3. Four-Wave Frequency Mixing in Gases

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With 14 Figures

Coherent light can be generated in the vacuum ultraviolet by third-harmonic generation and sum-frequency mixing in gaseous nonlinear media. Conversion efficiencies of up to several percent allow the generation of kW and even MW peak powers in nanosecond pulses, and linewidths as low as 0.05 cm\(^{-1}\) are known. Such four-wave mixing extends the range of high-resolution laser spectroscopy, and is useful in plasma diagnostics and laser chemistry. In this chapter, theoretical treatment is followed by detailed description of experimental techniques and a survey of results.

3.1 Background

In recent years methods of nonlinear optics have been successfully used to extend the tuning range of lasers into other spectral regions. In the visible and the infrared spectral regions this has generally been done by sum- or difference-frequency mixing in suitable nonlinear crystals. In the vacuum ultraviolet (vuv) spectral region, however, most solids become opaque and can no longer be employed, because the generated wave of interest is absorbed and cannot escape the nonlinear medium. It is for this reason that sum frequency mixing in the vuv region is generally carried out in gaseous media using four wave parametric processes.

This chapter focuses on the resonant and nonresonant third-harmonic generation and sum-frequency mixing in gaseous nonlinear media which in recent years have been investigated rather extensively [3.1 – 3]. Four-wave frequency mixing in gases has become increasingly interesting as a powerful technique for extending high-resolution laser spectroscopy into the vuv, where many atoms, and a number of the most important small molecules such as H\(_2\), CO, NO and others, have some of their most prominent absorption features. Several applications of this kind have already been reported [3.4, 5].

Harmonic generation in gases was first reported by New and Ward [3.6, 7] and by Rado [3.8]. The conversion efficiencies in these early experiments were extremely low and the results were therefore of little practical interest until Harris and coworkers [3.9 – 11] showed that the conversion efficiency in a gaseous system can be raised by several orders of magnitude using a two-component system. Similar to an idea which was originally demonstrated by
Bey et al. [3.12] in mixtures of liquids, Harris and coworkers showed that phase matching in gases can be achieved by a suitable mixture in which one component has been selected according to its nonlinear susceptibility, whereas the other component has to provide the appropriate overall refractive index for phase matching of the gas mixture. A further significant improvement was established by Bloom et al. [3.13], Hodgson et al. [3.14], and Leung et al. [3.15], who demonstrated that the conversion efficiency may be raised by exploiting a two-photon resonant enhancement of the nonlinear susceptibility. The latter effect was first discussed by Maker and Terhune [3.16] for third-order nonlinear processes in solids and liquids.

With gaseous systems, conversion efficiencies up to a few percent have so far been reported. Peak powers as high as 1 MW have been obtained for nonresonant harmonic generation using fixed-frequency high-power solid-state lasers [3.17, 18]. In order to indicate the technical development, a few further important achievements should be mentioned. Powers of the order of 10 W from nonresonant sum-frequency mixing of dye lasers were reported by Hilbig and Wallenstein [3.19, 20]. Peak powers of about 400 W at the wavelength of Lyman alpha have been obtained by Langer et al. [3.21] from nonresonant harmonic generation in phase-matched krypton argon mixtures, using a high-power dye laser. By using two-photon resonant sum frequency mixing of dye lasers, peak powers of the order of 10 W have been achieved by Wallace and Zdasiuk [3.22] and by Junginger et al. [3.23] in magnesium. Higher peak powers have been obtained by Freeman et al. [3.24] and Mahon and Tomkins [3.25] in mercury. Scheingraber and Vidal [3.26, 27] reported powers as high as 500 W in a strontium-xenon mixture using a tunable, excimer laser pumped dye laser. With a repetition rate of 100 s\(^{-1}\), this corresponds to a time-averaged power of 0.5 mW. Most recently, Hilbig and Wallenstein [3.28] have achieved powers as high as 0.5 – 3 kW by two-photon resonant sum and difference frequency mixing in mercury covering selected spectral regions between 109 and 196 nm.

Even higher peak powers, of typically 10 MW, have been achieved by combining the methods of nonlinear optics with high-power excimer lasers as amplifiers [3.29, 30]. Most recently, peak powers as high as 1 GW (pulse duration: 10 ps) have been demonstrated in the vuv where in the latter system a synchronously pumped, mode-locked dye laser was used in the initial nonlinear system [3.31, 32]. A significant advantage of gaseous nonlinear media over nonlinear crystals is that the gaseous media are not destroyed by laser-induced electric breakdown.

All of the preceding systems have used pulsed lasers to generate fundamental waves of sufficiently high input power. The first cw system was reported by Freeman et al. [3.33], who achieved a two-photon resonant enhancement of the nonlinear susceptibility by applying large magnetic fields to a strontium-xenon mixture. By means of a superconducting solenoid, they tuned the nonlinear medium into resonance with the incident cw beams [3.34] and obtained \(10^7\) photons/s (\(10^{-11}\) W, linewidth: 6 GHz). More recently, Timmer-