An Atom Trap Trace Analysis System for Measuring Krypton Contamination in Xenon Dark Matter Detectors

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We have developed an atom trap trace analysis (ATTA) system to measure Kr in Xe at the part per trillion (ppt) level, a prerequisite for the sensitivity achievable with liquid xenon dark matter detectors beyond the current generation. Since Ar and Kr have similar laser cooling wavelengths, the apparatus has been tested with Ar to avoid contamination prior to measuring Xe samples. A radio-frequency (RF) plasma discharge generates a beam of metastable Ar which is optically collimated, slowed, and trapped using standard magneto-optical techniques. We detect the fluorescence of single trapped $^{40}$Ar atoms with a signal to noise ratio of 5. The measured system efficiency of $3 \times 10^{-9}$ for Ar corresponds to an expected Kr in Xe sensitivity at the ppt level.

I. INTRODUCTION

Large volume liquid xenon (LXe) detectors are leading the field of dark matter direct detection with the best sensitivity achieved by the XENON100 experiment. A two-fold improvement in sensitivity is projected by next generation LXe experiments, such as XENON1T, which will use more than three tons of Xe as target. Xe is extracted from the atmosphere with a typical krypton (Kr) contamination at the part per million (ppm) level. The Kr contamination contributes background events through the radioactive isotope $^{85}$Kr, which undergoes $\beta$-decay with a 687 keV end point and 10.8 year half life. This background from $^{85}$Kr strongly limits the detection sensitivity of LXe dark matter detectors. The sensitivity reach of XENON1T requires a Kr/Xe contamination better than 0.5 ppt. Cryogenic distillation is an established technology for Xe purification from Kr at the part per million (ppm) level. However, practical and fast measurements at or below such extremely low levels of contamination are beyond the scope of conventional methods such as low level decay counting or accelerator mass spectroscopy. Hence, we have proposed the development of the atom trap trace analysis (ATTA) system described in this paper.

ATTA systems are based on laser cooling, trapping, and counting of single atoms. The extraordinary selectivity of the ATTA method is a result of the high number of resonant photon-atom interactions that are used to slow and capture an atom. Detection of other atomic or molecular species can be excluded at the 90% confidence level even at parts per quadrillion sensitivity. The photons are generated by a narrow bandwidth laser which is tuned to an isotope-specific optical transition. The setup which we have developed will allow for a rapid and reliable measurement of Kr concentrations in Xe at the ppt level, by trapping the most abundant (57%) isotope $^{84}$Kr. We can then infer the $^{85}$Kr concentration since the isotopic abundance of $^{85}$Kr ($1.5 \times 10^{-11}$) is known. A desired contamination of less than one $^{85}$Kr in $10^{23}$ Xe atoms thus corresponds to a ppt level contamination of $^{84}$Kr in Xe. Ultra-sensitive Kr trace analysis is also used for geological dating and studies of transport processes in the atmosphere, oceans, and groundwater monitoring nuclear-fuel reprocessing activities, and may be of interest for the rare gas industry to guarantee the purity of gases used in a variety of applications.

This paper is organized as follows. We first give a description of our ATTA device, which consists of the laser and vacuum subsystems, the RF discharge source and a single atom detection setup. An image of the full system is shown in Fig. 1. In the following section we experimentally characterize the parameters which determine the overall system efficiency, and demonstrate the ability to detect single atoms with a good signal to noise ratio. Finally, we discuss transitioning from argon (Ar) used for initial system characterization and testing to Kr measurements and show that, assuming the same overall efficiency for Kr as for Ar, ppt sensitivity of $^{84}$Kr detection in Xe is expected in $\sim 1$ hour of measurement time.

