From Helium-6 to Krypton-81

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Abstract. A new method of ultrasensitive trace-isotope analysis has been developed based upon the technique of laser manipulation of neutral atoms. It has been used to count individual ^{85}Kr and ^{81}Kr atoms present in a natural krypton sample with isotopic abundances in the range of 10^{-11} and 10^{-13}, respectively. The atom counts are free of contamination from other isotopes, elements, or molecules. The method is applicable to other trace-isotopes that can be efficiently captured with a magneto-optical trap, and has a broad range of potential applications.

COMMINS & RADIOACTIVE ATOMS

Professor Eugene Commins has had a long "friendship" with radioactive atoms, particularly with isotopes of noble gases. This friendship has been an extremely rewarding one, not only for his own scientific quests but also for the generations of students who followed his footsteps. Just as biological genes are passed along a family tree and affect the health and appearance of many generations down the branches, scientific "genes" are inherited through apprentice relationships and affect one's scientific taste, style, and sometimes preferred techniques. The following is a brief sketch of the scientific family branch that I am attached to.

Our story began in 1958 when Gene and his thesis advisor, Polykarp Kusch, measured the magnetic moment of ^{6}He ($t_{1/2}$ = 0.8 s). They did not observe any effects that would have risen if ^{6}He had a magnetic moment, so they concluded that its spin was probably zero [1]. Through this experiment Gene had his first working experience with radioactive atoms. It also marked the beginning of Gene's distinguished career on searching for anomalous effects.

Later at Berkeley, Gene and his students, including Frank Calaprice, did a series of now classic experiments measuring various correlation effects in the β-decays of ^{19}Ne ($t_{1/2}$ = 17 s) [2]. These measurements were used to test fundamental symmetries and to probe the underlying mechanisms of Weak Interaction.

Frank continued the research on ^{19}Ne at Princeton, where he was joined by Stuart Freedman, a younger graduate from Gene's group. Together they measured the angular distribution of positrons emitted in the decay of polarized ^{19}Ne as a test for the 2nd-class Weak Interaction [3].

When laser trapping of atoms was developed, Stuart, then at Argonne, recognized that radioactive atoms trapped in a Magneto-Optical Trap (MOT) provide an exceptionally good source for the type of experiments done so far with ^{19}Ne trapped in a cell. I was a new student in Stuart's group, and was assigned to work on this idea for

my thesis [4]. Being at the right place and the right moment, I joined the game of trapping radioactive atoms, and have been in it ever since.

In the following section, I will describe the work of my group at Argonne on the laser trapping of ^{81}Kr ($t_{1/2}$ = 230 kyr) atoms. A more detailed description of this work has been published in reference [5]. It is interesting to note that Gene has worked on ^{6}He, ^{19}Ne and ^{35}Ar ($t_{1/2}$ = 1.8 s). It is my pleasure to continue the family business working on a noble gas isotope down the periodic table.

Table 1. Quests on radioactive noble gases -- a family tradition.

Members	Time	Location	Isotope	Reference
Kusch & Commins	1958	Columbia	He-6	[1]
Commins & Calaprice	1969	Berkeley	Ne-19	[2]
Calaprice & Freedman	1975	Princeton	Ne-19	[3]
Freedman & Lu	1994	Berkeley	Na-21	[4]
Lu & Du	2001	Argonne	Kr-81	[5, 6]

ATOM TRAP TRACE ANALYSIS (ATTA)

Much can be learned from the concentrations of the ubiquitous long-lived radioactive isotopes. W. Libby and coworkers first demonstrated in 1949 that trace analysis of ^{14}C ($t_{1/2}$ = 5.7 kyr, isotopic abundance = 1×10^{-12}) can be used for archaeological dating [7]. Since then, two well established methods, Low-Level Counting and Accelerator Mass Spectrometry [8], have been used to analyze many other trace-isotopes at about the parts-per-trillion level and to extract valuable information encoded in the production, transport, and decay processes of these isotopes. The impact of ultrasensitive trace-isotope analysis has reached a wide range of scientific and technological fields.

We have recently developed a new method, Atom Trap Trace Analysis (ATTA) [5, 6], and utilized it to analyze two rare krypton isotopes, ^{81}Kr ($t_{1/2}$ = 230 kyr, isotopic abundance = 6×10^{-13}) and ^{85}Kr ($t_{1/2}$ = 10.8 yr, isotopic abundance ~ 1×10^{-11}). ^{81}Kr is produced in the upper atmosphere by cosmic-ray induced spallation and neutron activation of stable krypton isotopes. It is an ideal tracer for dating ice and groundwater that are older than 100,000 years, which is beyond the range of ^{14}C-dating. ^{85}Kr is a fission product of ^{235}U and ^{239}Pu, and has been used as a general tracer to study air and ocean currents, date shallow groundwater, and monitor nuclear-fuel reprocessing activities. Due to its high mobility, it may be used as a leak sensor to check the seals of nuclear fuel cells and nuclear waste containers.

