Femtosecond to Attosecond Optics

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Abstract: We continue to observe strong progress moving from femtosecond to attosecond optics. Attosecond pulses are now available in many different labs with additional gating techniques which somewhat relax the requirements for the pulse duration of the intense infrared laser pulses. The pulse repetition rate still sets a limit on the signal-to-noise ratio for attosecond pump-probe measurements, and therefore, other streaking techniques have resulted in time-resolved measurements with sub-10-attosecond accuracy. Recent improvements on high-power ultrafast diode-pumped solid-state and fiber lasers will offer alternative sources for megahertz attosecond pulse generation. First proof-of-principle experiments have been demonstrated.

Watching ultrafast processes in real time has been the driving force to generate increasingly shorter laser pulses. In 2009, we have seen that many more laboratories around the world can generate single attosecond pulses, as new techniques have relaxed the pulse-duration requirements from intense Ti:sapphire laser systems. The shortest pulses today are still about 80 as long and were demonstrated in 2008 [1] using intense ≈3.3-fs pulses and spectral filtering in the XUV. Previously, more pulse energy in the nanojoule regime was generated with a combination of polarization gating and a somewhat longer intense laser pulse of 5 fs generating 130-as pulses [2]. The polarization gating technique was further improved in combination with an intense two-color field which breaks the symmetry and only supports one high harmonic recombination event per cycle, which is referred to as the double-optical gating (DOG) technique. Pulses as short as 107 as with an 8-fs intense pulse have been generated [3], [4]. The requirements on the intense laser pulse have been further relaxed with the generalized DOG (GDOD) technique generating 148-as pulses, starting with much longer pulses of 28-fs duration—clearly in the many cycle regime at 800 nm [5].

A general challenge remains, however, because the signal-to-noise ratio for typical attosecond pump probe measurements is reduced by at least five orders of magnitude compared with the femtosecond regime since the pulse energy (as high as 1 nJ) comes at a much lower pulse repetition rate of 1 kHz instead of 100 MHz. To date attosecond measurements have been done in combination with streaking techniques mapping time to energy [6] or time to angular momentum (referred to as attosecond angular streaking) [7]. The latter approach has allowed for the fastest measurement that has ever been made directly in the time domain and which sets an upper limit in the tunneling delay time—even without using any attosecond pulses [8].

It is typically assumed that electrons can escape from atoms through tunneling when exposed to strong laser fields, but the timing of the process has been controversial and far too rapid to probe in detail. The Keller group has used attosecond angular streaking [7] to place an upper limit of 34 as and an intensity-averaged upper limit of 12 as on the tunneling delay time in strong field ionization of

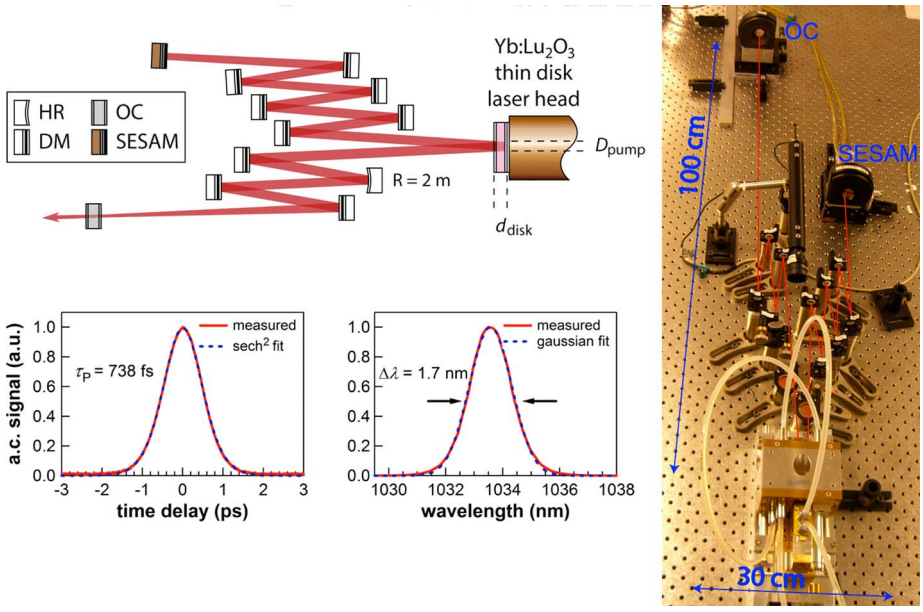

Fig. 1. SESAM mode-locked $Yb:Lu_2O_3$ thin-disk laser with 141-W average output power at a pump power of 349 W, which results in an optical-to-optical efficiency of 40%. The pulses have a time–bandwidth product of 0.35 at the full output power: which is very close to an ideal soliton pulse (i.e., 0.315), as expected from soliton mode locking. (HR: high reflector; OC: output coupler; DM: dispersive mirror; R: radius of curvature.)