II. EXPERIMENTAL APPARATUS

A simplified sketch of the ATTA system is shown in Fig. 2. The light for the atomic beam slowing, transverse cooling, and the magneto-optical trap (MOT) is generated by a semiconductor-based laser system. The gas sample is expanded into an ultra-high vacuum system with a base pressure $< 10^{-9}$ torr for cooling and trapping. During gas flow, the pressure in the source chamber is maintained at 0.6 mtorr, while the pressure in the MOT chamber remains at $10^{-8}$ torr. A beam of metastable atoms is created by passing the injected gas sample through a radio-frequency (RF) discharge region, where inelastic collisions with electrons and ions form a plasma and excite the ground state atoms. The atomic beam is collimated by two-dimensional optical molasses, slowed by radiation pressure with Zeeman tun-
FIG. 1. The atom trap trace analysis (ATTA) system at Columbia University.

FIG. 2. ATTA system schematic. Selected components and laser beam directions (red arrows) are shown. Metastable atoms are generated and cooled by a RF source with an attached cold finger, collimated by three transverse cooling stages, TC1-TC3, decelerated with the Zeeman slowing technique, and captured in the magneto-optical trap (MOT) for detection.

ing, and captured in the MOT. The metastable atoms that are periodically trapped in the MOT are detected and counted by their fluorescence with an avalanche photodiode (APD). For a known Xe gas inflow rate and overall system efficiency, the number of detected metastable $^{84}$Kr* atoms can be used to determine the $^{85}$Kr/Xe ratio. To avoid system contamination, the apparatus has been set up and tested exclusively with $^{40}$Ar.

A. Laser System

As $^{40}$Ar and $^{84}$Kr have no hyperfine structure, they can be cooled and trapped without the use of optical repumping. A semiconductor laser setup supplies the laser beams used for the transverse cooling stages, Zeeman slowing, and MOT, as shown in Fig. 3. The external cavity diode laser (ECDL) with an output power of 50 mW is custom-built in the Littman configuration (diode: Sacher SAL-0840-060). The laser output is amplified by a tapered amplifier (Eagleyard EYP-TPL-0808-01000) and delivered to the optical table via a high power polarization maintaining optical fiber.

Approximately 20 mW of the laser power after the fiber is split off for the locking system. We use the Doppler-free heterodyne spectroscopy technique to lock the ECDL to the required wavelength. For this purpose, a strong pump beam is detuned $+160$ MHz and amplitude modulated by double passing an acousto-optical modulator (AOM), and a weak probe beam is phase modulated by a resonant electro-optical modulator (EOM). A lock-in amplifier processes the signal detected by a photodiode. This provides the error signal for a piezo driver controlling the ECDL grating. The emission wavelengths needed for the closed cooling transitions are $811.7542$ nm for $^{40}$Ar* ($4^3P_2 - 4^3D_3$) and $811.5132$ nm for $^{84}$Kr* ($5^3P_2 - 5^3D_3$). The natural linewidths of both atomic transitions are $\sim 2\pi \times 6$ MHz.

The main portion of the laser power after the fiber is used for slowing, cooling and trapping. An AOM shifts the frequency of the laser by $+74$ MHz, and this first order beam is used for magneto-optical trapping. The unchanged zero-order beam is shifted by $+74$ MHz with the second AOM and is used for the transverse cooling stages. The unshifted beam double-passes a $-115$ MHz detuned AOM for an effective detuning of $-310$ MHz. This beam propagates along the Zeeman slower axis towards the source, and slows the atomic beam.

The optical coupling into the tapered amplifier and fiber optics introduces laser intensity fluctuations, which limit the signal to noise ratio of single atom detection. To minimize these fluctuations, a custom-made proportional-integral-derivative (PID) control circuit with a photodiode stabilizes the laser intensity of the trapping beams by feedback to the MOT AOM.

B. Vacuum System

The vacuum system consists of the following main components: sample reservoir, source chamber, three transverse cooling chambers, and detection chamber. A simplified sketch is shown in Fig. 4. Three turbopumps, backed by a single turbopump station with a dry diaphragm pump, differentially pump the system. A separate turbopump station evacuates the reservoir chamber. A manual all-metal ultra-fine leak valve separates the reservoir chamber from the source chamber, and allows for the pressure in the reservoir chamber to be maintained at about 0.6 mtorr during operation. A gate valve separates the first and second transverse cooling chambers. Pneumatic valves on the outputs of the three main turbopumps allow for breaking vacuum to the backing pumping station while maintaining vacuum in the system, and prevent the loss of vacuum in case of a power failure. Pirani and cold-cathode gauges monitor the base pressure in the reservoir chamber, source chamber, second transverse cooling chamber, and detection chamber. The base pressure is maintained at $< 10^{-8}$ torr in all
chambers.

