Measurement of high conversion efficiency to 123.6-nm radiation in a four-wave-mixing scheme enhanced by electromagnetically induced transparency

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We report measurements of the absolute yield of 123.6-nm vacuum ultraviolet (VUV) radiation produced in a resonant four-wave-mixing scheme in krypton enhanced by electromagnetically induced transparency. One of the mixing fields, in the ultraviolet (UV) at 212.55 nm, was in two-photon resonance with the $4p^6\ {}^1S_0-4p^5 5p[0,\frac{1}{2}]$ transition of krypton, while a second field (the coupling field) at 759 nm was resonant with the $4p^5 5p[0,\frac{1}{2}]$-$4p^5 5s[1,\frac{1}{2}]$ transition. Electromagnetically induced transparency has been predicted to be induced by this coupling field and to enhance the efficiency of the generation of the field at 123.6 nm, on the $4p^5 5s[1,\frac{1}{2}]$ to $4p^6\ {}^1S_0$ transition. This was confirmed by measurements of the absolute VUV radiation yield using a calibrated photodiode in the limit of a large density-length product. Energies of ~30 nJ were produced, which gave a conversion efficiency, from the coupling field energy of ~1%. Higher yields are thought to be achievable by increasing the coupling laser intensity, the path length of the medium, and by the use of a transform-limited UV pulse of the same duration as the coupling field.

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Schemes employed to generate coherent radiation at vacuum ultraviolet (VUV) wavelengths have commonly made use of four-wave mixing in atomic gases conducted off resonance in order to avoid reabsorption of the generated light. Typical conversion efficiencies in the region $10^{-5}-10^{-6}$ have been achieved for input powers in the range 1–5 MW [1,2]. These schemes have been widely used in linear spectroscopy but the relatively low pulse energies generated have largely precluded other uses. Applications such as nonlinear spectroscopy, photolithography, and photochemistry, for which coherent radiation at longer wavelengths has been used widely, have been difficult, if not impossible, in the VUV. More powerful sources of narrowband and coherent radiation have therefore been sought.

The third-order nonlinearity of the medium ($\chi^{(3)}$) controls the mixing of the three applied fields, and though this can be enhanced by resonance (for example two-photon resonance [3,4]) the generated field must be far from resonance to avoid reabsorption. The dispersion of the medium at resonance also varies steeply, causing disruption to the phase matching. Earlier work using two resonant lasers and a third near-resonant laser mixing scheme in mercury vapor reported high conversion efficiency (up to 5%) into the VUV at 130.2 nm [5]. This high conversion efficiency was attributed by the authors to the use of collimated unfocused beams over a 1-m path length and two-photon resonant and near single-photon resonant enhancement of $\chi^{(3)}$ for all fields.

In 1990 Harris, Field, and Imamoglu proposed that the phenomenon of electromagnetically induced transparency could be used to suppress the absorption of generated light which is in resonance with an atomic transition while also improving the dispersive properties of the medium [6]. The advantages of this type of scheme are that the nonlinear susceptibilities governing the coupling of light into the generated field are still resonantly enhanced through constructive interference. In contrast the linear susceptibility (governing absorption and dispersion) undergoes destructive interference. Changes in the dispersive profile of the medium improve phase matching by increasing the coherence length. Experiments have followed that demonstrate the viability of this method of wave mixing. In 1993 Jain *et al.* observed electromagnetically induced phase matching in a four-wave-mixing scheme in lead, with a measured conversion efficiency of 2.4×10^{-8} when referenced to the power of the coupling laser [7]. Recently Merriam *et al.* [8] achieved near unity conversion efficiency in a lead four-wave-mixing scheme using the principles of electromagnetically induced transparency (EIT). This scheme generated a field in the far ultraviolet (UV) at 186 nm using three near resonantly tuned transform-limited lasers. This type of mixing scheme relies upon the creation of a maximal coherence by a pair of transform-limited laser pulses resonant with atomic levels in a Λ configuration. A third field, close to resonance with a fourth energy level, mixes with this coherence to generate a new field with very high efficiency. It is not obvious how to extend this kind of scheme to the generation of significantly higher frequency fields (e.g., <130 nm) due to the absence of atoms with suitable energy-level configurations.

