Tunable Coherent Vacuum-Ultraviolet Generation in Atomic Vapors*

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Two tunable dye lasers have been used to generate coherent radiation at a tunable sum frequency in the vacuum-ultraviolet spectral region. Spectral ranges of ~ 3500 cm^{-1} around 1895 Å and ~ 1200 cm^{-1} around 1798 Å have been covered. These ranges may easily be extended. Large ($\sim \times 10^4$) enhancements of $2\nu_1 + \nu_2$ sum mixing are seen when $2\nu_1$ is tuned to a double-quantum allowed transition from the ground state in Sr vapor.

The original work by Harris and co-workers[1] on extending nonlinear optical-mixing methods into the vacuum-ultraviolet (vuv) spectral range has raised great interest in such techniques. We have employed tunable dye lasers to take advantage of the resonant enhancements of the nonlinearities in atomic vapors in order to generate easily detectable, tunable, coherent, vuv radiation via third-order sum mixing. With the use of several combinations of dyes, spectrally narrow vuv beams were generated and continuously tuned from 1778 to 1817 Å (a range of ~ 1200 cm^{-1}) and from 1833 to 1957 Å (a range of ~ 3500 cm^{-1}). Tunable coherent radiation in this range will be useful for a variety of absorption and resonance fluorescence studies in simple atoms and molecules.

The resonant enhancements occur when the input lasers are tuned to double-quantum transitions. With a single dye laser, we have observed that the intensity of third harmonic generation (THG) in Sr vapor increases by many orders of magnitude when the fundamental frequency is tuned to the half-frequency of a two-photon allowed transition. Since only two photons are needed for this resonance, one is free to mix in the light from a second dye laser with no constraint on its frequency, thereby generating tunable output at the frequency $2\nu_1 + \nu_2$, where ν_1 is the frequency of the fixed laser at the half-frequency of the double-quantum transition, and ν_2 is the frequency of the tunable laser.

A block diagram of the experiment is shown in Fig. 1. The Molectron UV-1000 nitrogen laser produces ~ 1-MW, 7.5-nsec long pulses at ~ 15 pulses/sec. The collinear, orthogonally polarized dye laser beams leaving the Glan prism have powers between 15 and 100 kW each. The dye lasers incorporated optics[2] that narrowed the spectral bandwidths to ~ 0.1 and ~ 1 cm^{-1}, respectively. The beams were focused, via a 33-cm focal-length lens, into the center of a simple Sr vapor cell made of nickel and heated to the 800–900°C range. Helium gas at pressures from 100 to 700 Torr and two $\frac{1}{4}$-in. apertures placed 25 cm apart (the approximate length of the hot zone) prevented the Sr vapor from diffusing to, and coating, the Pyrex input and LiF output windows. The output light could be observed either directly or through a McPherson model 225 vuv spectrometer using an EMR 541GX-08-18 solar-blind photomultiplier. This detector has no response to direct beams from the dye lasers with $\lambda > 4500$ Å.

The importance of resonant enhancement was dramatically illustrated when a single, linearly polarized beam from a sodium-fluorescein dye laser was tuned from 5337 to 5710 Å. The vuv frequency-tripled light displayed enormous resonant enhancements—the signal increasing $\sim 10^4$ times from its weak, off-resonance value—at four wavelengths: 5380, 5409, 5605, and 5681 Å. These wavelengths correspond to exact half-frequencies of the following doubly excited, even-parity states of Sr: $(5p^2)\ ^1S_0$, $^1D_2$, $^3P_2$, and $^3P_0$, respectively. An energy-level diagram of Sr showing one such $5p^2$ state is given in Fig. 2. Strong THG was also observed when a rhodamine 6-G laser was tuned to 5757 Å, the half-frequency of the singly excited $5s5d\ ^1D_2$ state. Smaller resonant enhancements were noted when a 7-diethylamino-4-methylcoumarin laser was tuned to the half-frequencies of the $5s8s\ ^1S_0$, $5s7d\ ^1D_2$, $5s9s$

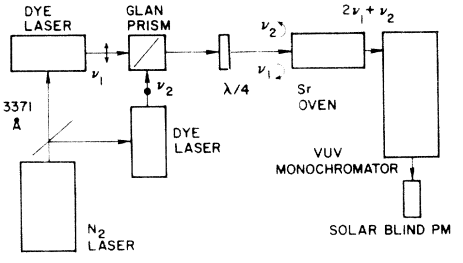

FIG. 1. Block diagram of the experiment.

