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Survey of Lasers for Spectroscopic Use: Optical and Ultraviolet

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Survey of lasers for spectroscopic use: optical and ultraviolet

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The principles and operation of narrow-bandwidth and mode-locked tunable dye lasers are described and the extension of their effective tuning ranges by harmonic generation and stimulated Raman scattering is discussed. Methods of producing high-power ultraviolet radiation using excimer lasers and techniques for vacuum u.v. generation by frequency mixing are also described.

1. Introduction

In many respects, the laser is an ideal spectroscopic source of visible and ultraviolet radiation. The spectral brightness of lasers can be many orders of magnitude greater than that of incoherent sources. Because of their very small bandwidths, continuously tunable single-mode lasers permit resolving powers exceeding those of conventional spectrometers. The capability of mode-locked lasers to deliver intense pulses of light with pulse widths of 10^{-13} s allows the study of ultra-fast transient phenomena. Furthermore, when visible lasers are used in combination with nonlinear optical frequency mixing techniques such as harmonic generation, intense continuously tunable radiation is available throughout the visible, ultraviolet and vacuum ultraviolet (v.u.v.) regions of the spectrum, where atoms and molecules have strong electronic resonances.

The visible and near infrared regions of the spectrum are dominated by the dye laser which, by suitable choices of dyes, is continuously tunable from 340 nm to 1000 nm. The more recently developed excimer gas lasers provide intense radiation over limited bandwidths throughout the ultraviolet region. As well as being useful sources of tunable u.v. radiation, they have found application as optical pumps for dye lasers and as amplifiers for radiation produced by the generation of the second and third harmonics of dye laser radiation, thereby permitting the efficient production of tunable v.u.v. radiation by optical frequency mixing.

2. Narrow-bandwidth dye lasers

2.1. *Continuous-wave dye lasers*

A dye laser may be made to operate continuously by pumping with a c.w. argon or krypton ion gas laser. A typical configuration of such a system when operating as an oscillator is shown in figure 1. The pump beam is focused by a concave mirror into the dye, which is in the form of a free-flowing jet of thickness *ca.* 100 μm. In the absence of tuning elements, the bandwidth of the laser output is *ca.* 3 nm. Pumping efficiencies of 25–30 % can be achieved, and output powers of up to 1 W can be produced in the region 400–1000 nm by suitable choice of dye and pumping laser.

If narrow bandwidths and frequency tuning are required, frequency-selective elements must be inserted into the resonator. Bandwidths of *ca.* 0.1 nm are obtained by using a single dispersing prism or a birefringent filter and the laser can be tuned continuously across the gain bandwidth of 50–100 nm by simply rotating the tuning element. If very narrow line widths are to be obtained, the longitudinal mode structure of the laser resonator must be taken into account. The spectrum of the output of a laser oscillator consists of a number of longitudinal modes equally spaced in frequency by $c/2L$, where L is the length of the resonator. To isolate a single mode requires the use of several frequency-selecting elements in the cavity, and in general Fabry–Perot etalons with different free spectral ranges are used in conjunction with a

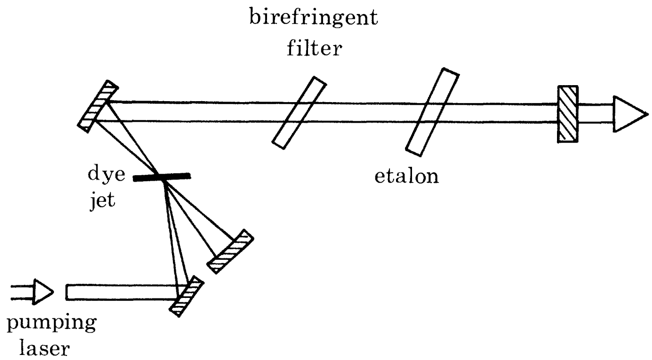

Figure 1. Schematic diagram of a c.w. dye laser oscillator.

