High-Repetition Rate Broadly Tunable Femtosecond Sources

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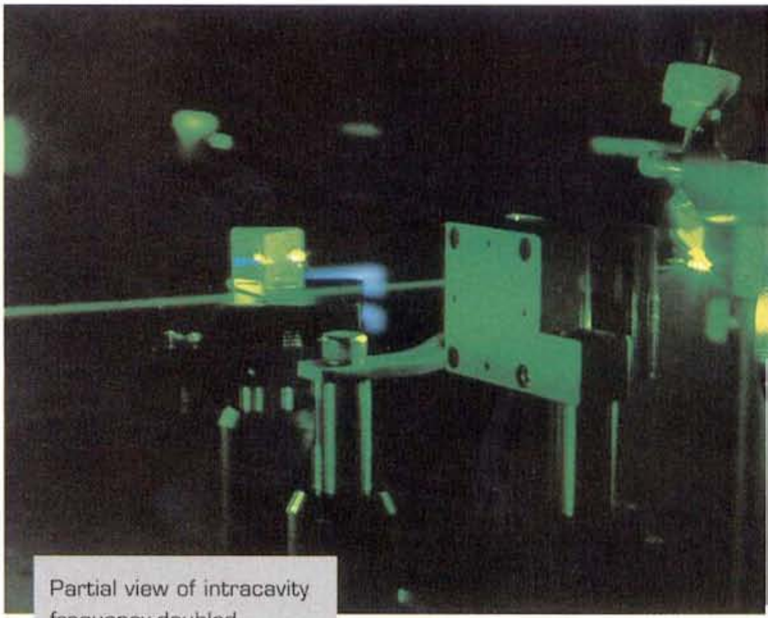
High-repetition rate, broadly tunable femtosecond sources are particularly important for the study of ultrafast processes because high repetition rates yield higher signal-tonoise ratios in such experiments, and because broad tunability allows a greater variety of materials and processes to be studied. Since the first demonstration of the broadly tunable femtosecond optical parametric oscillator (fsec OPO) several years ago,[1] dramatic advances in the development of such sources have occurred. Initially, only the Rh6G fsec dye laser was available as a pump source. This made the operation of the fsec OPO very difficult and the output power relatively low. With the development of the relatively high-power modelocked Ti:sapphire lasers,[2] the situation has changed fundamentally. Using the fs Ti:sapphire laser as a pump, high repetition rate fsec OPOs producing hundreds of milliwatts broadly tunable from 900 nm to over 3.5 μm have been demonstrated.[3-5] This development paved the way for the introduction of commercial fsec OPOs.[6]

Recent developments have involved the extension of such sources to new wavelength regions through the use of new nonlinear optical crystals and techniques. In this brief report, we summarize recent results on the highly efficient conversion of the Ti:sapphire laser-pumped fsec OPO output into the visible through intracavity doubling[7] and the first operation[8] of a KTA ($KTiOPO_4$) fsec OPO that can potentially operate in the important 3-5 μm region.

We have demonstrated a Ti:sapphire laser pumped intracavity-frequency-doubled KTP ($KTiPO_4$) OPO that generates a total of up to 230 mW of sub-100 fsec pulses tunable in the visible.[7] The OPO is aligned as a ring cavity with a 47 μm-thick Brewster-cut β-BaB_2O_4 (BBO) crystal in an additional focus. The BBO crystal does not greatly complicate the operation of the OPO. Due to the BBO crystal's large second-harmonic generation (SHG) phasematching bandwidth around the zero group velocity mismatch point at 1.47 μm, tuning the frequency-doubled OPO output in the range of 1.1-1.6 μm (SHG 550-800 nm) requires no adjustment of the BBO phase-matching angle. We have demonstrated continuous tuning of the SHG from 550-660 nm limited only by optics available at the time of the experiment. The potential tuning range is from ~ 475 nm up to the wavelength of the fsec Ti:sapphire pump laser, which is typically about 800 nm. The second-harmonic pulse train exhibits excellent stability and, as demonstrated by the real-time interferometric autocorrelation, the pulses are chirp-free. Regardless of the transverse mode structure of the Ti:sapphire pump laser, one can achieve an exceptionally clean TEM_{00} mode for the OPO which is imparted to the intracavity frequency-doubled beam.



Partial view of intracavity frequency-doubled femtosecond KTP optical parametric oscillator, and output in the visible.

The transparency range of the recently developed KTA crystal extends to 5.3 μm. Using KTA—instead of the usual KTP crystal in a Ti:sapphire laser-pumped fsec OPO with a linear cavity configuration including intracavity dispersion compensating prisms—we have achieved tuning from 1.29-1.44 μm in the signal branch and from 1.83-1.91 μm in the idler branch,[8] again limited only by the optics available at the time. Calculations show that the KTA OPO can be tuned to 5.3 μm using appropriate optics. The shortest signal and idler pulses we measured were 85 fsec and 150 fsec, respectively. Shorter pulses should be possible using thinner crystals and better dispersion compensation. Similar to the frequency-doubled OPO, the stability and mode quality (TEM_{00}) of the KTA OPO are excellent.

By using different crystals, pump sources, and frequency-conversion techniques, OPOs will ultimately extend the range of high-repetition rate broadly tunable femtosecond pulses even further into the infrared and the ultraviolet.

ACKNOWLEDGMENTS

This work is supported by Joint Services Electronics Program and the National Science Foundation.