Shorter wavelengths can be generated using relatively high-energy two-photon resonances in a four-wave-mixing scheme [Fig. 1(b)], but at the cost of large detunings from any single-photon resonance for these two-photon transitions. This detuning can, in principle, be compensated for by using EIT and high density-length products to enhance frequency mixing. In 1993 Zhang, Hakuta, and Stoicheff [9] reported their results from a four-wave sum-mixing scheme using electromagnetically induced transparency in atomic hydrogen. The conversion efficiency to the VUV (102.6 nm)

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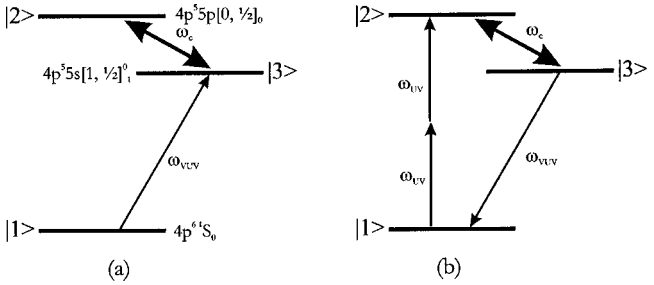

FIG. 1. Level scheme in krypton. (a) shows a scheme for EIT, giving transparency at 123.6 nm by application of 759-nm coupling field ω_c. (b) shows the four-wave-mixing scheme with two-photon pump field at 212.55-nm (ω_{UV}) and 759-nm coupling field (ω_c) with light generated at 123.6 nm (ω_{VUV}).

was measured to be 1×10^{-4}, limited by the rather low density-length products available in the atomic hydrogen medium [9].

Here we report an observation of ~1% conversion efficiency into the VUV at 123.6 nm, the highest reported efficiency in an EIT-enhanced four wave-mixing scheme for wavelengths less than 130 nm. The scheme employed is a four-wave-mixing scheme in room-temperature krypton gas (natural isotopic abundance and 99.995% purity). The relevant atomic energy levels can be reduced to the three levels shown in Fig. 1. EIT can occur in this ladder configuration comprising a ground state $4p^6\ {}^1S_0$ ($|1\rangle$), an excited state $4p^5 5s[1,\frac{1}{2}]$ ($|3\rangle$) dipole coupled to $|1\rangle$, and an excited state $4p^5 5p[0,\frac{1}{2}]$ ($|2\rangle$) which is dipole coupled to $|3\rangle$ but not to $|1\rangle$. The medium, optically deep at ω_{VUV} (123.6 nm), can be rendered transparent by a field ω_c (758 nm). The basic EIT ladder scheme of Fig. 1(a) can be incorporated within a resonant four-wave-mixing scheme, Fig. 1(b), by the introduction of a two-photon resonant field at 212.55 nm (ω_{UV}), applied in two-photon resonance with the $|1\rangle$-$|2\rangle$ transition. There is a large single-photon detuning of this field from the nearest intermediate state of 33 000 cm^{-1}. For resonant conditions the absorption and coherence length of the medium will be highly dependent on the coupling laser intensity. As the 758-nm pulse energy is increased they will become many orders of magnitude larger when the coupling laser Rabi frequency exceeds the Doppler width of the 123.6-nm transition. The absorption at 123.6-nm is suppressed by EIT due to destructive interference on the absorption pathway while the mixing process remains resonantly enhanced due to constructive interference [10]. A significant enhancement in the frequency up-conversion efficiency is therefore predicted.

