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High-Sensitivity Laser Spectroscopy with Atoms from a Cooled Helium Jet

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DISCLAIMER

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Abstract

We have developed a cryogenic He-jet system which efficiently transports radioactive atoms produced on-line at the Argonne National Lab Tandem-Linac Accelerator away from the production region and forms them into a cool atomic beam. This atomic beam will be probed with high sensitivity laser spectroscopy using the photon burst method. The ultimate goal of this work is to determine the sizes, shapes, and magnetic moments of short-lived nuclei through their atomic hyperfine structure. Preliminary measurements with the He-jet system and the adaption of the photon burst method to this new geometry are described.

I. Introduction

The purpose of the work described here is to determine the sizes, shapes and magnetic moments of short-lived nuclei. These are produced in very limited quantities at nuclear reactor and accelerator facilities. We accomplish this by measuring the hyperfine structure of neutral atoms formed with these nuclei. In order to extend the sensitivity of the measurements we are using the photon burst method[1] with which it is possible to measure high resolution spectra with only a few atoms per second. This will allow us to make nuclear determinations in regions which are not accessible to less sensitive methods. Hyperfine structure measurements with radioactive atoms have been made by several groups using a variety of laser spectroscopic techniques. Overviews of this field are given in three relatively recent review articles[2] and it was the primary topic of a recent conference.[3]

There are two basic parts to our experimental apparatus. The first is a cryogenic He-jet transport chamber in which nuclei produced with heavy-ion reactions are thermalized, neutralized, and formed into a cool atomic beam. This chamber is located on a beamline of the Argonne National Lab Tandem-Linac Heavy-Ion Accelerator. The second basic part is a laser spectroscopy system housed in a radiation-shielded adjacent room. Light from a c.w. tunable dye laser passes through a hole in the shielding wall and interacts with radioactive atoms transported by the He-jet. An offset locking scheme[4] is used to generate a digital frequency difference scale accurate to better than 1 MHz and scan the dye laser reproducibly over selectable frequency ranges as statistics for spectra from the tenuous atomic beam accumulate.

We have studied the He-jet system by measuring efficiencies for and distributions of radioactive atoms produced on-line by detecting their nuclear decay. We are also studying the statistical properties of light resonantly scattered by single atoms in a thermal atomic beam chamber using a light collector, photomultiplier tubes (PMT's), and pulse sorting electronics identical to that used with the He-jet chamber. Laser spectroscopy measurements with radioactive atoms produced on-line will be made in the coming year.

In Sec. II of this paper the He-jet system, and the measurements made thus far are described. In Sec. III the photon burst method as adapted to the He-jet geometry is described. Since, in this geometry, each atom scatters hundreds of photons weak decays to metastable states and the effect of light on an atom's motion become important and are briefly discussed. All measurements have been made either with the barium resonance line (5535Å) or sodium D2 resonance line (5890Å).

II. The He-Jet System

A drawing of the He-jet system is shown in Fig. 1. In the upper chamber a heavy-ion beam from the accelerator passes through on 8 mg/cm^2 Ta entrance foil and produces radioactive nuclei in one of several targets. The target cell contains 1 atmosphere of flowing He gas in which the recoiling nuclei stop and are neutralized. The helium gas

