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ATTA – A new method of ultrasensitive isotope trace analysis

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Abstract

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. This method is free of contamination from other isotopes and elements and can be applied to various different isotope tracers for a wide range of applications. The demonstrated detection efficiency is 1×10^{-7}. System improvements could increase the efficiency by many orders of magnitude.

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1. Introduction

We report a new atom-counting method, named atom trap trace analysis (ATTA) [1]. This method is based on laser manipulation of neutral atoms and has been successfully applied to count ^{85}Kr and ^{81}Kr (Table 1) in natural krypton samples. 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, does not require a special operation environment, and its apparatus can be made transportable for applications that require on-site analysis.

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2. Experimental techniques

Our design (Fig. 1) is based on a type of magneto-optical trap (MOT) system that had been used to trap various metastable noble gas atoms [2]. Trapping krypton atoms in the $5s[^3/_2]_2$ metastable level (lifetime $\approx$40 s) is accomplished by exciting the $5s[^3/_2]_2$–$5p[^5/_2]_3$ transition using laser light with a wavelength of 811 nm generated by a Ti–Sapphire ring laser. The laser frequency, after shifting by acousto-optical modulators (AOMs) to account for the isotope shift, is locked to a reference absorption line of the abundant ^{83}Kr atoms in a cell. The krypton gas sample is injected into the system through a 0.1 mm diameter nozzle, around which a dc discharge is maintained. A fraction of about 1×10^{-4} of the atoms are excited into the $5s[^3/_2]_2$ level by the discharge and, after exiting the

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

	Half-life (yr)	Atmospheric isotope abundance	Main production source
^{85}Kr	10.8 [11]	$\sim 1 \times 10^{-11}$ (low-level counting [6])	Nowadays a fission product of U and Pu
^{81}Kr	2.3×10^5 [11]	$(5.9 \pm 0.6) \times 10^{-13}$ (low-level counting [7]) $(5.3 \pm 1.2) \times 10^{-13}$ (AMS [8])	Cosmic ray-induced spallation and neutron activation of stable Kr isotopes

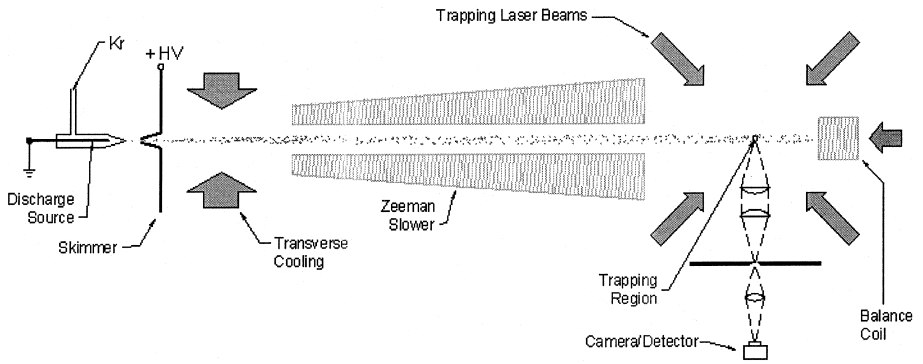

Fig. 1. Schematic layout of the atom beamline. Total length of the apparatus is about 2.5 m.

discharge region, remain in this metastable level until they hit 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 [3], and loaded into an MOT [4]. Atoms remain trapped on 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 into the vacuum system gives a demonstrated detection efficiency of 1×10^{-7}, which results from a combination of metastable production efficiency (estimated value $\geqslant 10^{-4}$), atomic beam divergence loss (estimated value $\geqslant 10^{-3}$), and the fraction of metastable atoms that are captured while passing through the trap (estimated value $\geqslant 10^{-1}$).

In the trap, a single atom scatters resonant photons at a rate of 10^7 s^{-1}, of which 1% are collected, spatially filtered to reduce background light, and then focused onto an avalanche photodiode (EG&G, SPCM-AQ-212) with a specified photon counting efficiency of 25%. For single atom detection, each trapping laser beam is set at 1 cm diameter and 2 mW/cm^2, and the magnetic field gradient of the MOT set at 16 G/cm along its axis. Under these conditions, fluorescence from a single atom induces a signal of 16 kilo-counts per second (kcps) while the background level is 3.4 kcps (Fig. 2). These laser and field parameters differ significantly from those needed for an optimum trap loading rate (3 cm beam diameter, 10 mW/cm^2 and 8 G/cm field gradient). The latter trap loading conditions generate too much background light to permit single atom detection so we switch at about 2 Hz between the optimal parameters for loading and detection.