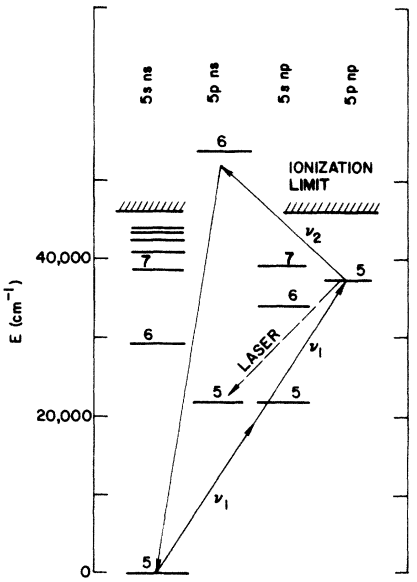

FIG. 2. Partial energy-level diagram of Sr atoms, showing resonantly enhanced tunable $2\nu_1+\nu_2$ generation using a $5p^2$ intermediate state.

$^1S_0$, and $5s8d\,^1D_2$ states. The shortest vuv wavelength obtained in this way was 1550 Å.

Using light at 5757 Å for THG, the vuv signal increased about 5 times when a few hundred Torr of xenon gas was added to the Sr vapor maintained at ~10 Torr. The addition of more Xe resulted in a sharp decrease of the vuv signal. We attribute this behavior to the dependence of the coherence length on the Xe pressure.[1]

By means of a simple technique to eliminate THG, a single-frequency tunable vuv light source was made using two dye lasers in the configuration of Fig. 1. Circularly polarized light cannot be frequency tripled in isotropic media since angular momentum would not be conserved.[3] A linearly polarized dye laser (ν_1) was tuned to 5757 Å and the THG signal was nulled using a $\lambda/4$ plate to convert the light to circular polarization. A second input tunable dye laser (ν_2) was then added collinearly by means of the Glan prism. The $\lambda/4$ plate converted some part of this light to circular polarization in the opposite sense, allowing strong vuv generation at $2\nu_1+\nu_2$. Tuning ν_2 sweeps the output at $2\nu_1+\nu_2$ over a tuning range as wide as that of ν_2. This tuning was verified for various combinations of dyes with the use of the vuv monochromator and also, in the case of direct photomultiplier detection, by sweeping the generated output wavelength over various known absorption lines such as one in CH_3I at 1927 Å.[4]

In Table I we present results for the tuning ranges we have covered. Table I also shows the tuning ranges to be expected through the use of other combinations of four commonly used dyes.[5] In each case, one laser is fixed at a double-quantum resonance while the other is tuned over its complete tuning range.

The nature of the resonant enhancement of the nonlinearity may be easily understood by considering the nonlinear susceptibility, $\chi^{(3)}$, which describes the sum mixing of the type $\nu_3=2\nu_1+\nu_2$. The nonlinear susceptibility may be expressed as[6]

$$\chi_{ijkl}^{(3)}\propto\sum_{\substack{g,m\\n,o}}N_g\frac{\langle g|\mu_i(3)|o\rangle\langle o|\mu_j(2)|n\rangle\langle n|\mu_k(1)|m\rangle\langle m|\mu_l(1)|g\rangle}{(\nu_{og}-\nu_3)(\nu_{ng}-2\nu_1)(\nu_{mg}-\nu_1)} \tag{1}$$