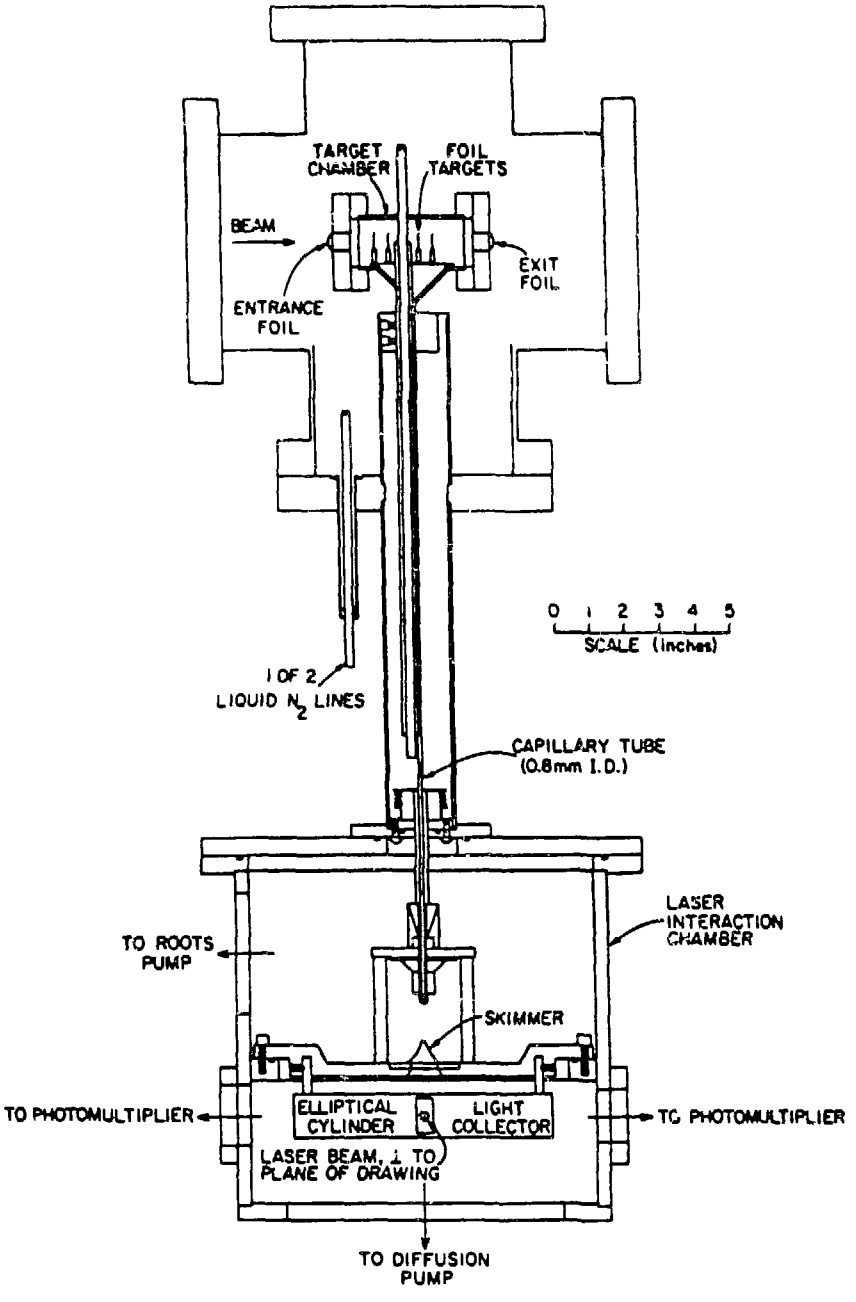

Figure 1

carries the atoms through a 50 cm long, 0.8 mm I.D. copper capillary. The capillary exit chamber is pumped by a 500 l/s Roots blower which maintains a pressure of 0.2 Torr. The He gas is cooled to 78°K before it enters the target cell and this cell and the capillary are kept at the same temperature by flowing liquid nitrogen. The principle loss mechanism is that free atoms stick to the sides of the target cell and capillary. Cooling the gas and the apparatus to 78°K increases the diffusion time for this to occur and hence reduces the losses.[5] It has the further advantage that the thermal velocities are reduced which increases the number of atoms available for a Doppler-free atomic beam as described below. The transport time through the capillary is about 10 ms and the gas in the target cell changes in about 1.5 s. The 1 atmosphere pressure differential across the capillary is sufficient for choked flow. Consequently, at the capillary exit, the flow velocity is equal to the velocity of sound at 78°K which is about 500 m/s for He.

After leaving the capillary, a free jet is formed[6] allowing the "isentropic core" of the jet to further cool as the gas expands. The transported radioactive atoms then pass through a 0.6 mm diameter skimmer into a region of better vacuum ($\sim 10^{-3}$ Torr) where they form an atomic beam. This atomic beam is perpendicularly intersected by laser light as it passes through a high efficiency light collector which is described in the next section. The laser-atomic beam interaction region is about 1 cm long and resonantly scattered light from this region is directed by the light collector to two PMT's.

The absolute transport efficiency of the system is difficult to determine due to uncertainties in nuclear production cross sections. Measurements with the (^{12}C, xnyp) reaction at 55 MeV on natural Ni targets compared with calculations using the compound nuclear code "Alice"[7] indicate that $\gtrsim$30% of the nuclei produced in the target cell are transported. The measurements were made by placing an aluminum catcher foil under the capillary exit, removing the catcher foil after a fixed number of incident heavy ions had passed throughout the target cell, and recording the β-delayed γ activity from the source collected. Placing either positive or negative potential on the catcher foil had no measurable effect on the activity collected indicating that it was transported as neutral objects.

