

Four-wave parametric mixing in optically inverted barium ions

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Optically inverted barium ions in the $6p_{(1/2)}$ excited state served as the nonlinear medium for doubly resonant four-wave parametric generation of coherent radiation at 167 nm. The excited barium ions were produced via selective autoionization. Stimulated-emission photons at 650 nm corresponding to the $6p_{(1/2)}-5d_{(3/2)}$ transition were mixed with input 451-nm laser photons tuned to the $6p_{(1/2)}-6f_{(5/2)}$ two-photon resonance to produce the 167-nm output photons.

If an atom is excited to an autoionizing state, it will in general decay to a multitude of final ionic states with corresponding distribution of ejected electron energies. Under some conditions, preferential decay to specific excited ionic states occurs. Recently, Bokor *et al.*¹ produced laser action in barium ions by exploiting this principle. Neutral barium was excited to an autoionizing state of the configuration $6p_{(3/2)}np$ with $n \geq 12$ by stepwise two-photon laser excitation. This class of autoionizing states was found to selectively decay to yield excited Ba^+ in the $6p_{(1/2)}$ state. We refer to this behavior as selective autoionization. Amplified spontaneous emission (ASE) was observed at 493 and 650 nm corresponding to the $6p_{(1/2)}-6s_{(1/2)}$ and $6p_{(1/2)}-5d_{(3/2)}$ transitions, respectively. In this Communication, we report the utilization of the excited $Ba^+ 6p_{(1/2)}$ ions produced in this way as a nonlinear medium for four-wave parametric generation of coherent radiation in the vacuum ultraviolet region of the spectrum.

A schematic energy-level diagram showing the relevant barium states and the optical excitation scheme is shown in Fig. 1. The autoionizing states of interest belong to the nominal configuration $6pnp$ and were excited via the $6snp$ resonant intermediate state. In the experiments reported here, the $n = 12$ configurations were used. The apparatus consisted of a barium vapor cell mounted with its output at the entrance slit of a 1-m focal length, normal incidence, vacuum ultraviolet monochromator. An evacuated chamber containing optics for the measurement of backward propagating vacuum ultraviolet emissions was fitted to the input of the cell. The input laser beams were obtained from pulsed dye lasers pumped by the second harmonic of a Nd:YAG laser and standard nonlinear optical techniques. Ultraviolet radiator tunable near 247 nm was produced with an en-

ergy up to 1 mJ per pulse at 10 pps and a 2-cm^{-1} bandwidth. This beam was used for excitation of the $6s^2-6s 12p$ transition in neutral Ba. Visible radiation tunable near 450 nm was produced with an energy of up to 10 mJ per pulse at 10 pps and a 0.5-cm^{-1} bandwidth. This beam was used for the second step in the excitation scheme, $6s_{(1/2)}12p-6p_{(3/2)}12p$.

The transition $6s_{(1/2)}12p-6p_{(3/2)}12p$ involves primarily a one-electron, nearly unity oscillator strength transition of the ion $6s_{(1/2)}-6p_{(3/2)}$, with the $12p$ Ryd-

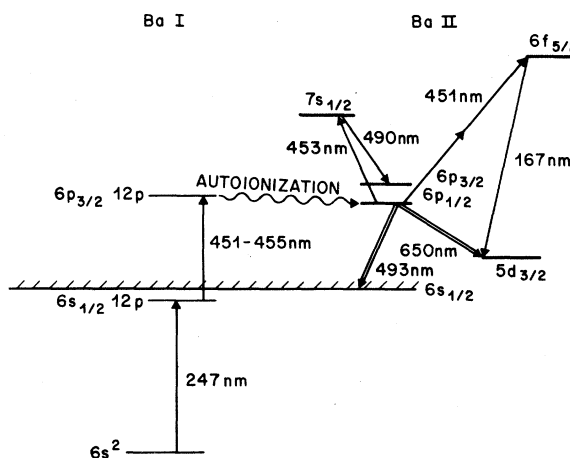


FIG. 1. A schematic energy-level diagram showing the relevant neutral and first ionic states of barium. The autoionization $6p_{(3/2)}12p \rightarrow 6p_{(1/2)} + e^-$ provides an inversion in the ion; the resulting ASE is observed at 650 and 493 nm (double arrows). This process remained saturated as the laser driving the $6s_{(1/2)}12p \rightarrow 6p_{(3/2)}12p$ was tuned from 451–455 nm. As explained in the text, this allowed the production of ASE and four-wave mixing output in the ion using only the original lasers used in the pumping of the autoionization level in the neutral.

