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ABSTRACT

An ultra-high brightness laser system has been developed to study the spectroscopy and dynamics of molecules and clusters in the vuv-xuv spectral region. The laser utilizes pulse amplification of a single-mode ring dye laser, frequency doubling, and four-wave mixing in a pulsed jet. Pulse energies of >100mJ in the visible and $>10^{11}$ photons/pulse in the vuv-xuv have been obtained. The bandwidth of the laser has been measured to be 91MHz in the visible and 210MHz in the xuv.

I. Introduction

In recent years there have been many advancements in vacuum ultraviolet (vuv) and extreme ultraviolet (xuv) light sources. These include improved continuum sources, synchrotron radiation, laser produced plasmas and four-wave mixing of pulsed ultraviolet lasers. All of these methods have their own advantages and disadvantages depending on the application involved.

In our laboratory, vuv and xuv light is used to study the spectroscopy and dynamics of photoionization processes of small molecules and clusters. Such experiments have very stringent requirements on the light source for the following reasons. First, individual transitions to be observed could have narrow linewidths (typically <200MHz in the absence of fast decay channels). Second, direct photoionization cross sections are generally small, from $10^{-17}cm^2$ to less than $10^{-20}cm^2$. In addition, for certain species (e.g. clusters) it is hard to obtain large number densities for investigation. Thus, a source of high intensity and excellent spectral purity is needed for a good signal-to-noise ratio. Finally, these studies use a wide range of photon energies thus requiring tunability over the broad vuv and xuv regions.

The laser system presented in this paper is designed to meet these needs. It has a nearly transform-limited bandwidth of <100MHz in the visible for a 7ns (FWHM) pulse.

It also has a high pulse energy of typically around 100mJ in the visible and 30mJ in the ultraviolet with good beam quality in order to achieve a good conversion efficiency into the vuv and xuv regions. With the use of frequency tripling and mixing techniques it has been tuned over a broad range in the vuv and xuv producing fluxes of $>10^{11}$ photons/pulse in a bandwidth of 210MHz ($0.007cm^{-1}$).

The basic technique used in the construction of this laser system is pulse-amplification of a single-mode dye laser. This approach was first demonstrated by M. M. Salour[1] using a nitrogen laser as the pump laser. Megawatt power in the visible was later achieved using the second harmonic of a Nd:YAG laser as pump.[2] Recently several groups have extended these narrowband sources to the vuv.[3,4] We describe here the aspects of the design and performance of our system that have significantly distinguished this laser from those reported previously by providing a source of exceptionally high spectral brightness and versatility.

II. Design

Nearly all the techniques used in this system have been demonstrated previously. However, the way the different components are combined is what makes our set-up unique. In brief, we start with a narrowband c.w. ring dye laser which is then pulse amplified by the second harmonic of a Nd:YAG laser. This is accomplished with a three-stage, four-pass dye amplifier chain built in our laboratory. The beam of pulsed visible photons is then sent through a frequency doubling crystal and the uv photons generated are used to make vuv and xuv radiation via third harmonic generation or four-wave mixing in a pulsed atomic or molecular beam[5,6,7]. A schematic of the system is shown in Figure 1.

The starting point of the system is a Coherent 699-29 Autoscan ring dye laser pumped by an argon ion laser. This gives a well characterized light beam of 1MHz bandwidth continuously tunable over the whole visible spectrum. The dye laser is optically isolated from the amplifier chain with a permanent magnet Faraday isolator to prevent mode-hopping induced by the feedback of amplified spontaneous emission (ASE) from the amplifiers.

The second key component of the laser system is the injection seeded Nd:YAG laser(Quantel model 592). An important limiting factor on the bandwidth of the system is the temporal profile of the amplified dye laser pulse. This, in turn, is determined mainly by the profile of the pump laser pulse. By utilizing injection seeding, thereby making the Nd:YAG laser output single-longitudinal-mode, the temporal profile of the pump and the dye lasers are made as smooth as possible resulting in the narrowest bandwidth. Use of the injection seeded Nd:YAG laser also produces a more stable uv and xuv power. This is again due to the single-mode operation which eliminates the mode-beating induced modulations seen in the temporal profile of multimode lasers.

The dye cells used are similar to the prism cells developed by D.S. Bethune for an excimer pumped dye laser.[8] These are side-pumped cells with the pump beam being internally reflected into the dye. Similar systems have recently been used to amplify ultrashort pulses with high gain.[9,10] The prism type dye cell has certain advantages associated with it. The dye is pumped from all sides so the gain can be made uniform to preserve the spatial quality of the seeding beam. There is no window damage by the pump laser because of the large pumping area. Finally, the amplified beam's spatial profile is decoupled from that of the pump beam. This enables the system to be used with pump sources that have very different beam shapes.

