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Single-shot spectra of temporally selected micropulses from a mid-infrared free-electron laser by upconversion

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We demonstrate the measurement of single-shot spectra of temporally selected micropulses from a mid-infrared free-electron laser (FEL) by upconversion. We achieve the upconversion of FEL pulses at 11 μm using externally synchronized Nd:YAG or microchip laser pulses at 1064 nm to produce sum-frequency mixing signals at 970 nm, which are detected by a compact CCD spectrometer without an intensifier. Our experimental system is very cost-effective, and allows us to obtain the laser spectra of selected micropulses at any temporal position within a single macropulse from an oscillator-type FEL.   © 2012 Optical Society of America
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Since the first realization of lasing in 1976 [1], free-electron lasers (FELs) have been attracting lots of interest in various research areas due to their large tunabilities in wavelength [2]. In order to apply the FEL pulses for processes involving resonances [3], precise knowledge of laser spectrum as well as pulse duration is particularly important. Moreover, since the lasing mechanism of FELs is essentially different from that of conventional lasers, such information on FEL pulses themselves sheds crucial light for the profound understanding of lasing mechanism of FELs. This can be particularly true for the oscillator-type FELs, which exhibit a dual pulse structure. A macropulse with a duration of microsecond to millisecond contains thousands of micropulses with a duration of femtosecond to picosecond at a time interval of hundreds of picosecond to several nanosecond. We point out that most of the FELs working at the mid-infrared (MIR) wavelength range are the oscillator type. For this reason, the pulse measurements of MIR FELs are almost always carried out by averaging over many macropulses, each of which contains thousands of micropulses. For instance, even a single-macropulse measurement of the FEL spectra, which is usually considered to be easy, is not so easy in the MIR range, and it requires the use of an array-type mercury cadmium telluride (MCT) or pyroelectric photodetectors whose resolution is limited by the low pixel density (typically 64–256 elements). Moreover, although the use of such array-type photodetectors allows us to obtain the MIR FEL spectrum for a single macropulse, it is still an average over thousands of micropulses within a single macropulse, since the time response of those detectors are rather slow. As a result, although the MIR FEL spectra of temporally selected micropulse(s) are much more valuable to monitor the laser spectra during the spectroscopic experiments and to understand the detail of the lasing process, such study does not exist in the literature.

The purpose of this Letter is to report the measurement of single-shot spectra, for the first time to our knowledge, of *temporally selected micropulse(s)* from an MIR FEL by upconversion using an externally synchronized laser source at 1064 nm. Although the upconversion of MIR pulses with visible or near-infrared (NIR) pulses was first reported quite some time ago [4,5], it is much more recent that the idea has been applied for the MIR [6–9]. Related to the present work we note that the upconversion of tetrahertz radiation from an FEL by an NIR continuous-wave laser has been recently reported in [10].

We would like to note that whether the upconversion technique works equally well for the case of MIR FELs is not *a priori* obvious due to the following reasons: First, our MIR pulses from the FEL form a train of several thousands of micropulses, while in all the works mentioned above the MIR pulses are isolated. The peak intensity one can safely use for an isolated pulse may be too much for a pulse train, and can damage a nonlinear crystal for upconversion. Second, we do the synchronization of two independent lasers and hence there is some timing jitters, while in all the above works both MIR and NIR pulses are from the single light source and hence the synchronization is nearly perfect. To reduce the problem of timing jitters we employ the nanosecond or sub-nanosecond lasers at the expense of wasting most of the pulse energy due to the mismatch of the pulse durations between the MIR and NIR pulses. Third, due to the use of a NIR laser source at 1064 nm the sum-frequency mixing (SFM) signals in our case appear around 970 nm where the sensitivity of Si-based CCD photodetectors is usually quite low (<10% of its maximum value, which typically appears around 500–700 nm), while the sensitivity of the detector is much higher if one uses, for example, a Ti:Sapphire laser at the expense of its cost.