Our previous krypton experiments were conducted using a gas jet [11]. We demonstrated a large ($\sim10^4$) increase in the VUV yield attainable by using a resonant coupling laser in an optically deep sample. In the experiment reported here a krypton-filled cell was used which permitted the accurate determination of path length and density and an increase in the density-length product (NL) of the krypton medium to values much greater than those achievable in the gas jet. The gas jet had densities 10^{15}–$10^{16}\,\mathrm{cm}^{-3}$ of krypton with path lengths 0.1 to 0.5 cm ($NL\sim10^{15}\,\mathrm{cm}^{-2}$), approximately the same NL values as the maximum used in earlier four-wave-mixing schemes in hydrogen [9]. In contrast the 1-cm gas cell was operated in the pressure range 0–10 mbar ($0<NL<2.5\times10^{17}\,\mathrm{cm}^{-2}$). This increase in NL allowed us to access much higher conversion efficiencies.

In these experiments the UV field was provided by a XeCl excimer pumped dye, which was then frequency doubled to produce a pulse of length 16 ns, energy 150 μJ, and 1.5-GHz bandwidth [11]. The exact two-photon resonance frequency was determined by the measurement of photoionization in a separate krypton gas cell. The coupling field at 759-nm was provided by a titanium-sapphire amplified optical parametric oscillator pumped by a single-mode, frequency-doubled Nd:YAG (yttrium aluminum garnet) laser. This 759-nm coupling field was a single-mode, near-transform-limited pulse (<250 MHz, 4 ns) which was kept to within ±1 GHz of the resonance by reference to laser-induced fluorescence in a krypton hollow cathode lamp. The relative jitter in arrival time of the laser pulses was minimized using computer-controlled active feedback to around ±2.5 ns, which is short compared to the 16-ns pulse length of the UV field, and so the infrared (IR) pulse was timed to coincide within ±2.5 ns of the peak of the UV intensity.

The two beams were directed into the 10-mm-long krypton cell. The UV laser field was focused at the interaction region by a 330-mm focal length lens giving a confocal parameter of ~100 mm. This ensured that the intensity of the UV radiation was uniform throughout the 10-mm-long cell. At the interaction region the coupling beam diameter was larger to provide uniform intensity throughout the focused region of the UV beam, with the coupling beam size changing very little over the length of the cell. These beam sizes were measured with a charge-coupled device (CCD) array placed at the interaction plane. The coupling beam was measured to be 2.3 ± 0.1 mm [full width at half maximum (FWHM)] in diameter at the interaction region, and the UV beam 0.10 ± 0.02 mm (FWHM) in diameter at the interaction region

The rear window of the cell consisted of a 3-mm-thick polished MgF_2 disk that transmitted the generated VUV radiation into a 1-m Seya mounted grating spectrometer (see Fig. 2). The spectrometer was pumped by an oil diffusion pump and maintained at a pressure of 10^{-5} mbar. The grating was a 1200 lines/mm aluminum-coated ruled grating, with a 998.8-mm concave radius and a magnesium fluoride coating to prevent oxidation of the surface (supplied in 1998). The absolute VUV radiation intensity was measured using a calibrated photodiode at the exit slit of the monochromator. The aluminum oxide photodiode was issued by NIST as a transfer standard detector calibrated in 1987 [12]. The detector was essentially solar blind and produced extremely low dark current and noise characteristics. Radiation incident on the photocathode generated photoelectrons in the film and an anode biased to 60 V generated an electric field that enhanced the collection of the electrons emitted from the surface. The signal was measured on a fast oscilloscope (Tektronix TD620). Further measurements of the relative scaling of the VUV pulse intensity (but not the absolute energy) with coupling laser intensity were made using a solar

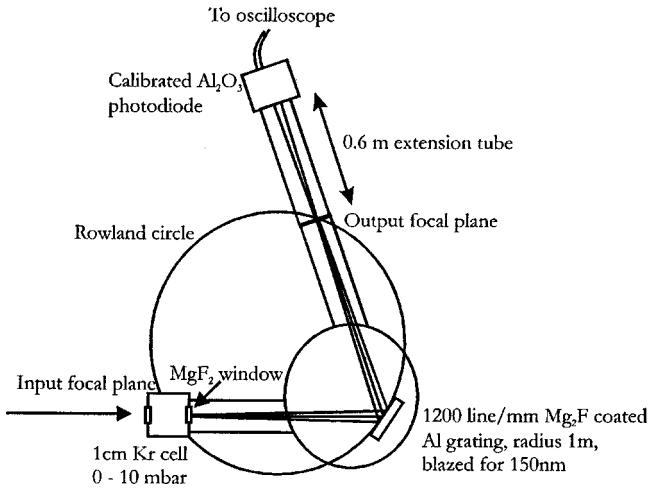

FIG. 2. Schematic of experiment showing the spectrometer, with 60-cm extension on output port. The Rowland circle of the grating is indicated.

