearly resolved in the lower figure.

Before we go on to describe paths 5 and 6 in Fig. 1, in which electrons rather than photons are detected, two comments are in order. First, in many cases, intermediate states, such as in paths 1, 3, and 4 in Fig. 1, can be a hindrance rather than a help to single-atom detection. If the emission from these states is slow, the atom gets stuck in these "metastable" states and cannot be excited again. This problem does occur in sodium and barium but is not so severe as to prevent single-atom detection. In some other atoms, particularly more-complex ones, the problems are more severe, and detection of individual atoms by fluorescence methods will be more difficult. (Of course, if single-atom detection on the average is all that is desired, such atoms can be removed and replaced by new ground-state atoms.)

Second, it is conceivably possible to do single atom-detection by using the energy liberated in the collisions (the dashed lines in paths 1 and 3). Often this energy is converted directly into kinetic energy of the buffer gas. The local temperature and pressure rise propagates outward as an acoustic wave from the atom and is detectable by sensitive microphones. This is called optoacoustic or photoacoustic spectroscopy. Although detection of small absorptions has been achieved with this technique,⁵ to our knowledge it has not been demonstrated yet on the single-atom scale.

IONIZATION METHODS

Paths 5 and 6 in Fig. 1 represent a totally different concept in single-atom detection. In these methods, laser-induced ionization of the atom occurs, creating a free electron and an ion. These charged particles are then detected by electrodes placed near the interaction region.

The first experiments using resonance-ionization spectroscopy (RIS) (path 5) for single-atom detection were done by Hurst *et al.* at Oak Ridge National Laboratory in 1976.⁶ In their scheme, cesium atoms were ionized directly by the same high-intensity pulsed-laser beam that provided the first step of excitation. A buffer-gas mix of argon and methane was added to the detection chamber so that the

single electron released by photoionization of one atom led to an avalanche of 10^4 electrons, as in a gas-proportional counter. These experiments actually were done about a year before the Minnesota and USSR experiments using resonance fluorescence. Thus the Oak Ridge group was really the first to achieve true single-atom detection using a laser.

One of the disadvantages of the RIS method is that photoionization cross sections are small compared with cross sections for transitions between bound states. Thus a high-power pulsed laser (on the order of a kilowatt) was required. Bekov *et al.* at the Institute of Spectroscopy in Moscow pointed out that the laser-power requirements could be reduced by several orders of magnitude if the second laser beam excites the atom instead to a high-lying state near the ionization limit. After the two-step excitation by the pulsed lasers, a pulsed electric field on the order of 10 kV/cm is applied to the atom. This is sufficient to cause spontaneous ionization of the highly excited atoms only, as indicated in path 6 of Fig. 1. The ion is then accelerated and detected by an electron multiplier tube. This method is called selective laser excitation and field ionization by its proponents. Bekev *et al.* detected single sodium and ytterbium atoms by this method in 1978.⁷

COMPARISON OF METHODS

None of the six schemes shown in Fig. 1 is clearly superior to the others. The two ionization methods have the advantage that they are applicable to a wider variety of elements. Usually ionization occurs rapidly, before the atom has a chance to decay to metastable intermediate states. At least half the elements could be detected with single-atom sensitivity by both ionization methods. One disadvantage of the ionization schemes, however, is that pulsed lasers are required. Unless the atoms are moving very slowly or the laser repetition rate is very high, some atoms will go undetected during the time between laser pulses. A second disadvantage is that the atoms are destroyed in the detection process (they become ions). Certain applications,

such as the single-atom velocity measurements described below, cannot be done with these methods. The fluorescence schemes all have the advantage of using low-power continuous dye lasers. In addition, the detection process is nondestructive. The main disadvantage of the fluorescence methods, however, is the limited applicability. The many cycles of excitation and emission that are required provide plenty of opportunities for the atom to get stuck in a metastable intermediate state. As a result, most atoms from the middle of the periodic table are not good candidates because they have multiple ground states. Only the alkali and alkaline earth elements appear favorable for true single-atom detection by fluorescence methods.

A variety of interesting applications should be possible now that single-atom detection has been successfully demonstrated. We have already mentioned high-resolution spectroscopy on rare isotopic species. Two other applications that we will discuss in this article are measurements of the velocity of single atoms and the possibilities of applying these methods to look for quarks. Other exciting applications include the detection of solar neutrinos and super-heavy elements, measuring evaporation from surfaces directly, studying slow transport processes and chemical reactions, and investigating the statistical fluctuations of atoms in a small volume.

