Atom trap trace analysis

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A new method of ultrasensitive isotope trace analysis has been developed. This method, based on the technique of laser manipulation of neutral atoms, has been used to count individual ^{85}Kr and ^{81}Kr atoms present in a natural krypton gas sample with isotopic abundances in the range of 10^{-11} and 10^{-13}, respectively. The method is free of contamination from other isotopes and elements, and can be applied to many different isotope tracers for a wide range of applications.

Keywords: ATTA, atom trap trace analysis, magneto-optical trap, ^{85}Kr, ^{81}Kr

We report a new and widely-applicable method, Atom Trap Trace Analysis (ATTA) [1], which promises to enhance the capability and expand the applications of ultrasensitive isotope trace analysis. Our work so far has focused on the long-lived ^{85}Kr and ^{81}Kr isotopes, whose properties are listed in table 1. ^{85}Kr has been used as a general tracer to study air and ocean currents, date shallow groundwater [2], and monitor nuclear-fuel reprocessing activities [3]. ^{81}Kr is an ideal tracer for dating ancient groundwater and ice on the time scale of 10^5–10^6 years [4]. Suitable techniques for the aforementioned applications must be capable of identifying these rare isotopes in a natural sample. Presently, only two techniques, Low-Level Counting (LLC) [5] and Accelerator Mass Spectrometry (AMS) [6], have been able to satisfy this stringent requirement. Laser-based techniques, such as Resonance Ionization Spectrometry (RIS) [7] and Photon Burst Mass Spectrometry [8], have the potential of being simple and efficient. Using RIS, Hurst and coworkers were able to count ^{81}Kr atoms in an enriched sample with over 50% efficiency [9].

The newly-developed ATTA method, based upon laser manipulation of neutral atoms, has been successfully applied to count ^{85}Kr and ^{81}Kr in natural krypton sam-

Table 1
The properties of ^{85}Kr and ^{81}Kr.

	Half-life (yr)	Atmospheric isotope abundance	Main production source
^{85}Kr	10.8	$\sim 1 \times 10^{-11}$ LLC [2]	A fission product of U and Pu
^{81}Kr	2.3×10^5	$(5.9 \pm 0.6) \times 10^{-13}$ LLC [5]	Cosmic-ray induced spallation and
		$(5.3 \pm 1.2) \times 10^{-13}$ AMS [6]	neutron activation of stable Kr

ples. ATTA differs significantly from all previous methods in that it is free of any contamination from different isotopes or elements. Therefore, ATTA can tolerate impure gas samples, and does not require a special operation environment. Moreover, the apparatus can be made transportable for applications that require on-site analysis.

Our design is based on a type of magneto-optical trap system that had been used to trap various metastable noble gas atoms [10]. Trapping krypton atoms in the $1s_5$ metastable level (lifetime = 40 s) is accomplished by exciting the $1s_5$–$2p_9$ transition using laser light with a wavelength of 811 nm generated by a Ti:Sapphire ring laser. Krypton gas is injected into the system through a 0.1 mm diameter nozzle, around which a DC discharge is maintained. About 1×10^{-4} of the atoms are pumped into the $1s_5$ level by the discharge and, after exiting the discharge region, remain in this metastable level until they collide with walls. Two-dimensional transverse cooling is used to reduce the atomic beam divergence and amplify the atom flux in the forward direction by a factor of 20. The thermal (300°C) atoms are then decelerated with the Zeeman slowing technique [11], and loaded into a magneto-optical trap [12]. Atoms remain in the trap for an average of 1.8 s when the vacuum is maintained at 2×10^{-8} Torr. This trap system can capture the abundant ^{83}Kr (isotopic abundance = 11.5%) atoms at the rate of 2×10^8 s^{-1}. The ratio of the capture rate to the injection rate gives a total capture efficiency of 1×10^{-7}. In the trap, a single atom scatters resonant photons at a rate of 10^7 s^{-1}, of which 1% are focused onto an avalanche photodiode. With each trapping laser beam set at 1 cm diameter and 2 mW/cm^2, the fluorescence from a single atom induces a signal of 16 kcps (kilo-counts per second). These laser parameters are significantly different from those needed for an optimum trap loading rate (3 cm diameter, 10 mW/cm^2 intensity). In order to meet the two different requirements, we switch at about 2 Hz between the optimal parameters for loading and for detection, with each phase lasting for about 0.25 s.

