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METASTABLE KRYPTON BEAM SOURCE VIA TWO-PHOTON PUMPING TECHNIQUE

WILLIE WAI-YEUNG WONG AND LINDA YOUNG

ABSTRACT

Metastable beams of rare gas atoms have wide applications in chemical analysis of samples, as well as in aiding understanding of fundamental processes and physical attributes. Most current sources of metastable rare gas atomic beams, however, are limited in their flux density, which greatly reduces their utility in applications such as low level trace analysis and precision measurements. Previous work has demonstrated feasibility of metastable krypton production via two-photon pumping, and this paper extends that possibility into beam form. Further optimization on this scheme, moreover, promises 100-fold increase of metastable krypton flux density over that of an rf-driven discharge.

INTRODUCTION

Metastable beams of rare gas atoms have a wide range of applications in atom lithography (Berggren, Bard, & al., 1995), cold collision physics (Walhout, Sterr, Orzel, Hoogerland, & Rolston, 1995), and rare isotope detection (Chen et al., 1999). Its use in trace analysis (Chen et al., 1999) is especially promising in identification of samples both chronologically and geologically. Moreover, the same technique can be applied to chemical "branding" of objects with inert substances. Whereas the substance will not react with the object, the high internal energy of the metastable state, once achieved, enables easy detection (Chen et al., 1999). In previous studies, the metastable beams were extracted from the excitation products of a DC (Lu, Hoogerland, Milic, Baldwin, & Buckman, 2001) or rf (Chen et al., 2001) discharge, with a metastable fraction of less than 0.1%. For many applications, especially that of trace analysis, it is desirable to increase the metastable fraction to achieve higher efficiencies (Chen et al., 2001).

In this paper we extend a previously explored optical method (Young, Yang, & Dunford, 2002) to produce a beam of metastable krypton. The metastable atoms, denoted Kr*,

are of the 5s, J=2 metastable state. Contrasted to the previous electron based methods, which are non-discriminatory between different excitation states, the specifically targeted photons produce a higher fraction of the desired Kr*. Because of its odd parity and J=2 angular momentum state, three E1 photons are needed to arrive at the Kr* state from the even, J=0 ground state (Young et al., 2002). Here we use the same excitation pathway as the Young et al. (2002) paper, namely resonant two-photon excitation at 124 nm and 819 nm, with an ensuing 760 nm decay.

MATERIALS AND METHODS

The basic design for the experiment can be seen in figures 1 and 2. The apparatus consists of two stainless-steel vacuum chambers. The first one is the production chamber, and the second one the probe. The two figures represent, respectively, the processes in each of the chambers. A beam of krypton is produced in the primary production chamber by drawing a vacuum in the production chamber and letting in krypton gas through a capillary array affixed to the end of a guiding tube. With a turbo-molecular pump running on the production chamber, we can maintain a steady pressure of 4 mTorr in the chamber with an inlet pressure of 7 Torr. The two values vary approximately linearly in the high pressure regime (2 mTorr and above) and the chamber pressure decreases more rapidly than the inlet pressure in low pressures (up to 1 mTorr). The capillary array has an aspect ratio of one to forty, thereby restricting the angular divergence of the atomic beam to $5x10^{-4}$ sr. By constricting the aperture of the capillary array to 5 mm, we ensured the beam's ability to traverse the distance between the two chambers (approximately 25 cm) without interacting with the walls of the conducting tubes (approximately 7 cm in diameter). The Kr* is induced in the krypton gas through resonant optical pumping.

As seen in figures 1 and 3, we introduce the ultra-violet radiation of 123 nm via a UV resonance lamp. We flow a mixture of 10% Kr and 90% He gas at 0.6 Torr through the

krypton is optically pumped to the 5p J=2 state, there is a 77% branching probability that it will settle into the Kr* state. Detection of the 760 nm fluorescence is accomplished by a focusing lens at 5 cm from the interaction region that projects the fluorescence onto the photodiode which is fitted with a interference filter that passes about 50% of the 760 nm photons. A measurement at 800 nm showed the photodiode to be sensitive to 0.4 mA of current output per 1 mW of input power, or that each picoampere of current corresponds to about 10^7 incident photons per second.

Once the presence of Kr* is established, more precise counting can be accomplished through the 811 nm cyclical absorption-emission of the Kr* as seen in figure 2. In the probe chamber, a 811 nm diode laser crosses the krypton beam. The cyclical transition provides a good indication of the number of Kr* that arrive in the second chamber.



Figure 1: Experimental Setup I: Production of Metastable Krypton Beam

lamp and inject a 65W microwave to excite the gas mixture for ultraviolet emission. The lamp itself has a magnesium fluoride exit window for UV transmittance. The values are, again, from the Young et al. (2002) paper, and adjusted to have maximum value on the XUV detector (seen in figure 1). The 819 nm photons are sent in from a Ti:Sapphire ring laser, operating typically at 300 mW. To reduce noise signals from the various detectors, however, we chop the infrared laser at 1275 Hz, to get an average power of around 150 mW. Using an external lens, we can broaden the laser beam size to 6 mm, comparable to that of the atomic beam size. The beams are sent in normal to the atomic beam path to reduce the effects of Doppler shifting and provide maximal signal at resonance.