SINGLE-ATOM VELOCITY MEASUREMENTS

In 1976, Bartlett and She discovered that, by modifying conventional laser-Doppler-velocimetry (LDV) techniques, they could measure the velocity of a single dust particle moving with the wind at a distance of 100 m.⁸ They used two parallel beams rather than crossed beams, as in the LDV method. In their method (called LTV), a burst of light is scattered by a dust particle when it passes through each laser beam. A telescope and a photon counter located near the laser detect two bursts of photons for each dust particle passing through the two laser beams. A clipped digital correlator computes the correlation function of

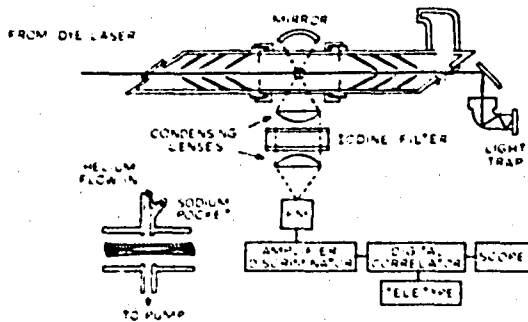


Fig. 3. The experimental setup for single-atom velocity measurements.

the detected counts in real time. The time of flight, T , of the particle between the two beams can be determined directly from the correlation function of scattered light on the oscilloscope. The transverse velocity of the particle can then be calculated using $v = d/T$, where d is the measured separation of the two laser beams.

The impressive sensitivity obtained with individual dust particles at large distances led us to expect that the velocity of individual atoms could be measured at short distances.⁹ The success of the Minnesota group in detecting bursts of fluorescence from single atoms confirmed our expectations. We built the apparatus shown in Fig. 3 for the purpose of measuring the velocity of single sodium atoms. Some care was taken to reduce the level of stray laser light reaching the detector with the use of Brewster windows, light traps, and baffles. The detectable stray light in this cell is about 10^{-11} of the incident laser light when the cell is evacuated. When the cell is filled with helium buffer gas at 200 Torr, flowing vertically downward, a small amount of additional stray light that is due to Rayleigh scattering results.

A few sodium atoms are seeded into the helium flow by heating a side pocket containing sodium metal. As the sodium atoms drift through the two laser beams, we expect to see a burst of fluorescence as an atom crosses each beam. Although the total efficiency of our lens-mirror and photomultiplier system is only 1% (one count detected per 100 photons emitted), theoretical calculations predict that each detected burst will contain a number of counts under a variety of flow conditions.

In our initial experiments we have been using 130- μ m-diameter laser beams of about 1-mW power, each with a separation of $d = 875 \mu\text{m}$. The flow speed, as determined by experiments using dust particles, is about 80 cm/sec in the focal region. With this apparatus we have observed correlation functions in real time such as that shown in Fig. 4. The center of the second bump in Fig. 4 gives the apparent time of flight, $T = 900 \mu\text{sec}$, from which we calculate that $v = 97 \text{ cm/sec}$. The observed number of counts in each burst, about 30, is in good agreement with theoretical predictions.⁹

Unfortunately, we cannot say for sure that we have measured the velocity of a single atom in Fig. 4. At the pressures and beam sizes of this experiment, diffusion contributes as much to the atom's observed motion as the mean flow velocity. In fact, we cannot be absolutely sure that both bursts of counts were due to the same atom. It is possible that one atom diffused sideways after it passed through the first beam and missed the second beam. Sometimes we do observe single-bump

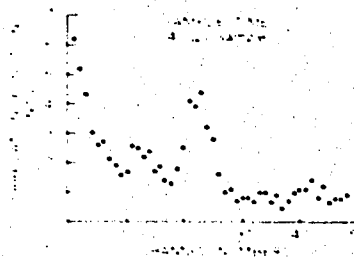


Fig. 4. A correlation function obtained with the apparatus of Fig. 3. The laser beams were 130 μm in diameter and separated by 870 μm . The total measurement time was 4 msec (100 samples).

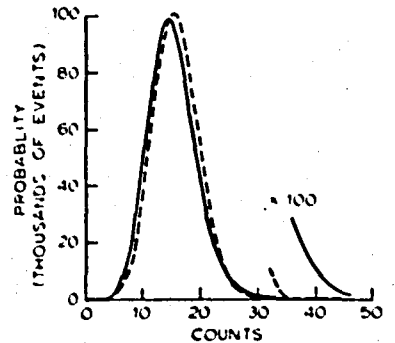


Fig. 5. Probability distributions for photons arriving in 250- μ sec time arrivals: laser off resonance (dashed line) and laser on resonance (solid line) with sodium atoms.

correlation functions indicating only a single burst from an atom. Although we have reduced the sodium density to prevent the appearance of two atoms in a measurement time, we cannot rule out the possibility that Fig. 4 may represent two different atoms rather than one atom traversing two laser beams. We can be sure, however, that each burst observed is due to a single atom's passing through a laser beam.