Similar measurements were made with the ^{122}Sn (^{12}C, xn) reaction at 75 MeV bombarding energy. By replacing collector foils masked by a 1 mm diameter circular aperture at the light collector position below the skimmer, we found that the overall transport efficiency from target cell to Doppler-free atomic beam was about 1%. By varying the size of the circular aperture over the catcher foil, we established that the effective temperature of the source of atoms at the top of the skimmer is roughly 1°K.

It should be emphasized that at present all of the results for the He-jet system have been inferred from measurements of nuclear decay of the transported species. Since we use ultrapure He gas which is precooled to 78°K and then passed through a series of molecular sieve filters before entering the target cell, it seems unlikely that there is significant formation of molecules within the target cell. Previous studies[5] are consistent with this statement. However the definitive test of observing resonance fluorescence from radioactive atoms produced in the target cell and transported by the He-jet, has not yet been made.

III. Photon Burst Method

The rate at which nuclei can be produced begins to decrease rapidly as one moves into regions far from nuclear stability. Since these regions are of primary interest, high sensitivity measurement techniques are clearly desirable. We are using the photon burst method to enhance the sensitivity of resonance fluorescence. The basic idea is that, for certain atomic transitions, a single atom can scatter hundreds of photons which will be bunched into a time period equal to the transit time through the laser light. Hence by using the time correlation of detected photons, the number of events from background sources (e.g. aperture scattered laser light or photoelectrons from background radioactivity) can be reduced or eliminated.

In earlier measurements,[1] a thermal atomic beam intersected laser light in a small ($\sim$1 mm diameter) region at the focus of a reflecting ellipsoid of revolution. An aperture restricted the entrance to a PMT located at the second focus. Although the short interaction region limited the average number of detected photons to about 1, measurements were still posible with atom fluxes down to about 10/s. Autocorrelation in the dark counts of the PMT set this lower limit.

We are using a different geometry for work with the He-jet chamber which is shown in Fig. 2. A ribbon-shaped beam of laser light perpendicularly intersects the atomic beam over a 1 cm long region at the common line focus of a reflective double elliptical cylinder. The cylinder is 2.5 cm long and its ends are capped with reflecting plates. The long interaction region makes it possible to detect many more photons per atom than in previous work. Resonantly scattered light is focused by the ellipses through slits at opposing foci and then carried to PMT's by light pipes. The overall light collection and detection efficiency of this system is about 3% for a dipole radiation pattern of green light using EMI 9658 PMT's. Background due to aperture scattered laser light is about 2000/s per milliwatt of power and the dark count rate of the tubes is a few hundred per second when cooled to -20°C.

At present, studies with this technique have been made in an off-line reference atomic beam chamber using Na and Ba atoms. This chamber uses a thermal atom source (oven) and houses a light collector identical to that used in the He-jet chamber. The reason for using the reference chamber for these studies is that accelerator beam time is very limited and we have not developed a satisfactory method of seeding the cryogenic He-jet with stable atoms. The primary difference in the atomic beams produced by the He-jet and

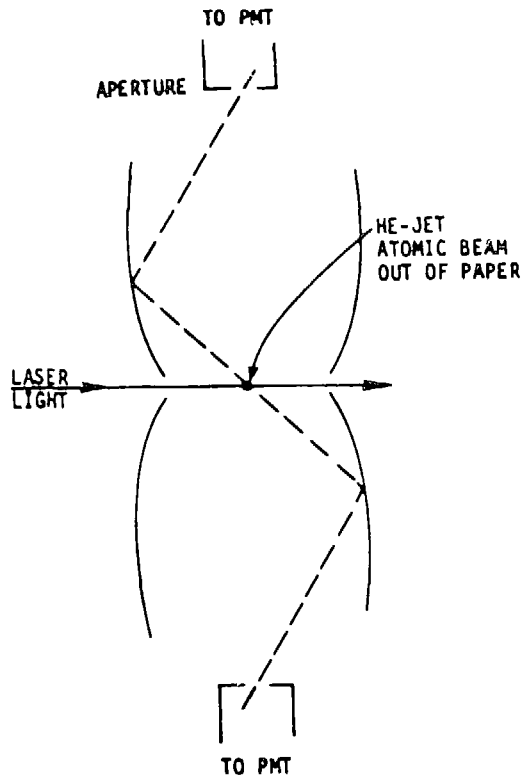

Figure 2

by evaporation from an oven is that atoms from the He-jet have a fixed velocity equal to the He flow velocity at the entrance to the skimmer whereas an oven produces atoms with a distribution of velocities. For evaporated barium atoms the average transit time across 1 cm is 25 μs and for barium atoms from the He-jet the transit time is fixed at about 15 μs.

