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Extreme Ultraviolet Picosecond Pulses

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Extreme ultraviolet picosecond pulses

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Nonlinear optical frequency conversion has provided the capability of generating coherent radiation in the extreme ultraviolet region of the spectrum. Various combinations of laser systems, nonlinear media and nonlinear interactions have been used to generate radiation at wavelengths as short as 38 nm. The use of higher order processes (e.g., fifth and seventh order frequency mixing) has proved especially fruitful in extending coherent radiation to this spectral region. The conversion efficiencies of nonlinear interactions of different orders are compared and the role of competing processes in limiting harmonic power is discussed.

1. Introduction

One of the continuing efforts in laser development has been the extension of coherent radiation to progressively shorter wavelengths. Recent years have seen steady progression through the visible to the ultraviolet (u.v.), vacuum ultraviolet (v.u.v.) and most recently to the extreme ultraviolet (x.u.v.), bringing us almost to the soft X-ray range. The wavelength ranges corresponding to these regions are illustrated in figure 1*a*. These advances have proceeded in two main areas. On the one hand, numerous primary lasers have been developed that operate at single wavelengths or in restricted spectral bands over a wide range in the visible, u.v. and v.u.v. The wavelengths of some of the more important lasers are indicated in figure 1*b*. More or less simultaneously, techniques for shifting laser radiation to different parts of the spectrum through nonlinear optical frequency conversion have been developed. These techniques have been used to generate coherent radiation throughout much of the u.v. and v.u.v. and have allowed coherent radiation to be extended to the x.u.v. The wavelength range that is currently covered by optical frequency conversion is indicated in figure 1*c*. In this paper I shall discuss the use of nonlinear optical interactions for generating coherent radiation in the x.u.v., i.e. at wavelengths shorter than about 100 nm.

2. Nonlinear optical frequency mixing

Optical frequency conversion occurs because of nonlinear polarizations that are induced in a transparent medium by intense laser radiation. In the electric dipole approximation the nonlinear polarization can be expressed as

$$P_{\mathrm{nl}} = \chi^{(2)}E^2+\chi^{(3)}E^3+\chi^{(4)}E^4+\chi^{(5)}E^5+\ldots \qquad (1)$$

Here the coefficient $\chi^{(n)}$ is the nth order nonlinear susceptibility and E is the electric field of the incident light wave. Because the nonlinear polarization involves products of oscillating fields, it will contain terms that oscillate at frequencies that are sum and difference combinations of those contained in the incident radiation. Each of these Fourier components will in turn radiate a new field at its own frequency.

In the wavelength range above 200 nm, second harmonic generation and three wave sum frequency mixing can be done very effectively in highly nonlinear anisotropic crystals. At wavelengths below about 200 nm, suitable nonlinear crystals are opaque and conversion must be done in transparent gases. Because of the inversion symmetry of gaseous media, only odd order nonlinear processes are allowed by electric dipole interactions. The power in the generated wave depends on the pump intensity, nonlinear susceptibility, and value of the phase mismatch between the generated field and the nonlinear polarization. We can illustrate these

