PRODUCTION OF INTENSE, COHERENT, TUNABLE, NARROW-BAND LYMAN-ALPHA RADIATION*

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ABSTRACT

Nearly transform limited pulses of 1216 Å radiation have been generated by sum frequency generation in 0.1 to 10 torr of mercury vapor. The summed input beams, consisting of photons at 3127 Å and 5454 Å originate in 1 MHz band-width ring-dye laser oscillators. The beams are amplified in pulsed-dye amplifiers pumped by the frequency doubled output of a Nd:YAG laser. The 3127 Å photons are tuned to be resonant with the two-photon 6^1S to 7^1S mercury transition. The VUV radiation can be tuned by varying the frequency of the third non-resonant photon. We have also observed difference frequency generation at 2193 Å and intense fluorescence from the 6^1P state at 1849 Å. We have studied the intensity and linewidth dependance of the 1849 Å fluorescence and 1216 Å sum frequency signals on input beam intensity, mercury density, and buffer gas pressure and composition.

INTRODUCTION

We are currently involved in a number of investigations which require a coherent radiation source near the hydrogen Lyman-α wavelength at 1216 Å. These include a measurement of the two-photon ionization cross section of atomic hydrogen at Lyman-α, technology for cooling and trapping of atomic hydrogen, and measurements of the heights and widths of the (n=2) Feshbach and Shape resonances in H^-. These experiments require a source that is narrow-band (preferably transform limited), broadly tunable, bright, and coherent. We have constructed such a source and measured the effects of mercury density, phase matching conditions, and strong input fields on the output intensity and linewidth.

VUV PRODUCTION

The Lyman-α radiation is produced by summing in mercury vapor two photons at 3127 Å and a third photon at 5454 Å. The input beams originate in 1 MHz band-width ring-dye lasers. They are amplified in pulsed-dye amplifiers which are pumped by the doubled output of a Nd:YAG laser. The resulting beams have transform-limited linewidths of 300 MHz. The beams were focussed with a 25 cm focal length achromatic lens to a 100 μm spot in a cell containing 0.1 torr of mercury. The intensities of the beams at the focal point were 2×10^9 W/cm^2 at 3127 Å and 1×10^{10} W/cm^2 at 5454 Å. The mercury cell construction was similar to the cell described in reference 1. In addition to the sum frequency generation, we also observed difference frequency generation at 2193 Å and fluorescence from the 6^1P state at 1849 Å.

For most of the measurements reported here, the mercury cell was operated at a temperature of 180°C. A few of the measurements were taken at 150°C. Absorption measurements with a low pressure mercury lamp determined that this corresponds to a mercury density of 10^{15} cm^{-3}.

INTENSITY AND LINEWIDTH OPTIMIZATION

The peak Lyman-α intensity occurred when the 3127 Å beam was tuned 0.5 cm^{-1} to the blue of the low intensity resonance wavelength of 31964.1 cm^{-1}. The 1849 Å and 2193 Å signals were maximum when the 3127 Å signals remained fixed at 31964.1 cm^{-1}. The full width at half maximum of the 1849 Å fluorescence signal as a function of the wavelength of the 3127 Å beam was 2.6 cm^{-1}, which is indicative of power broadening. Our results can be compared to the strontium measurements of Scheingraber and Vidal[2] who reported a similar blue Stark shift of the resonant frequency. Mahon and Tomkins[1] have reported an observation of the Stark broadened absorption linewidth in mercury at these intensities, but did not report the blue shift in the resonance frequency.

To further investigate the saturation of the 6^{1}S to 7^{1}S transition, we measured the fluorescence signal at 1849 Å as we varied the mercury cell temperature, input beam intensity, and buffer gas pressure and composition. The 1849 Å signal is proportional to the population of the 7^{1}S state since it results from a cascade from the 7^{1}S state to the 6^{1}S ground state through the 6^{1}P state.

The intensity of the fluorescence signal decreased by a factor of 5 when the 5454 Å beam was allowed to pass through the cell. This indicates that the branching ratios for sum and difference frequency generation are large enough to significantly reduce the population of the 7^{1}S state. Hence, although Smith et al.[3] noted a problem with stimulated emission at 1.0 μm resulting from 7^{1}S to 6^{1}P transitions in a plane wave geometry, this will not be a problem for Lyman-α output in a tight focussing geometry like ours. In order to maximize the 1849 Å signal, the remaining measurements were made without the 5454 Å beam being present.
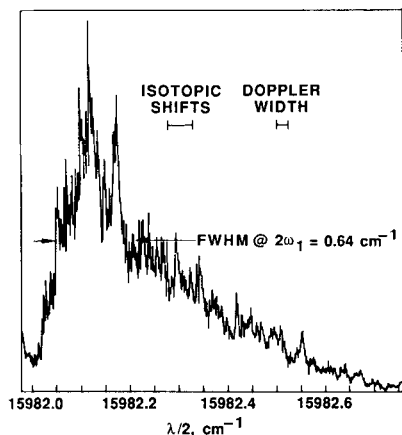

ISOTOPIC DOPPLER
SHIFTS WIDTH

FWHM @ 2ω_1 = 0.64 cm^{-1}

15982.0   15982.2   15982.4   15982.6
$\lambda/2$, cm^{-1}

Figure 1. Fluorescence as a function of detuning of the 3127 Å input beam.

