

Temporal characterization of mid-IR free-electron-laser pulses by frequency-resolved optical gating

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We performed what we believe are the first practical full-temporal-characterization measurements of ultrashort pulses from a free-electron laser (FEL). Second-harmonic-generation frequency-resolved optical gating (FROG) was used to measure a train of mid-IR pulses distorted by a saturated water-vapor absorption line and showing free-induction decay. The measured direction of time was unambiguous because of prior knowledge regarding free-induction decay. These measurements require only 10% of the power of the laser beam and demonstrate that FROG can be implemented as a pulse diagnostic simultaneously with other experiments on a FEL.

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Free-electron lasers (FEL's) lase at wavelengths not easily accessible with conventional lasers.^{1,2} Several user facilities provide mid-IR and far-IR radiation,² and short-wavelength facilities are planned. Most FEL's use radio-frequency linear accelerators as the electron beam source and produce synchronously pumped picosecond laser pulses tens of wavelengths long.¹ FEL users perform a variety of time-resolved spectroscopy experiments including pump-probe, photon-echo, and transient-grating measurements, whose temporal and spectral resolution is limited by the laser pulse length and spectral width, respectively, and can be degraded if the time-bandwidth product is not optimal. Spectral and autocorrelation measurements provide a partial description of the laser pulses, but both present and future experiments will benefit significantly from more comprehensive pulse characterization.

Frequency-resolved optical gating (FROG) is a technique for measuring the complete electric field of an optical pulse: both intensity and phase versus time or frequency.³ FROG involves spectrally resolving the signal pulse in an autocorrelation measurement. Various second- and third-order nonlinearities and beam geometries have been used for the autocorrelation measurement in FROG. Previously we used a (third-order) polarization-gate FROG arrangement to measure mid-IR FEL pulses.⁴ The utility of this approach was limited, however, because the nonlinear medium, germanium, depolarized the light, yielding excessive polarizer-leakage background. Also, the entire FEL beam power was required for these measurements. We present here significantly improved measurements of FEL pulses by second-harmonic-generation (SHG) FROG.⁵ These measurements were made on the mid-IR FEL at Stanford University⁶ at a wavelength near 5 μm . Although other researchers measured the temporal and spatial dependence of the intensity and phase of longer, microwave FEL pulses,⁷ to our knowledge these are the first practical full-characterization pulse measurements performed in the mid IR or on an ultrashort-pulse FEL. The measured pulses were ~ 1 ps long and were perturbed by a water-vapor absorption line and thus exhibited free-induction decay.

The SHG FROG measurement required only 100 nJ of energy, 10% of the laser power, and occurred simultaneously with other experiments that used the rest of the beam. Thus SHG FROG is well suited as a real-time diagnostic for the FEL and could replace the autocorrelator and spectrometer currently used.⁸

The FEL beam comprises groups of thousands of picosecond, 2- μJ micropulses, each separated from successive pulses by ~ 80 ns.⁶ These micropulse groups, or macropulses, are typically 2 ms in duration and recur every 100 ms. Variations in the pulse can occur for micropulses early in the macropulse, so we restricted our measurements to the central 0.5 ms of the macropulse and averaged these pulses over 1–4 macropulses for each point in the FROG trace.

Figure 1 is a schematic of the SHG FROG apparatus. Because the SHG FROG trace is the set of spectra versus delay in a SHG intensity autocorrelation, array detectors are desired. Mid-IR array detectors are becoming available and appear ideal for multishot (and single-shot) FROG measurements of IR pulses. We did not possess one, however, so we used a single-element mercury cadmium telluride detector and scanned both optical delay and wavelength mechanically. The optical delay consisted of a retroreflector upon a translation stage driven by a stepper motor. A second stepper motor set the wavelength of a McPherson monochromator with 30-cm focal length and a 5-cm grating. A coated calcium fluoride plate split the laser beam into two before the delay line, and a paraboloidal mirror with 150-mm focal length (along the beam path) focused and crossed the two input beams in the 2-mm-thick AgGaSe_2 doubling crystal. The 10% of the FEL beam power directed into the FROG device yielded a typical micropulse energy at the SHG crystal of 100 nJ. The spot size was ~ 100 μm , and the external crossing angle was 4° , so parallax across the spot was negligible compared with the picosecond-scale optical pulse lengths. The detectivity of the mercury cadmium telluride detector varies (linearly) with the wavelength, but the scan range was $< 2\%$ of the wavelength and the noise in the trace was 5% of its peak value, so no detectivity correction was used. Also, the SHG conversion efficiency did not

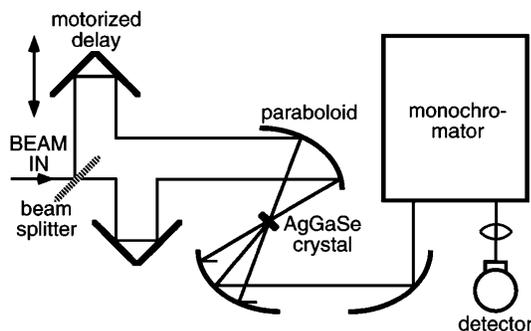


Fig. 1. Schematic of the mid-IR FROG apparatus, consisting of a SHG intensity autocorrelator, the output signal of which was fed into a monochromator. Both optical delay and wavelength were scanned mechanically, and each point in the FROG trace was averaged over several thousand optical pulses.

vary significantly over the laser bandwidth. An iris between the crystal and the monochromator blocked any SHG from the individual beams, and a large detector amplifier offset was subtracted from the traces after data acquisition, yielding slightly negative values for some points. These values were set to zero before the FROG inversion algorithm was run.

