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Optical production of metastable krypton

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Abstract

We have investigated a new scheme for excitation of the 5s, $J = 2$ metastable level of Kr ($5s[3/2]_{J=2}$) which can be readily extended to other rare gases. In the scheme, an ultraviolet (UV) lamp is used to create a population of Kr atoms in the $5s[3/2]_{J=1}$ level in a gas cell. The excited atoms are then pumped to the $5p[3/2]_{J=2}$ level, using 819 nm light from a Ti:sapphire laser, from which they decay to the metastable state with a branching ratio of 77%. We made two striking observations: (1) the laser power required to saturate the second step decreases markedly as a function of gas cell pressure, and (2) the UV photon flux is converted with very high efficiency ($\approx$10%) to metastable atom flux. A Monte Carlo study of the scattering of UV photons in the cell reproduces the trends observed. The understanding achieved points to the design of a higher flux source of metastable atoms.

1. Introduction

Metastable rare gas atoms have found wide use in fields spawned by the revolution in laser manipulation of atoms, such as cold collision physics [1], optical lattices [2], atom lithography [3], rare isotope detection [4], and, most recently, Bose–Einstein condensation (BEC) [5]. The high internal energy of the metastable atom enables both novel detection schemes in BEC studies [5] and localized surface damage of suitable resists [3], and thus provides a niche beyond similar studies with ground-state atoms. For these studies, the metastable rare gas atoms are generally produced in beam form by direct extraction from a DC [6] or rf [7] discharge. Such methods generally produce beams with a metastable fraction of $<$0.1%. Earlier metastable sources developed for molecular beam studies have used direct electron bombardment of a neutral beam in a crossed [8] or coaxial [9, 10] excitation geometry, with even lower metastable fractions. For many of the applications, it is simply desirable to have an increased metastable beam fraction, but for the single atom counting of rare isotopes [7] where the sample size is limited, it is essential.

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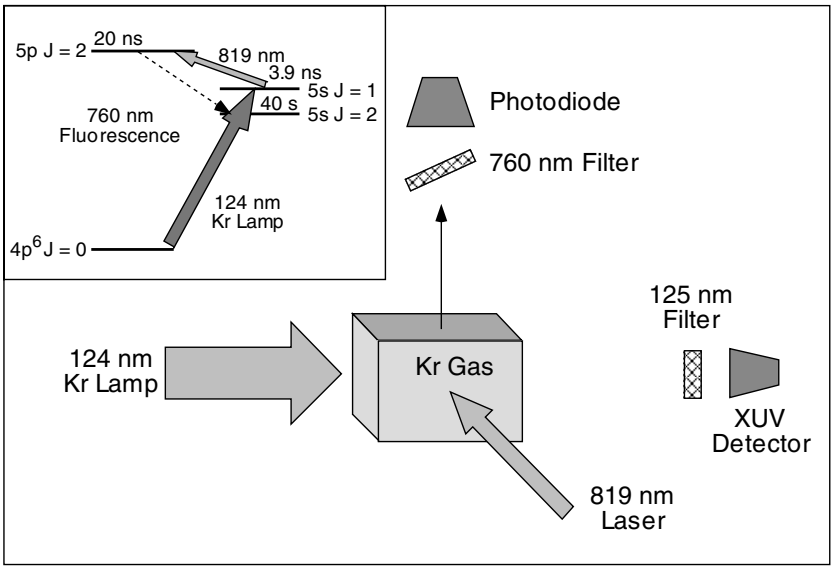

Figure 1. Schematic of the experimental apparatus. The inset shows the energy levels of the Kr atom that are relevant to the experiment. The abbreviated notations 5s ($J = 1$), 5s ($J = 2$) and 5p ($J = 2$) are used here and in the text for the $5s[3/2]_{J=1}$, $5s[3/2]_{J=2}$ metastable and $5p[3/2]_{J=2}$ levels, respectively.