The experimental setup is shown in Fig. 1. The Kyoto University free-electron laser (KU-FEL) operates at 11 μm with a repetition rate of 1 Hz [11]. Each macropulse has a duration of ~1.5 μs and contains several thousands of micropulses with a duration of about 0.7 ps and an interval of 350 ps between them. As for the 1064 nm laser we employ either the *Q*-switched Nd:YAG (LOTIS TII, model LS-2136, multimode, 20 ns pulse duration, 50 ns jitter) or actively *Q*-switched microchip lasers (Standa, STANDA-Q1, single-mode, 0.8 ns pulse duration,
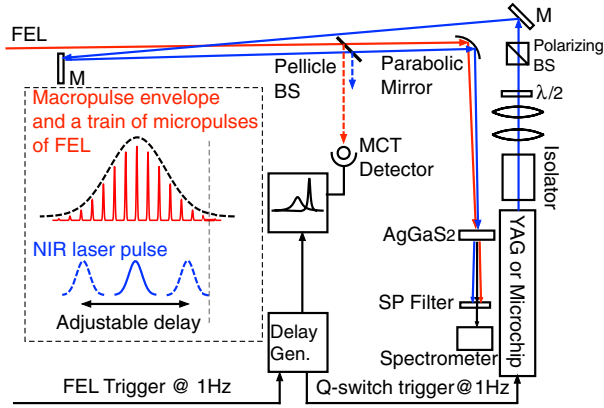
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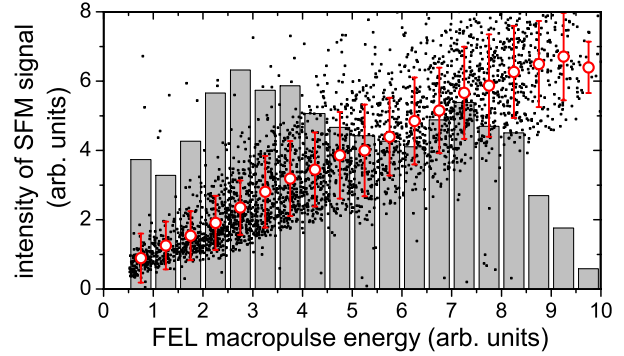
Fig. 1. (Color online) Experimental setup.

Fig. 2. (Color online) Correlation between the intensity of SFM signals and FEL macropulse energy. Small black dots, Raw data; Gray columns, Frequency of events; Red circles and error bars, Averaged SFM signal intensity and error bars.

<350 ps jitter). In either case the Q-switch of the 1064 nm laser is triggered by the emission timing of KU-FEL with some controlled delay through the delay generator (SRI, DG645). Note that the timing jitter of the NIR lasers is not a serious problem for our case, since their pulse durations are larger than the interval between the micropulses of FEL.

Unless otherwise noted, we employ the Nd:YAG laser for upconversion. A half-wave plate and a polarizing beam splitter are introduced for the 1064 nm pulse so that its polarization axis becomes parallel to that of the FEL pulse. Then, both beams are focused onto the $AgGaS_2$ crystal (type I, 2 mm thickness, 37 degrees cut angle) using a gold-coated off-axis parabolic mirror with a focal length of 15 cm. The SFM bandwidth for the 2 mm crystal is about 0.7 μm at 11 μm, which is much larger than the bandwidth of the retrieved FEL spectrum, ~0.3 μm (see Fig. 3(d)), which ensures that our choice of the crystal thickness is appropriate. The energy of FEL pulses is 4 mJ per macropulse, while that of the 1064 nm pulses is 5 mJ per pulse at the $AgGaS_2$ crystal.

A rough estimation shows that each macropulse of KU-FEL contains approximately 4000 micropulses and hence the energy of each micropulse is about 1 μJ. Since the diameters of the two beams at the crystal are measured to be about 1 mm for both, the peak intensities of the FEL micropulse and 1064 nm pulse are about 130 MW/cm^2 and 25 MW/cm^2, respectively. The relatively low peak intensities prevent not only damage on the crystal but also distortion of the spectrum by self-phase modulation in the crystal. The two incident beams have a small cross angle of about 2.5° so that we can spatially separate the SFM signals from the incident beams. After the $AgGaS_2$ crystal we place a shortpass filter with a cut-off wavelength at 1000 nm to block the 1064 nm pulse. For the detection of SFM signals, we employ a commercial compact spectrometer (OceanOptics, model HR4000CG-UV-NIR) with a slit width of 5 μm and grating of 300 lines/mm (0.75 nm spectral resolution). The spectrometer is also synchronized with the laser system. As a reference we pick up a small portion of the FEL pulse by a Pellicle beam splitter to monitor the temporal shape and energy of the macropulse by an MCT detector, and store the data with a two channel digital oscilloscope.

As a first test we have measured the SFM signal intensity as a function of FEL macropulse energy. The results are shown in Fig. 2. The shot-to-shot change of the 1064 nm pulse energy is <20%. Regardless of the large shot-to-shot fluctuation of FEL macropulse energies, we can clearly see the nearly linear dependence, as we expect. Recall that the SFM signal intensity is proportional to the micropulse intensity (and with a good approximation macropulse energy) of FEL. By calibrating the spectrometer sensitivity at ~970 nm, we estimate the SFM efficiency to be about 0.2%–0.6%.