berg electron acting as an observer.² Using focused excitation, this transition was easily depletion broadened³ to a width of up to 10 nm. Under these conditions, selectivity in the autoionization process was preserved, and ASE at 650 nm was produced over this entire tuning range. This fact allowed off-resonant production of the $6p_{(1/2)}$ ions and subsequent excitation of these ions to higher-lying states using the same input laser. Two such processes were investigated. When the input laser was tuned to exact resonance with the $6p_{(1/2)}-7s_{(1/2)}$ transition at 452.6 nm, strong ASE on the $7s_{(1/2)}-6p_{(3/2)}$ transition at 490 nm was observed, indicating efficient transfer of population from the $6p_{(1/2)}$ to $7s_{(1/2)}$ level. The $6p_{(1/2)}$ level was sufficiently depleted that the 650- and 493-nm ASE outputs were fully quenched. Well-collimated output at 490 nm was observed from both ends of the cell, with a wavelength which was identical (to within our measurement accuracy of ± 0.02 nm) to that given for the $7s_{(1/2)}-6p_{(3/2)}$ transition in Moore.⁴ Further, the wavelength of the output was independent of the density of barium, and the wavelength of the excitation laser.

In the second case, the visible laser was tuned near 451 nm, corresponding to the $6p_{(1/2)}-6f_{(5/2)}$ two-photon transition in Ba^+ . In this case, the 650-nm ASE was not quenched; in fact, no significant decrease in 650-nm output was observed for any tunings of the visible laser in this vicinity. Nevertheless, strong collimated emission at 167 nm was detected in the forward direction. The peak output was estimated to be 10^{12} photons per pulse. Because no emission was detected in the backward direction, this output cannot be due to ASE and is instead attributed to a doubly resonant four-wave parametric interaction involving the excited Ba^+ as the nonlinear medium. Here, two of the input 451-nm photons mix with the internally generated 650-nm ASE photons to produce the 167-nm output photons. This process closely resembles recently investigated examples of four-wave parametric oscillation⁵ with the important difference that the large nonlinearity arises from the doubly resonant susceptibility of the excited $Ba^+ 6p_{(1/2)}$ ions.

The parametric nature of this process is further illustrated by the data of Fig. 2. Here, the output emission at 167 nm was monitored as a function of the input laser wavelength near 451 nm for two different temperatures of the barium vapor cell. Op-

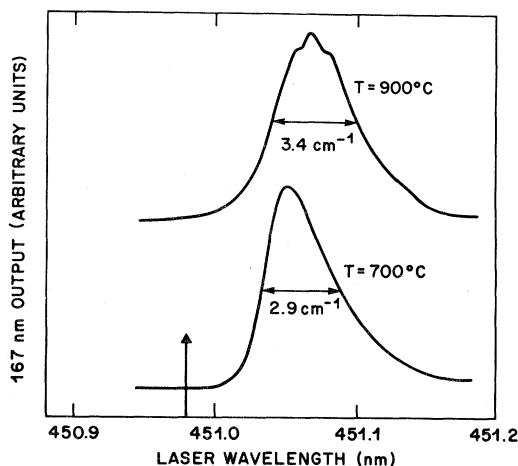


FIG. 2. Plot of the intensity of emission at 167 nm as a function of the input wavelength near 451 nm for two different temperatures of the barium cell. The arrow marks the laser wavelength for exact two-photon resonance as given by Moore. The tuning curve is seen to be density dependent, broadening and red shifting with increased barium density.

timum output is achieved for a laser tuning of approximately 7 cm^{-1} to the red side of the true two-photon resonance as given in Moore.⁴ The tuning curve is seen to be density dependent, broadening the red shifting with increased barium density. This type of excitation spectrum is characteristic of phase-matching behavior in four-wave mixing processes.⁶

In conclusion, excited $Ba^+ 6p_{(1/2)}$ ions produced by decay of the neutral $Ba^+ 6p_{(3/2)} 12p$ autoionizing state have been exploited as a nonlinear medium for the efficient generation of coherent vacuum ultraviolet radiation. Up to 10^{12} photons per pulse at 167 nm were generated at 167 nm. These results indicate the potential of the selective autoionization mechanism acting in combination with additional nonlinear processes for the generation of high-brightness coherent radiation in the deep ultraviolet region of the spectrum.

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