In this paper we investigate an optical method for excitation of the 5s, $J = 2$ metastable level[2] of Kr ($5s[3/2]_{J=2}$) [11]. This level will also be denoted as Kr*. The new method has potential for significantly increasing the Kr* fraction over that available from the non-selective electron-based methods. Because this level has odd parity and angular momentum $J = 2$, three E1 photons are required for excitation from the even parity, $J = 0$ ground state. There are several pathways to the metastable state. The simplest is to use two photons to reach the even parity $5p[3/2]_{J=2}$ level, which will spontaneously decay into the target metastable state as shown in the inset of figure 1. Either non-resonant two-photon excitation [12], or two-photon resonant excitation can be used to reach this level. The non-resonant two-photon excitation requires 215 nm radiation and the excitation efficiency is dependent on the square of the intensity, making cw laser methods quite complex. For resonant two-photon excitation 124 and 819 nm photons are required. The initial 124 nm transition also presents a challenge for laser technology [13]. Thus, using lasers for both steps is undesirable as a starting point of a complex experiment. Instead, we employ a Kr resonance lamp to provide the 124 nm photon for the first step. While VUV rare gas resonance lamps [14] have been used extensively to initiate photochemical reactions, they have not previously been used in this application to our knowledge [15][3].

[2] The notation $nl[K]_J$ refers to j–l coupling where n and l are the principal and orbital quantum numbers, K is the sum of the angular momentum of the core plus the orbital angular momentum of the promoted electron and J is the total angular momentum.

[3] This paper describes the use of a hydrogen resonance lamp to produce a beam of metastable H atoms, with a flux of 10^6 s^{-1}.

2. Experimental details

The experimental apparatus is shown schematically in figure 1. Ultraviolet (UV) radiation from the lamp is incident on a cell of Kr gas where it drives the transition to the 5s ($J = 1$) level. The intensity of the UV photons is measured using a XUV photodiode detector which is fitted with a filter with a peak transmission at 121.8 nm and a width of 12.75 nm. The 819 nm excitation from the 5s ($J = 1$) level to the 5p ($J = 2$) level is provided by a chopped, single-mode, cw Ti:sapphire laser. In the final step of the scheme, the 5p level decays to the metastable 5s ($J = 2$) level with a branching ratio of 77%. The 760 nm fluorescence emitted in this decay provides a signature for the creation of a metastable atom and is monitored through a lock-in amplifier using a photodiode fitted with an interference filter. The filter has a bandpass of about 8 nm and is angle tuned to provide a ≈50% peak transmission at 760 nm for photons incident normal to the detector. This system provides a measure of the rate of production of metastable atoms.

The resonance lamp provides lines at 123.6 and 116.5 nm. It has an exit window made of magnesium fluoride and uses a mixture of 10% Kr and 90% He gas which is continuously flowing. We typically work with 70 W of microwave power and a pressure of 0.8 Torr. These parameters were chosen to optimize the two-photon resonance signal by maximizing the signal from the photodiode (see figure 1). Under these operating conditions the 123.6 nm line is highly self-reversed. This was studied by measuring the intensity of the 123.6 nm radiation surviving transit across the cell as the Kr pressure in the cell was increased. These data could be understood assuming that the spectral distribution of the lamp was such that only Kr atoms in the far wings of the Doppler velocity profile could provide significant absorption. To obtain a quantitative estimate of the degree of self-absorption we developed a simple model for the photon scattering in the cell in an attempt to understand the dependence of the detector signal on the pressure in the cell. The model is based on a parametrization of the properties of a self-absorbing lamp discussed in the review by Cowan and Dieke [16]. By comparing the results of the model with the pressure attenuation data we estimate that there is a 'valley' in the centre of the resonance line from our lamp with a width of about 10 GHz and an amplitude of ≈10% of the peaks. During our experiment, we typically measure a photon flux of 10^{17} photons s^{-1} sr^{-1}, with a FWHM divergence of 0.2 rad to give a total lamp output of $\approx 2 \times 10^{15}$ photons s^{-1}, in reasonable agreement with earlier work [17].

Despite the self-reversal of the UV lamp, strong 760 nm fluorescence signals were readily obtained as a function of laser frequency. Resonances were measured at ≈10 levels of laser power for each gas cell pressure. These were fit to Gaussians, to obtain the peak height and the width for a given laser power. The plot of the peak height (y) versus power (x) saturates and we determine the saturation power by fitting these curves to

$$y = P_1 \frac{x/P_2}{1 + x/P_2}$$

where P_1 is the magnitude of the metastable signal and P_2 is the saturation power for the 819 nm transition at a given UV intensity. Comparing saturation curves at different cell pressures leads to some interesting observations. The first is that, at higher pressures, the curves do not completely saturate; rather, the signal shows a slow linear increase with increased laser power. This effect is believed to be associated with power broadening and an expansion of the effective interaction volume. We are studying this phenomenon but, in the following, we will not consider this very high power regime further. A more striking observation is the large variation of the saturation power, P_2, as a function of pressure which is presented in figure 2(a). Note that there is an order of magnitude drop in the saturation power as the pressure is increased from 0.5 to 10 mTorr. We also observe (see figure 2(b)) that the magnitude of the metastable signal, P_1, increases by a factor of 20 as the cell pressure is increased from 0.1 to 10 mTorr.