relatively low-dispersion element such as a birefringent filter. By suitable choice of finesse, thickness and orientation of the etalons, a single mode of the resonator can be isolated. However, continuous tuning of a single mode laser requires simultaneous variation of the resonator length L and the transmission maxima of the etalons. The cavity length can be varied either by mounting one of the mirrors on a piezoelectric transducer or by placing a plane parallel plate called a galvo plate within the resonator at close to the Brewster angle. By tilting the galvo plate the optical length of the resonator and hence the frequencies of the modes can be scanned. The transmission maxima of the etalons can be varied using piezoelectric transducers, by tilting or by pressure changes. For a laser employing two etalons, a single mode may be tuned continuously over a limited range (*ca.* 30 GHz) by scanning the cavity length and the etalon with the smaller free spectral range. Tuning over the entire laser bandwidth requires simultaneous tuning of all the frequency-selective elements and of the cavity length and this requires sophisticated control systems.

Fluctuations in the absolute frequency of a single-mode laser are caused by statistical fluctuations in the density of the dye solution and by acoustic vibrations in the resonator, and limit the frequency stability to *ca.* ± 10 MHz. This can be reduced by comparing the frequency of the dye laser with a resonant frequency of an external temperature-stabilized reference cavity whose resonant frequencies are in turn controlled by reference to a stabilized He–Ne laser (Gerhardt & Timmermann 1977). By using the error signal to drive a piezoelectric transducer attached to a lightweight end mirror of the dye laser, the frequency can be stabilized to *ca.* 100 Hz.

When a laser is pumped above threshold, the gain is depleted by saturation. In a laser oscillator, a standing wave is created within the resonator and gain depletion is greatest at the antinodes, whereas there are regions of unsaturated gain at the nodes. This spatial hole-burning promotes hopping to adjacent modes and can only be controlled by the use of several high-finesse frequency selective elements with consequent optical losses and reduction in laser output power. This problem is overcome in a ring dye laser in which a travelling wave propagates in one direction. A diagram of a ring laser is shown in figure 2. Spatial hole-burning is eliminated and since dyes may be regarded as homogeneously broadened, single-mode selection is achieved with fewer low-finesse frequency selecting elements in the cavity. Consequently, cavity losses

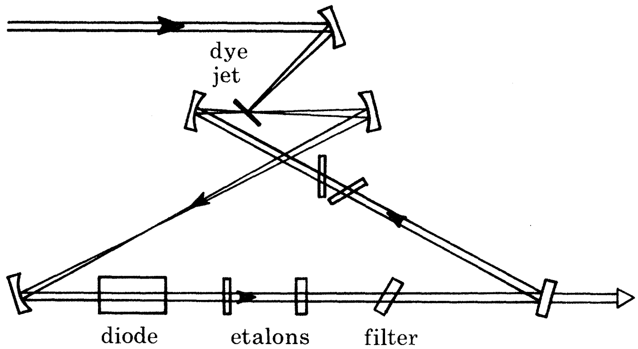

FIGURE 2. A frequency stabilized ring dye laser.

are reduced and output powers increased. To ensure unidirectional operation, an optical diode is usually inserted into the cavity. This consists of a Faraday rotator and birefringent crystal in series. Since the polarization rotations produced by each component are cumulative in one direction, but of opposite sign in the other direction, any polarization-dependent elements in the cavity such as Brewster-angled galvo plates will ensure unidirectionality. Commercially available frequency tunable ring lasers have short-term frequency stability of *ca.* 1 MHz and output powers of *ca.* 1 W over a continuous tuning range of 30 GHz.

