Normalization of the single atom counting rate in an atom trap

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The single atom counting rate of a rare isotope and the loading rate of another stable isotope with an abundance over 10 orders of magnitude larger are measured in one atom trap. The linear correlation between the measured counting/loading rates is examined to determine the ^{84}Kr/^{82}Kr and ^{85}Kr/^{83}Kr ratios of a Kr gas sample. Experiments show that the relative uncertainty is reduced to 1.3% when the single atom counting rate of ^{85}Kr is normalized by the measured ^{83}Kr loading rate. The measurement of the normalized single atom counting rate can be used to determine extremely low (10^{-16}–10^{-11}) isotope abundance. This normalization method is robust and can also be applied in other atomic systems.

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Because of the unique chemical and physical properties of noble gases, radioactive isotopes ^{85}Kr ($t_{1/2} = 10.8y$), ^{39}Ar ($269y$), and ^{81}Kr ($2.29 \times 10^5 y$) are ideal tracers for environmental samples like groundwater or ices [1,2] (and references therein). Extremely sensitive detection of ^{85}Kr and ^{39}Ar is also important in dark matter detection experiments where liquid xenon [3] or liquid argon [4] detectors are used. Trace ^{85}Kr or ^{39}Ar in the liquid detectors will present serious background noise, which must be eliminated. Atom trap trace analysis (ATTA) [5] is an emerging technique able to measure extremely low isotopic abundance (parts in a trillion or less) through selectively trapping rare isotopes by laser cooling. ATTA measurement of ^{81}Kr has been applied in dating one-million-year old groundwater [6], and recent progress has reduced the necessary krypton sample size from 100 μL (standard temperature and pressure) to about 5–10 μL [1]. ATTA detection of ^{39}Ar has also been demonstrated [7] though the efficiency still needs to be improved for practical applications.

The single atom counting rate of a particular isotope is measured in ATTA to derive its abundance in the sample. Because the trapping efficiency slowly drifts over time due to various changes of the experimental conditions, the loading rate of a control isotope needs to be measured for normalization. ^{85}Kr has been used as the control isotope in the measurement of ^{81}Kr [8]. Since the natural abundances of ^{81}Kr and ^{85}Kr are close, the single atom counting rates of both rare isotopes can be measured to derive the ^{81}Kr/^{85}Kr ratio. Because ^{85}Kr is completely depleted in old samples, before the ATTA measurement of ^{81}Kr/Kr [6], the sample must be mixed with a control gas in which the ^{85}Kr/Kr abundance is known or has been measured by low level counting [9]. However, this complicates the sample preparation and may introduce additional errors. This method can not be applied for the ^{39}Ar/Ar measurement since there is no other rare Ar isotope with an abundance close to ^{39}Ar.

Hence it is desirable to use a stable isotope as a control isotope for normalization. However, because the difference in abundance between stable and radioactive isotopes is so big, the atom counting technique can not be applied for stable isotopes, and it is not trivial to measure the loading rate of stable isotopes with sufficient precision (<5%). Recently, Jiang *et al.*, measured the loading rate of trapped ^{83}Kr (natural abundance 11.5%) through the quenching of the trapped atoms. Isotope abundance measurements of ^{85}Kr and ^{81}Kr with an uncertainty of 7–10% has been demonstrated [1]. However, the method is sensitive to the intensity and alignment of the quenching laser. Therefore, great efforts have been made to stabilize them during the experiment, and the instrument needs to be frequently calibrated with a standard sample. Here we present a different approach to directly measure the loading rate of the stable isotopes using a "quench-and-capture" process. The reliability of this method is tested by measurements of the ^{82}Kr/^{84}Kr and ^{85}Kr/^{83}Kr ratios of a Kr gas sample. It provides an effective way to determine the trapping efficiency and can be generally applied in other atomic traps.

The cold krypton atom trap apparatus is based on the one that has been presented in [10]. In brief, metastable Kr* atoms ($5s[3/2]_2$) are produced through radio frequency discharge in a liquid-N_2 cooled ceramic tube. The Kr* atomic beam is transversely cooled by a 811 nm laser beam and then slightly focused in a two dimensional magneto-optic trap (2D-MOT). The Kr* beam flux is about 10^{16} atoms/s/sr in the trapping chamber which is 2 m downstream from the discharge. Kr* atoms are slowed down in a Zeeman slower and then trapped in a MOT in the trapping chamber. Typically, about 10^8 cold Kr atoms can be trapped simultaneously in the center of the MOT.