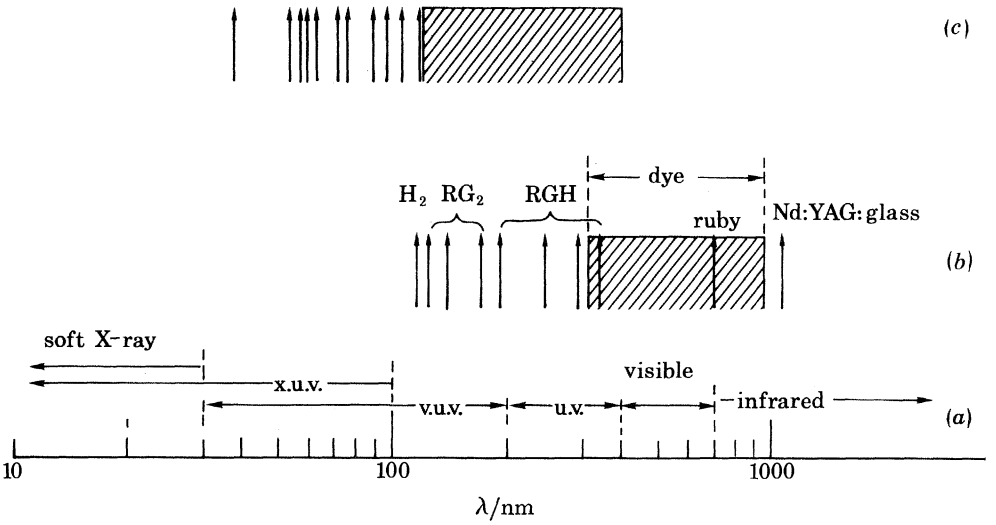

FIGURE 1. (*a*) Designation of wavelength ranges between 10 nm and 1000 nm. (*b*) Wavelengths of some primary lasers. RG_2, rare gas excimer laser; RGH, rare gas halide laser. (*c*) Additional wavelengths covered by nonlinear frequency mixing.

dependences for the different order interactions by considering the harmonic generation process. In generating x.u.v. radiation, the incident beam is usually focused to take advantage of the increased interaction that accompanies higher intensities. If we assume that the focused beam has a Gaussian profile, the power in the qth harmonic is given by

$$P_q = \frac{2^{q-1}\,\pi^{q+1}}{q n_1 n_q c^{q-1}} \left| \frac{N}{n_1 - n_q}\,\chi^{(q)} \right|^2 |b\Delta k\, F_q(b\Delta k)|^2 \times \left(\frac{4P_1}{b\lambda_1}\right)^{q-1} P_1 \qquad (2)$$

where $b = 2\pi\omega_0^2/\lambda$ is the confocal parameter of the focused beam, ω_0 is the spot size of the beam waist (1/e field radius), P_1 is the power in the incident beam and $\Delta k = k_q - qk_1$ is the wavevector mismatch (k_i is the wavevector of the ith wave). F_q is given by

$$F_q = \int_{-\xi'}^{\xi} \frac{e^{-\frac{1}{2}ib\Delta k\,\xi''}}{(1+i\xi'')^{q-1}}\,d\xi'', \qquad (3)$$

and the nonlinear medium extends from $-\xi'$ to $+\xi$, where $\xi = 2z/b$. The quantity $(n_1 - n_q)/N$ is the dispersion per atom, independent of gas pressure, while the entire dependence on gas pressure, phase mismatch and focusing is given by the quantity $|b\Delta k F_q(b\Delta k)|^2$. For a beam that is focused tightly, so that the depth of focus is much smaller than the extent of the nonlinear medium, this quantity is given by

$$\begin{aligned} |b\Delta k F_q|^2 &= \left(\frac{4\pi}{(q-2)!}\right)^2 (\tfrac{1}{2}b\Delta k)^{2q-2}\, e^{b\Delta k} && \Delta k < 0 \\ &= 0 && \Delta k \geqslant 0. \end{aligned} \qquad (4)$$

The dependence of the power in the third, fifth and seventh harmonics on $b\Delta k$ for tight-focused geometry is shown in figure 2*a*. The power in each of the harmonics is optimum for a negative value of Δk, given by $b\Delta k = -(2q-2)$, while no harmonic power is generated for positive values of Δk. These properties are characteristic of conversion in a focused beam and arise from the change in propagation vector as the beam passes through the focus. This change in k-vector can be compensated for by negative dispersion in the medium, but for positive dispersion the phase change causes harmonic power generated in the first half of the focus to be cancelled in the second half. A second geometry that has also proven useful in x.u.v. generation is one of a beam focused into a semi-infinite medium extending to one side of the beam waist. The quantity $|b\Delta kF_q|^2$ is shown for this geometry for the three harmonics in figure 2*b*. Harmonic conversion is still optimized at negative values of Δk, but some conversion is now possible for positive dispersion.