The presence of Kr^{*} is inferred from the 760 nm fluorescence emitted when the krypton atom decays from the 5p J=2 state to the metastable state (see figure 3). Once the



Figure 2: Experimental Setup II: Detection and Counting of Metastable Krypton in the Beam

RESULTS

The results of the experiment seem to confirm the feasibility of metastable rare gas atomic beam production through optical methods, yet much work is still needed to surpass the production rate of current rf-discharge methods. At the sensitivity available, we are able to observe a discernable signal down to pressures of 1.5 mTorr. The signal is established by scanning the Ti:Sapphire laser across a 0.007 nm band centered at the resonance frequency of 819.2303 nm. Because of the velocity distribution of the thermal atoms in the beam, it is expected, and observed, to have a Gaussian profile with respect to frequency. The limit observed at 1.5 mTorr is due to the background noise of the instruments, such as the electrical noise associated with the amplifiers,

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Figure 3: The excitation pathway to arrive at the 5s J=2 state. First excite the ground state with an ultra-violet photon to 5s J=1, and excite that within 4ns to 5p J=2 with an infrared photon. Finally, the atom decays into the metastable state by emitting an infrared fluorenscence with 77% branching probability.

and the random photon scatterings originating from the UV lamp. At below 1.5 mTorr, the noise is much larger than the signal, which was ascertained when the sum of repeated data runs approaches a flat line. At the central peak at 1.5 mTorr, we observed a signal size of 1.3 pA. At higher pressures, the increase in signal size is comparable to the UV absorption observed in the Young et al. (2002) paper. For example, at 4.1 mTorr, we observed one of 1.4 pA.

Calculations assuming isotropic emission of the 760 nm fluorescence, zero attenuation of the signal from the Kr* gas present, and zero attenuation from the lens material, as well as an idealized lens showed a reasonable figure. The lens is placed 5 cm from the interaction region, and the aperture on the photodiode is 11.6 mm in diameter, which computes to a 4x10⁻² sr observation window. Factoring in our previously calculated 1pA to 10⁷ photons per second conversion, and the 50% attenuation of the pass filter, we arrive at 10¹⁰ photons per second total as a lower bound. On the other hand, if the lens failed to focus at the infrared wavelength, an upper bound established by that is on the order of 10¹² photons per second. Running in similar conditions as the rf-discharge (Chen et al., 2001), our preliminary, yet to be optimized production rate (4x10¹⁰photons per sec) is already approaching the value of the rf (10^{11}) within an order of magnitude.

DISCUSSION AND CONCLUSION

At 4 mTorr pressure, the thermal atoms still have Doppler profiles in the direction normal to the beam. Yet in this beam form, we collected data that are similar in form to, albeit smaller in value than the data presented in Young et al. (2002). As mentioned earlier, this apparatus is far from optimized. By extrapolation using the data from Young et al. (2002), it is very possible that we can produce Kr atoms at pressures well below 1.5 mTorr; however, we are currently limited by noise from observing the presence of Kr* atoms at lower pressures. The main source of the noise is from the higher energy transitions of krypton gas in the UV lamp, which produces infrared photons in resonance with those we are trying to observe. Because the UV lamp is not a directed source of light, infrared photons have high probability of surviving multiple reflections inside the stainless steel chambers. This might be overcome in future experiments by painting the inside of the chamber black, or if technology permits, the use of UV lasers.

Because of the 50 cm distance between the two chambers, only at pressures below 0.1 mTorr will the quenching interactions between Kr and Kr* yield a mean free path that allows any significant amount of Kr* to survive into the second chamber. At such pressure, the predicted level of signal falls lower than instrument noise, and is not detectable with our current apparatus, so tests in this regime were inconclusive. As the pressure drops, moreover, the transverse thermal velocity becomes smaller, and so do the wings of the Doppler profile. Currently we have a Gaussian whose width is 700 MHz. In low pressures, the resonant frequency will approach a line frequency, thus enabling a frequency specific scan of fine structure and/or isotopic difference. This can enable us to perform rare isotope trace analysis by loading specific metastable isotopes into a magneto optical trap.

Future optimization for the system will first focus mainly on the noise reduction. Besides painting the interaction chamber black, as mentioned above, there is also the possibility of optimizing the UV lamp. In the experiments documented in the paper, the readings on the UV detector indicated an abnormally small absorption of the UV light by the krypton gas in the chamber, as compared with the data in the Young et al. (2002) paper. Also, the net UV production is smaller than expected (Young et al., 2002). It is possible that with our current configuration, the UV producing region of the lamp is not at its best position possible, (Young et al., 2002) such that a significant portion of the UV radiation is absorbed by a cushion of krypton gas near the window of the lamp. All such optimization must be taken into consideration before a fully efficient source of metastable rare gas atoms can be made available.

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REFERENCES

Berggren K K, Bard A, et al (1995). Science 269, 1255.

- Chen C Y, Bailey K, Du X, Li Y M, Lu Z.-T, O'Connor T P, Young L, & Winkler G. (2001) *Rev. Sci. Instrum.*, 72, 271.
- Chen C Y, Li Y M, Bailey K, O'Connor T P, Young L, & Lu Z.-T. (1999) *Science*, *286*, 1139.
- Lu W, Hoogerland M D, Milic D, Baldwin K G H, & Buckman S J. (2001) *Rev. Sci. Instrum.*, 72, 2558.
- Walhout M, Sterr U, Orzel C, Hoogerland M, & Rolston S L. (1995). *Phys. Rev. Lett.*, *74*, 506.
- Young L, Yang D, & Dunford R W. (2002) J. Phys. B, 35, 2985.