2.2. *Pulsed dye lasers*

C.w. dye lasers provide very narrow line widths and are very useful as sources for linear spectroscopy. However, for nonlinear spectroscopic processes and harmonic generation for u.v. and v.u.v. generation, higher output powers are desirable. This requires the use of pulsed lasers, pumped either by flashlamps or by other lasers. An inevitable consequence of using pulsed systems is that the line width of the laser is increased and is at best Fourier transform limited. For a 10 ns pulse, this corresponds to a minimum bandwidth of *ca.* 50 MHz. At $\lambda = 600$ nm, this corresponds to a resolving power of 10^7, comparable with that of a high-quality Fabry–Perot etalon used in conventional high-resolution spectroscopy. Flashlamp-pumped dye lasers can produce pulses of 100 ns–100 μs duration, and for some applications they constitute an economic alternative to c.w. lasers, which require expensive ion lasers for pumping. However, narrow bandwidth operation with long pulses is hampered by thermally induced inhomogeneities in the dye medium, but by restricting the duration of the output pulse to a few hundred ns, line widths of less than 1 GHz and even a single longitudinal mode can be obtained (Gale 1973). Flashlamp-pumped dye lasers can provide high-energy output pulses (*ca.* 10 J), average output powers of up to 100 W (Jethawa *et al.* 1978) and are also suitable as amplifiers. The relatively long pulses make these lasers very suitable for the production of ultra-short pulses by mode-locking (see §3).

The pumping of dye lasers with short (*ca.* 10 ns) pulses of radiation from a N_2 laser (λ = 337 nm) excimer laser (e.g. XeCl, λ = 308 nm) or a harmonic of a Q-switched neodymium YAG laser ($\lambda_{2\omega}$ = 530 nm, $\lambda_{3\omega}$ = 355 nm) provide convenient methods of generating tunable laser radiation throughout the visible and ultraviolet spectral regions. In the most widely used transverse geometry, the pumping radiation is focused with a cylindrical lens into the dye cell, where it is absorbed in a thin layer very close to the entrance window. As for the c.w. dye lasers, frequency selection and tuning can be achieved by the use of etalons, but gratings offer greater simplicity of tuning. If the dye laser beam is expanded to fill the tuning grating, this ensures the maximum resolving power and reduces the laser line width. Beam expansion also decreases the

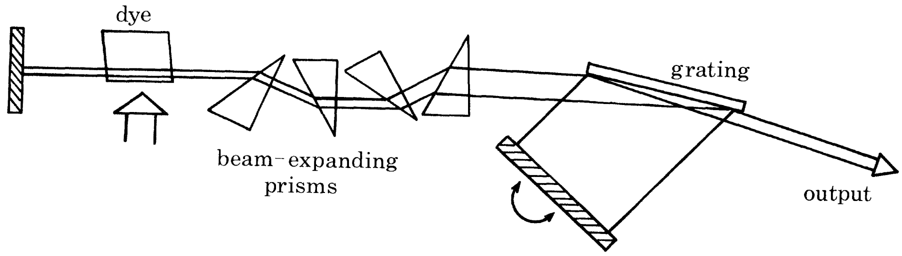

FIGURE 3. Configuration of a laser-pumped tunable dye laser with grazing-incidence grating and beam-expansion prisms.

power density on the grating and prevents optical damage. The beam may be expanded by a telescope (Hänsch 1972) or by using the grating at near-grazing incidence (Soshan *et al.* 1977), as shown in figure 3. This latter technique has the advantage of simplicity of construction and the capability of producing very narrow line widths that are continuously tunable over a wide spectral range. However, at grazing incidence the efficiency of holographic gratings is very poor. Consequently, high pumping powers are required, and can give rise to undesirable thermally induced optical inhomogeneities in the dye. This problem can be alleviated by the use of multiple-beam expanding prisms, which permit an increase of the grazing angle, reduction of cavity losses and hence of pumping power. This design permits the use of very short laser cavities with consequent wide spacing of longitudinal modes and increased number of cavity loops during the short pumping pulse. This increases the frequency spacing of the longitudinal modes, aiding single mode selection by additional intracavity elements. In addition, the increased number of cavity transits reduces the transmission bandwidths of the frequency-selective elements below their passive values, enabling fewer or lower-finesse elements to be used. Line widths of *ca.* 1 GHz can be produced by using a grating and mirror as shown in figure 3. By replacing this mirror with a Littrow grating, the spectrum may be further narrowed so that it is dominated by a single mode. Line widths of *ca.* 300 MHz and time-averaged values of 750 MHz have been reported for this technique. The line width of a laser may be further reduced to values close to the Fourier transform limit by passing the radiation through a high-finesse (*ca.* 200) confocal Fabry–Perot interferometer external to the laser cavity. Line widths of 100 MHz can be produced by this method. Continuous smooth tuning of the laser without the external interferometer can be achieved by pressure tuning. The tuning elements (including air-spaced etalons, if used) are enclosed in a pressure-tight chamber and by pressurizing with either N_2 or a more refractive gas such as propane, a laser operating in the visible region may be tuned by up to *ca.* 4000 GHz (Wallenstein & Zacharias 1980).