The fluorescence intensity as a function of the frequency of the 3127 Å input beam is shown in Figure 1. The measured 0.64 cm^{-1} AC Stark broadened linewidth is significantly larger than either the linewidth of the input beam, the Doppler broadening, or the isotopic frequency shifts.

Figure 2 is a plot of the intensity of the 1849 Å fluorescence as a function of the temperature of the mercury cell. Increasing the mercury cell temperature beyond 145°C reduces the fluorescence output. Absorption measurements with a low pressure mercury lamp show a monotonic decrease in cell transmission of 2537 Å light up to 220°C. This suggests that the 7^{1}S population decrease is caused by an increase in an excited state collisional process such as excimer formation.[4]

We further investigated the saturation of the two photon absorption by monitoring the 1849 Å fluorescence as a function of input beam intensity. Figure 3 is a plot of this signal as a function of the energy of the 3127 Å pulses. The smooth curve in the figure is a fit to the functional form $I = \alpha E^x$, where I is the florescence signal and E the pulse energy in mJ. The best fit was for $\alpha = 0.99$ and $x = 1.8$. The fact that the functional dependance does not deviate from the expected low intensity quadratic form implies that the occupation probability of the 7^{1}S state is much less than 1. In other words, the cascade from the 7^{1}S state is sufficiently fast to inhibit fluorescence reduction even though the input intensities are large enough to produce
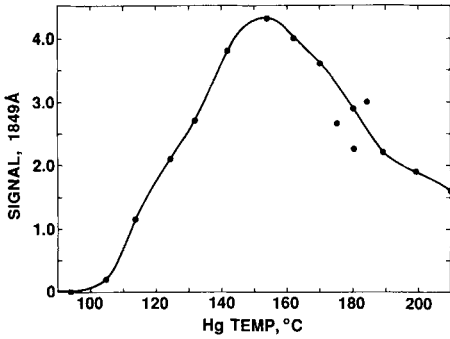
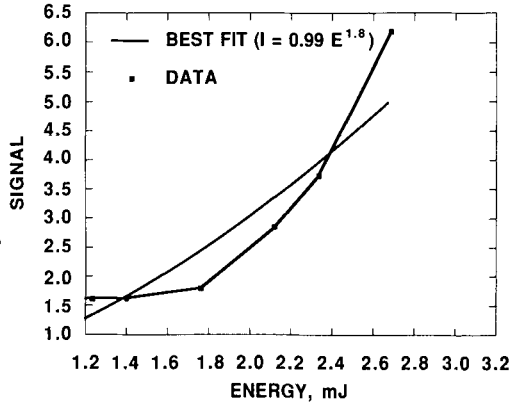Figure 2. 1849 Å fluorescence as a function of mercury cell temperature.

Figure 3. Fluorescence signal as a function of 3127 Å pulse energy.

significant power broadening and AC Stark shifts. This is different from the observations of Scheingraber and Vidal[2] in strontium, where a dip in third-harmonic generation is attributed to significant population in the upper two-photon resonant state.

Switching buffer gas from helium to krypton resulted in a slightly less intense 1849 Å signal at a given input power and cell temperature. However, the fluorescence had the same dependance on the input intensity for both gasses. Varying the krypton or helium buffer gas pressure resulted in no detectable change in the florescence. However, we do expect the partial pressure of krypton to have a strong effect on the sum frequency generation. Krypton is negatively dispersive in this wavelength regime, affecting the phase matching conditions of the sum frequency process.

CONCLUSIONS

In summary, sum frequency generation in mercury with transform limited input beams provides a powerful spectroscopic source for production of tunable, bright, narrow-band, coherent radiation near the hydrogen Lyman- α transition. Although the VUV output increases rapidly with mercury temperature, apparent excimer formation limits the maximum temperature to $145°C$.

With 3 mJ 300 MHz linewidth beams at 3127 Å and tight focussing, the power broadening is larger than the isotopic frequency shifts, the Doppler width, or the input beam linewidth. This ensures that the beams interact with each atom in the mercury column. In spite of the high intensity of the input beams, there is evidence that the population of the 7^1S state is not significant enough to reduce the intensity of sum frequency generation.

REFERENCES

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