During measurement, part of a gas sample is transferred to the reservoir chamber, which has a volume of 0.63 l. Typical sample volumes are ~1 l STP. A pressure transducer (with a quoted accuracy of 0.1% of full scale) attached to the reservoir is used to measure the total sample size and gas consumption rate. A capacitive manometer is used to monitor the pressure in the source chamber with a quoted accuracy of 0.5%, as the discharge efficiency is relatively sensitive to the source pressure. The inner surface of the detection chamber is painted black using ultra-high vacuum compatible paint (AZ Technology, MLS-85SB) to minimize scattering of the cooling and trapping beams. A residual gas analyzer connected to the detection chamber is used to monitor the vacuum composition.

C. RF Discharge Source

A beam of atoms is generated by expanding the gas sample into the vacuum system. Efficient ultraviolet laser sources to drive noble gas cooling transitions from the ground state are currently not available, hence metastable atoms with optically accessible transitions have to be generated. A RF discharge source ignites and sustains a low-pressure plasma in the presence of the gas flow. Inelastic collisions with the plasma electrons and ions excite a fraction of the effusing atoms into the long-lived metastable state.\textsuperscript{10}

A schematic of the source is shown in Fig. 5. The design parameters are chosen based on Refs.\textsuperscript{10,11} The gas flows through an electrically insulating and thermally conductive tube made of aluminium nitride (AlN). A Pt100 temperature sensor is mounted directly on the AlN tube. The brass shield is 11.1 cm long with an inner diameter of 6.35 cm. The copper coil is 7.6 cm long with 16 windings, made from 18 American wire gauge (AWG) bare copper wire, and has an inner diameter of 3.4 cm. Holes were drilled through the brass shield through which the ground and RF power wire are connected. The flexible coil is soldered to the shield at one end, and a Macor ceramic spacer keeps it centered. A copper cold finger connected to a pulse tube refrigerator (Iwatani PDC08) is used to cool the gas to ~ 160 K. This increases the fraction of Kr atoms below the capture velocity of the Zeeman slower by more than a factor of three. Cryogenic high-vacuum compatible grease on the connections between the copper pieces increases the cooling efficiency.

The RF signal is generated by a voltage controlled oscillator. It passes through a voltage controlled variable attenuator, and is subsequently amplified by an RF amplifier (Mini-Circuits ZHL-50W-52-S). The amplified signal is fed into the source by a SubMiniature version A (SMA) cable feedline. Care has to be taken to match the impedances of the amplifier and the source to maximize the fraction of the RF power deposited into the plasma, as well as to minimize heating.\textsuperscript{12}

To ignite the plasma source, the RF power is first increased until a bright discharge is visible through the rear viewport, then the power is reduced. We observe hysteresis between the low-brightness mode at ignition and lower RF powers, and the high brightness mode at higher RF powers.\textsuperscript{13} The metastable atom flux is significantly increased when the source operates in the high-brightness mode; running the discharge at the minimal power required for this mode reduces heating. Typical operating

FIG. 3. Solid-state laser system schematic. An external cavity diode laser (ECDL) is locked to a gas reference cell using saturated absorption spectroscopy. After amplification the laser light is distributed to the transverse cooling (TC) stages, magneto-optical trap (MOT), and Zeeman slower (ZS) with the appropriate intensities, polarizations, and frequency detunings. AOM: acousto-optical modulator; EOM: electro-optical modulator.