Three dye amplifiers are used in our design. The first cell has a bore size of 1mmφ x 20mm long. The second cell, which is double-passed, has a 3mmφ x 30mm long bore. The final amplifier is 6mmφ x 60mm long. We find that by adjusting the dye concentration such that one dye absorption depth at the pump wavelength equals approximately the bore diameter the best output beam profile and power combination are obtained. In this design the amplifier gain is changed by varying the pump beam intensity at each stage.

Telescopes consisting of a lens-pinhole-lens combination and magnifying powers of x1, x3 and x2 are used to shape the c.w. dye laser beam profile to fill the bore of the first, second and third amplifiers respectively. The pinholes serve as spatial filters to maintain a diffraction-limitied profile for the dye beam as well as to control the growth of ASE which could deplete the gain in the amplifiers.[2]

VUV generation is done in a pulsed jet of atoms or molecules. This process has been described in detail previously.[7,11,12] Here we list a few of its features. First, there are no exit windows for the tripled light so there is no short wavelength cut-off. Second, the length of the tripling region is not necessarily greater than the confocal parameter of the beam so the tripling medium does not have to be negatively dispersive.[13] Finally, for the shorter wavelengths the problem with self-absorption is reduced. One change we have made compared to our previous vuv generation experiments is the use of a large travel PZT driven pulsed valve.[14] This new home-made valve (operated with a 1mm diameter nozzle) allows for a larger throughput than the commercial valves that we used previously giving nearly a factor of ten increase in number density in the tripling medium which in turn results in a greatly improved tripling efficiency. Together with a higher uv power, up to two orders of magnitude improvement in xuv signal has been seen with the new valve.

The last component to be discussed here is the 1m normal incidence monochromator (McPherson model 225) used to seperate the photons of different wavelengths present in the laser beam. While the monochromator reduces the vuv-xuv beam intensity by a large factor it introduces a tremendous amount of experimental versatility making it an invaluable component in the laser system. The monochromator offers two major conveniences. The first, as mentioned above, is that it seperates out the fundamental light beams, permitting the use of vuv and xuv photons without interference of the powerful uv and/or visible beams. The second is that the curved grating recollimates the vuv-xuv beam allowing an optimal spatial overlap of the photon beam with our molecular beam in the ionization chamber. Optical damage to the grating is minimized by expanding the uv beam to fill the size of the grating.

III. Results

A. Visible

Through three stages of the amplifier chain a gain of approximately 10^8 is obtained with a Nd:YAG pump laser energy of 450mJ per pulse at 532nm. The pump energy is distributed in the ratio of 1:8:91 resulting in a gain of 10^3, $3x10^3$ and 33 for the three stages respectively. The laser system has been operated in the visible from 562nm to 620nm using R590 dye in the c.w. dye laser and various dyes in the amplifiers. The power output curves for these dyes under normal operating conditions are shown in Figure 2. The maximum output obtained so far has been 120mJ at 570 nm, the peak of R590 dye. However, at this power the beam contains some hot spots in its spatial profile. Under operating conditions that result in a uniform beam, the pulse energy is approximately 10% less than the maximun energy obtainable. ASE is depleted by the amplified c.w. beam to less than 10% of the measured energy over most (~90%) of the tuning range. Near the edge of the range the ASE is effectively subdued by using a different dye in the final amplifier. The final near-field beam profile is a circle 6mm in diameter with hard edges and a few diffraction rings. The diffraction rings come from the aperturing of the near-Gaussian beam by the bore of the amplifier cells. These rings can be minimized by reducing the c.w. laser beam diameter. However, this causes a loss in output power of 20% or greater. A compromise between spatial beam quality (both uniformity and extent of diffraction rings) and pulse energy is required.

The temporal profiles of the Nd:YAG and pulsed dye laser beams are shown in Figure 3. The dye pulse shows significant deviation from the near Gaussian profile of the pump beam. The fast rise in the front end of the pulse is caused by the exponential gain of the amplifiers. Double passing the second amplifier contributes to the slight broadening at the top. The steep fall and shortening of the pulse to 6.5ns is due to gain saturation in the second and final amplifiers. These factors will contribute to the spectral broadening of the pulse discussed below.