Our trace comprised 42 delays over a range of 14 ps and 64 wavelengths over a range of $0.08 \mu\text{m}$. A computer running the LabView program controlled the stepper motors and acquired the data. Several thousand micropulses were averaged (both by the detector bandwidth and by the computer) for each point in the FROG trace. The computer stepped through an entire delay scan and then moved to the next wavelength. These 42×64 scans require ~ 15 min each. Simultaneously operating during the FROG measurement was an unrelated nonlinear-spectroscopy experiment using the other 90% of the beam power.

Figure 2(a) shows an experimental SHG FROG trace of $\sim 5.13\text{-}\mu\text{m}$ wavelength FEL pulses. The trace is symmetric, as required for a SHG FROG trace, indicating that the laser remained stable over the scan. Additive trace noise was $\sim 2\%$ of the trace peak value, and multiplicative noise (which is due to laser power fluctuations) was $\sim 5\%$. Although SHG FROG traces are not as easy to interpret as PG FROG traces,⁷ we can see that the shape of this trace is clearly not an ellipse as it would be for a transformed-limited Gaussian pulse.

One can obtain information directly from the FROG trace without running the FROG algorithm. In particular, the marginals of the SHG FROG trace (the integral of the trace over either delay or wavelength) correspond to the spectral autoconvolution and the temporal autocorrelation.⁵

Running the FROG algorithm,⁹ which yields the pulse intensity and phase, currently requires a $2^n \times 2^n$ array with each axis increment equal to the reciprocal of the other axis range. We extended the trace to 128×128 by zero padding the delay axis and interpolating the wavelength data points. In the presence of significant noise, the performance of the FROG algorithm improves if the FROG trace is first low-pass filtered.¹⁰ In our case this involved two-dimensional Fourier transforming of the trace and setting all points

in the transform plane outside a radius of $0.26 \times N/2$ (where N is the number of delays and wavelengths) to zero. This operation suppressed the trace's high-frequency noise, which was clearly not relevant to the pulse, but also slightly broadened the actual trace features, by at most a few percent in our measurement.

Figure 2(b) shows the retrieved pulse's temporal intensity and phase profiles. The algorithm actually returned the time-reversed replica of Fig. 2(b), but the direction of time is ambiguous in SHG FROG, and the physics of the pulse shape described below dictates that Fig. 2(b) has the correct direction. The time reversal involves reversing the intensity versus time and reversing and negating the phase. The instantaneous frequency is thus reversed with no negation. The rms width of the main peak is 0.7 ps, and that of the whole pulse is 1.1 ps. The inset compares the computed autocorrelation when the retrieved intensity is used with the measured autocorrelation computed by finding the experimental delay marginal.

The retrieved spectrum in Fig. 2(c) has a rms width of 12 nm, giving a reconstructed rms time-bandwidth product of 1.0. It also has an absorption dip at $5.14 \mu\text{m}$, which is within monochromator-calibration uncertainty of a known absorption line at $5.148 \mu\text{m}$.¹¹ This is one of many lines of water vapor between 5 and $7 \mu\text{m}$ (Ref. 11) that affect the FEL beam as it travels through the atmosphere. The observed dip was broadened for several reasons: the absorption was highly saturated, a weaker (but also saturated) line was within the observed width, and (to a lesser extent) the trace was filtered as described above. Only the last 3 m of the beam path before the doubling crystal were in open air; the rest of the path was in vacuum. Because the line is narrower than the pulse spectrum, the water molecules continue to oscillate and emit coherently after the initial pulse has passed for a time corresponding to the reciprocal of the absorption line width (including any broadening). This action is known as free-induction decay, and it appears in Fig. 2(b) as the small trailing pulse in the time domain.

The dotted curve in Fig. 2(c) is the reconstructed spectral phase. (We time reversed the spectral profile by negating the spectral phase and leaving the spectrum unchanged.) We also deconvolved the experimental trace spectral marginal (a nearly direct measurement of the spectrum) to compare with the reconstructed spectrum (points).

A better check on the convergence of the algorithm is a comparison of the experimental and retrieved FROG traces. Figure 2(d) shows the retrieved FROG trace, which agrees well with the experimental trace in Fig. 2(a).

Figure 2(e) shows the predicted intensity and phase of the pulse resulting from free-induction decay assuming a slightly chirped Gaussian pulse distorted by a Lorentzian absorption line. The absorption wavelength, decay rate, and strength were adjusted to match the curves in Fig. 2(b). Although the leading edge of the main pulse in Fig. 2(b) is not Gaussian, this model accurately predicts both the intensity and the phase.

