Summary

This thesis comprises an experimental study on optical parametric oscillators (OPO), sum-frequency mixing devices and their spectroscopic applications. These non-linear optical devices are sources of coherent radiation and can be used to extend the wavelength range of existing lasers. In general, they are composed of a non-linear crystal inside an optical resonator. Inside the crystal an applied optical field (a beam of laser light) induces polarisation effects; the molecular dipoles in the crystal will start to oscillate at the same frequency as the applied field. This dipole oscillation in turn leads to the emission of an electro-magnetic (EM) wave at the same frequency as the applied EM field. When the incident light field is sufficiently strong, non-linear effects occur; i.e. the dipole-oscillation is no longer harmonic, but will contain an-harmonic terms. Hence, this polarisation will generate an EM field that corresponds to these an-harmonic terms, resulting in light at other frequencies.

In an OPO a powerful laser-beam (the pump) is converted into two new wavelengths (the signal and idler wave) while conserving energy and momentum. Frequency mixing, on the other hand, is the opposite process; two wavelengths are combined in a non-linear medium to generate the sum or the difference of the frequencies of the incident waves, again conserving momentum and energy. By ”adding up” two photons, higher energy photons can be generated, viz. sum-frequency mixing can be used to generate ultra-violet light from visible light. Difference-frequency mixing can be used to produce infra-red light, as low energy photons are generated. These techniques can be applied to generate coherent light in a broad range of wavelengths in the electro-magnetic spectrum. Using solid state lasers in combination with non-linear techniques radiation in a large wavelength range, starting from 200 nm up to 5 \( \mu \)m, can be generated.

The application of these non-linear optical systems depends strongly on the output characteristics of the generated radiation, such as wavelength, pulse-length and bandwidth. For spectroscopic and many other applications often a narrow-bandwidth is desirable. This thesis is focused on the generation of narrow-bandwidth radiation by both pulsed and continuous-wave devices.
In chapter 2, a tunable single-longitudinal-mode (SLM) OPO system is described, that can be electronically scanned over 2.5 cm$^{-1}$. To reduce the bandwidth of the OPO system, a configuration with a narrow-bandwidth seed oscillator is used. The narrow-bandwidth seed-oscillator contains a grating at grazing incidence angle for maximum wavelength selectivity. To obtain sufficient power, the output of the seed oscillator is injected in an amplifier, the power OPO. This seeder-amplifier system provides a stable narrow bandwidth output power (about 11 mJ in 1.5 ns pulses when pumped with 355nm pulses from a Nd:YAG laser) with an overall efficiency of 14.5%. This device is demonstrated to be widely applicable in spectroscopy.

In chapter 3 a novel design for a nanosecond OPO is reported. The design involves a ring cavity-configuration with a grazing-incidence grating. This travelling-wave OPO is pumped by the third harmonic of multi-mode as well as single-mode Nd:YAG lasers. The observed bandwidth of 0.5 GHz at a pulse duration of 1.3 ns when pumped with the single-mode laser is close to the Fourier-transform limit. The configuration of a frequency-selective ring cavity is compact and walk-off compensated and exhibits a relatively low threshold. It could serve as a seed-oscillator for a power OPO similar to the system described in chapter 2.

In chapter 4 a new and efficient method to generate the third harmonic of any single continuous-wave (cw) laser is discussed. It is demonstrated in a device to generate tunable UV-radiation by frequency-tripling of a Ti:Sapphire (Ti:S) ring laser. An external cavity locked to the Ti:S is used to enhance the output intensity and to generate the second harmonic. Subsequently, this second harmonic light is coupled into a second enhancement cavity, together with the fundamental light. To fulfil resonance conditions for both waves simultaneously –which is not trivial because of dispersion in the crystal– this cavity is equipped with dispersion compensating elements. By locking both the fundamental and the second harmonic light of the Ti:S laser simultaneously, deep UV-light is generated in a BBO crystal. Up to 175 mW of output power near 272 nm has been produced, starting from 2.1 W light at the fundamental wavelength.

Chapter 5 comprises the application of a second harmonic cavity for spectroscopy on samarium. A high resolution laser-induced fluorescence study has been performed on thirteen ground-state transitions, using a frequency-doubled Ti:S laser in the wavelength range 350 - 450 nm. From each spectrum isotope shifts and
hyperfine structure constants are derived. Analyses of the data on the isotope shifts in King plots result in a determination of the nuclear parameter $\lambda^{A,A'}$ (related to the change in mean square nuclear radii) for the various isotopes.