plus the sum of similar terms with the frequencies permuted. The summation in Eq. (1) is taken over all states of the nonlinear medium. N_g is the density of occupied states in the quantum state g, and $\langle n|\mu_k(\alpha)|m\rangle$ is the dipole matrix element of the kth component of $\mu(\alpha)$ between states m and n, where $\mu(\alpha)$ is the dipole operator component parallel to the input electric field at ν_α. In the frequency denominators, ν_{ng} is the energy difference (in units of frequency) between states n and g. Equation (1) shows the resonant enhancement which may be obtained by setting $2\nu_1$ equal to ν_{ng}. The use of a two-photon resonance has important advantages.[7] If ν_1, ν_2, or ν_3 is set equal to a resonance frequency, $\chi^{(3)}$ will be resonantly enhanced but the input light will be strongly absorbed. Similarly, if ν_3 equals a resonance frequency, the output light at the sum frequency is absorbed. In addition, it is hard to phase match[1,8] if ν_1, ν_2, or ν_3 is close to an allowed single-photon resonance. If, however, $2\nu_1$ equals the frequency of a double-quantum allowed transition, the light can only be absorbed by the relatively weak two-photon absorption. But Eq. (1) shows that the resonant enhancement of $\chi^{(3)}$ can be just as strong as for single-photon resonances. To realize the full potential of the resonant enhancement, the laser linewidth must be as narrow as the material linewidth, which may be

TABLE I. Tuning ranges in the vuv obtainable with various combinations of dyes with λ_1 tuned to the half-frequency of a two-quantum transition in Sr. The underlined entries show dye combinations used and tuning ranges achieved in this work. The other entries show the tuning ranges that would be covered with the remaining combinations of these four dyes.

Dye	7-Diethylamino-4-methylcoumarin	Mixture Coumarin 6 and Coumarin 102	Sodium Fluorescein	Rhodamine 6G
Resonant State λ_1	$5s7d\ ^1D_2$ 4779A	$5s5d\ ^1D_2$ 5032A	$5p\ ^1D_2$ 5409A	$5s5d\ ^1D_2$ 5757A
7-Diethylamino-[a] 4-methylcoumarin 4648-4525A	1578-1609A	1632-1666A	1710-1746A	1778-1817A
Mixture Coumarin 6 [b] and Coumarin 102 5000-5350A	1617-1651A	1674-1712A	1756-1797A	1827-1872A
Sodium Fluorescein 5337-5710A	1657-1685A	1710-1747A	1795-1836A	1870-1914A
Rhodamine 6G 5680-6111A	1682-1718A	1744-1783A	1833-1875A	1907-1957A

[a] One may tune 7-diethylamino-4-methylcoumarin to wavelengths shorter than 4400 Å, but for sum mixing, phase matching becomes impractical below 4648 Å since λ_2 approaches the main Sr resonance line at 4607 Å.

[b] Ref. 5.

$\sim 0.1\ cm^{-1}$ for atomic vapors.

In order for a resonantly enhanced term of the type expressed in Eq. (1) to make a large contribution to $\chi^{(3)}$, the matrix elements must correspond to allowed transitions. On this basis one expects a very strong effect from tuning $2\nu_1$ into resonance with the singlet $5p^2$ states, since the one-electron $5s$-$5p$ transition has an oscillator strength of order unity, and the two-photon resonance from the ground state to a $5p^2$ state can be viewed as two successive virtual transitions of this type, e.g., $5s^2\,^1S_0 \rightarrow 5s5p\,^1P_1^0 \rightarrow 5p^2\,^1S_0$. However, the large enhancement observed for the triplet states $^3P_2$ and $^3P_0$ of the $5p^2$ configuration is unexpected because the $\Delta S = 0$ selection rule is violated.

Another manifestation of the large $5s$-$5p$ two-photon cross section was the appearance of superradiant laser beams at 6566, 6550, and 4811 Å, corresponding to the transitions $5p^2\,^1S_0$-$5s5p\,^1P_1^0$, $5p^2\,^1D_2$-$5s5p\,^1P_1^0$, and $5p^2\,^3P_2$-$5s5p\,^3P_2^0$, respectively, when these upper levels were pumped via two-photon absorption. However, the two-photon absorption did not prevent large resonant enhancement of the THG or sum mixing.