Bandwidth measurements were done with two different confocal etalons: a 300MHz FSR etalon with a finesse of 100 and a 1.5GHz FSR etalon with a finesse of 130. In both cases the c.w. and pulsed signals through the etalons were measured simultaneously as the c.w. dye laser was scanned. The pulse bandwidth, measured with R590 dye in the amplifiers, was found to be 91MHz (see Figure 4). In comparison, the Fourier transform-limited bandwidth for a Gaussian pulse 7ns wide is 32MHz. The discrepancy between these two numbers is consistent with that observed in other pulse-amplified single-mode systems.[2] An important contributing factor to the larger than transform-limitied bandwidth is the exponential dye gain that produces the non-Gaussian temporal profile of the pulse described above. This profile, in turn, is dependent on the amplifier dye. Thus slightly different bandwidths are observed in the other dyes used. No power broadening of the bandwidth is seen. In fact, a gain narrowing effect of roughly 5-10% occurs when the pump power is increased. Hence the entire origin of the broadening is still not well understood. Frequency pulling of the amplified beam was also seen. The amplified beam was shifted by a few MHz to the blue of the c.w. beam. This effect has been observed previously[15] and attributed to a Stark shift by the high electric field produced in the dye by the pump laser. However, in our laser the pump beams are not focused in the dye cells so the electric field strength is much lower then in conventional dye cells. We have observed that this shift is configuration and dye dependent. For high absolute accuracy the shift must be measured for the experimental conditions used. The exact cause of the shift will be a focus of future experiments.

The present scanning capabilities are set by the 699-29 laser. These include a minimum step interval of 1MHz and a slowest scanning speed of 10MHz per second. An independently controlled scanning speed would be most desirable especially for experiments with poor signal to noise.

B. Ultraviolet

Second harmonic generation is done with either an INRAD 5-12 autotracking unit or a Quanta-Ray WEX1 utilizing KDP crystals. The maximum conversion efficiency measured (after reflection losses at optical surfaces were accounted for) was 40% with a well collimated beam. During operation, when the beam must be slightly diverging for the doubling units to track, the conversion efficiency is around 30-35%. The uv beam profile is very similar to that of the visible beam with the diffraction rings being more pronounced. Extension to short uv wavelengths at around 220nm is done by mixing the frequency doubled dye with the 1064nm beam. Here, pulse energies of about 5mJ have been obtained. At the moment this conversion is limited by the divergence and large spot size of the ir beam ($dia_{ir} > 2dia_{uv}$). We expect even higher conversion with an optimized matching of the uv and ir beams.

C. VUV & XUV

The efforts to produce high power single-mode visible and uv light with good spatial control are to ensure a good conversion efficiency in generating single-mode vuv and xuv radiation. A 25cm focal length lens is used to focus the input beams at the pulsed molecular jet several mm downstream from our home-made pulsed nozzle source. The generated radiation and the fundamental beam(s) are dispersed by the differentially-pumped monochromator. This source has been tuned from wavelengths around 124nm to wavelengths around 74nm. A Quanta-Ray DCR-2 Nd:YAG/PDL dye laser system is used as the second laser when mixing of two lasers is called for. Table I summarizes the wavelengths we have studied to date, showing the generation scheme used, the generating medium and the incident pulse energies. These wavelengths are chosen to match our current ultra-high resolution studies on Kr and H_2 systems.[16] Continuous tuning is accomplished either by scanning the cw dye laser (MHz resolution) or the second laser ($1cm^{-1}$ resolution). Other wavelengths can be obtained easily by choosing the appropriate input wavelengths and generating medium. The narrowest laser bandwidth is achieved by non-resonant third harmonic generation of the near-transform limited beam. To test the spectral resolution we scanned the 4p-7s $^2P_{3/2}(J=1)$ transition of ^{86}Kr in a collimated atomic beam with a 30MHz Doppler width at 94.5nm using this source (see Figure 5). The vuv monochromator was set at 1st order. Detection was by ionization of the excited Kr atom with a second uv laser beam maintained at well below 1mJ to minimize power broadening. After correcting for contributions from Doppler broadening we calculate that the xuv bandwidth is 210MHz.

The high brightness of our uv beam and the improved valve helped to increase the vuv-xuv output compared to previously obtained signals by as much as two orders of magnitude. The output reported in Table II is what is available at the input to the monochromator. The number of photons/pulse was derived from measurements of direct ionization of NO made with a single ion chamber.[17] Typical usable fractions of photons are from 20% at λ>120nm to ~1.5% at 74nm.

In some cases, we have observed gas breakdown at the focal point of the uv beam as a result of its high brightness. This causes spectral broadening of the vuv beam. The problem is remedied by increasing the distance between the pulsed nozzle and the laser focal point. This necessarily reduces the output power of the vuv and must be taken into account when using the source. A complete summary of the laser's performance is given in Table II.

IV. Summary

We have described a laser system capable of continuously scanning over much of the visible, uv, vuv and xuv spectral regions with high pulse energy and a near transform-limited bandwidth. A second pulse-amplified single-mode dye laser system is under construction to replace the broadband dye laser in the mixing schemes to make xuv light. This should provide full single frequency coverage of the entire spectrum discussed above. Future plans include the use of high power excimer lasers to facilitate efficient pumping of dyes in the blue-green region, use of tunable solid-state amplifiers and the use of high repetition rate pump lasers for higher average power.