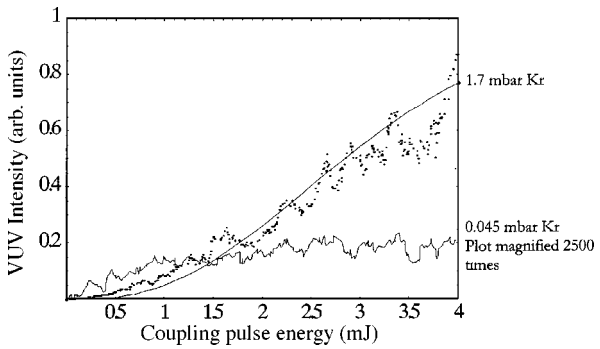

FIG. 3. Experimental dependence of the generated VUV pulse intensity (in relative units) as a function of the coupling laser pulse energy for a krypton pressure of 1.7 mbar (discreet points) and 0.045 mbar (solid line). The data for 0.045 mbar has been multiplied by a factor of 2.5×10^3 to allow comparison with the higher pressure data. The VUV intensity was measured on a shot-to-shot basis by a photomultiplier tube. Note that there is substantial shot-to-shot noise due to fluctuations in the UV laser intensity, which accounts for the noise in the plot. The solid line shows the corresponding theoretical plot for these conditions, i.e., 10-mm cell, $5\times10^{16}\,\mathrm{cm}^{-3}$ density. The absolute scaling of the VUV signal is arbitrary and has been adjusted to be approximately the same magnitude as the experimental data.

blind photomultiplier tube (PMT) which replaced the photodiode. The PMT was used due to its superior sensitivity and dynamic range compared to the photodiode.

The sensitivity of the photodiode to light at the generated wavelength, 123.58 nm, was extrapolated from the quantum efficiency curves provided in [12] to be 0.0075 ($\pm$0.001) electrons per photon. In order to minimize saturation due to space-charge effects a large VUV spot was used. The VUV was nominally focused at the exit slit of the monochromator and so the photocathode was mounted 600 mm back from this plane. The signal in this position was seen to increase by a factor of greater than 100 compared to that with the detector at the exit focal plane of the VUV, indicating the strong saturation resulting from the small beam area.

The low sensitivity of the detector dictated that in the calibration experiment the VUV signal had to be maximized by using the highest available laser power (4 mJ per pulse). The highest VUV signals were found at the highest coupling intensities available (see Fig. 3). For coupling pulse energies of 4 mJ the optimum VUV generation was found at $\sim$2 mbar.

The dependence of VUV, intensity on IR power for krypton pressures of 1.7 and 0.045 mbar is plotted in Fig. 3. The 0.045-mbar data is multiplied by a factor of 2.5×10^3 for comparison of the energy scaling to be made. This large difference in energy scales confirms the large increase in VUV yield when using higher NL products. The low density-length data (0.045 mbar) is quantitatively similar to those results obtained with a gas jet [11]. The data for 1.7 mbar show good qualitative agreement with a calculation using the method outlined in [10] (shown for the 1.7-mbar data) which is also shown on the same figure as a smooth solid line. This theoretical model assumes steady-state conditions, with the atomic data for krypton used for the decay rates, monochromatic radiation, Doppler broadening, and propagation through a uniform density cell (more suited to the current arrangement than for the gas jet). The VUV yields at higher coupling intensities are much greater than for lower density-length product regimes in the gas jet [11] or the cell, which peak at much lower coupling pulse intensities (Rabi frequencies). For example, the VUV yield reaches its maximum value at around 1-mJ coupling pulse energy for 0.045 mbar of krypton (see Fig. 3). Given the limitations of the coupling laser intensity available to us experimentally it was found that the maximum VUV intensity, though not necessarily the VUV efficiency, was found at $\sim$2 mbar. Above 2 mbar of pressure the intensity of the VUV generated falls off again as the coupling pulse intensity necessary to overcome the preparation energy and to create transparency leads to a reduction in the nonlinear susceptibility, governing the coupling of energy into the VUV field. This complex interplay between the optimum NL product and the coupling intensity will be the subject of further investigation.