In the course of tuning ν_2, we observed further strong vuv signal enhancement in the form of resonances. These additional enhancements appear to correspond to the sweeping of $2\nu_1 + \nu_2$ over various excited states lying above the first ionization limit. The location of such states is currently a subject of research.[9] With a $5p^2$ state acting as the intermediate double-quantum resonance, additional enhancement would *a priori* be expected to occur for such autoionizing states in Sr as $6s5p\,^1P_1^0$ (see Fig. 2), $5d5p\,^1P_1^0$, etc., which are optically connected to both the ground state and the $5p^2$ state. Using $5p^2$ and other known double-quantum resonances, it should be possible to elucidate the quantum numbers of the autoionizing levels.

We have further observed that the strongest vuv generation occurs when rather special conditions of resonance are met. For instance, the vuv signal generated at 1791 Å, with ν_1 tuned to 5757 Å and ν_2 tuned to 4742 Å so that $\nu_1 + \nu_2$ equals the frequency of the $5s7s\,^1S_0$ state, is stronger than all other observed vuv signals. vuv power measurements were performed under these conditions. With the laser power at ν_1 measured at 16 kW and the laser power at ν_2 measured at 1.6 W at the entrance to the Sr oven, the vuv power measured was 5.2×10^{-5} W. The Sr vapor pressure was 25 Torr, and 460 Torr of Xe was added for phase matching. The laser at ν_2 was attenuated by a factor of 1.1×10^4 to avoid saturating the detector. The power conversion efficiency from 4742 to 1791 Å was thus 3.3×10^{-5}. The vuv power output was measured to be linear in the power at ν_2 and quadratic in the power at ν_1 for powers low enough not to saturate the detector. Assuming that this linearity applies for much

higher powers, the unattenuated input power at ν_2 of 18 kW produces a vuv signal of 4×10^9 photons/pulse.

In summary, we have used the resonant enhancement of the nonlinear optical response in Sr vapor to produce tunable vuv radiation. Extension of the tuning range should be possible through the use of other vapor systems such as mercury and cadmium, and other dye lasers and frequency-doubled dye lasers. This method should produce diffraction-limited coherent light, tunable with a <0.1 cm^{-1} bandwidth from ~2300 Å, the present limit of frequency-doubled dye lasers, to ~800 Å.

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Time Dependence of Electron Scattering in a Beam-Plasma System*

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The time dependence of fast-electron production in a beam-plasma system is observed experimentally. We find that the fastest particles are produced in pulses which are correlated with the quenching of one mode and the onset of a second lower-frequency faster phase velocity of oscillation.

Heating of target plasmas by an intense electron beam is a potentially attractive method of forming a fusion plasma. Recent works have treated both experimental and theoretical aspects of this problem.[1,2] Since at least a part of the energy transfered to a target plasma by such beams is through the well-known two-stream instability, it is important to understand the details of the energy transfer by plasma wave generation. In the present note we will discuss observations of the time dependence of the scattering process in a moderate-energy beam-plasma system. We find direct evidence for the correlation of faster modes and the generation of faster particles. The scattering process exhibits a sequential time dependence; the quenching of the most intense (dominant) mode is correlated with both the excitation of less-intense (subdominant) lower-frequency modes of oscillation and the generation of fast-particle pulses.

The experimental system has been described previously.[3] It consists of a beam-formed hydrogen plasma with density between 10^9 and 10^{10} cm^{-3}, beam energy between 500 and 1500 V, and beam currents between 1 and 20 mA. A strong axial magnetic field (~3400 G) renders the electron motion effectively one dimensional. The spectrum is observed with electrostatic loops surrounding the plasma, and the electron distribution is measured with a retarding-field energy analyzer.

The dispersion for the plasma column in the frequency and parameter range of interest to the present experiment is shown in Fig. 1(a). The dispersion was measured interferometrically using one fixed and one moving probe. We should note that it exhibits a slight negative curvature. Lower-frequency modes are slightly faster.

The relative stability of modes on this branch of the dispersion can be controlled with the beam