For rare isotopes (^{81}Kr or ^{85}Kr), due to their extremely low abundance (10^{-12}–10^{-11}), only individual atoms will be trapped. Fluorescence of a single atom captured in the MOT is collected and imaged on an electron-multiplying charge-coupled device (EMCCD). The insets of Fig. 1 illustrate the recorded images with and without a ^{85}Kr atom in the trap. The fluorescence signal within the region of interest where the single atom may appear is integrated, a sample of which is shown in Fig. 1. The presence of a single ^{85}Kr atom is characterized by a step signal above the background. Statistics of the single atom signal shows that the signal-to-noise ratio is about 15.

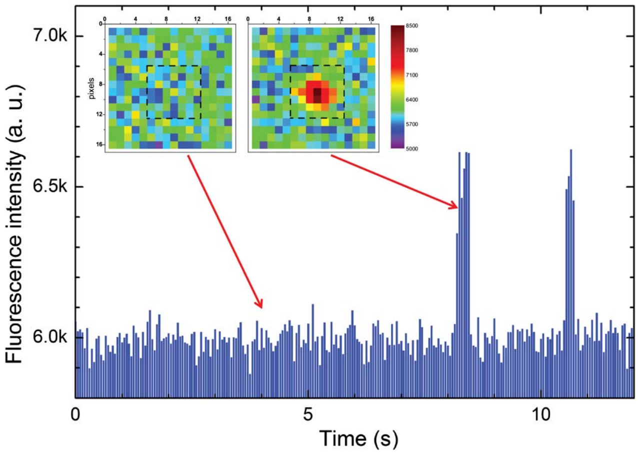

Fig. 1. (Color online) Signal from a single ^{85}Kr atom captured in the MOT. The insets show the images of the trap with and without a ^{85}Kr atom.

The single atom counting rate drifts because of miscellaneous fluctuations in the discharge, laser power and beam quality, etc. It therefore needs to be normalized with the measurement of the loading rate of a stable isotope with a known abundance such as ^{83}Kr. However, the loading rate of ^{83}Kr is 10 orders of magnitude larger than that of ^{85}Kr, and thus must be measured differently. When the ^{83}Kr atoms are trapped, the number of trapped atoms grows very fast and soon reaches a balance between the trap loading and the loss due to collisions from the background gas in the vacuum and other trapped atoms. Such process can be described with a simplified equation:

$$dN/dt = L - \alpha N - \beta N^2, \quad (1)$$

where N is the number of trapped atoms, L is the loading rate of the MOT, α is the coefficient of the collision loss due to background gas, and β is the effective coefficient of the trap loss due to collisions among trapped atoms. The collision coefficients vary with the background pressure, the size and profile of the cold atom cloud, laser beam shape, etc., and are thus difficult to measure accurately. By measuring the fluorescence signal from the ^{83}Kr atoms in our trap, we estimate that the loading rate of cold ^{83}Kr is typically 10^{10} s^{-1}, the number of atoms N in equilibrium (when $dN/dt = 0$) is on the order of 10^8, and the lifetime of trapped atoms is a few hundred milliseconds. It indicates that the loss rate due to background collision (αN) is less than a few percent of L. In this case, within the first few milliseconds after the trap has been turned "on", the rate of the number of trapped atoms, dN/dt, is approximately equal to L while the loss from collisions is negligible.

Therefore, the loading rate L of the trap can be derived from the slope of $N(t)$ in the first few milliseconds. However, it is not easy to turn "on" the trap sufficiently quickly. A simple way to turn the trap on and off is to chop the trapping laser (811 nm) in the MOT. The laser beam can be switched off within a few microseconds using an acousto-optic modulator (AOM). But in order to maximize the loading rate in ATTA measurements, the beam waist of the trapping laser must be over 2 cm. It takes milliseconds for an atom to arrive at the center of the MOT from the edge of the trapping region. This delay prevents us from measuring the fast build up of the cold atoms in the MOT.