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References

1. M.M. Salour, Opt. Commun., **22**, 202 (1977).
2. P. Drell and S. Chu, Opt. Commun., **28**, 343 (1979).
3. L. Cabaret, C. Delsart and C. Blondel, Opt. Commun., **61**, 116 (1987).
4. C.H. Muller III, D.D. Lowenthal, M.A. DeFaccio and A.V. Smith, Opt. Lett., **13**, 651 (1988).
5. A.H. Kung, Opt. Lett., **8**, 24 (1983).
6. J.Bokor, P.H. Bucksbaum and R.R. Freeman, Opt. Lett., **8**, 217 (1983).
7. R.H. Page, R.J. Larkin, A.H. Kung, Y.R. Shen and Y.T. Lee, Rev. Sci. Instrum., **58**, 1616 (1987).
8. D.S. Bethune, App. Opt., **20**, 1897 (1981).
9. C. Rolland and P.B. Corkum, Opt. Commun., **59**, 64 (1986).
10. R. Falcone, private communication.
11. C.T. Rettner, E.E. Marinero, R.N. Zare and A.H. Kung, J. Phys. Chem., **88**, 4459 (1984).
12. D.S. Bethune and C.T. Rettner, IEEE J. Quantum Electron., **23**, 1348 (1987).
13. G.C. Bjorklund, IEEE J. Quantum Electron., QE-11, 287 (1975)
14. D. Proch, T. Trickl, to be published.
15. C. Wieman and T.W. Hansch, Phys. Rev. A, **22**, 192 (1980).
16. T. Trickl, M. Vrakking, E. Cromwell, Y.T. Lee and A.H. Kung, to be published.

17. J.A.R. Samson, **Techniques of Vacuum Ultraviolet Spectroscopy**, (John Wiley & Sons, Inc., New York, 1967), p 266.

TABLE I: EXAMPLES OF WAVELENGTHS GENERATED

WAVELENGTH (nm)	ν (cm^{-1})	ν_{vis} (cm^{-1})	Amplifier Dye	ν_{uv}(=2ν_{vis}) Energy (mJ)	Generating Scheme	Generating Medium
123.6	80917	17578	R590	35*	$3x\nu_{uv}$	CO
117.8	84910	16982	Kiton Red	30	$(2x\nu_{uv})+\nu_{vis}$	Xe
116.5	85847	17169	R610 (basic)	20	$(2x\nu_{uv})+\nu_{vis}$	Xe
100.1	99894	16649	R640	15	$3x\nu_{uv}$	Ar
98.2	101868	16978	Kiton Red	30	$3x\nu_{uv}$	Ar
95.1	105146	17524	R590	35	$3x\nu_{uv}$	Xe
80.4	124344	ν=17768	R590	4‡	$2x\nu_{uv}+\nu'_{uv}$	Xe
		ν'=17242†	R590	15		
74.2	134792	17768	R590	4‡	$3x\nu_{uv}$	Xe

*$\nu_{uv}=\nu_{ir}+\nu_{vis}$ (ν_{ir}=Nd:YAG fundamental)

†Second laser

‡$\nu_{uv}=2\nu_{vis}+\nu_{ir}$

TABLE II: LASER SYSTEM PERFORMANCE

	VISIBLE	U.V.	VUV-XUV
ENERGY	>100mJ	>30mJ	$>10^{11}$ photons/pulse†
BANDWIDTH (FWHM)	91MHz	<140MHz*	210MHz
PULSE LENGTH	7ns	<7ns	<7ns
PRESENT TUNING RANGE	562-620nm	281-310nm 222-240nm	74-124nm
TEMPORAL PROFILE	Near Gaussian	Near Gaussian	Near Gaussian