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To obtain the most reliable longitudinal and transverse mode control in a laser-pumped dye laser, very low pumping pulse energies (*ca.* 1 mJ) are required. The efficiencies of such oscillators operating close to threshold are low (*ca.* 1 %) and for most applications amplification is necessary. Because pulses of several hundred mJ are available from commercially available excimer and Nd:YAG (second harmonic) lasers and because dyes have high efficiencies (*ca.* 20 %) throughout the visible and ultraviolet regions, amplification of 10^3–10^5 is readily achieved in three stages and output powers of several MW may be produced. Spectral and spatial filtering between oscillator and amplifiers is often necessary to suppress amplified fluorescence and improve the beam divergence of the laser (Wallenstein & Zacharias 1980). Continuous coverage of the spectral region 340–950 nm may be obtained by a suitable choice of dyes and pumping sources (Teller *et al.* 1981).

3. Mode-locked lasers

The minimum pulse duration that may be generated by a laser is given by the Fourier transform of the spectral bandwidth of the amplifying medium. For atomic lasers such as argon or krypton ion lasers, the line widths are relatively narrow (*ca.* 5–8 GHz) and pulse durations are limited to *ca.* 60 ps. For dye lasers with bandwidths of *ca.* 20 THz, pulses of *ca.* 50 fs duration should be feasible in principle. To obtain such ultra-short pulses, the longitudinal modes of a laser are coupled in phase either by active or passive techniques and give rise to a train of pulses.

A laser may be actively mode-locked by modulating the gain or the loss at the cavity loop frequency. For the argon and krypton ion lasers, an acousto-optic modulator operating either at the fundamental or a harmonic of the cavity loop frequency is placed within the resonator and produces pulses of *ca.* 60 ps duration with average powers of several watts. Mode-locked ion lasers may be used to pump a dye laser whose cavity loop frequency is tuned to that of the ion laser. This is referred to as synchronous pumping. Under these circumstances pulses of 1–10 ps duration with output powers of 10–50 mW and peak powers of 50–300 W can be produced over the range 450–850 nm. Output powers of up to 1.5 kW may be obtained by using a cavity dumping technique. A acousto-optic quartz element is inserted in the dye laser cavity, and when energized with ultrasonic pulses, most of the intracavity energy is diverted out of the resonator. Repetition rates up to 3 MHz and pulse durations of *ca.* 15 ps can be produced in this way.

Passive mode-locking techniques are simpler in that they do not require the use of electro- or acousto-optic modulators and their associated electronics. A saturable dye is inserted into the laser cavity, and, if the intracavity intensity is sufficiently high, the dye is saturated and its transmission increases. The highest intensity fluctuations of the laser field therefore experience less loss and are preferentially amplified. Saturation of the absorption and gain both serve to shorten the pulse to its ultimate duration set by the properties of the dyes.