Under the higher NL conditions (1.7 mbar $=5\times10^{16}\,\mathrm{cm}^{-2}$) of the results also shown in Fig. 3 it should be noted that the coupling laser energy required to provide a Rabi frequency larger than the Doppler width (0.5 μJ) is much less than that to satisfy the coupling laser preparation energy [14] (about 0.5 mJ).

In the calibration experiment the pulse energy was 4 mJ giving a Rabi frequency of $\sim10\ \mathrm{cm}^{-1}$ for the coupling laser, as compared to the Doppler width of the VUV transition, which was only $0.03\ \mathrm{cm}^{-1}$. The pressure of krypton, 2 mbar, gave an NL value of $5\times10^{16}\,\mathrm{cm}^{-2}$. Under these conditions the output of the photodiode was a pulse of 10 ns with a pulse height of 2 mV measured into 50 Ω. This corresponds to 2.5×10^6 electrons per pulse, giving a VUV pulse energy of 0.5 nJ.

In order to determine the actual VUV pulse energy generated in the cell, several factors need to be taken into account, including the efficiency of the grating and the transmission of optics between the region of generation of the VUV and detection. Taking into account the ruling effi-

ciency, the reflectivity of aluminum, and the blaze angle, the efficiency of the grating was calculated to be $10\pm5\%$. It should, however, be noted that additional losses might occur due to the deterioration of the grating surface with time and VUV absorbing impurities in the vacuum chamber.

The rear window of the cell was made of magnesium fluoride. This material is often used for VUV work, due to its transmission of short wavelength light and nonhygroscopic nature. However, 123.58-nm light is near the limit of its transparency dictating the accurate measurement of the transmission of the window at 123.58 nm. The window was placed in the output arm of the spectrometer and the attenuation of the signal was measured using the photomultiplier operated in the linear regime. The rear window of the cell was replaced for this run by a similar MgF_2 window. The window used in the calibration experiment was found to transmit 15% of the incident VUV radiation.

When the attenuation of the grating and the window are taken into account the energy of the VUV generated at the interaction region is calculated to be 33 ± 10 nJ. This value of the VUV pulse energy is probably an underestimate. Absorption by residual oil molecules in the vacuum chamber and on the grating has not been taken into account. Some saturation of the signal at the photodiode is also a possibility. Larger displacements of the photodiode from the focal plane than 600 mm were impractical leaving the VUV spot still significantly smaller than the 6×6 mm^2 spot size used for the calibration of the device. Therefore some signal saturation, further reducing the measured VUV energy, cannot be ruled out.

The energy of the infrared pulse which interacts in the four-wave-mixing scheme is that portion of the infrared light that overlapped the smaller (0.1 mm diameter) region in which the UV pulse was focused in the cell. This was determined to be 5 μJ, corresponding to a conversion efficiency into the VUV of $\sim1\%$ from the coupling laser.

The energy of the VUV field generated will ultimately be limited by one of two factors. The first is that the 212.55-nm photoionization channel can destroy coherence induced by the coupling field [10]. Another limiting factor is the driving of the $|1\rangle$-$|2\rangle$ coherence due to the generated field as discussed by Deng *et al.* [13]. They found that the optimum density-length product for a four-wave-mixing scheme in rubidium was 6×10^{14} cm^{-2}, limited by the destructive interference effects of the generated field. Our optimum density-length product, 5×10^{16} cm^{-2} is clearly much greater than that found by Deng *et al.*, and is probably due to the transform-limited nature of our coupling laser creating better transparency. There is, however, a maximum limit to the value of the density-length product of the medium when the preparation energy exceeds that provided by the coupling laser [14].