†Measured at 98nm

*Estimated

FIGURE CAPTIONS

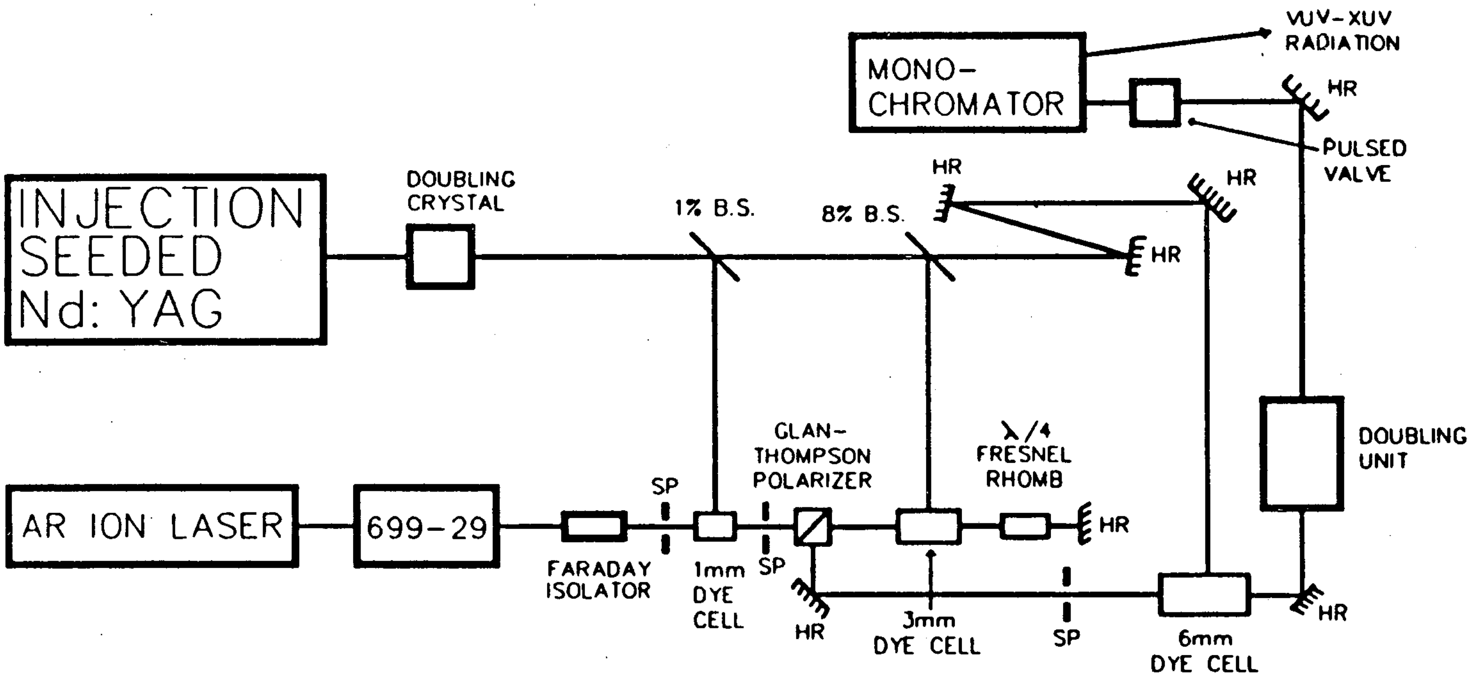
Figure 1. Laser system schematic.(BS=beam splitter, HR=high reflecting mirror, SP=spatial filter/telescope)

Figure 2. Output response curves of the dye laser for various dyes (*denotes basic solution).

Figure 3. Upper trace: Temporal profile of the injection seeded Nd:YAG laser pulse(FWHM=10ns). Lower trace: Temporal profile of the dye laser pulse(FWHM=7ns).