ATTA is a new laser-based atom-counting method. Our design is based on a type of MOT system [9] that has been used to trap various metastable noble gas atoms. Trapping krypton atoms in the $5s[^3/_2]_2$ metastable level (lifetime $\approx$ 40 sec) is accomplished by exciting the $5s[^3/_2]_2$ - $5p[^5/_2]_3$ transition. Two repump sidebands are generated via additional AOMs to optically pump the atoms into the F=13/2 level for ^{85}Kr and F=11/2 level for ^{81}Kr where they can be excited by the trapping light. In the analysis, a krypton gas sample is injected into the system through a discharge region, where about 1×10^{-4} of the atoms are excited into the $5s[^3/_2]_2$ level via electron impact

excitation. The thermal (300°C) atoms are then transversely cooled, decelerated with the Zeeman slowing technique, and loaded into a MOT. Atoms remain trapped for an average of 1.8 sec as the vacuum is maintained at 2×10^{-8} Torr. This trap system can capture the abundant ^{83}Kr atoms at the rate of 2×10^{8} sec^{-1}. The ratio of the capture rate to the injection rate gives a total capture efficiency of 1×10^{-7}.

With expected capture rates between 10^{-3} sec^{-1} and 10^{-2} sec^{-1} for the rare krypton isotopes, the system must be able to detect a single atom in the trap [10]. In the trap, a single atom scatters resonant photons at a rate of ~10^{7} sec^{-1}, of which 1% are collected, spatially filtered to reduce background light, and then focused onto an avalanche photodiode with a photon counting efficiency of 25%. In order to achieve a high capture efficiency and a clean single-atom signal, the setup is switched at 2 Hz between the different parameters optimized for capture and for atom counting. The resulting fluorescence signal of a single atom is 16 kcps (kilo-counts per second) while the background level is 3.4 kcps (Fig. 1).

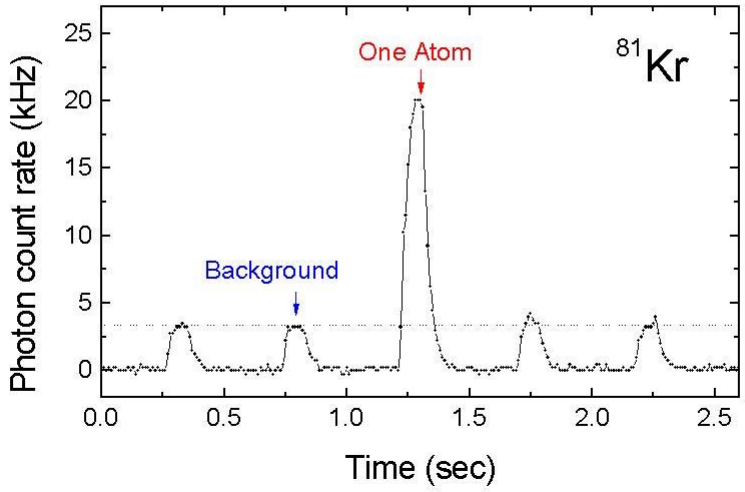

Figure 1. Signal of a single trapped ^{81}Kr atom. The photon counter is only open during the detection phase. Single atom signal ≈ 1600 photon counts, background ≈ 340 photon counts.

We have trapped and counted ^{85}Kr and ^{81}Kr atoms from a natural krypton gas sample. The frequency settings of the trapping laser and the two sidebands are in good agreement with previous spectroscopic measurements obtained using enriched ^{85}Kr gas and enriched ^{81}Kr gas. We have also mapped the atom capture rates versus laser frequency (Fig. 2). Furthermore, repeated tests were performed under conditions in which a ^{85}Kr (^{81}Kr) trap should not work, such as turning off repump sidebands and tuning the laser frequency above resonance. These tests always yielded zero atom counts, which shows that the recorded counts are solely due to laser-trapped ^{85}Kr (^{81}Kr) atoms.