FIG. 4. Vacuum system schematic. The reservoir chamber is connected to the source chamber via an ultra-fine leak valve. Differential pumping stages ensure a sufficient pressure gradient between source chamber, transverse cooling chambers (TC1, TC2, TC3), and magneto-optical trap (MOT). The vacuum is characterized by a pressure transducer (D), capacitive manometer (C), combined Pirani/cold cathode gauges (PIG), and a residual gas analyzer (RGA).
FIG. 5. Cross-section of the RF source. The gas expands through an AlN tube which is cooled by a cold finger. A plasma is generated by an RF signal applied to the copper coil, producing metastable atoms. Temperature monitored by the Pt100 resistive platinum temperature sensor. 

parameters are 15 W of applied RF power at 118 MHz, at a pressure of 0.6 mtorr and temperature of $-110^\circ$C.

D. Single Atom Detection Setup

The low contamination level of Xe by Kr after cryogenic distillation leads to an expected average MOT population of less than one atom. Thus, the system must have the ability to efficiently detect single trapped atoms.

A sketch of the single atom detection setup is shown in Fig. 6(a). We employ six anti-reflection coated lenses between the MOT and detector to gather and focus the fluorescence photons. The first three lenses, used for light collection and collimation, are mounted in vacuum. The other three lenses are mounted outside the vacuum in a lens tube. The 150 $\mu$m pinhole acts as a spatial filter, and a bandpass filter rejects background light. A 810$\pm$5 nm band-pass filter (BP) is used to remove ambient light entering the lens tube. The fluorescence photons are detected by an avalanche photodiode (APD, Perkin-Elmer single photon counting module SPCM-AQRH-12) mounted on a translation stage. Both the lens tube and APD are mounted in a light-tight enclosure. We employ a microcontroller failsafe to protect the APD from overexposure, using the Arduino open-source electronic prototyping platform. The failsafe also relays the TTL photon count signals of the APD to a DAQ card (National Instruments PCI Express 6321) connected to a computer. Software records the count frequency.

A single atom trapped in the MOT scatters approximately $10^7$ photons per second into the full solid angle of $4\pi$ sr. The effective solid angle of our setup for light collection is 1.8%, as determined by ray-tracing simulations taking into account the 180 $\mu$m$^2$ detector area. Including reflection losses, detector efficiency and transmittance of the BP (0.58), the ideal fluorescence signal without spatial filtering is 31 kHz per atom.

III. CHARACTERIZATION OF THE INSTRUMENT

A. System Efficiency

To estimate the consumption rate, the reservoir is filled with gas which is then allowed to flow out through the ultra-fine leak valve. The pressure decrease in the reservoir is measured while maintaining the source chamber pressure at 0.6 mtorr. Using the ideal gas law, we esti-
mate the consumption rate to be $6 \times 10^{16}$ atoms/s.

To characterize the production of metastables by the RF source, a fluorescence laser was set perpendicular to the atomic beam direction in the first transverse cooling region. The detuning of the laser beam was varied and the fluorescence light of atoms of different transverse velocity classes was measured with a CCD camera. As the AIN tube inner diameter is small compared to the longitudinal distance of 8.53 cm from the source exit, the most probable longitudinal velocity of the Ar atoms could be estimated from geometry to be 283 m/s, corresponding to a temperature of 129 K. Since the design capture velocity of our Zeeman slower is 245 m/s for Kr and 250 m/s for Ar, source cooling increases the capture fractions from 18% for Kr and 6% for Ar (at 400 K), to 68% and 33%, respectively. A metastable atom flux of $6 \times 10^{11}$ atoms/s with an angular flux density of $9 \times 10^{13}$ s$^{-1}$ sr$^{-1}$ is generated. Ar atoms are excited by the RF discharge source with an efficiency of $\sim 10^{-5}$.

The first transverse cooling stage increases the atom number captured by the MOT by a factor of $20$, the second transverse cooling by a factor of $2$. The rapid transverse expansion of the slowed atom beam in the 4.5 cm after the Zeeman slower exit reduces the efficiency of the 3rd transverse cooling. With only the horizontal beam of the 3rd transverse cooling stage used, a 20% increase in the trapped atom number can be observed. Optimizing all experimental parameters while preserving a small trapping beam detuning and high MOT field gradient, allows for $10^7$ atoms to be trapped at a time. Switching off the Zeeman slower field reduces the atom number in the MOT to $< 10^3$.