A block diagram of the pulse sorting and timing electronics is shown in Fig. 3. A pulse from either PMT starts a clock in the master counter for a time τ. During this time interval the number of detected photons is counted. At the end of this interval the multiplicity of the event (i.e. the number of detected photons) is strobed through a router to an PDP 11/03 computer. This computer also controls the frequency of the tunable dye laser through a CAMAC interface. As the laser is scanned many spectra are accumulated, each corresponding to a unique multiplicity. The number of detected photons in each PMT is also recorded by separate slave counters for each event and it is therefore possible to require one or more photons from <u>each</u> tube in order for the event to be accepted by the computer. Requiring at least one photon pulse from each PMT greatly reduces background in burst spectra due to autocorrelation in the dark counts of a PMT. The master counter can be operated in two modes. In mode 1 a photon pulse turns on a clock for time τ_1, each successive pulse is counted, and when the clock reaches τ_1 the multiplicity of the event is strobed to the computer. The pulse counter is then reset and the clock is turned off. In mode 2 operation each successive pulse also resets the clock. In this mode the counter runs continuously as long as there are no time intervals longer than τ_2 between photon pulses. The reasons for these two timing schemes are discussed below.

Suppose that a two-level atom spends time t_1 in laser light producing a total rate of detected photons R when the laser is tuned to resonance. The probability that this atom yields x detected photons is given by the Poisson distribution:

$$P_x = \frac{(Rt_1)^x}{x!} e^{-Rt_1} . \qquad (1)$$

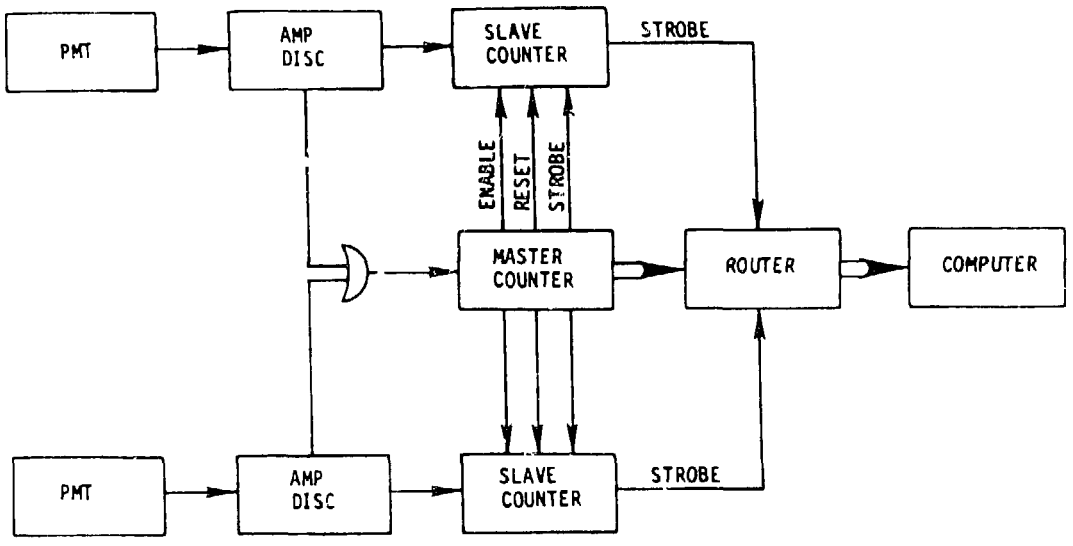

Figure 3

Assuming a lifetime for spontantous emission of 10 ns, a 20% population inversion and a 5% light collection and detection efficiency, R is about 10^6/s. Since for an atomic beam from the He-jet crossing a 1 cm laser light interaction region, $t_1 \sim 15$ μs, the average number of detected photons per atom is about 15. In this situation it is sensible to use the coincidence counter in mode 1 for which the clock is turned on for a fixed time interval of 15 μs. The characteristic time, τ_1, used to distinguish bursts from atoms is then the transit time through laser light. The rate of x-fold events due to uncorrelated background is:

$$N_x^{(1)} = \left[\frac{N}{1+N\tau_1} - \right] \frac{(N\tau_1)^{x-1}}{(x-1)!} e^{-N\tau_1} \qquad (2)$$

where N is the total background rate. The quantity in brackets is the rate at which the counter is enabled and the remaining factors comprise the Poisson probability for x-1 random pulses once the counter is enabled. Since N is typically 10^3/s to 10^4/s, bursts due to random background pulses are negligible in spectra for large multiplicities and the background is due entirely to correlated dark counts.