Pulsed dye lasers may be passively mode-locked if the pumping times are long enough for the pulse-shortening processes to be fully effective. Flashlamp-pumped dye lasers fulfil this requirement with pumping times of *ca.* 1 μs and have sufficient intracavity power to saturate dyes. Pulse of 2–10 ps duration may be produced over the spectral range 450–700 nm by this technique (Arthurs *et al.* 1972; Mialocq & Goujon 1978). C.w. dye lasers can be mode-locked by focusing into the dye, which is held in contact with one of the cavity mirrors. The shortest pulses

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(*ca.* 0.5 ps) are obtained by using Rhodamine 6G and DODCI as amplifier and absorber respectively, and the system is tunable over the range of 600–615 nm. A greater tuning range can only be achieved with an increase of pulse duration. Shorter pulses can be obtained by operating the c.w. dye laser in a ring configuration and replacing the dye cell by free-flowing jet (as shown in figure 4). Pulses as short as 90 fs have been generated by this method, but the tuning range is further restricted (Mourou & Sizer 1982).

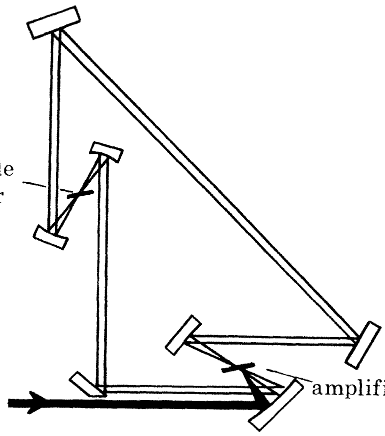

FIGURE 4. A c.w. mode-locked ring dye laser.

4. Second harmonic generation

One of the most convenient and efficient methods of extending the tuning range of dye lasers into the ultraviolet spectral region is by the generation of the second harmonic frequency by sum-frequency mixing in a suitable nonlinear crystal. For efficient conversion, the fundamental and second harmonic waves should propagate in phase through the nonlinear medium and this requires that the refractive indices at both frequencies should be equal. This phase-matching condition may be achieved in a birefringent crystal when the ordinary and extraordinary refractive indices are the same in a specified direction relative to the optical axis of the crystal. In the special case where this occurs in a direction perpendicular to the optical axis (90° phase-matching), the fundamental and second harmonic waves propagate collinearly through the crystal, and the conversion efficiency is maximized. If the frequency of the input laser is tuned, the crystal must also be tuned to maintain phase-matching conditions. For 90° phase matching, this is achieved by changing the temperature of the crystal since the difference between the ordinary and extraordinary refractive indices is dependent on both temperature and wavelength. Otherwise, tuning by changing the angle of incidence of the fundamental laser beam is most convenient, but the temperature of the crystal must be stabilized. By suitable choice of laser dye and nonlinear crystal, the effective tuning range of dye lasers may be extended to 217 nm, thereby effectively giving continuous tuning from 217 to *ca.* 1000 nm. Since the conversion efficiency is proportional to the power density of the fundamental laser, high-power lasers are favoured. However, by focusing a low-power laser beam into the crystal, the conversion efficiency may be increased. Intracavity operation further enhances the efficiency and since most resonators for ring dye lasers are designed to have an auxiliary beam waist, the positioning of the crystal at that point offers a convenient method for generating c.w. frequency-tunable, narrow-bandwidth laser radiation in the ultraviolet region of the spectrum. Output

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powers of up to 100 mW can be produced and by suitable choice of dye, the spectral range between 217 and 450 nm can be covered. For mode-locked lasers, extracavity second harmonic generation is most efficient. Conversion efficiencies of up to 1% and average output powers of 10 mW can be obtained.

The lower wavelength limit may be reduced to less than 200 nm by non-degenerate three-wave mixing in which two lasers of frequencies ω_1 and ω_2 are mixed in a nonlinear crystal to generate ω_3 such that $\omega_1+\omega_2 = \omega_3$ (Dunnings 1978). In particular, in laser-pumped dye lasers, the pumping laser and the tunable dye laser may be mixed to generate tunable ultraviolet radiation (Blit *et al.* 1977; Massey & Johnson 1976). Since the output power is proportional to the product of the power densities of the two lasers, the greater power of the pumping laser gives rise to increased ultraviolet power. However, the line widths of pumping lasers are usually broad and the generation of very narrow line widths by this technique may require the use of two narrow bandwidth lasers with consequent increased complexity.