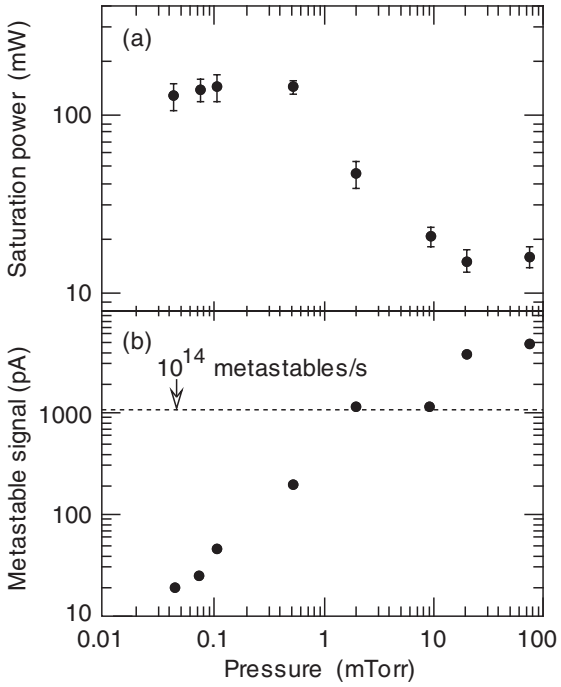

Figure 2. (a) Saturation power as a function of the Kr pressure in the cell. (b) Signal intensity as a function of pressure.

The absolute production rate corresponding to the metastable signal is estimated using the manufacturers' specifications for the detector efficiency and filter transmission. Also, the solid angle collection efficiency was derived from a Monte Carlo simulation of the UV–laser interaction region and detection geometry. Uncertainties in the absolute detection efficiency and the size of the laser–UV interaction region, together with possible systematic errors due to misalignment of the apparatus, mean that our estimate is only good to a factor of two. The resulting scale factor is shown in figure 2(b) where the dashed horizontal line corresponds to a production rate of 10^{14} Kr^* s^{-1}, or a UV-to-metastable conversion efficiency of $\approx 5\%$.

3. Monte Carlo simulation

We have developed a Monte Carlo program which models our experimental situation as a cube of Kr gas ($3 \times 3 \times 3$ cm), with the lamp (1×1 cm) incident on one face and the laser (4×4 mm) crossing perpendicularly at a distance of 1 cm from the face. Our code is based on the work of Jong-Sen Lee [18]. Photons are generated using an assumed frequency and angular distribution at the lamp. We trace each photon's progress through the gas cell until it is absorbed on a wall or is lost because it participates in a laser–UV two-photon absorption ('laser absorption') leading to formation of a metastable Kr atom. In the 23% of decays which do not lead to the formation of Kr^*, the atom cascades to the ground state with emission of an UV photon that continues to scatter. The Kr atoms are characterized by a Doppler velocity distribution and by the distribution of isotopes corresponding to the natural abundance. The energies of the scattered photons are modified by the kinematics of their interactions with the Kr gas atoms. Photon scattering leads to a steady-state population of atoms in the 5s ($J = 1$) level.

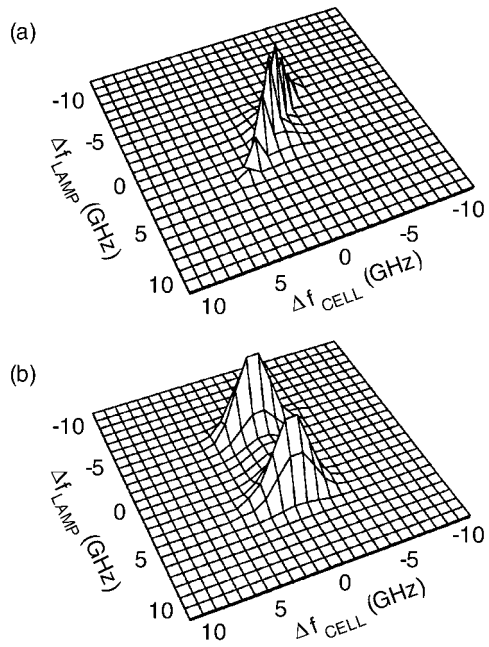

Figure 3. Monte Carlo results giving the correspondence between the offset frequency of a lamp photon $\Delta f_{\mathrm{Lamp}} = f_{\mathrm{Lamp}} - f_0$, to the offset frequency $\Delta f_{\mathrm{Cell}} = f_{\mathrm{Cell}} - f_0$ of a scattered photon. Here, f_0 is the transition frequency from the $5s[3/2]_{J=1}$ level to the ground state. In order to determine the frequency response of the system, the distribution of the photons emitted by the lamp is taken to be flat and ranges from +12 to −12 GHz relative to the resonance frequency. (a) The Kr pressure is 10^{-5} Torr, (b) the Kr pressure is 5×10^{-4} Torr.