The situation is somewhat different for an atomic beam produced by an oven. In this case the average transit time for a barium atom across 1 cm is 25 μs but the distribution of velocities about this average must be considered. Since the transit time is not fixed, it is not as useful as a correlation time. Coincidence times $\tau_1 >> 25$ μs are required to ensure that the photon chain is terminated by the atom leaving the laser light rather than the clock timing out. Allowing the latter possibility greatly complicates the analysis of the statistics of detected photons. Fortunately a burst is also characterized by a second time interval, the average time between detected photons (t_2) produced by an atom on resonance. For allowed transitions this is roughly 1 μs. Aside from atom pushing effects (described later) t_2 is independent of the transit time. This second characteristic time can be utilized by resetting the clock with the arrival of each additional photon pulse within a burst (i.e. using the coincidence counter in mode 2). If the coincidence time $\tau_2 \geq 10t_2$, the probability that a detected photon pulse produced by an atom does not reset the clock is $\leq 5\times10^{-5}$. With this arrangement the coincidence counter correctly records the multiplicity of detected photons from each atom regardless of transit time.

The rate of x-fold events from random background pulses is given by

$$N_x^{(2)} = [Ne^{-N\tau_2}] (1-e^{-N\tau_2})^{x-1} e^{-N\tau_2} \qquad (3)$$

for mode 2 statistics. The first quantity in brackets is the rate at which the counter is enabled and the remaining factors comprise the probabilty that the counter is retriggered exactly x-1 times.

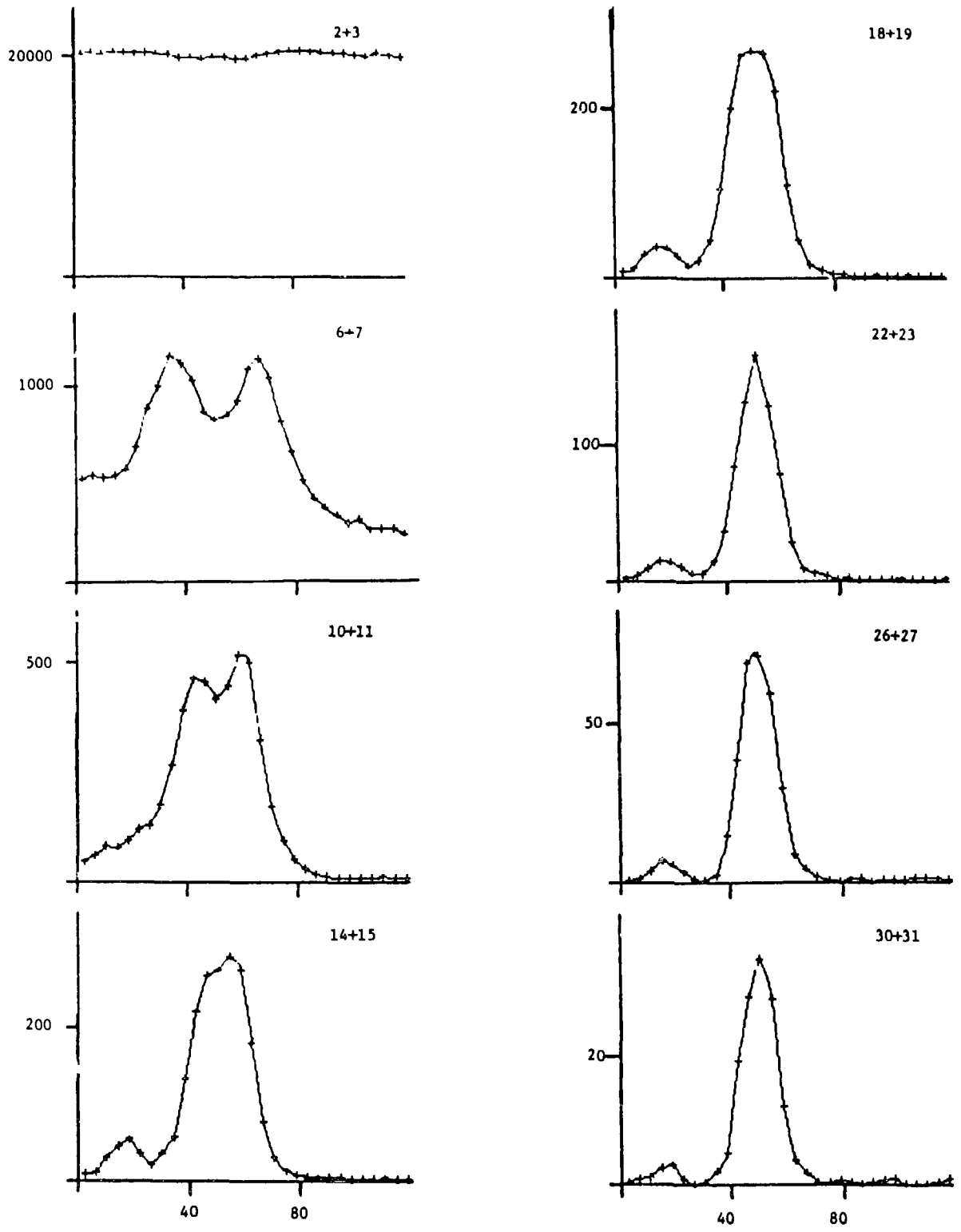
2+3
20000
18+19
200
6+7
1000
22+23
100
10+11
500
26+27
50
14+15
200
30+31
20
40
80
40
80