To determine the loading rate of the MOT, a CCD camera is set up at one of the viewports of the MOT chamber to detect the fluorescence of the atom cloud. On another viewport, a photodiode is mounted to provide increased temporal resolution. The brightness of the CCD image and the photocurrent generated by the photodiode are converted to the number of photons emitted from the trapped atoms. From this, the MOT size and number of trapped atoms, $N(t)$, are derived. The trap parameters are modeled by

$$\frac{dN(t)}{dt} = L - \frac{N(t)}{\tau} - \beta n N(t)$$

with loading rate $L$, quadratic collision term $\beta$, MOT lifetime $\tau$, and constant density $n$. Fitting Eq.(1) for trap loading and decay measurements, we determined a loading rate of $2 \times 10^9$ atoms/s. Given our consumption rate, the overall efficiency for our setup is $3 \times 10^{-9}$ for $^{40}$Ar*. The expected efficiency for $^{84}$Kr* is $6 \times 10^{-9}$.

Table I summarizes the performance data of the complete system when tested with $^{40}$Ar*, as well as the loading rate enhancement factors due to the transverse cooling stages.

| Parameter                        | Value |
|----------------------------------|-------|
| gas consumption rate             | $6 \times 10^{16}$ atoms/s |
| metastable atom flux             | $6 \times 10^{13}$ atoms/s |
| angular flux density             | $9 \times 10^{13}$ atoms/s/sr |
| source efficiency                | $10^{-5}$ |
| MOT loading rate                 | $2 \times 10^8$ atoms/s |
| overall efficiency               | $3 \times 10^{-9}$ |
| flux enhancement: TC1: $20\times$, TC2: $2\times$, TC3 (horizontal): $1.2\times$ |

**Table I. Performance data of the ATTA device for $^{40}$Ar*.**

**B. Single Atom Detection**

To demonstrate fluorescence detection with single atom resolution, the efficiency of the setup was reduced by blocking the Zeeman slower laser beam as well as the transverse cooling light. Thus, only a few atoms at a time are captured in the MOT. Figures 6 (b,c) show a plot of the APD count rate over 50 seconds with an integration time of 60 ms, as well as the associated histogram. Discrete steps of 7.4(1) kHz are visible in the photon count rate. These discrete signal levels correspond to zero, two and three trapped atoms in the MOT, the single atom signal to noise ratio is 5. The atomic beam fluorescence and stray reflections of the trapping laser beams contribute a background of 22 kHz, while the dark count of the APD module is negligible at 0.3 kHz. The additional Zeeman slower laser beam contribution to the APD background during the Kr measurements should be compensated by the absence of the atomic beam fluorescence. The average lifetime of a single metastable atom trapped in the MOT is 0.71(1) seconds. The MOT magnetic field as well as the detuning of the trapping beams were not changed for detection, while the trapping beam intensity was reduced by a ND filter with a transmission of 0.2. Using the full trapping beam intensity, while increasing the loading rate, also increases the APD background significantly. We therefore will implement switching of the beam intensity between a loading and detection mode.

**IV. DISCUSSION**

We have characterized and optimized the Kr in Xe ATTA system using Ar, to avoid contamination by Kr. To the best of our knowledge, the role of Xe as carrier gas in the production of metastable Kr atoms in a RF discharge has not yet been investigated. However, systematic study using Ne, Kr and Xe as carrier gases for Ar has been conducted, where Xe yielded the optimal fractional metastable atom population of $2 \times 10^{-4}$. Assuming similar source efficiency and loading rates for $^{40}$Ar* and $^{84}$Kr*, and taking into account that the Zeeman slower design is optimized for $^{84}$Kr*, we expect to detect 0.8 $^{84}$Kr* atoms per hour at a ppt Kr contamination level in Xe. Raising the efficiency of the transverse cooling stages by increasing the available laser power as well as installation of a two-dimensional MOT in the second transverse
cooling zone should further improve the counting rate, and therefore system sensitivity.

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