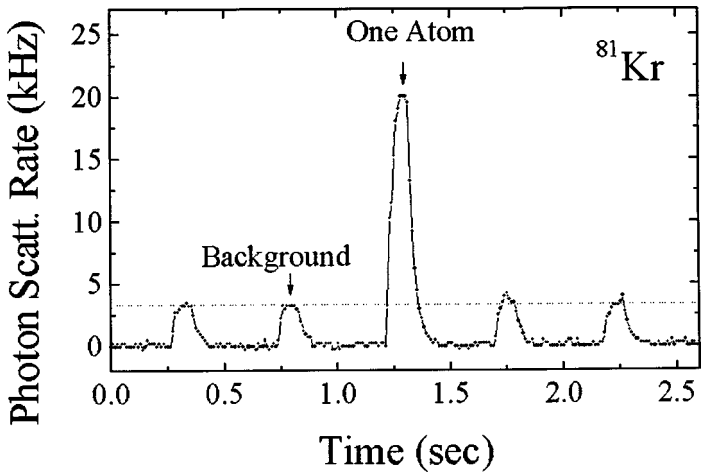

Figure 1. Signal of a single trapped ^{81}Kr atom. During loading time, the photon-count rate was low because the counter was blocked for protection from over-exposure.

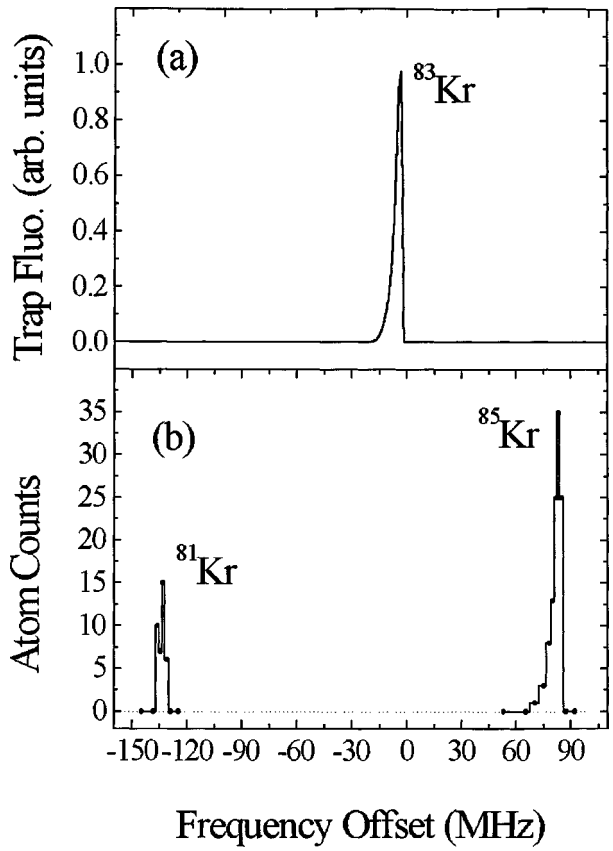

Figure 2. (a) Fluorescence of trapped ^{83}Kr atoms vs. laser frequency. Fluorescence was measured with a low-gain photodiode detector. (b) Number of ^{81}Kr and ^{85}Kr atoms counted vs. laser frequency. Each data point represents the number of atoms counted in 3 h for ^{81}Kr, and 0.5 h for ^{85}Kr.

We have trapped and counted ^{85}Kr and ^{81}Kr atoms from natural krypton gas. Figure 1 shows a typical signal of a trapped ^{81}Kr atom. The frequency settings of the trapping laser are in good agreement with a previous spectroscopic measurement using enriched ^{85}Kr and ^{81}Kr gas [13]. We have also mapped the atom capture rate vs. laser frequency (figure 2), and showed that the maximum loading was achieved with the frequency detuned approximately 4 MHz below resonance. Furthermore, repeated tests were performed under conditions in which a ^{85}Kr or ^{81}Kr trap should not work, and have always yielded zero atom counts. Thus, we have shown that the recorded counts are due to laser-trapped ^{85}Kr and ^{81}Kr atoms, and that no background atom-counts from other isotopes or elements have been observed.

Previous efforts to develop a laser-based technique have encountered serious problems with contamination from nearby abundant isotopes. ATTA is immune from isotope contamination for several reasons: fluorescence is only collected in a small region (0.5 mm diameter) 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; a trapped atom allows a long observation time; and trapping allows the temporal separation of capture and detection so that both capture efficiency and detection sensitivity can be optimized.

At one particular setting, we measured capture rates of ^{83}Kr, ^{85}Kr, and ^{81}Kr, which were $(1.5 \pm 0.3) \times 10^{8}$, $(1.9 \pm 0.3) \times 10^{-2}$ and $(1.3 \pm 0.4) \times 10^{-3}$ s^{-1}, respectively. If we assume the same detection efficiency for all three isotopes, then we get isotopic abundances of $(1.5 \pm 0.4) \times 10^{-11}$ for ^{85}Kr and $(1.0 \pm 0.4) \times 10^{-12}$ for ^{81}Kr, which are in good agreement with previous measurements using other methods [5,6]. In the future, we intend to calibrate the capture efficiencies with enriched samples of known isotopic abundances, and correct for any isotope-dependent effects.

Acknowledgements

We thank Carl Wieman and Walter Kutschera for stimulating discussions. This work is supported by the U.S. Department of Energy, Nuclear Physics Division, and L.Y. by the Office of Basic Energy Sciences, Division of Chemical Sciences, under contract W-31-109-ENG-38.

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