5. Raman scattering

Frequency shifting of powerful lasers by stimulated Raman scattering in molecular gases provides an effective method of extending the tuning range of lasers. By focusing the laser beam of frequency ω_L into a gas cell, radiation of frequencies $\omega_L \pm n\omega_R$ is produced. The three most commonly used gases are H_2, D_2 and CH_4 with Raman shifts (ω_R) of 4155, 2987 and 2917 cm^{-1} respectively, at pressures of several tens of atmospheres. Conversion efficiencies into the first Stokes frequency ($\omega_L-\omega_R$) of 10% and first anti-Stokes frequency ($\omega_L+\omega_R$) of the order of 1% can be obtained for input powers of 1–10 MW, but higher-order scattering processes are significantly less efficient and can result in a deterioration in beam quality (Loree *et al.* 1979).

It is interesting to note that *spontaneous* anti-Stokes scattering may be used to produce a tunable, narrow-bandwidth, short-wavelength source suitable for spectroscopic use. Anti-Stokes scattering from the $2s^1S$ excited state of He with the use of frequency-tunable dye lasers provides radiation of *ca.* 1.5 cm^{-1} bandwidth tunable over *ca.* 40000 cm^{-1} around 56.9 nm and has been used in v.u.v. spectroscopic studies of potassium (Harris 1977).

6. Excimer lasers

Excimer lasers provide high output powers in the u.v. and v.u.v. regions of the spectrum and enable laser spectroscopic techniques that were originally applied in the visible region by using dye lasers to be extended to shorter wavelengths. An excimer is a molecule with a bound excited electronic state and dissociative ground state, as shown in figure 5. Of particular importance as lasers are the diatomic noble gas molecules (e.g. Xe_2) and the noble gas halides (e.g. KrF). With the exception of XeF and XeCl, which have weakly bound ground states and give rise to emission spectra with vibrational structure, noble gas and noble gas halide excimers have repulsive ground states and have unstructured continuous emission bands. A list of noble gas halide lasers and their wavelengths is shown in table 1. The pumping power density required to pump a laser increases dramatically as the transition frequency is increased, and consequently noble gas halide excimer lasers are pulsed and are pumped by low-inductance transverse electrical discharges, by high-current electron beams or by electron beam sustained discharges. Homonuclear noble gas excimer lasers are pumped exclusively by electron beams.

Of the noble gas excimer lasers, Xe_2 ($\lambda \approx 172$ nm) is the most convenient to use because of low damage thresholds for laser optics at shorter wavelengths. A repetitive (0.5 Hz) laser, tunable over the region 170–175 nm with output powers of 1 MW, has been developed (Edwards *et al.* 1979). Noble gas halide lasers pumped by u.v. pre-ionized avalanche discharges offer a very convenient source of high-power u.v. radiation. XeCl lasers ($\lambda = 308$ nm) with high repetition rates (10–100 Hz) and output pulse energies of 100–500 mJ are commercially available and are very suitable for pumping dye oscillator and amplifier laser systems. Continuous coverage of the range 330–975 nm is possible with appropriate dye changes with conversion efficiencies ranging from 18% in the near ultraviolet to *ca.* 5% in the infrared (Teller *et al.* 1981).

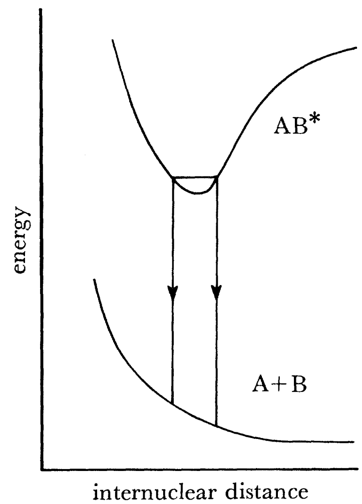

FIGURE 5. Generalized potential energy diagram of an excimer AB*.