In order to turn the trap on/off quickly, in addition to the 811 nm trapping laser which is kept on, a 810 nm "quench" laser with a beam diameter of 0.5 mm is used, aligned perpendicular to the atomic beam and overlapping with the cold atomic cloud. According to the diagram of the energy levels shown in Fig. 2, the 810 nm laser will transfer the Kr atoms from the metastable $5s[3/2]_2$ level to the $5p[5/2]_2$ level. Through spontaneous decay, the Kr atoms will go to the $5s[3/2]_1$ state and then to the ground state $^1S_0$ by emitting a 877 nm photon and a 124 nm photon. When the "quench" laser is on, no cold Kr atoms can be accumulated in the MOT. When it is turned off, which is realized within a few microsecond using an AOM, the cold atomic cloud will build up immediately in the MOT. The number of trapped cold Kr atoms can be detected by measuring the scattering fluorescence from the cloud using a photo diode. As shown in Fig. 2, the fluorescence signal from the trapped atoms increases first linearly, and when more atoms are trapped, the loss due to collisions increases and then the number of trapped atoms reaches an equilibrium. The loading rate of cold Kr atoms can be derived from a linear fit of the rising curve in the first few milliseconds.

The quantitative capability of the "quench-and-capture" method was first verified by measuring the trap loading rates of two stable isotopes ^{84}Kr and ^{82}Kr, which should be proportional to respective isotope abundances (56.99% and 11.59%). A series of measurements have been carried out under different experimental conditions with the same Kr gas sample (Nanjing Special Gas Inc.). In each measurement, the loading rates of both isotopes were measured. We switched between two isotopes every few minutes to eliminate the slow drift of the trapping efficiency. The statistical uncertainty of each measured loading rate is about 2%. The results are shown in Fig. 3(a). The averaged ratio of the ^{84}Kr/^{82}Kr loading rates is determined to be 4.87 ± 0.01, and agrees well with the value of 4.92 calculated from the natural

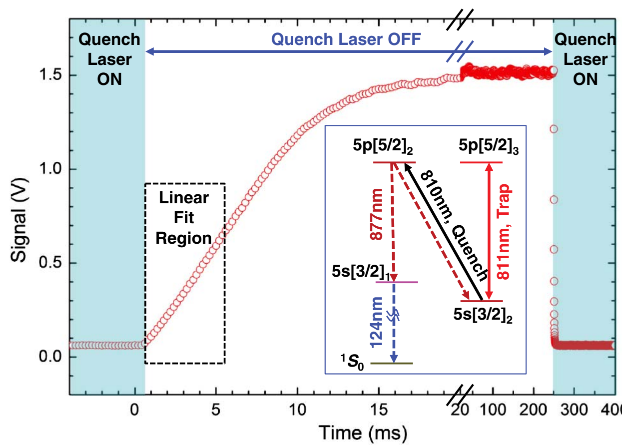

Fig. 2. (Color online) Capture rate measurement using the "quench-and-capture" method. The fluorescence signal of the trapped ^{83}Kr atoms is shown when the quench laser is switched on and off. The inset shows the diagram of the lowest energy levels of Kr.

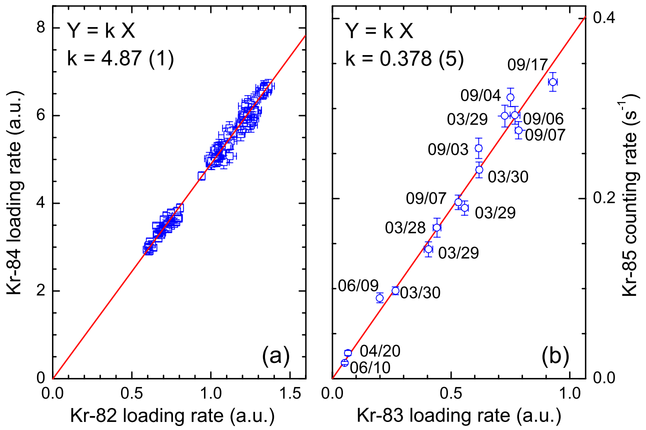

Fig. 3. (Color online) (a) Trap loading rates of ^{84}Kr and ^{82}Kr using a same Kr gas sample and (b) correlation between the ^{85}Kr counting rate and the ^{83}Kr loading rate. The dates given beside the data points show when the measurements were taken.

abundances. The residual systematic difference (1%) may result from imperfect optimization of the trapping laser frequency for either isotope. We also note that the lack of hyperfine structure ($I = 0$) for the even isotopes of the noble gases helps enable a clean comparison and the expected correlation with isotope abundance. It is not easy to compare to the isotopic abundances for those isotopes that have hyperfine structures because the loading rate also depends on hyperfine repumping lasers.