As a first step toward gaining an understanding of photon scattering in our cell, we consider an idealized lamp which emits photons with a flat distribution in a frequency extending 12 GHz to either side of resonance. This study was done to determine the frequency response of the system. Later these results were folded with realistic lamp distributions to obtain the final Monte Carlo results. For each scattering event in a small volume near the intersection of the laser and UV light beams we record the energy of the photon emitted by the lamp and the energy of the scattered photon. At low pressure there is no light trapping and so only photons from the lamp that are within the Doppler linewidth of the cell are scattered and the scattered photons have the same energy as the lamp photons. This is illustrated in figure 3(a) which is for a Kr pressure of 10^{-5} Torr. It shows a linear relationship between lamp photon energy and scattered photon energy. At higher pressure, a more complicated situation exists as illustrated in figure 3(b) which is for a Kr pressure of 5×10^{-4} Torr. This case is characterized by photon trapping and there is little correspondence between incident and scattered photon energies. The lamp photons emitted near the centre of the Doppler resonance undergo resonance scattering near the lamp and only have a small probability of reaching the interaction region. Lamp photons emitted far from the resonance centre have a better chance of undergoing their first scatter in the interaction region. The lab energy of these photons will, in general, be shifted by the scattering and those photons that are shifted towards the centre of the resonance can become trapped in that region. The spectral distribution of the trapped photons tends toward the Doppler line profile expected of the Kr gas in the cell as shown in figure 3(b).

The photon scattering Monte Carlo program was then used to characterize the UV photon intensity in the interaction region as a function of Kr pressure in order to understand the

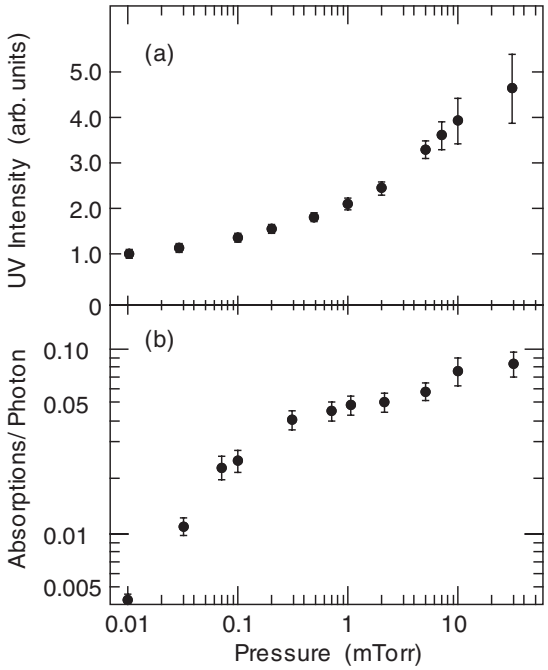

Figure 4. Results of the Monte Carlo simulation. Error bars are statistical uncertainties associated with the simulation. (a) UV intensity in the interaction region as a function of pressure. (b) Fraction of lamp photons which result in the creation of a metastable Kr atom (laser absorbed) as a function of pressure.

saturation power (P_2) data of figure 2(a). In the simplest model, the transition probability for the two-photon absorption (UV + laser) depends on the product of the intensities of the two light sources. In this case, the UV intensity must increase with cell pressure as a complement to the laser saturation power shown in figure 2(a). For these studies we folded the Monte Carlo results with a more realistic initial photon distribution corresponding to our self-reversed lamp. The simulation accounts for the metastable production process in an approximate way. We assume saturation conditions are met for the two-photon transition to the $5p[3/2]_{J=2}$ state within the laser beam, i.e. if a UV photon excites a Kr atom in the region of the cell traversed by the laser beam, we assume that the laser drives a transition to the 5p $J = 2$ level which sometimes decays to the metastable state and the UV photon is lost. The result of this simulation is given in figure 4(a), which shows the relative UV intensity in the interaction region resulting from photon scattering. The important result here is that, due to the trapping of the UV photons in the interaction region, the UV intensity increases with pressure. This is qualitatively what we expect although in the simulation the UV power increases by only about a factor of 5 as the pressure increases from 0.01 to 30 mTorr (see figure 4(a)) whereas in the experiment the saturation power decreases by an order of magnitude over this pressure range (see figure 2(a)).