The bandwidths of ArF, KrCl and KrF lasers are *ca.* 100 cm^{-1} and the beam divergence is *ca.* 10 mrad when operated with a stable resonator. The beam divergence may be reduced to close to the diffraction limit (*ca.* 50 μrad) by the use of unstable resonators (C. E. Webb, personal communication), although with a reduction in output power. Frequency control of the lasers is more difficult and various methods using intracavity etalons, gratings and prisms have been investigated. Tuning of a range of *ca.* 1 nm with a line width of 0.3 cm^{-1} can be achieved by using a grating at grazing incidence, but the beam dimension must be controlled by the use of apertures, with a resultant substantial reduction in output power (Caro *et al.* 1982). Frequency selection by the use of multiple etalons can give a line width of 0.1 cm^{-1} (Goldhar *et al.* 1980), but alignment and tuning of such a resonator is very complicated. The pulse duration of most discharge-pumped excimer lasers is limited to *ca.* 10 ns by discharge instabilities and restricts the number of cavity loops available for amplification of the desired radiation mode. The use of several high-finesse frequency-selective elements introduces losses and consequent lengthening of the build-up time of the laser; this results in a severe reduction of output energy to a few hundred mJ and amplification is normally desirable.

An alternative to direct control of the excimer laser is the use of the laser as an amplifier of radiation of the required spectral quality and divergence, which can be generated by a dye laser system and harmonic generation. Although the signal injected into the amplifier must

TABLE 1. WAVELENGTHS AND OUTPUT POWERS OF LABORATORY EXCIMER LASERS

excimer	wavelength/nm	typical output power/MW
Ar_2	126	—
Kr_2	146	—
Xe_2	172	5
ArF	193	25
KrCl	222	1
KrF	248	50
XeCl	308	25
XeF	351	20

compete with amplified spontaneous emission within the amplifier, input powers of *ca.* 100 μW are sufficient. The single-pass gain of a discharge-pumped excimer laser is *ca.* 500–1000, but overall gains of up to 10^8 are possible by injection-locking of a laser oscillator, preferably one with an unstable resonator that can provide superior beam divergence. However, this requires careful matching of the injected beam profile to the lowest-order spatial mode of the unstable resonator, and frequency tuning of the injected signal requires simultaneous tuning of the length of the unstable resonator in order to maintain efficient discrimination against higher-order transverse modes and to avoid mode pulling (Bigio & Slatkine 1981). The power of this approach has been demonstrated (Pummer *et al.* 1982) by the construction of an ArF laser system with an output power of greater than 40 MW, an output pulse energy of 300 mJ, a spectral width of less than 260 MHz, absolute frequency control to within 1.8 GHz, a beam divergence of *ca.* 5 μrad, and repetition rates up to 10 Hz. A similar high-brightness system has been developed for KrF at 249 nm (Hawkins *et al.* 1980).