In order to check the statistics of burst sizes and occurrence frequencies, we measured probability distributions for the number of counts in a sample time (250 μsec) larger than a typical atom's transit time across one beam (130 μsec). To simplify the analysis, only one laser beam was used in these experiments. When the laser was tuned off resonance, we obtained the distribution represented by the dashed line in Fig. 5. This distribution is exactly Poisson, corresponding to random noise and no bursts. When we tuned the laser back to resonance, we saw a non-Poisson distribution, as indicated by the solid line in Fig. 5. Note that on-resonance samples with 30-50 counts occur much more frequently. These sample times must have contained bursts of counts from single sodium atoms. Computer calculations that take into account the measured density of sodium atoms agree well with these observations.

We are currently working on measurements with larger beam diameters and faster flow speeds, in which diffusion plays a lesser role. Only after

these experiments are completed will we be able to say that single-atom velocity measurements have been made unambiguously. As further improvements are made, we hope that flow speeds of 10^4 - 10^5 cm/sec will be measurable with this type of apparatus, setting the stage for wind-tunnel applications. Another interesting application is the measurement of the diffusion coefficients themselves. The correlation functions obtained with one laser beam at low flow speeds, where diffusion dominates, could be used to determine the diffusion coefficients of atoms in a buffer gas directly.

SPECTROSCOPIC SEARCH FOR QUARKS

One of the most exciting applications of single-atom detection is an optical search for quarks, the fundamental subunits of elementary particles. The theory of quarks, which dates back to 1964, is now well established. According to this model, the quarks will have noninteger charge: $\pm 1/3$ or $\pm 2/3$ times the charge of the electron. A variety of quarks searches, including cosmic-ray, mass-spectrometer, accelerator, and magnetic-levitation experiments, has been conducted. However, until two years ago, no experiment produced any reliable evidence for the existence of free quarks. By this time theorists had developed models that predicted that quarks can only exist inside elementary particles and not alone by themselves. Then, in 1977, Larue *et al.*¹⁰ of Stanford University reported evidence for fractional charges in units of $1/3e$ in superconducting Millikan-type experiments they had been working on for over a decade. Still, two years later, no solid confirming evidence, except from their own experiments,¹¹ has been published.

Most of the principal researchers involved with single-atom detection with lasers have expressed interest in looking for quarks by spectroscopic means. This idea was mentioned in the original paper by Fairbank *et al.*¹ and has appeared in subsequent publications by others. A number of proposals have been submitted by these researchers,

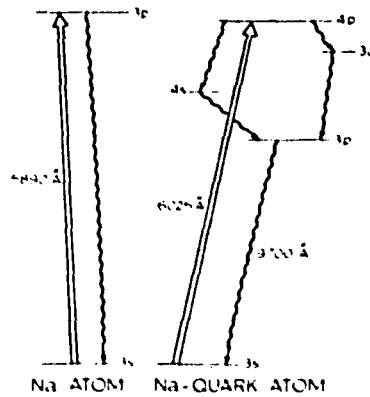


Fig. 6. Energy levels of a sodium atom and a sodium atom with a $-1/3e$ quark bound to the nucleus.

but not one has yet been funded. The mood of the physics community still seems to be one of skepticism, particularly among high-energy theorists, whose models are now in conflict with the Stanford experiment.

The proposed optical quark searches would look for quark atoms, that is, atoms that have a fractional nuclear charge because an extra quark is bound to the nucleus. Since we can now detect single atoms, we should be able to find the one exotic quark atom that might be in a sample. The predicted energy levels of a sodium atom with a $-1/3e$ quark bound to the nucleus are shown in Fig. 6, as an example. Note that the fractional change in nuclear charge has a pronounced effect on the energy levels: it reduces the spacings by almost a factor of 2 in this case. The first excitation step for these quark atoms would have to be at 9700 or 6025 Å, instead of 5890 Å as in normal sodium. The second step could involve any of the methods shown in Fig. 1. In the resonance-fluorescence scheme, for example, when the excitation is at 6025 Å, the photons at either 6025 or 9700 Å could be used for detection.

Since no accurate calculations from first principles have been done for quark atoms, the energy levels shown in Fig. 6 are only approximate. They were obtained by extrapolation of experimental data for the sodium isoelectronic sequence. The uncertainty in the wavelengths shown is on the order

of 100 Å. This is typical of estimates for other $-1/3e$ quark atoms. For $+1/3e$ quark atoms, however, interpolation rather than extrapolation is required, and accuracies of a few angstroms can be obtained.

One of the advantages of optical quark searches is that, if resonances appearing to be quark atoms are found, they can be tested unambiguously by looking for transitions to higher states.

By using the observed resonance wavelength, the higher quark-atom transitions can be predicted very accurately. A successful optical quark search should leave no doubt as to whether or not a free quark has been found. These types of experiments, therefore, are ideal as confirmation tests of the existence of free quarks indicated by the Stanford experiments.

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