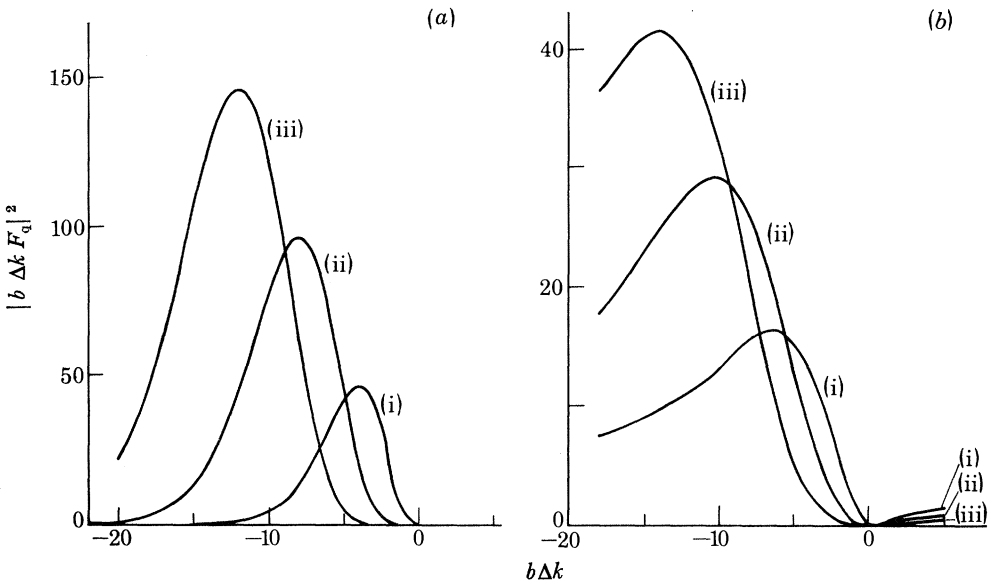

FIGURE 2. Variation of phase optimization integral with $b\Delta k$ in a single component medium. (*a*) Infinite medium: (i), third harmonic; (ii), fifth harmonic; (iii), seventh harmonic. (*b*) Semi-infinite medium: (i), third harmonic; (ii), fifth harmonic; (iii), seventh harmonic.

Harmonic generation is a special case of sum frequency generation, and phase matching conditions for the sum frequency processes are the same as those discussed above. Phase matching requirements for difference frequency processes can require negative, zero or positive mismatch and can be determined from analyses such as that given by Bjorklund (1975).

3. MATERIAL CONSIDERATIONS

Generation of x.u.v. radiation has been done primarily in the rare gases, but some results have been reported in metal vapours and atomic hydrogen as well. As a class, metal vapours tend to have large nonlinear susceptibilities because of the large oscillator strengths of their discrete transitions. X.u.v. radiation usually lies in the photoionization continuum of these media, making coupling to the harmonic wavelength relatively small unless advantage can be

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

taken of autoionizing resonances. Absorption due to photoionization in metal vapours is relatively weak but can become important if the interaction length or density become too large.

Rare gases, on the other hand, are transparent farther into the x.u.v. Helium, for example, is transparent to 50.4 nm and Ne, Ar and Kr all have their photoionization thresholds below 100 nm. The rare gases tend to have smaller nonlinear susceptibilities than do metal vapours, requiring higher pump intensities. Also, continuum absorption in the rare gases is generally stronger than in metal vapours. The rare gases are, however, better suited for x.u.v. generation since they are more compatible with the differential pumping geometries that are commonly used at wavelengths below 105 nm because of the scarcity of window materials.