The most important practical limit to VUV energy will be the available input UV pulse energy. The UV pulse energy at the interaction region was measured to be 60 μJ with a pulse length of over 16 ns. The coupling laser pulse duration was less than 4 ns, therefore the effective UV energy available to the mixing process, within the coupling pulse duration, was 15 μJ. Given that the generated VUV radiation was estimated to have an energy of 33 nJ, this corresponds to an efficiency, from the UV, of 0.2%. An UV pulse of similar length to the coupling pulse would produce much larger VUV energies. Another shortcoming of our UV laser is its bandwidth, 1.5 GHz, which was not optimally matched to the coupling field. This should also be taken into account when analyzing the relative efficiency of the scheme compared to the UV pulse energy, as matched UV and IR pulses are required for optimum conversion efficiency. Although only 60-μJ UV pulses were available to us, far higher energy transform-limited UV pulse energies are, in principle, achievable. For example, Bergeson *et al.* [15] produced 3-mJ transform-limited pulses at 210 nm with a pulse length and bandwidth well matched to the requirements of the present experiment.

In conclusion we have measured substantial VUV energies associated with high conversion efficiencies of about 1%, in an EIT-enhanced VUV frequency up-conversion scheme. Future investigation will examine the VUV yield when two temporally matched transform-limited pulses are used with a longer path-length cell. Conversion efficiencies significantly larger than 1% should be possible, with VUV pulse energies perhaps greater than 10 μJ if higher power transform-limited UV pulses are used.

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[1] J. F. Reintjes, *Nonlinear Optical Parametric Processes in Liquids and Gases* (Academic, New York, 1984).

[2] C. R. Vidal, *Tuneable Lasers*, edited by L. F. Mollenauer and J. C. White, Topics in Applied Physics Vol. 59 (Springer-Verlag, Berlin, 1987), Chap. 3.

[3] G. Hilbig and R. Wallenstein, IEEE J. Quantum Electron. **QE-19**, 194 (1983).

[4] J. P. Marangos, N. Shen, H. Ma, M. H. R. Hutchinson, and J. P. Connerade, J. Opt. Soc. Am. B **7**, 1254 (1990).

[5] C. H. Muller III, D. D. Lowenthal, M. A. DeFaccio, and A. V. Smith, Opt. Lett. **13**, 651 (1988).

[6] S. E. Harris, J. E. Field, and A. Imamoglu, Phys. Rev. Lett. **64**, 1107 (1990).

[7] M. Jain, G. Y. Yin, J. E. Field, and S. E. Harris, Opt. Lett. **18**, 998 (1993).

[8] A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, Opt. Lett. **24**, 625 (1999).

[9] G. Z. Zhang, K. Hakuta, and B. P. Stoicheff, Phys. Rev. Lett. **71**, 3099 (1993).

[10] J. C. Petch, C. H. Keitel, P. L. Knight, and J. P. Marangos, Phys. Rev. A **53**, 543 (1996).

[11] C. Dorman and J. P. Marangos, Phys. Rev. A **58**, 4121 (1998).

[12] U.S. Department of Commerce, National Bureau of Standards, Report of Test, P.O. C3XD 9607400 (1987).
[13] Lu Deng, W. R. Garret, M. G. Payne, and D. Z. Lee, Opt. Lett. **21**, 928 (1996).
[14] S. E. Harris, and Z.-F. Luo, Phys. Rev. A **52**, R928 (1995).
[15] S. D. Bergesson, A. Balakrishnan, K. G. H. Baldwin, T. B. Lucatorto, J. P. Marangos, T. J. McIlrath, T. R. O'Brian, S. L. Rolston, C. JU. Sansonetti, Jesse Wen, N. Westbrook, C. H. Chen, and E. E. Eyler, Phys. Rev. Lett. **80**, 3475 (1998).