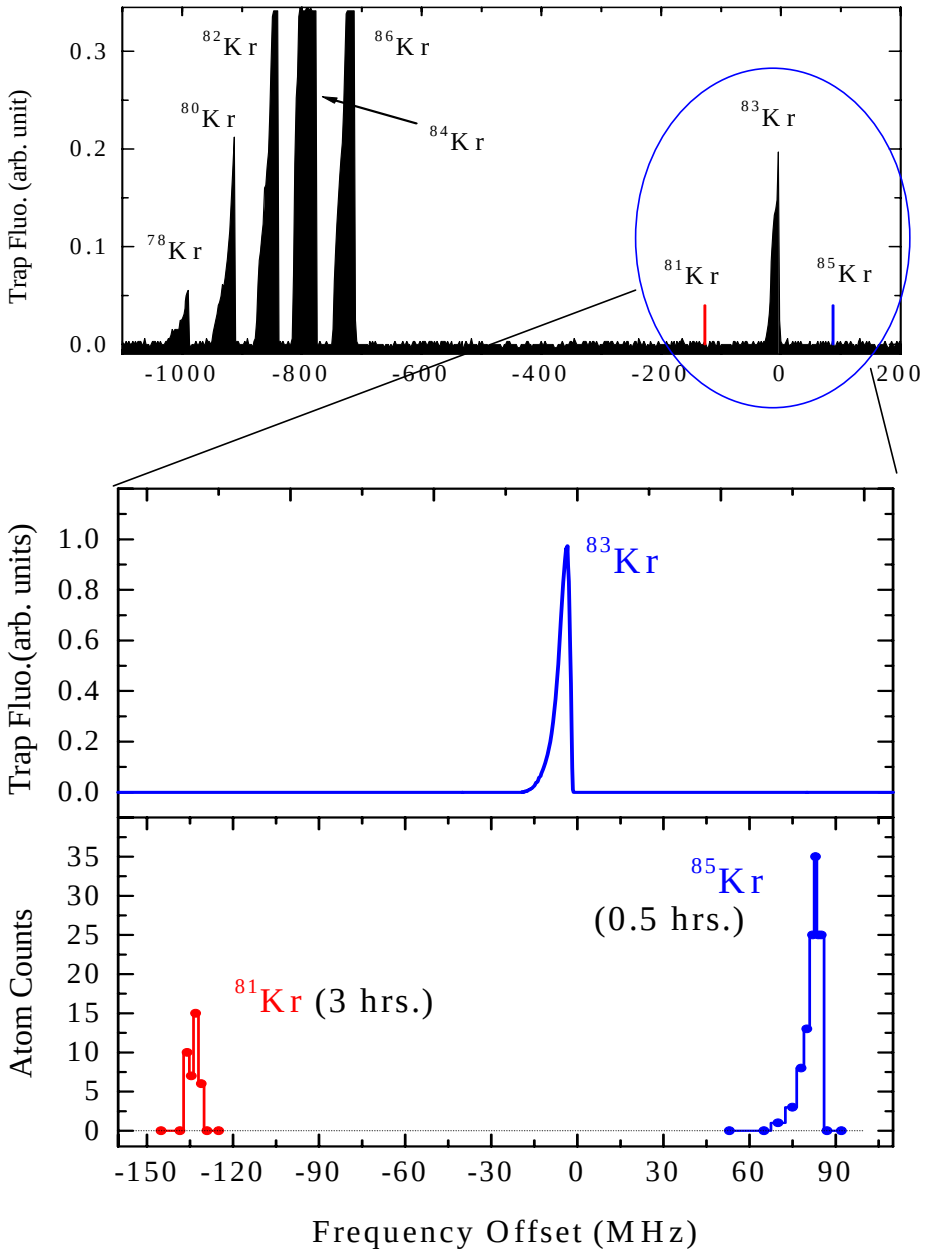

Figure 2. (a) Fluorescence of trapped krypton atoms. Dark bands are the signal of stable isotopes measured with a low-gain photo-diode detector. Line markers mark the positions of the two rare isotopes. (b) Fluorescence of trapped ^{83}Kr atoms versus laser frequency. (c) Number of ^{81}Kr and ^{85}Kr atoms counted versus laser frequency. Each data point represents the number of ^{81}Kr atoms counted in 3 hours, and ^{85}Kr atoms counted in 0.5 hours.

Previous efforts to develop a laser-based technique have encountered serious problems as a result of contamination from nearby abundant isotopes or isobars. ATTA is immune from the contamination for several reasons: fluorescence is only collected in a small region (ϕ 0.5 mm) around the trap center; a trapped atom is cooled to a speed below 1 m/s so that its laser induced fluorescence is virtually Doppler-free; the long observation time (>100 ms) allows the atom to be unambiguously identified (S/N $\approx$ 40); and trapping allows the temporal separation of capture and detection so that both capture efficiency and detection sensitivity can be optimized. Our design also provides additional features, such as chopping off the atomic beam before detecting the trapped atom

The capture rate of our system depends on the discharge current, laser power, and optical alignment. At one particular setting, we measured capture rates of ^{83}Kr, ^{85}Kr, and ^{81}Kr, which were $(1.5\pm0.3)\times10^{8}$ sec^{-1}, $(1.9\pm0.3)\times10^{-2}$ sec^{-1}, $(1.3\pm0.4)\times10^{-3}$ sec^{-1} respectively. If we assume the same detection efficiency for all three isotopes, then we get isotopic abundances of $(1.5\pm0.4)\times10^{-11}$ for ^{85}Kr and $(1.0\pm0.4)\times10^{-12}$ for ^{81}Kr, which are in good agreement with previous measurements performed using other methods. The capture efficiencies can be calibrated with enriched samples of known isotopic abundance to correct for any isotope-dependent effects and measure isotopic ratios in unknown samples. For example, in ^{81}Kr-dating, a known amount of ^{85}Kr can be mixed into the sample, thus allowing the ^{81}Kr abundance be extracted by measuring the ratio of ^{81}Kr / ^{85}Kr.

Our system has achieved an overall efficiency of 1×10^{-7}. Use of this system to measure the abundance of ^{85}Kr to within 10% would require 2 hours and a krypton sample of 3 cm^3 STP while measurement of ^{81}Kr to within 10% would require 2 days and a sample of 60 cm^3 STP. This limits the current system to atmospheric applications where large samples of gas are available. Improvements, such as a liquid-nitrogen cooled discharge source and recirculation of krypton gas, are presently under investigation.

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