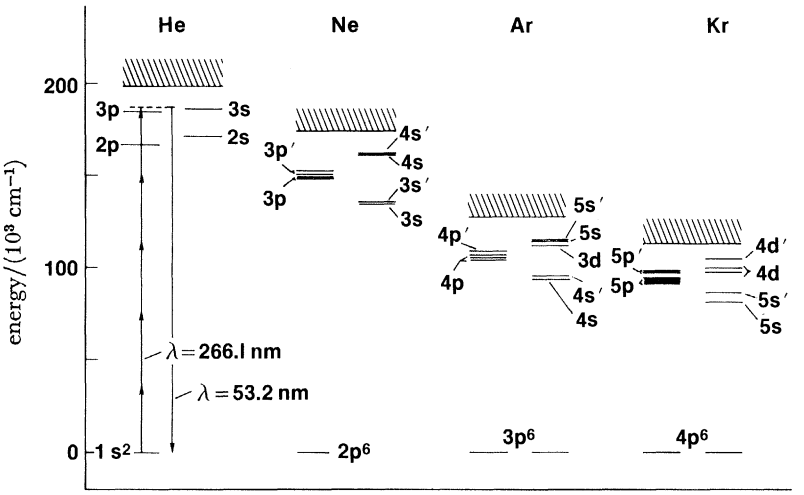

FIGURE 3. Energy level diagram of four of the rare gases. Fifth harmonic conversion under conditions of negative dispersion in helium is illustrated.

Energy level diagrams of four of the rare gases are shown in figure 3. The negative dispersion needed for optimization of harmonic and sum-frequency processes can be obtained by choosing a medium such that the fundamental wavelength lies below all the excited levels while the generated wavelength lies just above a dispersive resonance. Such a situation is illustrated for fifth harmonic conversion in He. Generally, negative dispersion can be found for some extent of wavelengths above each odd parity level. Phase optimizing conditions can be found in one or more of these gases for generation of radiation over much of the range between 50.4 and 100 nm.

The conversion efficiency can also be increased if the incident frequencies or combinations or multiples of them are chosen to coincide with a suitable level in the medium. For third order processes the most important enhancements come from two-photon resonances with unallowed transitions because these are not accompanied by strong absorption at either pump or generated wavelengths. In the higher order processes, additional absorption-free resonances are also possible.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

4. Experimental results

The various nonlinear processes that have been reported for x.u.v. generation, along with the types of laser systems used, appropriate nonlinear media and wavelength range covered are summarized in table 1. Various combinations of laser systems, nonlinear media and types of nonlinear interaction have been used to generate coherent radiation at many discrete wavelengths down to 38 nm.

Table 1. Nonlinear processes used for x.u.v. generation

order	process	laser	λ/nm	nonlinear medium	reference
3	3×354.7 nm	Nd:YAG	118.2	Cd	Kung *et al.* (1972)
	$2\times266.1+1064$	Nd:YAG	118.2	Ne	Reintjes *et al.* (1978)
	$2\times266.1+532$	Nd:YAG	106.4	He, Ne	Reintjes *et al.* (1978)
	3×308	XeCl	102.7	Ar	Reintjes (1979)
	$2\times243+486$	dye	97.2	H	Troshin *et al.* (1978)
	3×268.8	Nd:glass	89.6	Hg	Slabko *et al.* (1977)
	3×266.1	Nd:YAG	88.7	He, Ne, Ar	Harris *et al.* (1973); Reintjes *et al.* (1978)
	3×171	Xe_2	57	Ar	Hutchinson *et al.* (1976)
5	$4\times266.1-532$	Nd:YAG	76	He, Ne	Reintjes *et al.* (1978)
	$4\times266.1-1064$	Nd:YAG	70.9	He, Ne	
	$4\times266.1+1064$	Nd:YAG	62.6	He, Ne	
	$4\times266.1+532$	Nd:YAG	59.1	He, Ne	
	5×266.1	Nd:YAG	53.2	He, Ne, Ar, Kr	
7	7×266.1	Nd:YAG	38	He	
9	9×1060	Nd:glass	118	Na	Grozeva *et al.* (1977)

4.1. *Third order processes*

Third order processes have been used to generate radiation at wavelengths as short as 57 nm. These are the lowest order allowed processes in gases and as such are usually expected to be the strongest of the nonlinear interactions. They have been used extensively for frequency conversion at longer wavelengths in the v.u.v. and visible and were also the first to be used for generation in the x.u.v.

