Two-color facility based on a broadly tunable infrared free-electron laser and a subpicosecond-synchronized 10-fs-Ti:sapphire laser

G. M. H. Knippels, M. J. van de Pol, H. P. M. Pellelman, P. C. M. Planken, and A. F. G. van der Meer

FOM-Institute for Plasma Physics Rijnhuizen, P.O. Box 1207, 3430 BE, Nieuwegein, The Netherlands

Received August 3, 1998

Subpicosecond synchronization between a mirror-dispersion-controlled 10-fs Ti:sapphire laser and the Free-Electron Laser for Infrared Experiments has been achieved. The measured intensity cross correlation between the two lasers is consistent with a jitter of only 400 fs rms. The wide and continuous tunability of the free-electron laser (FEL; 4.2–300 μm) combined with ultrashort pulse duration of six optical cycles and high pulse energy of several tens of microjoules makes a series of two-color experiments possible in a previously inaccessible wavelength range. We demonstrate these capabilities by performing a two-color pump–probe experiment to study carrier cooling in GaAs. A FEL tuned from 8 to 17 μm is used as the pump, and a synchronized Ti:sapphire laser pulse serves as the probe.

In studies of fast dynamics in semiconductor systems many different nonlinear optical techniques are used. A well-known technique is the pump–probe technique, in which one measures the lifetime of an excited state by monitoring the absorption change of a weak probe pulse when the delay is scanned with respect to a strong pump pulse. The use of a second independently tunable frequency for the probe pulse in this pump–probe scheme strongly enhances the versatility of the technique and allows for a new class of experiments. Typical two-color sources are optical parametric oscillators and optical parametric generator–optical parametric amplifiers pumped by Ti:sapphire lasers, which are capable of generating two optical pulses with different frequencies. With the current status of these systems it is possible to generate IR wavelengths of up to 10 μm easily, and also, at the cost of strongly decreasing pulse energy, up to 15 μm.

With the use of a free-electron laser (FEL), the IR wavelength range that can be covered, compared with that of an optical parametric generator–optical parametric amplifier, is extended from the mid IR to several hundred micrometers. This wavelength range is important for the study of intraband absorption, phonons, and intersubband absorption in semiconductors. In addition to this extended wavelength range, FEL’s such as the Free-Electron Laser for Infrared Experiments (FELIX, Nieuwegein, The Netherlands) offer several other attractive features. Besides their broad wavelength coverage, FEL’s are rapidly tunable (by a factor of 4 in less than 1 min) and generate pulses with energies as great as 50 μJ. This high pulse energy, combined with an ultrashort pulse duration of only a few optical cycles, leads to peak powers of more than 100 MW. Also important is the possibility of adapting the optical pulse duration of the FEL (typically more than a factor of 10) and optimizing it for the system under study. Another particular characteristic of the FEL is its time structure, which is rather different from that of Ti:sapphire-based systems. It consists of a burst of picosecond-long micropulses separated by 1, 20, or 40 ns. This burst of micropulses has a typical length of 5 μs (the macropulse) and is repeated at 10 Hz.

In this Letter we report what we believe to be the first successful subpicosecond synchronization of a Kerr-lens mode-locked 10-fs Ti:sapphire laser system (Femto Source Pro; Femto Lasers, Vienna, Austria) to a FEL. Previous attempts to synchronize a different FEL with a regeneratively mode-locked Ti:sapphire laser led to several picoseconds of jitter. The Ti:sapphire laser described here was pumped by 5 W of 532-nm radiation from an intracavity frequency-doubled Nd:YVO₄ laser (Millenia V; Spectra-Physics, Mountain View, Calif.) and employed so-called chirped-mirror technology to achieve reliable and stable production of ultrashort optical pulses. We modified the cavity by mounting a tiny (5-mm-diameter, 1-mm-thick) high reflector in the short arm of the cavity on a piezoelectric transducer with 10-μm range. This piezo-controlled mirror was used for active cavity stabilization. We used a fast Si photodiode (−3 dB at 1.5 GHz) to monitor the 100-MHz optical pulse train. The 1-GHz component of this signal and a reference signal of the 1-GHz clock of the FEL were combined in a double-balanced mixer, the output of which served as the error signal for driving the piezoelectric transducer. The unity gain bandwidth of the loop was 1 kHz, determined primarily by the piezoelectric transducer drive electronics and the loop filter. A schematic layout of the feedback loop is given in Fig. 1. The use of the 10th harmonic of the 100-MHz round-trip frequency in the feedback loop provides better locking because of the reduced influence of amplitude–phase-noise ratio in the loop.

When the FEL and the Ti:sapphire laser were frequency locked to each other, we found the temporal overlap by simply scanning a phase shifter. We used type I sum-frequency generation in a 100-μm-thick AgGaS₂ crystal (MolTech, Berlin, Germany) at a FEL wavelength of 9 μm to measure the degree
Fig. 1. Schematic layout of the synchronization electronics and optics. An ultrastable rf clock (SMG 801.0001.52; Rohde & Schwarz, Munich, Germany) running at 250 MHz is multiplied by 4 to yield 1 GHz. A rf splitter splits the 1-GHz signal into two. One arm serves as the reference clock input for the double-balanced mixer (DBM), and the other part drives the electron gun and the accelerating structures that generate the electron beam that pumps the FEL. A fast silicon diode (Si-PIN) detects the 100-MHz Ti:sapphire pulse train and filters out the 1-GHz component, which is mixed with the 1-GHz clock in the double-balanced mixer, yielding the phase difference. After passing through a 1-MHz low-pass filter (LPF) and a loop filter (unity gain at 1 kHz), the error signal is sent to a piezoelectric transducer (PZT) driver for adjustment of the cavity length. A phase shifter is used to control temporal overlap. BS, beam splitter; BPF, bandpass filter.