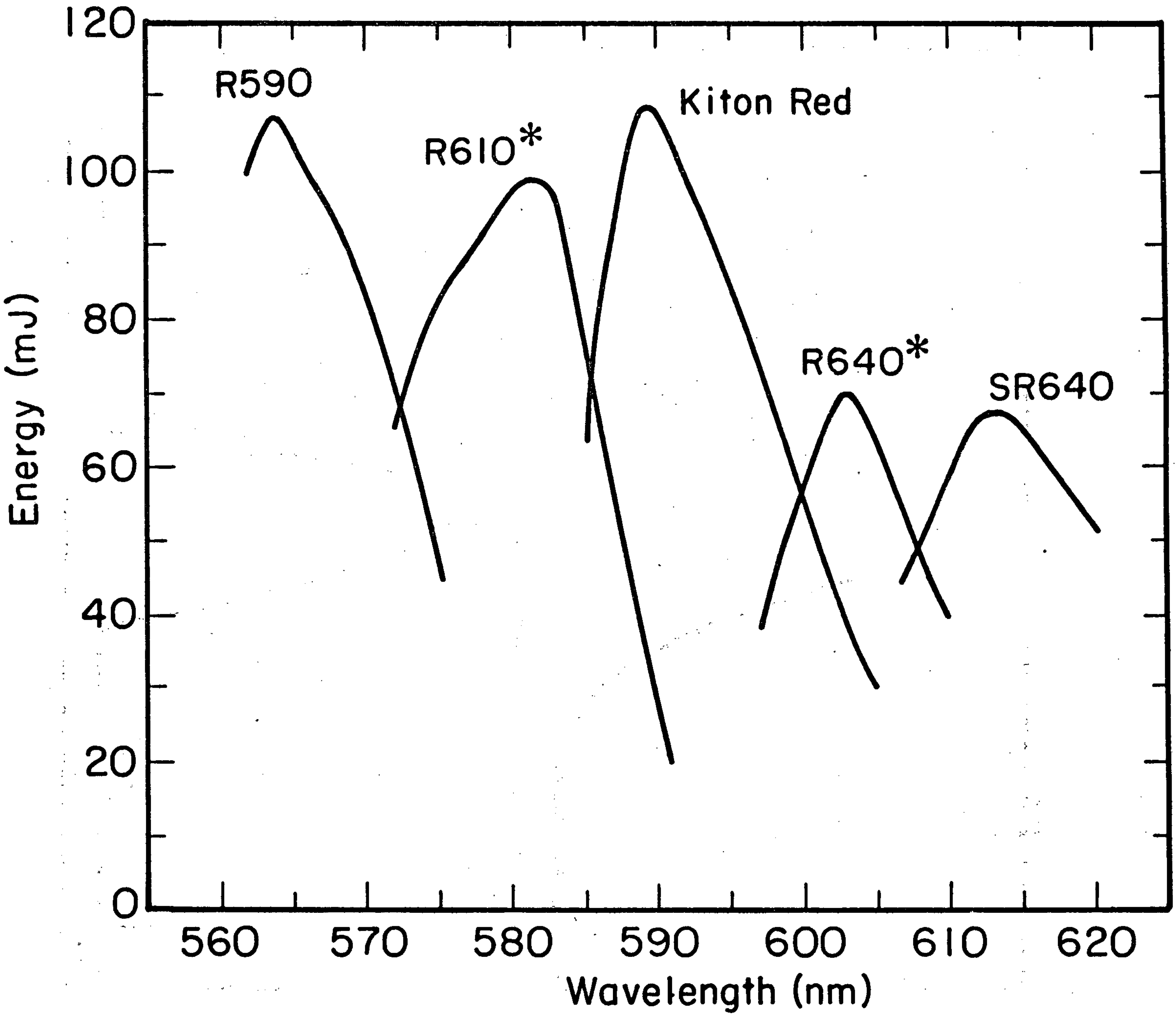
Figure 4. Scan of pulsed dye laser through a 300MHz etalon with R590 dye in the amplifiers (FWHM=91MHz).

Figure 5. Scan of the 4p-7s $^2P_{3/2}$(J=1) transition of Kr at 94.5nm. FWHM=210MHz(0.007cm^{-1}).