Third harmonic conversion and sum frequency generation of harmonics of Nd:YAG and Nd:glass lasers have been observed in both rare gases and metal vapours. The fourth harmonic of a Nd:YAG laser at 266.1 nm has been tripled in He, Ne and Ar, giving radiation at 88.7 nm. The fourth harmonic of the Nd:YAG laser has also been mixed with the lower harmonics to give radiation at 106.4 and 118.2 nm. Tripling of the third harmonic of a YAG laser has also been observed. Third harmonic conversion of the fourth harmonic of a Nd:glass laser in Hg vapour has been reported. Here, the interaction was enhanced by tuning the output of the glass laser at 1.06 μm so that its fourth harmonic matched a two-photon resonance in Hg.

Third harmonic conversion of radiation from various excimer lasers have also been reported. Although these results did not involve the use of picosecond pulses, they are of interest in this topic. The lasers themselves have potential for generating picosecond pulses either directly through operation as mode-locked oscillators or as amplifiers for picosecond pulses generated in other lasers.

Radiation from a xenon excimer laser at 171 nm was used to generate radiation at 57 nm by third harmonic conversion in Ar. Again the laser radiation was adjusted to take advantage of a two-photon resonance in the argon to enhance the conversion process. Rare gas excimer lasers have been demonstrated to lase at wavelengths as short as 125 nm (argon excimer laser) and continued extension of these techniques should provide radiation to as short as 41.7 nm by third order conversion processes.

Very recently we have also demonstrated third harmonic conversion of radiation from a XeCl laser to produce radiation at 102.7 nm. Several rare gas halide lasers are available operating at wavelengths ranging from 350 to 193 nm, and further development of third order conversion of these sources should provide coherent radiation at wavelengths ranging from about 117 to 64.3 nm.

4.2. *Higher order processes*

Higher order interactions can be used to generate still shorter wavelengths because they allow a larger step along the frequency scale to be taken in a single process. Fifth order interactions have been used for generating radiation at several wavelengths between 53.2 and 76 nm. Pump radiation for these processes was derived from a mode-locked Nd : YAG laser followed by two stages of second harmonic conversion. Fifth harmonic conversion of the YAG fourth harmonic pulses was used to produce radiation at 53.2 nm. Six wave sum-and-difference-frequency mixing were used to generate radiation at wavelengths out to 76 nm. In these interactions the frequency of the generated radiation was given by $\omega_g = 4\omega_{\text{u.v.}} \pm \omega_i$, where ω_i corresponds to either the fundamental or second harmonic of the Nd : YAG laser. In this set of experiments this interaction was the strongest because the two stages of second harmonic generation that followed the YAG laser were so efficient that they severely depleted the fundamental and second harmonic intensity. Seventh harmonic conversion of fourth harmonic pulses from a mode-locked Nd : YAG laser has been used to generate radiation at 38 nm, the shortest wavelength at which coherent radiation has yet been reported. Finally, ninth harmonic generation has been used to convert Nd : glass laser radiation directly to 118.2 nm.

The advantages that the higher order processes offer in terms of short wave generation may not be realized if their efficiency is too low. Because the higher order processes depend more strongly on the pump power than do the lower order ones it can be anticipated that they can be comparable with or stronger than the lower order ones at sufficiently high input intensities. These processes are therefore ideally suited to the high peak powers that can be obtained in picosecond pulses. In liquids and solids, intensities are restricted to values of the order of 10 GW/cm^2 by dielectric breakdown, and lower order processes have always been observed to dominate higher order ones in these media. Much higher input intensities are possible in the low pressure gases that are used for x.u.v. generation, and favourable conditions may exist in these media for effective use of the higher order processes.

In order to investigate the relative importance of interactions of different order, third and fifth harmonic conversion in helium have been compared (Reintjes & She 1978). Fifth harmonic power at 53.2 nm was observed to exceed third harmonic power at 88.7 nm for pump powers in the range from 6 to 300 MW as is shown in figure 4. It appears evident, then, that experimental conditions can be realized in low pressure gases in which conversion in higher order processes is as effective as conversion in lower order ones.

Maximum conversion efficiencies that have been reported in the x.u.v. are of the order of 10^{-5} for fifth and third harmonic conversion of YAG laser harmonics, corresponding to peak

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

pulse powers of the order of 1–10 kW at 53.2 and 88.7 nm. Ultimately, conversion must drop off for the higher order processes or the perturbation expansion of (1) will become invalid. In experiments that have been reported to date, seventh harmonic efficiency is about an order of magnitude lower than fifth, giving peak powers of about 100 W at 38 nm.