In Fig. 3(a) we present a few raw spectra of the SFM signals, each of which has been obtained by upconversion of temporally selected micropulses of the different macropulse. The timing of the 1064 nm pulses is set to be at about 1.2 μs after the peak of the FEL macropulse envelope (see the inset in Fig. 3(a)). Because the pulse duration of the Nd:YAG laser is 20 ns and the time interval between the FEL micropulses is 350 ps, the number of FEL micropulses we have selected for upconversion is about 57. In order to retrieve the MIR FEL micropulse
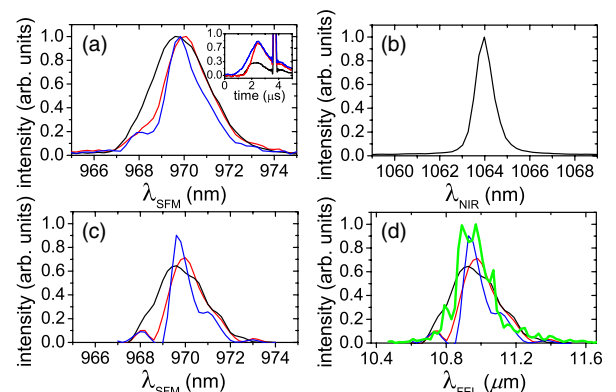

Fig. 3. (Color online) Retrieval of the spectra of FEL micropulses by upconversion. (a) Raw spectra of SFM signals. Three examples, normalized by their peak values, are shown. The inset shows the corresponding macropulse shapes. where the narrow pulse appearing at 3.5 μs is the 1064 nm pulse. (b) Spectrum of the microchip laser. (c) Deconvolved spectra of SFM signals from raw spectra in (a). (d) Retrieved MIR spectra of the temporally selected FEL micropulses and the MIR FEL spectrum (thick green curve) taken by the scanning-type monochromator (height not in scale).
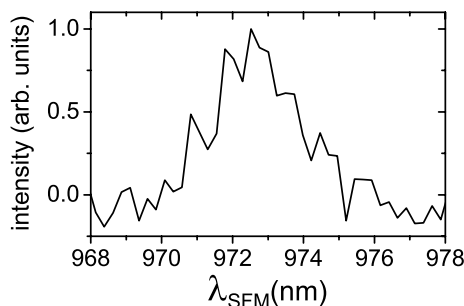
Fig. 4.　Raw spectrum of the SFM signal with the microchip laser.

spectra, we must perform the deconvolution. We have found that the spectral profiles of the multimode Nd:YAG ($\sim$0.45 nm bandwidth) and single-mode microchip ($<3.7 \times 10^{-3}$ nm bandwidth) lasers measured by our spectrometer are almost identical, as shown in Fig. 3(b). This implies that the spectral resolution of our spectrometer, 0.75 nm, is much worse than the linewidths of the Nd:YAG and microchip lasers, and the spectral profile shown in Fig. 3(b) is nothing but the instrumental function of the spectrometer, which we use for deconvolution. The deconvolved SFM spectra are shown in Fig. 3(c). Once the SFM spectra have been obtained by deconvolution, we can retrieve the spectra of the temporally selected FEL micropulses, λ_{FEL}, by using the relation of $\lambda_{\text{FEL}} = (\lambda_{\text{SFM}}^{-1} - \lambda_{\text{NIR}}^{-1})^{-1}$, where λ_{SFM} and λ_{NIR} are the wavelengths of the SFM signal and NIR pulse, respectively. The retrieved spectra of temporally selected FEL micropulses are shown in Fig. 3(d). For comparison we also show in Fig. 3(d) the MIR FEL spectrum taken by a scanning-type monochromator. The agreement is reasonably good.

In order to improve the temporal resolution, we now replace the 20 ns Nd:YAG laser by the 0.8 ns microchip laser. Since the effective pulse energy of the microchip laser is about half of that of the Nd:YAG laser, we move the AgGaS$_2$ crystal closer to the focal point, and the diameters of the FEL and 1064 nm beams are reduced to 0.5 mm, which results in the peak intensities of 500 MW/cm^2 and 50 MW/cm^2, respectively. Regardless of such high peak intensities we find that one hour laser irradiation at 1 Hz does not induce any visible damage on and in the crystal.

In Fig. 4 we show a raw spectrum of the SFM signal obtained with the microchip laser. Note that only $\sim$2 micropulses of FEL are selected for upconversion.

Although the SFM signals become much weaker, we believe that there is a lot of room for improvement through the replacement of the spectrometer by the one with higher sensitivity ($> \times 20$) and the grating by the one with much better ($> \times 5$) diffraction efficiency and spectral resolution.

In conclusion, we have demonstrated the measurement of single-shot spectra of temporally selected micropulses from the MIR FEL by upconversion. With the microchip (Nd:YAG) laser we are able to obtain the single-shot spectra of only $\sim$2 (57) micropulses of MIR FEL at any temporal position within a macropulse. The spectral resolution of our current system is $\sim$7 cm^{-1} ($\sim$10 cm^{-1}) with the microchip (Nd:YAG) laser, and it is mostly limited by the resolution of the spectrometer. If we use a spectrometer with a finer grating, say, 1200 lines/mm, we should be able to improve the resolution of the system to $\sim$1.2 cm^{-1}. The technique described in this work will be useful not only for the online monitoring of the FEL spectra but also for the investigation of the correlation between the properties of the electron beam and those of the FEL beam.

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