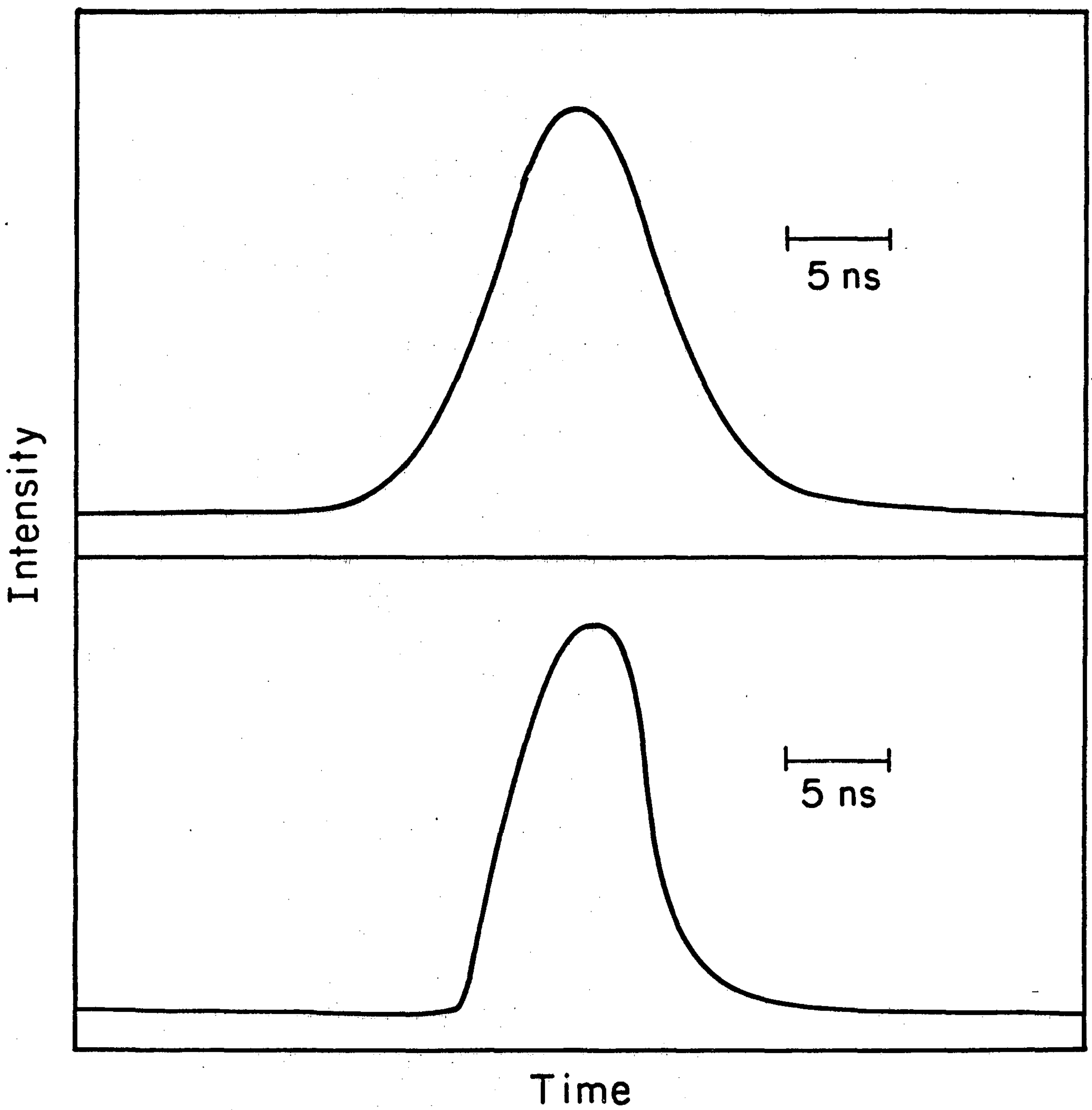
XBL 883-3124

Figure 1.