of jitter that was left between the two lasers in a direct optical cross-correlation experiment. The sum frequency was detected through a calcite polarizer that blocked the Ti:sapphire background. As shown in Fig. 2, we scanned a delay line to vary the optical delay between the two laser pulses, and the individual data points in the figure correspond to a 50-shot-averaged measurement over successive macropulses. The duration of the Ti:sapphire laser pulses was measured separately with an autocorrelator that was close to the AgGaS$_2$ sample and was found to be 10 fs FWHM (see the right-hand-side inset in Fig. 2). The IR FEL pulse duration was similarly measured with a CdTe-based autocorrelator$^6$ to be 435 fs FWHM in duration (see the left-hand-side inset). The jitter from the cross-correlation signal was thus estimated to be 400 fs rms ($\sim$900 fs FWHM, assuming uncorrelated Gaussian noise). The degree of synchronization that was achieved was present over many minutes, and the resulting system was competitive with other commercially available systems that allow locking of a Ti:sapphire laser to an external clock. For example, Spectra-Physics and Coherent systems typically specify a jitter of less than 3 ps rms ($\sim$6 ps FWHM) on a 60-s time scale or shorter for tabletop systems. In our case the FEL was located 40 m from the Ti:sapphire laser, and the excellent synchronization that was achieved demonstrates the intrinsically stable cavity design of the Ti:sapphire laser as well as the tight locking of the FEL to its 1-GHz clock. The large separation between the laser led to a slow thermal drift in synchronization of $\sim$1 ps per 30 min.

Although we demonstrated the synchronization of the Ti:sapphire laser to the FEL at an IR wavelength of 9 $\mu$m, this synchronization is limited only by the availability of suitable sum-frequency mixing crystals (and was verified at 7–13-$\mu$m wavelength with the same AgGaS$_2$ crystals). The synchronization is present throughout the whole FELIX wavelength (4.2–300 $\mu$m) range, since we directly locked the Ti:sapphire laser to the FEL’s internal rf clock and did not rely on any wavelength-dependent feedback mechanism.

In a separate experiment we used this synchronized two-color laser system to perform a preliminary pump–probe experiment in an n-doped GaAs sample. Since the sample was intentionally doped, the electron states at the bottom of the conduction band were filled. In the experiment we used the intense IR FEL pulse (pulse duration, 0.5–1 ps; energy, $\sim$1 $\mu$J per micropulse) to heat the electron gas by promoting electrons from lower to higher states in the conduction band, by means of an intraband absorption process. We then used the delayed Ti:sapphire pulse to measure a change in transmission of the interband transition at wavelengths near the bandgap of GaAs. In Fig. 3 the results at room temperature are given for different FEL wavelengths. The results clearly show a FEL-induced enhanced absorption that decays on a time scale of several picoseconds. The fast rising edge of the signal is another indication of the good synchronization between the two lasers. The transmission is decreased because after FEL excitation more states near the bottom of the conduction band to which electrons can be excited from the valence band at the Ti:sapphire wavelength become available. The decay time of the signal agrees with the typical cooling times of the hot electron gas, caused by longitudinal-optical–phonon emission.$^{10}$ The longer decay time in

Fig. 2. Measured optical cross correlation between the FEL running at 9 $\mu$m and the Ti:sapphire laser at 800 nm in a 100-$\mu$m-thick AgGaS$_2$ crystal. The inset on the left-hand side shows the background-free autocorrelation measurement of the 435-fs FEL pulse with a homebuilt autocorrelator based on CdTe,$^6$ and the inset on the right-hand side shows the 10-fs fringe-resolved autocorrelation function of the Ti:sapphire pulse. The derived jitter is 400 fs rms ($\sim$900 fs FWHM). The scan took approximately 2 min to record.

November 15, 1998 / Vol. 23, No. 22 / OPTICS LETTERS 1755
Fig. 3. Absorption change of the Ti:sapphire pulse through a 300-µm-thick bulk GaAs samples as a function of delay with respect to the IR pump pulse from the FEL. The different traces were taken at room temperature and correspond to different doping levels and different IR wavelengths and powers. Trace (a) was taken at the 16.7-µm wavelength and $3 \times 10^{17}$ cm$^{-3}$ doping, trace (b) at 8 µm and $3 \times 10^{18}$ cm$^{-3}$ doping, and trace (c) also at 8 µm and $3 \times 10^{18}$ cm$^{-3}$ doping but with 10 times more FEL power.

Fig. 3(c) probably is due to the occurrence of intervalley scattering of carriers at higher FEL intensities. Near zero delay there might also be increased transmission of the Ti:sapphire pulse owing to the ac Stark effect induced by the strong electric field of the FEL pulse. Note that, in principle, in a future experiment one could exploit the broad spectrum of the 10-fs Ti:sapphire pulse to measure the frequency-resolved cooling times over the 700–900-nm wavelength range.

We have successfully synchronized a passively mode-locked 10-fs Ti:sapphire laser to the FELIX free-electron laser. The two laser systems are separated by several tens of meters and share only a stable 250-MHz signal generator. The resulting measured jitter is only 400 fs rms. The system allows for a whole series of new two-color experiments, especially those in which high peak powers in the IR or tunable beyond the range covered with optical parametric amplifier–optical parametric generator systems is required.

This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie, which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek. H. P. M. Pellemans is grateful to the UK Engineering and Physical Sciences Research Council for financial support.

G. M. H. Knippels’s e-mail address is knippels@rijnh.nl.

References

4. See, for example, the specifications of the TOPAS-800 parametric generator–amplifier from Excel-Quantronix.