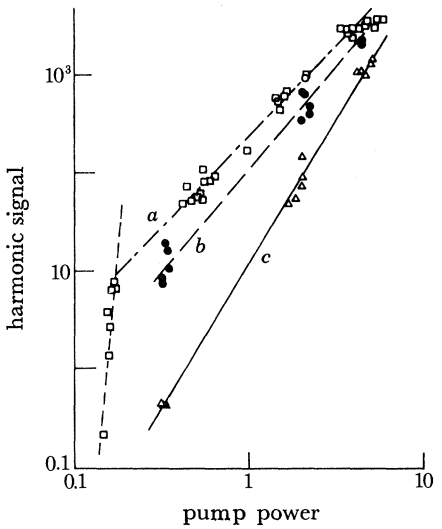

FIGURE 4. Comparison of fifth and third harmonic conversion in helium: (*a*) fifth harmonic, 25 Torr; (*b*) fifth harmonic, 18 Torr; (*c*) third harmonic, 18 Torr. (1 Torr ≈ 133.32 Pa.)

TABLE 2. LIMITING MECHANISMS IN OPTICAL FREQUENCY CONVERSION

saturation of excited state population through multiphoton absorption	disturbs phase matching, lowers ground state population, direct optical energy loss
Stark effect	alters magnitude of susceptibility
photoionization, breakdown	reduces ground state population, direct optical energy loss, disturbs phase matching
Kerr effect	alters phase matching conditions

5. COMPETING MECHANISMS

In addition to radiation at new wavelengths, the nonlinear polarization can result in a number of other effects that can disturb the frequency conversion process. Some of these other nonlinear effects are listed in table 2, along with their effect on the frequency conversion process. There are basically three ways that these competing processes affect the frequency conversion process. They can cause loss of energy at either the pump or generated wavelength, reduce the value of the nonlinear susceptibility or alter the phase matching conditions. The net result of these effects is to reduce the amount of power that can be generated in the frequency conversion process and to weaken its dependence on the pump power. Because the same transition dipole matrix elements are involved in both kinds of nonlinear processes, the efficiency in some cases can be limited to a maximum value that depends only on the ratio of matrix elements but not on the input power. These competing processes are much more important for frequency conversion in gases than in liquids and solids, and some form of limitation has been observed in virtually all attempts to achieve highly efficient conversion in

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

gases. The effects of such competing processes are evident in the fifth harmonic data in figure 4. At low laser powers, the fifth harmonic shows the expected fifth power dependence on pump strength. At laser powers above about 12 MW, the dependence is much weaker, varying approximately as the second power of the pump strength. Similar saturation is not evident in the third harmonic conversion.

Identification of the effects responsible for saturation in a given configuration is required so that parameters may be chosen to optimize conversion. Experimental and theoretical studies have shown that nonlinear refractive indices play an important role in limiting conversion efficiency in a wide range of frequency conversion processes. Evidence of saturation of the nonlinear susceptibility has also been reported and quantitative studies show that both types of processes are active in the saturation of fifth harmonic conversion.

6. Discussion

The use of nonlinear optical frequency conversion has provided a new source of coherent radiation in the extreme ultraviolet, a range that is currently covered by arc lamps and synchrotron radiation. Already the spectral brightness of the laser sources in this region is several orders of magnitude greater than the more conventional sources, and improvements are anticipated as the limiting mechanisms become better understood. Further development of the techniques discussed here, especially with regard to the use of tunable dye lasers for pump sources, should provide continuously tunable, highly monochromatic radiation in this range. Such radiation has potential for improved resolution in spectroscopic studies of molecules, atoms and surfaces of solids. Continued study of the effects discussed here should also give new insights into the interaction of intense radiation with matter, providing deeper understanding of processes that affect a wide range of nonlinear optical effects, not only in this spectral region but also in the visible and near u.v.

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PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES