Title

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DISCLAIMER

Ultra-high Resolution VUV-XUV Laser: Application to the Hyperfine Study of Krypton

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Abstract

A near-transform-limited VUV-XUV laser source with 210 MHz resolution has been constructed and used to study the hyperfine splitting, isotope shift, and lifetime of the three lowest ns (n=5,6,7) Rydberg states of Krypton.

Introduction

Transform-limited tunable lasers represent photon sources with the highest spectral purity and spectral brightness. The ability to deliver photons in the narrowest possible spectral width finds applications not only in high-resolution spectroscopy, but also in cases where efficient use of photons are desired. Examples of this are plenty in the field of chemical physics. Experiments involving selective excitation of molecules: stimulated-emission pumping studies, resonance-enhanced multiphoton excitation and ionization, laser-induced fluorescence, molecular beam excitation studies, and studies of molecules or impurities embedded in liquids, matrices, and crystals can all benefit from these lasers. Near transform-limited tunable pulsed lasers in the visible and near-uv have been available for several years.[2,3] Recently we have extended the tuning range of these types of lasers to the vuv and xuv region. In this article we shall describe the basic requirements and the approach we took for the laser development and a brief summary of the first application of this source: a hyperfine study of the ns Rydberg states of krypton. Details of the laser development and the krypton study have been submitted for publication elsewhere.[4,5]

Design Goals

The goals follow from the desire that the source will be used for photoionization and LIF study of small to intermediate sized molecules in a pulsed molecular beam setting:

* broad tunability, at least from 70 nm to 160 nm.;
* high spectral brightness, preferably transform-limited;
* high intensity, 10^{15} to 10^{16} photons per second at 10 Hz.;
* uniform spatial profile;
* user friendly.

If these goals are met the laser source not only will be able to serve as a universal probe of many chemical processes but also as a powerful excitation source to initiate such processes.

Approach

Since the first and foremost requirement for the source is broad tunability it is a simple choice to use the technique of four-wave mixing for the generation of vuv and xuv in the prescribed range. This well-established technique [6] has very broad wavelength coverage, potentially can be very efficient [7], and uses reliable commercial components. Successful exploitation of this technique, which involves high order nonlinear optical processes, requires careful control on the spectral, temporal, and spatial properties of the visible laser(s) used. For this reason the followings were chosen to constitute the major components of our system:

* a pulse-amplified CW single-longitudinal-mode dye laser for good frequency control;
* an injection-seeded Nd:YAG laser as pump laser for a smooth Gaussian temporal profile;
* side-pumped prism dye cells to preserve the spatial quality of the dye beam;
* a pulsed nozzle as the vuv generator for simplicity and flexibility in the choice of four-wave mixing medium;
* a 1-meter grating monochromator to disperse the vuv-xuv wavelengths from the incident visible and uv radiation.
* a Faraday optical rotator for isolation of the CW dye laser from the pulsed dye amplifiers;
* lens-pinhole-lens spatial filtering between successive stages of dye amplification to control amplified-spontaneous-emission growth.

Auto-tracked second-harmonic-generation crystals are used to convert the visible to the uv prior to the four-wave mixing stage. Table I is a summary of the performance of the laser. Entries to the table are self-explanatory. It is however

necessary to point out that we measure a frequency shift of 0 to ~20 MHz between the CW laser frequency and the amplified output frequency. This shift depends on the dye used in the amplifiers as well as the number of amplification stages used. At this point we are trying to understand the cause of this shift. But it is clear that for experiments where this shift is significant to the results it is necessary to determine the shift for each frequency measurement. Details of the characterization of the laser are provided in reference 4.

Table I: Summary of Laser Performance

	Visible	UV	VUV-XUV
Energy	100 mJ	30 mJ	1 uJ
Bandwidth (FWHM)	91 MHz	140 MHz	210 MHz
Pulsewidth	7 nsec	<7 nsec	~4 nsec
Temporal Profile	near Gaussian	near Gaussian	near Gaussian
Tuning Range (tested)	562-620 nm	281-310 nm 222-240 nm	74-124 nm
Frequency Shift	Amplifier configuration and dye dependent		

Hyperfine study of Krypton

Utility of this source has been demonstrated in a high-resolution study of the 4p-5s[3/2], -5s'[1/2], -6s[3/2], -6s'[1/2] and -7s[3/2] J=1 transitions of Kr spanning the spectral region from 123.6nm to 94.5nm. The ns states (n=5,6,7) were studied using 1 + 1 resonant multiphoton ionization where the ns states form the resonant intermediate. Calibrations using known I_2 frequencies yield an improved absolute calibration of the ^{86}Kr 4p-ns transition frequencies by more than an order of magnitude. The hyperfine parameters for the 5s[3/2] level are in very good agreement with those measured using interferometric techniques. This is the first time that determination of hyperfine parameters for the other levels listed above are possible. From these parameters we find that for n=5 the jj quantum numbers are good to 99.63 $\pm$ 0.06% while for n>5 the jj quantum numbers are good to at least 99.99%.

Isotope shifts for the more abundant (>0.1% natural abundance) isotopes were obtained. These shifts can be explained by the simple Bohr shift and the specific mass effect. To within our resolution we do not observe a volume effect on the shift. Lifetimes for the n=6 and 7 states which are longer than the duration of our laser pulse can also be measured. Our lifetime results differ substantially from calculations reported in the literature. These results should form a basis for the atomic theorists to obtain an improved set of wavefunctions for Kr.

Remark

The laser system described above meets most of the goals outlined in the beginning of this article. However, a major shortcoming is that the intensity falls short of the goal by 3 to 4 orders of magnitude. A second pulse-amplified single-mode dye laser system is under construction to provide a second frequency for resonance-enhanced four-wave mixing operation. This could improve the intensity by two orders of magnitude. Amplification in the vuv using inert-gas excimer systems or free-electron lasers is certainly possible. Other plans include use of tunable solid-state amplifiers (alexandrite, Ti-sapphire), use of high-repetition-rate pump lasers, and use of high power excimer lasers to facilitate efficient pumping of blue-green dyes.

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