7. FOUR-WAVE MIXING

The effective wavelength of operation of narrow-bandwidth dye lasers can be extended beyond the limit set by second-harmonic generation ($\lambda = 217$ nm) by four-wave mixing techniques ($\omega_4 = \omega_1+\omega_2+\omega_3$) of which third-harmonic generation ($\omega_4 = 3\omega_1$) is an example. The nonlinear medium in this case is a vapour or gas. Efficient conversion is promoted by the use of powerful lasers, high nonlinear susceptibilities of the medium and the achievement of phase matching. The nonlinear susceptibility is enhanced when a laser is tuned close to a two-photon resonance in the gas, but since this predetermines the input laser frequency, tunable radiation can only be produced if at least two input lasers are used. For a two-photon resonance $\Omega = 2\omega_1$, tunable radiation of frequency ω_4 may be produced by the interaction of the fixed frequency ω_1 and tunable frequency ω_2 by sum frequency generation ($\omega_3 = 2\omega_1+\omega_2$). Because the output power will be proportional to $I^2_{w_1} I_{\omega_2}$ in the absence of saturation, the laser beams are focused into the medium and under these circumstances the phase-matching condition for sum frequency generation requires that the gas is negatively dispersive, i.e. $\Delta k = k_4-2k_1-k_2 < 0$, where k is the angular wavenumber. The optimal value of Δk is determined by the confocal parameters of the focused beams, but for a given experimental arrangement Δk may be adjusted by mixing the negatively dispersive nonlinear gas with a positively dispersive buffer gas. For the process $\omega_4 = 2\omega_1-\omega_2$, the phase-matching condition is $\Delta k \gtreqless 0$ and this can be achieved for both positively and negatively dispersive single-component gases.

Four-wave frequency mixing in Xe, Kr and Hg has proved to be very effective in producing narrow bandwidth tunable intense v.u.v. radiation. In principle, a significant fraction of the spectral region 105–146.9 nm can be covered by sum frequency generation in either Xe or Kr since dispersion is negative and continuous tuning from 105 to 220 nm is possible by difference frequency generation. Non-resonant mixing in Xe and Kr of the fundamental and second harmonic frequencies of a dye laser and the second harmonic and 1.06 μm radiation from a Nd-YAG laser provides continuously tunable radiation over the range 110–210 nm with powers of up to 60 W (Hilbig & Wallenstein 1982*a*). The output power can be increased to 0.5–10 kW by resonance enhancement, and the two-photon transitions 5p—6p, 5p—7p and 5p—4f in Xe have been used for sum and difference mixing ($\omega_4 = 2\omega_1 \pm \omega_2$) yielding continuously tunable radiation in the ranges 73–101 nm and 129–220 nm (Hilbig & Wallenstein 1982*b*).

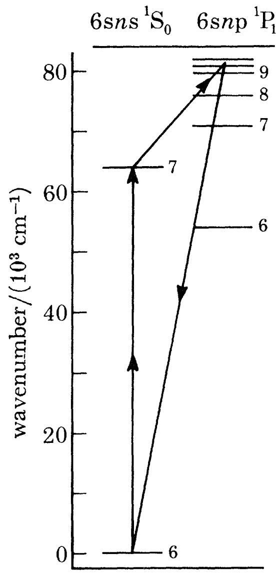

FIGURE 6. Energy-level diagram for resonant four-wave mixing in Hg.

By tuning to the 6s—7s two-photon transition of Hg, sum frequencies have been generated in regions around the np^1P_1 (n = 9–12) levels that enhance the nonlinear susceptibility in the region 119–127 nm, with efficiencies of up to 1 %, output powers of 10 kW and line widths of 0.02 cm^{-1} (Hilbig & Wallenstein 1982; Mahon & Tomkins 1982). (An energy level diagram is shown in figure 6.) Shorter-wavelength radiation may be produced by sum frequency generation with the narrow bandwidth (less than 260 MHz), low divergence (*ca.* 10 μrad) injection-locked excimer lasers described earlier, but the efficiency is reduced by the lack of window materials with high transmission at $\lambda \approx 110$ nm and by photoionization losses in the nonlinear medium. Tunable radiation with narrow bandwidths in the region of 64 nm (Pummer *et al.* 1982), 79 nm (Egger *et al.* 1982) and 83 nm (Egger *et al.* 1980) by non-resonant four-wave mixing, and output powers of up to 30 W have been produced in the absence of phase matching. It is interesting to note that the techniques described enable tunable laboratory sources to be constructed with a spectral brightness in the vacuum ultraviolet several orders of magnitude greater than that available from synchrotron sources and with effective resolving powers of up to 10^7, which is greater than that of the largest spectrographs.

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