Using the Monte Carlo simulation we also studied the surprisingly efficient yield of metastable atoms per UV photon. Here, we monitored the fraction of UV photons from the lamp that produced a laser absorption leading to production of a metastable Kr atom. The results are given in figure 4(b) and show an increase in the fraction of lamp photons which produce a metastable atom as a function of the cell pressure. The fraction tends to plateau at a value of about 10% at the highest pressures studied. These results can be compared to the experimental data on the magnitude of the metastable signal, figure 2(b). Again, we have

qualitative agreement as the simulation shows a relative increase of a factor of 3.4 in laser absorption as the pressure increases from 0.1 to 10 mTorr, which is less than the change in signal strength we observe experimentally over the same pressure range. However, the Monte Carlo results reinforce our experimental observation that the metastable production rate can approach 10% of the UV photon flux. We also observe that the conversion efficiency increases as the laser interaction is moved closer to the lamp.

4. Outlook

Based upon the understanding achieved through the Monte Carlo studies of the gas cell, we can intelligently design an optically pumped atomic beam of metastables. A high conversion efficiency of UV-to-metastables can be maintained by running the beam at a relatively high local density. The limit on the density will be set by the mean free path for Kr^*, which can undergo collisions with ground-state atoms which result in elastic scattering ($\sigma \approx 42$ Å^2), metastability exchange ($\sigma \approx 75$ Å^2) [19] and fine-structure changing (quenching) (for Ne $\sigma \approx 4 \times 10^{-4}$ Å^2) [20]. Metastability exchange and elastic scattering do not affect the metastable flux, but may be detrimental to the beam quality. The quenching collisions remove metastable atoms from the beam, but are expected to have a much lower cross section based upon analogy with data on neon [20]. For a mean free path of 50 cm, using the elastic collision cross section, we could operate at a pressure of 0.1 mTorr, corresponding to a conversion efficiency of ≈0.5–2.5% (the range determined by experiment and Monte Carlo simulation). The use of a capillary array to collimate the beam would eliminate the pathlength for UV photons through Kr gas before reaching the laser interaction region, which would in turn increase the efficiency by a factor of ≈2. In addition, the flux from a single lamp can be increased by a factor of ≈2 over that used during these measurements, when the lamp output had deteriorated due to window contamination. With these factors taken into account, we calculate a metastable flux ranging from 4×10^{13} to 2×10^{14} Kr^* s^{-1}. The UV Doppler profile of the beam would resemble that of the gas cell by crossing the UV beam at an angle relative to the atomic beam longitudinal velocity. Tuning of the wings of the lamp frequency distribution by varying the partial pressure of Kr can be used to optimize further the overlap for a given atomic beam velocity. The angular divergence of the beam would be determined by the capillary array (typical aspect ratio 1×40), with a value of 5×10^{-4} sr to give an angular flux of ≈0.8–4×10^{17} metastables s^{-1} sr^{-1}—a considerable improvement over the state-of-the-art rf discharge source, 4×10^{14} metastables s^{-1} sr^{-1} [7].

The metastable fraction can also be estimated, using our observations for pressures below 2 mTorr which show an excitation rate per atom of ≈60 Hz for those atoms which are in resonance with the laser. By applying the same 4× enhancement listed above, we would estimate an excitation rate of 240 Hz/atom. For a transit time of 1×10^{-4} s along a UV–laser interaction length of 2 cm, a metastable fraction of ≈2.4% is projected.

In conclusion, we have demonstrated a simple method using a UV resonance lamp and cw laser to produce metastable Kr. The high conversion efficiency of UV photons to metastable atoms is understood to be a result of increased UV photon intensity due to multiple scattering in the laser interaction region and occurs in a pressure regime where the UV optical mean free path is small, while the collisional quenching mean free path remains large. The UV resonance lamp can easily be run for Kr, Xe and Rn resonance lines, but Ar poses a problem because the short-wavelength resonance line at 105 nm creates defects in the LiF window. Modification of the present scheme to an atomic beam configuration is expected to lead to a notable improvement in the loading rate for metastable rare gas atom traps, with particular benefits in situations where the sample size is limited.

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