The effectiveness of the atom counting normalization is tested through the measurements of the ^{85}Kr counting rate and the ^{83}Kr loading rate. The ^{85}Kr counting rate was measured by continuous counting of the trapped ^{85}Kr atoms every 2 h and the ^{83}Kr loading rate was measured using the "quench-and-capture" method. In a series of measurements carried out over a period of half a year with the same Kr gas sample, miscellaneous experimental conditions were deliberately changed, leading to a considerable change in the trapping efficiency. The experimental results, together with the date, are shown in Fig. 3(b). The measured ^{85}Kr counting rates and the ^{83}Kr loading rates show excellent linear correlation. The uncertainty of the linear coefficient is about 1.3%, and agrees with the statistical uncertainty estimated from individual measurements.

It is worth noting that the present method is insensitive to variations of the laser power and alignments. Since the ^{83}Kr loading rate is measured when the 810 nm laser is off, the intensity of the quench laser is not critical and does not need to be stabilized. The power of the 811 nm laser used for trapping is well above the saturation intensity and we did not notice any considerable effects due to moderate power fluctuations of the 811 nm laser beam. As shown in Fig. 3(b), the effectiveness of the normalization remains satisfactory over several months. The present normalization method has been applied in the ATTA measurements of trace ^{81}Kr and ^{83}Kr, and may be used for ^{39}Ar counting. It can also be conveniently applied in those atom trap studies when a quantitative trapping efficiency needs to be measured.

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References

1. W. Jiang, K. Bailey, Z. T. Lu, P. Mueller, T. O'Connor, C. F. Cheng, S. M. Hu, R. Purtschert, N. Sturchio, Y. Sun, W. Williams, and G. M. Yang, Geochim. Cosmochim. Acta **91**, 1 (2012).
2. J. Welte, F. Ritterbusch, I. Steinke, M. Henrich, W. Aeschbach-Hertig, and M. K. Oberthaler, New J. Phys. **12**, 065031 (2010).
3. E. Aprile and T. Doke, Rev. Mod. Phys. **82**, 2053 (2010).
4. M. G. Boulay and A. Hime, Astropart. Phys. **25**, 179 (2006).
5. C. Y. Chen, Y. M. Li, K. Bailey, T. P. O'Connor, L. Young, and Z. T. Lu, Science **286**, 1139 (1999).
6. N. C. Sturchio, X. Du, R. Purtschert, B. E. Lehmann, M. Sultan, L. J. Patterson, Z. T. Lu, P. Muller, T. Bigler, K. Bailey, T. P. O'Connor, L. Young, R. Lorenzo, R. Becker, Z. El Alfy, B. El Kaliouby, Y. Dawood, and A. M. A. Abdallah, Geophys. Res. Lett. **31**, L05503 (2004).
7. W. Jiang, W. Williams, K. Bailey, A. M. Davis, S. M. Hu, Z. T. Lu, T. P. O'Connor, R. Purtschert, N. C. Sturchio, Y. R. Sun, and P. Mueller, Phys. Rev. Lett. **106**, 103001 (2011).
8. X. Du, R. Purtschert, K. Bailey, B. E. Lehmann, R. Lorenzo, Z. T. Lu, P. Mueller, T. P. O'Connor, N. C. Sturchio, and L. Young, Geophys. Res. Lett. **30**, 2068 (2003).
9. H. H. Loosli, M. Moll, H. Oeschger, and U. Schotterer, Nucl. Instrum. Meth. Phys. Res. **17**, 402 (1986).
10. C. F. Cheng, W. Jiang, G. M. Yang, Y. R. Sun, H. Pan, Y. Gao, A. W. Liu, and S. M. Hu, Rev. Sci. Instrum. **81**, 123106 (2010).