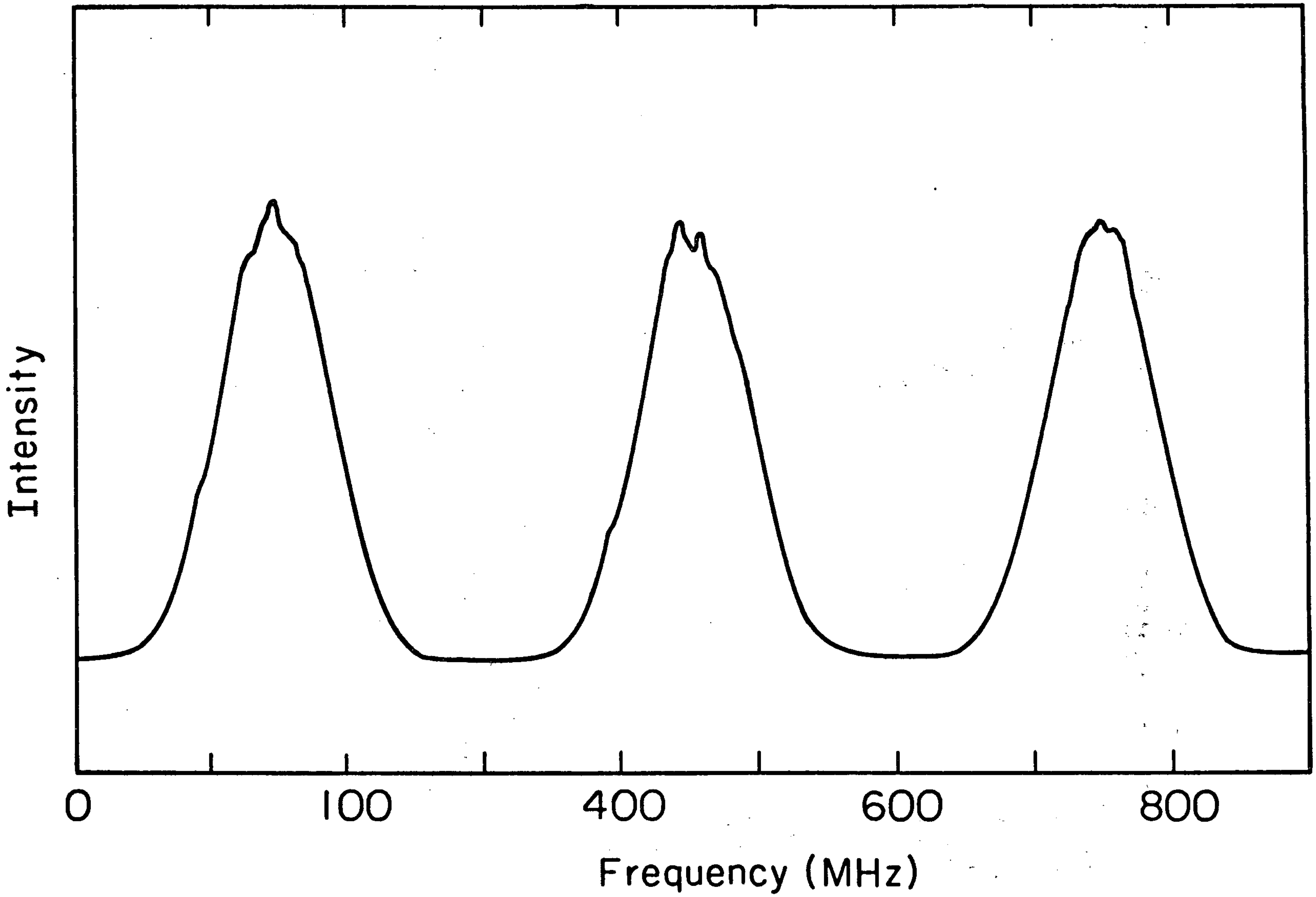
XBL 888-3125

Figure 2.



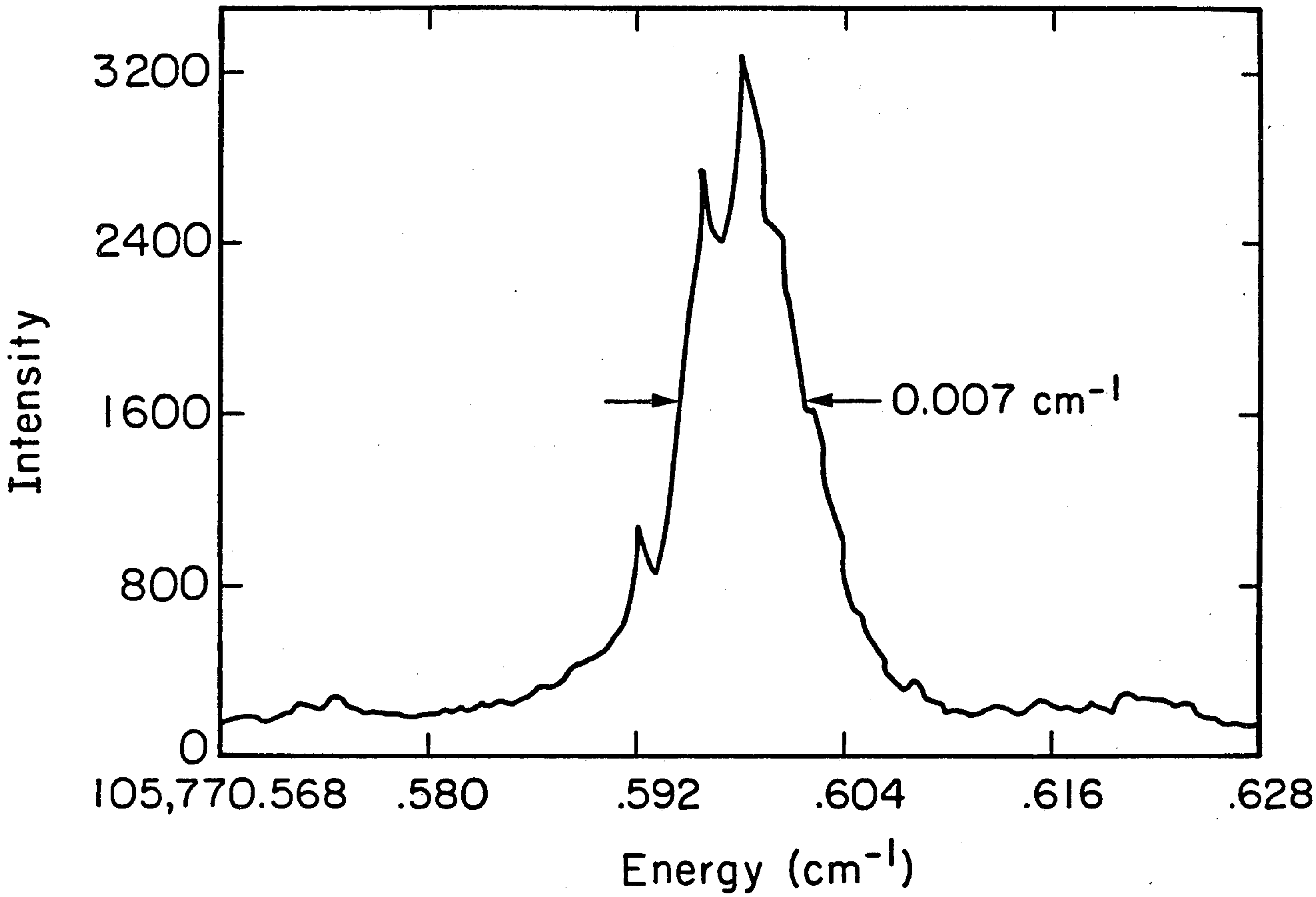
XBL 888-3128

Figure 3.



XBL 888-3127

Figure 4.



XBL 888-3126

Figure 5.