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High-Order Harmonic Generation

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Extremely high-order harmonic radiation generated from the interaction of intense (>10^{14} W/cm^2) subpicosecond laser pulses with dense gases offers research scientists a new source of coherent soft x-ray radiation. Harmonic radiation extending to the 33rd harmonic of 1064 nm radiation was first reported in 1989.[1] However, during the past year the most dramatic developments in this new area of nonlinear optics have occurred. Harmonic radiation extending below 7 nm was reported by several groups,[2] many of the observations were well described by both quantum and classical theories,[3] the coherence of this XUV source was demonstrated,[4] and coherent control of the output XUV polarization using a second field of different frequency was shown.[5]

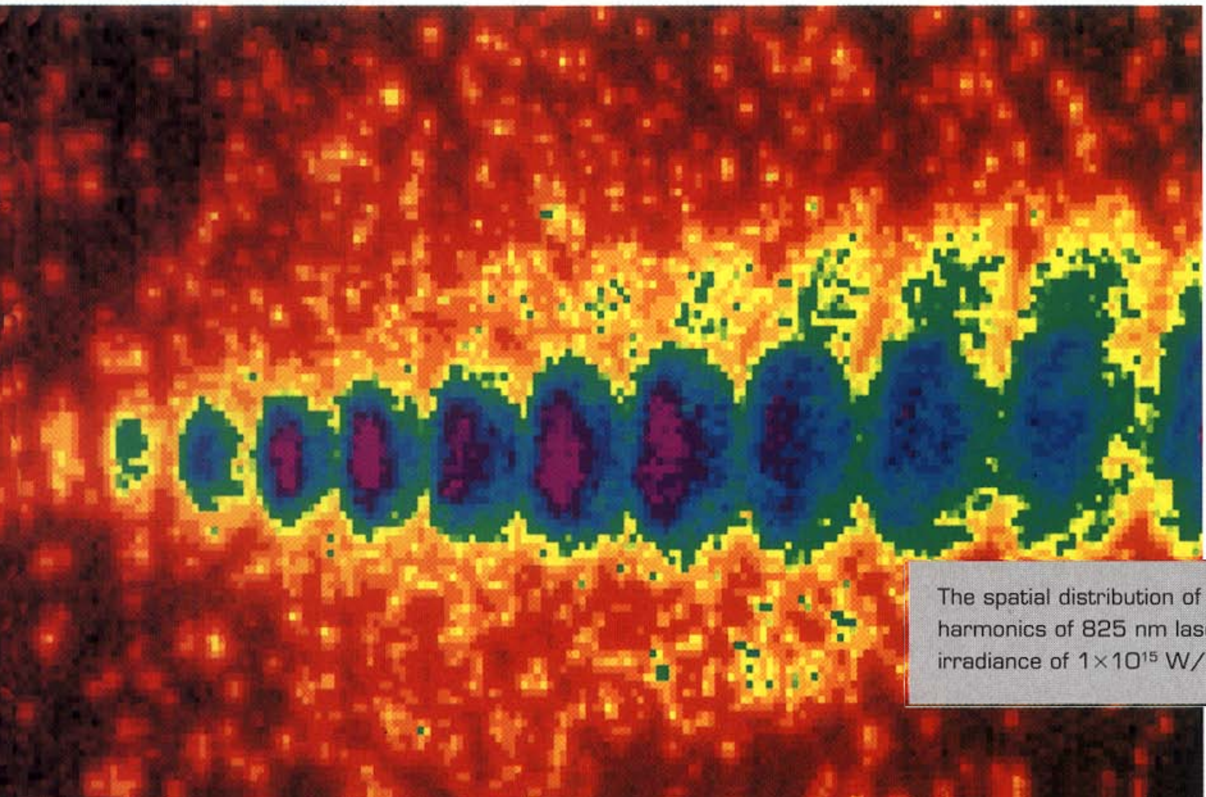
High-order harmonic emission occurs when a bound electron is excited to the continuum and driven by the intense laser field across the anharmonic atomic potential. The quasi-free electron can return to the ground state by emitting a photon of energy equal to the sum of the ionization potential, IP, and its kinetic energy. It can be shown both classically and quantum mechanically[3] that the maximum energy of the harmonic emission is approximately

$$\mathrm{IP} + 3\mathrm{Up},$$

where Up(eV)=9.33×10^{-14} I(W/cm^2) λ^2 (μm) is the electron's quiver energy, I is the laser intensity, and λ is the laser wavelength.

The macroscopic nature of harmonic emission complicates the single atom description. Phase matching between the harmonic field and the driving field of the laser influences the overall efficiency (< 10^{-6}) and coherence of the high-order harmonics[4]. The figure below shows the spatial distribution of the 41st to 65th harmonics of 825 nm laser light in neon at an irradiance of 1x10^{15} W/cm^2.[6] The smooth, essentially Gaussian distributions indicate the high degree of spatial coherence of the radiation that makes it possible to re-image this light to nearly the diffraction limit for use in applications.

An important distinction between harmonic generation and x-ray, laser-produced XUV radiation is the tunability of the harmonic spectrum. By tuning the incident laser wavelength or by selecting different harmonics, complete coverage from 7-100 nm may be achieved. We are currently using this tunable harmonic source for XUV spectroscopy of the rare gases. Although current harmonic generation systems are limited to the 10 Hz repetition rate of the laser, dramatic advances in kilohertz repetition rate-short pulse lasers will extend the average power of this new source of coherent XUV radiation.



The spatial distribution of the 41st to 65th harmonics of 825 nm laser light in neon at an irradiance of 1×10^{15} W/cm^2.