3. Results and discussion

We have trapped and counted ^{85}Kr and ^{81}Kr atoms from natural krypton gas (Fig. 2). The frequency settings of the trapping laser are in good agreement with previous spectroscopic

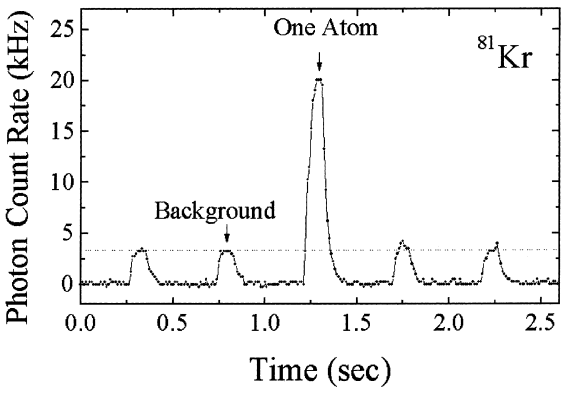

Fig. 2. Signal of a single trapped ^{81}Kr atom. During loading time, the photon count rate was low because the counter was blocked for protection from over-exposure.

measurements obtained using enriched ^{85}Kr gas and enriched ^{81}Kr gas [5]. We mapped the atom capture rate versus laser frequency (Fig. 3), and showed that the maximum loading was achieved with the frequency detuned approximately 4 MHz below resonance, consistent with the results observed on ^{83}Kr traps (Fig. 3(a)). Furthermore, repeated tests were performed under conditions in which a ^{85}Kr (^{81}Kr) trap should not work, such as tuning the laser frequency above resonance, and these tests have always yielded zero atom-counts. These tests show that the recorded counts are due to laser-trapped ^{85}Kr (^{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: the cooling and trapping effect itself is sharply isotope selective; 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, during which 10^3 scattered signal photons are collected; and trapping allows the temporal separation of capture and detection so that both capture efficiency and detection sensitivity can be optimized. With a 100 ms observation period, the signal of a single trapped

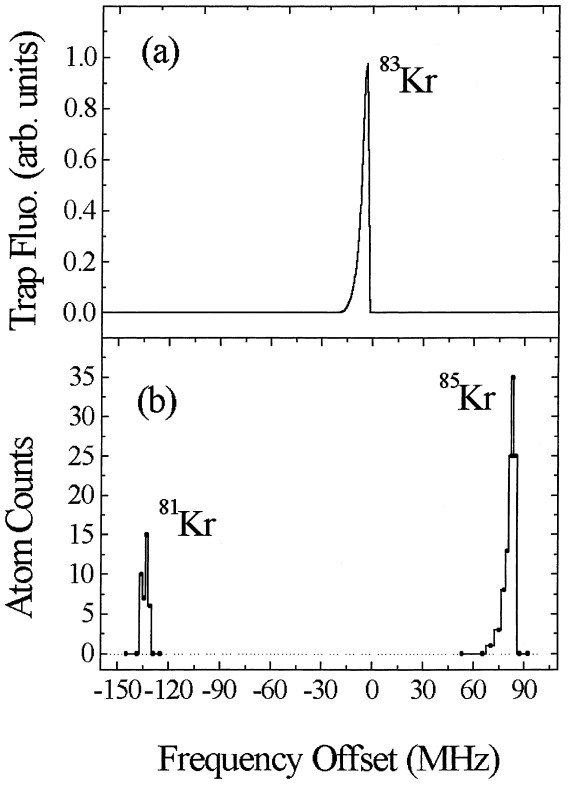

Fig. 3. (a) Fluorescence of trapped ^{83}Kr atoms versus laser frequency. Fluorescence was measured with a low-gain photodiode detector. (b) Number of ^{81}Kr and ^{85}Kr atoms counted versus laser frequency. Each data point represents the number of atoms counted in 3 h for ^{81}Kr, and 0.5 h for ^{85}Kr.

atom is 50 times the noise (1σ) of the background photons.

Besides isotopic selectivity, another important characteristic is detection efficiency. The efficiency of our system depends on the discharge current, laser power, as well as optical alignment. 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 [6–8]. 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.

4. Conclusion and outlook

Our system has achieved an efficiency of 1×10^{-7}, which limits the current system to atmospheric applications where large samples of gas are readily available. It is possible to raise the efficiency by many orders of magnitude through improvements such as cryogenic cooling in the discharge region and re-circulation of krypton gas. Another promising way to improve efficiency is to use an ultraviolet (UV) laser to excite the atoms to the metastable level via a two-photon transition. In one proposed scheme, krypton atoms are excited by a CW laser ($\lambda = 216$ nm) from the ground level to the $5p[^5/_2]_2$ excited level, from which about 10% of these atoms decay into the $5s[^3/_2]_2$ metastable level. We calculated [9] that 5% of the atoms can be transferred to the $5s[^3/_2]_2$ level in an arrangement, where atoms cross an intense laser beam (10 W over 100 μm^2) present at the center of a near-concentric power build-up cavity. Without the constraint from the discharge, the atoms can be collimated and cooled to liquid nitrogen temperature, thus further reducing the loss due to beam divergence. This scheme should improve the detection efficiency to about 10^{-3}. Further improvement is possible if the krypton atoms are recycled back to the source. Another proposal seeks to combine the high efficiency of the "atom buncher" [10] and the high selectivity of ATTA. In this scheme, krypton atoms are collected to a cold spot cooled by liquid helium inside an enclosed cell. A pulse of laser light is directed to the cold spot to release some atoms at low temperature. This is followed by another UV laser pulse to excite the released atoms to the $5s[^3/_2]_2$ level. The metastable atoms of interest are then captured into a trap.

ATTA can be applied to many different isotopes. Laser trapping is well established on alkali, alkali earth and noble gas elements. Trapping of other elements is generally more difficult due to both the complexity of their ground level structures and the lack of suitable UV lasers. Some of these problems could be overcome with future advances in UV laser technology.

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