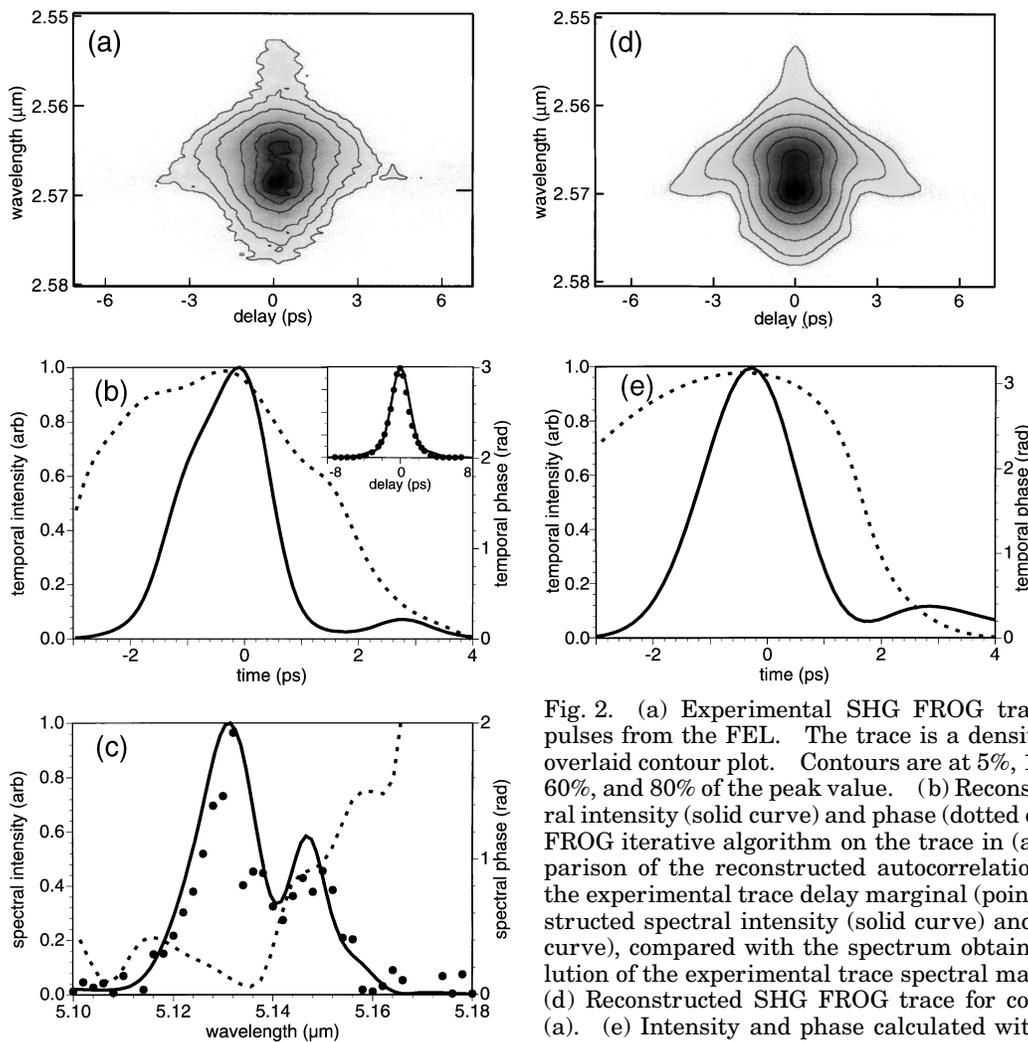


Fig. 2. (a) Experimental SHG FROG trace of 5.13- μm pulses from the FEL. The trace is a density plot with an overlaid contour plot. Contours are at 5%, 10%, 20%, 40%, 60%, and 80% of the peak value. (b) Reconstructed temporal intensity (solid curve) and phase (dotted curve) from the FROG iterative algorithm on the trace in (a). Inset: comparison of the reconstructed autocorrelation (curve) with the experimental trace delay marginal (points). (c) Reconstructed spectral intensity (solid curve) and phase (dotted curve), compared with the spectrum obtained by deconvolution of the experimental trace spectral marginal (points). (d) Reconstructed SHG FROG trace for comparison with (a). (e) Intensity and phase calculated with a model of a Gaussian pulse distorted by a narrow Lorentzian line.

In conclusion, SHG FROG is a practical real-time diagnostic for the mid-IR FEL. It requires only $\sim 10\%$ of the total laser power, the same as the currently used autocorrelator and monochromator, which offer significantly less information. Although our SHG FROG measurements required several minutes to acquire, newly available IR cameras will greatly reduce this time, permitting even single-shot operation. Finally, free-induction decay removed the time direction ambiguity in SHG FROG. Any known time-asymmetric property of the pulse will permit unambiguous measurements of the intensity and phase profile.

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