The role of carrier gases in the production of metastable argon atoms in a rf discharge

Kenneth Rudinger,[1,2,a)] Zheng-Tian Lu (卢征天),[1,2] and Peter Mueller[1]
[1]*Physics Division, Argonne National Laboratory, Argonne, Illinois 60439, USA*
[2]*Department of Physics and Enrico Fermi Institute, University of Chicago, Chicago, Illinois 60637, USA*

(Received 29 January 2009; accepted 4 March 2009; published online 27 March 2009)

We investigate the role of carrier gases in the production of metastable argon atoms in a rf-driven discharge. The effects of different carrier gases (krypton, xenon, neon, and helium), carrier gas pressures, and rf discharge powers are examined. A xenon carrier gas provides the greatest metastable population of argon, yielding an optimal fractional metastable population of argon $(\mathrm{Ar}^*/\mathrm{Ar})$ of 2×10^{-4} at 0.2 mTorr of xenon gas. The optimal krypton configuration yields 60% of the xenon-supported population at 1.5 times higher pressure. Neon and helium perform considerably worse probably due to their higher ionization potentials. © *2009 American Institute of Physics.*
[DOI: 10.1063/1.3105722]

Due to a lack of readily available vacuum-UV lasers that can excite argon atoms from the ground state (with the lowest-energy transition at 106.7 nm),[1] many spectroscopy experiments probe transitions from its metastable $4s^2[3/2]^\circ$ state. One method to populate this metastable state of argon atoms is to use a rf-driven discharge to excite the gas into a plasma via electron collisions.[2] However, when the argon gas sample size is limited, a different gas species is needed as a carrier gas to sustain the plasma discharge. For example, we aim to perform laser spectroscopy of the radioactive isotope argon 39 (half life=269 yr) of which a typical sample size deliverable from a nuclear reactor is ~5 nL STP obtained through neutron irradiation of a potassium fluoride sample. In this paper, we explore the effects of different carrier gases, carrier gas pressure, and rf discharge power on the fractional metastable population $(\mathrm{Ar}^*/\mathrm{Ar})$ of a small argon gas sample, which is determined using a laser spectroscopy method.

To determine the quantity of interest $(\mathrm{Ar}^*/\mathrm{Ar})$, we probe the metastable atoms by exciting the $4s^2[3/2]^\circ-4p^2[5/2]$ transition with an 811.5 nm laser beam. We note that the transmission of the laser beam passing through the discharge cell of length l containing the metastable argon atoms is given as

$$\frac{S}{S_0} = e^{-l/l_0}, \tag{1}$$

where S_0 is the initial laser power, S is the transmitted power, and l_0 is the characteristic attenuation length, which depends on the known transition properties and is inversely proportional to the density of metastable argon atoms. Taking into account both power broadening and Doppler broadening, we are able to calculate $\mathrm{Ar}^*/\mathrm{Ar}$ based on transmission measurements.[3] However, this simple absorption method is not sensitive to changes in laser attenuation below a few percent. To gain one to two orders of magnitude in sensitivity, FM saturated absorption spectroscopy is performed [as described in Hall *et al.* (1981)] (Ref. 4) and is calibrated against the simple absorption measurement.

The experimental setup is depicted in Fig. 1. The argon sample and a carrier gas are, via leak valves, bled into a discharge cell with a 3 cm diameter and a length of 10 cm. As measured by a residual gas analyzer, the pressure of the argon ranges from 2×10^{-3} to 2×10^{-1} mTorr; the pressure of the carrier gas ranges from 6×10^{-2} to 1.7×10^{1} mTorr. A rf coil around the cell driven at 76 MHz drives a plasma discharge in the cell. The carrier gas provides the density necessary to sustain the plasma. Electron impacts with argon atoms populate the metastable $4s^2[3/2]^\circ$ state. It is the population of this state that we are trying to optimize. Two diode lasers operating at identical wavelengths are used to examine this state. The probe beam is phase modulated at 20 MHz by an electro-optical modulator before passing through the glass cell with an intensity of 5 $\mathrm{mW/cm^2}$. (The transition has a saturation intensity of 1.26 $\mathrm{mW/cm^2}$).[3,5] After passing through the cell, it is focused onto a silicon photodiode. The pump beam with an intensity of 100 $\mathrm{mW/cm^2}$ is frequency shifted by 40 MHz and chopped at 57 kHz by an acousto-optic modulator before passing through the cell antiparallel to the probe beam. The signal from the photodetector is demodulated at 20 MHz by a frequency mixer and at 57 kHz

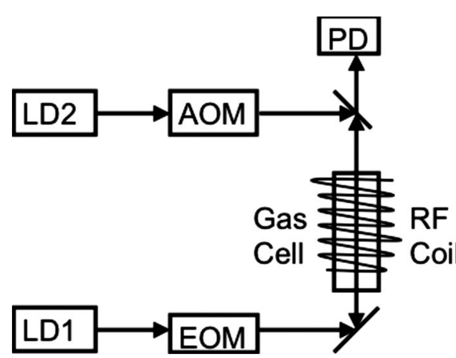

FIG. 1. Diagram of saturated absorption spectroscopy setup. LD, laser diode; EOM, electro-optic modulator; AOM, acousto-optic modulator; PD, photodetector.

a)Electronic mail: rudinger@anl.gov.