a helium atom in the nonadiabatic tunneling regime [8]. To achieve this the exact timing of an intense, close-to-circular polarized laser field in the two-cycle regime was exploited—similar to the hands of a clock. This measurement is exactly based on the definition of "time" by "counting cycles"—here, a fraction of a cycle is measured. The experimental results give a strong indication that there is no real tunneling delay time, which is also confirmed with numerical simulations using the time-dependent Schrödinger equation. This measurement was done over a Keldysh parameter variation of 1.45 to 1.17 for which we could apply a small Coulomb field correction using a semiclassical picture. At higher intensities, we would expect a stronger Coulomb correction which, however, was not observed experimentally last year. This may shed some light on the ongoing discussion when simple semiclassical pictures start to break down on an atomic scale. Theory is still catching up with these new experimental results, which is a general observation in high field physics experiments. Generally, the community needs better theoretical models with valid approximations that can describe the attosecond dynamics on an atomic and molecular level. Solving the time-dependent Schrödinger equation becomes an impossible task for larger targets. Single-atom and small-molecule experiments may help to find valid approximations that can correctly predict the detailed dynamics and can reduce the numerical complexity for future models.

So far, attosecond streaking techniques have been very successful and have demonstrated that new and unexpected experimental results can be obtained when the attosecond dynamics is observed on an atomic scale. More surprises are expected, and this is what makes it exciting and motivating to push forward the frontier in time-resolved measurement techniques. However, streaking techniques have limitations, and increased efforts are being dedicated toward attosecond pulse generation in the megahertz regime and/or higher pulse energies in the kilohertz regime to ultimately allow for attosecond pump-probe measurements, where one attosecond pump pulse initiates and another time-delayed attosecond pulse probes the fast process. For example, a 100-μJ pulse energy at 5-MHz pulse repetition rates requires an average power of 500 W. Ti:sapphire lasers are not suitable for such high average power [9], and currently, the most promising femtosecond laser systems are either based on fiber laser amplifier systems, SESAM mode-locked thin-disk lasers, or optical parametric amplifiers pumped by high-average-power picosecond thin-disk lasers [10]. To

date, a femtosecond fiber CPA system has generated 830 W [11], and a passively mode-locked Yb:Lu_2O_3 thin-disk laser has generated more than 120 W of average power [12], which was recently improved further to more than 140 W with an optical-to-optical efficiency of 40%. The thin-disk laser approach requires no amplifiers, the complexity is not higher than for low-power laser oscillators (see Fig. 1), and novel Yb-doped laser materials have a great potential for sub-100-fs pulse generation [13]. In 2009, we observed the first megahertz HHG generation using high-power Yb-doped fiber CPA systems [14], and we have successfully demonstrated a lower pulse energy requirement for HHG in photonic crystal fibers [15] such that pulse energies in the tens of microjoules are sufficient for efficient HHG, which is a value easily accessible by SESAM mode-locked thin-disk lasers [16], [17].

The rapid progress in high-average-power femtosecond laser technology will continue. The development of high average photon flux, compact, tabletop ultrafast VUV/XUV sources will enable new measurements in photoelectron imaging spectroscopy, surface science, metrology, and biology and could become key instrumentations for laser accelerators and high-repetition-rate free-electron lasers (FELs). The pulse energy of low-repetition-rate systems very often has to be increased to a level where space-charge effects (i.e., electromagnetic forces between the generated charged particles within the interaction volume) start to blur the dynamics to be investigated—simply because the minimal detectable signal requires a certain average photon flux. A high photon flux but low pulse energy will also be very important for surface science and condensed matter physics using, for example, angle-resolved photoemission spectroscopy (ARPES) (e.g., in [18]). Strongly correlated electron systems with their complex physics are one of the biggest challenges in condensed-matter physics today. For example, space-charge effects have been a serious limitation for time-dependent measurements in high-T_c superconductors in the VUV regime [19]. A higher photon flux at higher repetition rates with lower pulse energies would allow for time-resolved ARPES.

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