Figure 4

Selected burst spectra for the 5535A resonance line in natural Ba are shown in Fig. 4. These were taken using a 1 cm long interaction region with an average power density at roughly 5 mW/cm^2. The atomic beam flux was about 100 atom/s. The mode 2 coincidence time was 24 μs and a minimum of one photon from each PMT was required for a valid event. The horizontal scale for the figure is laser frequency in channel number where a one channel difference corresponds to 1.829 MHz. The multiplicities are grouped together by 2's, e.g. the events corresponding to multiplicities 2 and 3 are included in the same spectrum. Only alternate spectra (i.e. 2+3, 6+7, 10+11, . . .) are shown. The large peak near channel 54 is due to ^{138}Ba (72% abundant) and the small peak near channel 18 is due to ^{137}Ba, F=5/2 (11% abundant).

In these spectra the improvement in peak to background ratio for higher multiplicities is obvious. (Indeed there is no peak in the 2+3 spectrum.) The nonlinear dependence on the rate of detected photon R (given in equation 1) gives rise to a narrowing of the lineshapes for large x. At this laser intensity there is significant power broadening. The dips in the center of peaks in low multiplicites arise in the same way. The average number of detected photons per atom is approximately 12.

If the number of photons per atom were limited only by transit time through laser light, the probability for various multiplicites would be given by equation 1 appropriately averaged over the velocity distribution of the source. However for the barium resonance transition (and many others as well) the upper 6s6p $^1P_1^o$ level decays to the metastable 6s5d $^1D_2$ level as well as to the ground state. The detected photon chain ends when the atom is pumped into this level. The Ba data partially shown in Fig. 4 are not consistent with Poisson statistics and it is likely that a significant portion of the atoms are being optically pumped. If the light collection and detection efficiency can be measured with sufficient accuracy this may provide a means for measuring small branching ratios. We have generalized the Poisson distribution for this case and are studying this possibility.

A second effect which becomes important for long (1 cm) interaction regions in the change in an atom's velocity along the direction of the light.[8] We observe this through a shift in the frequency of the resonance. For strong, recycable transitions with light atoms (e.g. Na D2 F=2 - F=3) this effect actually limits the number of photons per atom and causes a marked asymmetry in the lineshapes. Since the burst spectra are sorted by the number of scattered photons, as the multiplicity x increases the centroids of the observed peaks move toward higher laser frequency. If this shift is small (as is the case for heavy atoms such as Ba) the shift in the centroid for multiplicity x is given by

$$\frac{\Delta\nu}{\nu_o} = \frac{1}{2}\,x\,\frac{h\nu_o}{\xi\, mc^2} \qquad (4)$$

where m is the mass of the atom, ν_o is the frequency of the resonance and ξ is the light collection and detection efficiency. These shifts have been observed in the Ba 5535A and Na 5890A lines and provide a measure of the light collection and detection efficiency, ξ.

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