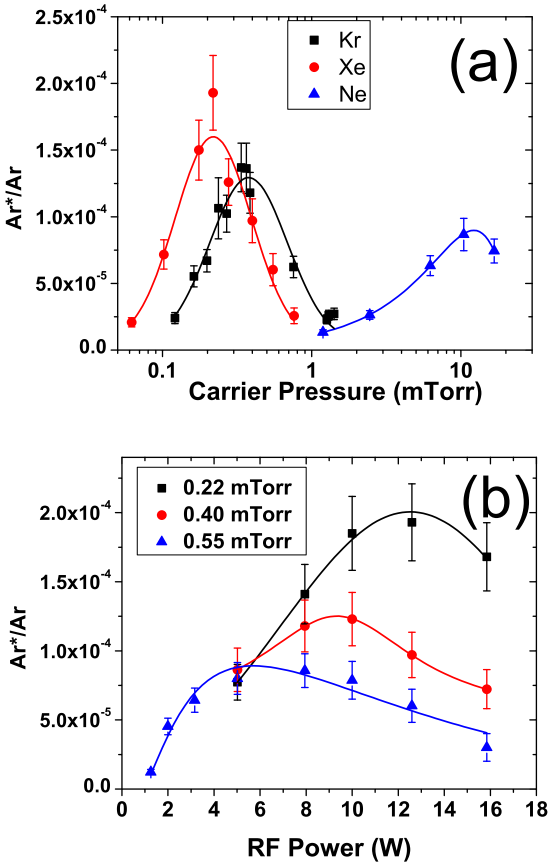

FIG. 2. (Color online) (a) Ar^*/Ar as a function of carrier gas pressure for all three carrier gases with a rf power of 12.6 W. Average argon pressure for krypton, xenon, and neon are 0.026, 0.019, and 0.019 mTorr, respectively. The maximum measurable neon pressure was 20 mTorr due to experimental constraints. (b) Ar^*/Ar as a function of rf power for three different pressures of xenon. Lines are added to guide the eyes.

by a lock-in amplifier; the lock-in output is recorded. Based on this output and the measured partial pressure of argon, we calculate Ar^*/Ar.

Figures 2(a) and 2(b) illustrate our findings. The overall qualitative response of fractional metastable population to carrier gas is similar for each gas, reflecting a balance between "good" (metastable-populating) and "bad" (metastable-depopulating) collisions. As carrier pressure initially increases, metastable population increases. Presumably lower carrier gas pressure provides lower plasma electron density, thereby driving down the good *e*-Ar collision rate. Beyond an optimal pressure (different for each gas), metastable population decreases. Higher carrier gas pressure raises the plasma electron density to the point where the bad *e*-Ar^* and carrier gas-Ar^* dominate over good collisions. Similar behavior is exhibited when pressure is held constant and rf power is varied, indicating that rf power similarly affects collision rates, as increased rf power results in increased electron density and energy. To simply maximize the metastable population, xenon should be used as the carrier gas, as it yields an optimal fractional metastable population of argon of 2×10^{-4} at 0.2 mTorr of xenon gas. The optimal krypton configuration yields 60% of the xenon-supported population at 1.5 times higher pressure. Neon and helium perform considerably worse probably due to their higher ionization potentials. The optimal neon configuration yields 40% of the xenon-supported population at 50 times higher pressure. A significantly smaller metastable population is supported by helium. A more detailed explanation of why each gas has a different optimal pressure as well as sustainable metastable population requires further theoretical investigation.

We thank Kevin Bailey and Thomas O'Connor for technical support and additional group members Cunfeng Cheng, Yun Ding, Brent Graner, Wolfgang Korsch, Ibrahim Sulai, William Trimble, and Reika Yokochi for helpful discussions and general support. This work was supported by the U.S. Department of Energy, Office of Nuclear Physics under Contract No. DE-AC02-06CH11357.

[1] Yu. Ralchenko, A. E. Kramida, J. Reader, and NIST ASD Team. NIST Atomic Spectra Database (version 3.1.5), 2008.
[2] C. Y. Chen, K. Bailey, Y. M. Li, T. P. O'Connor, Z.-T. Lu, X. Du, L. Young, and G. Winkler, Rev. Sci. Instrum. **72**, 271 (2001).
[3] C. J. Foote, *Atomic Physics* (Oxford University Press, New York, 2005).
[4] J. L. Hall, L. Hollberg, T. Baer, and H. G. Robinson, Appl. Phys. Lett. **39**, 680 (1981).
[5] A. A. Radzig and B. M. Smirnov, *Reference Data on Atoms, Molecules and Ions* (Springer, Berlin, New York, 1985).