Cavity dumping of an injection-locked free-electron laser

Susumu Takahashi, Gerald Ramian, and Mark S. Sherwin

Department of Physics and Institute for Terahertz Science and Technology,
University of California Santa Barbara CA 93106

(Dated: October 2, 2009)

Abstract

This letter reports cavity dumping of an electrostatic-accelerator-driven free-electron laser (FEL) while it is injection-locked to a frequency-stabilized 240 GHz solid-state source. Cavity dumping enhances the FEL output power by a factor of ~8, and abruptly cuts off the end of the FEL pulse. The cavity-dumped, injection-locked FEL output is used in a 240 GHz pulsed electron spin resonance (ESR) experiment.

PACS numbers: 41.60.Cr, 87.64.kh
High-power pulsed electron spin resonance (ESR) is an emerging technique to investigate fast dynamics of biological molecules\(^1\). At present, most high-power pulsed ESR spectrometers operate near the X-band frequency of 9.5 GHz with kilowatt-level power and \(\sim 100\) nanosecond time resolution. A trend in the evolution of next generation pulsed ESR is for higher magnetic fields and frequencies for both finer spectral and time resolution. Because the linewidth of ESRs tends to be extremely narrow, the source radiation must have a correspondingly narrow bandwidth and stable frequency. High-power pulsed ESR, using few-ns pulses to rapidly manipulate spins for spin-echo and related experiments, has been demonstrated at 95 GHz at Cornell using a kW-power klystron-based source\(^2\). Another klystron based-pulsed ESR system has been built at St. Andrews, UK\(^3\).

A bottleneck for a higher-frequency pulsed ESR spectrometer is a scarcity of sources with high power and narrow bandwidth. Commercially-available klystron-amplifiers operating between 200 and 300 GHz can currently generate several tens of watts of peak power. Gyrotron sources with \(>100\)W peak power below 200 GHz have been developed for dynamic nuclear polarization (DNP) and ESR experiments\(^4\). Gyrotrons generating several tens of watts of continuous wave (cw) output at 1 THz have also been made\(^5\). However, the frequency of a gyrotron is usually not tunable. There are also commercially-available solid-state based sources which generate 10s of mW near 240 GHz, and operate at higher frequencies with lower power. The solid-state sources typically consist of a microwave frequency synthesizer, microwave amplifiers and frequency multipliers. Free-electron lasers (FELs) are also good candidates as sources of tunable high-power pulsed radiation at terahertz and sub-terahertz frequencies. The UC Santa Barbara (UCSB) FEL\(^6\), driven by an electrostatic accelerator, produce several micro-second pulses of quasi-cw radiation whose frequency is tunable from 120 GHz to 4.7 THz. Lasing on a single-mode with sub-megahertz line-width has recently been demonstrated by using the recently-installed injection-locking system\(^7\). The UCSB millimeter-wave FEL (MM-FEL) typically generates hundreds of watts at 240 GHz, which is being used to develop a FEL-driven pulsed ESR spectrometer\(^8\).

In this article, we demonstrate the operation of a cavity dump coupler (CDC) in combination with injection-locking of the UCSB MM-FEL. This is a key achievement for FEL-powered pulsed ESR. The CDC does two important things. First it increases peak power to above 1 KW levels, and second it abruptly switches that power off. The latter is critical in improving the signal-to-noise ratio of the pulsed ESR spectrometer.
FIG. 1: (a) Schematic of the injection locking system for the UCSB MM-FEL. The injection source, the isolator and a tunable 100 MHz synthesizer are located in free-electron laser lab. The FEL outputs are sent to the users lab through vacuum optical transport system. (b) Reflectivity characteristics of optimized Si plate with 495 µm thickness.

Fig. 1(a) shows a schematic of the UCSB MM-FEL with the CDC. The MM-FEL which covers from 120 to 890 GHz employs a 6.25 m-long folded cavity, a Si plate coupler, and optical and THz access in a room-temperature high-vacuum housing. The coupler consists of a high-resistivity silicon (Si) plate. Because the band gap of Si is much greater than photon energy at THz frequencies, the Si plate is normally transparent to THz radiation. However, when the Si wafer is illuminated by a high flux of radiation with photon energy greater than the band gap, then the plasma frequency of excited carriers exceeds the THz frequency, and the Si plate becomes highly reflective. The excitation of carriers on Si surface is so fast that the actual switching time of the Si coupler is determined by the pulse characteristics of the excitation laser. The switch remains "on", for several microseconds, limited by the recombination time of the photo-excited carriers. This photo-activated semiconductor switching technique was first demonstrated with polycrystalline germanium and later silicon.8,9
addition, the technique is regularly used for a fast pulse slicer switch of THz waves in the UCSB users lab\textsuperscript{10,11}.

As shown in Fig. 1(a), the MM-FEL CDC system has a capability to change the angle of the Si plate with respect to the incident FEL radiation away from Brewster’s angle. This provides variable coupling to maximize the FEL output power in normal operation (\textit{i.e.} without cavity dumping). The thickness of the Si plate was carefully chosen as 495 um in order to provide adequate coupling as a function of angle as shown in Fig. 1(b). In use, the Si angle is remotely controlled by a stepper motor which rotates a center shaft connected to the Si plate and flat mirror to maintain constant angle and position of the output beam. In the FEL output port, we employ a quartz window whose thickness is optimized at 240 GHz for minimal reflection.

We employ a frequency-doubled Nd:YAG laser (Big Sky Laser CFR200\textsuperscript{12}) to switch the Si coupler. The laser emits pulses with 532nm of wavelength, $7\pm2$ ns full-width at half-maximum (FWHM) and $\sim200$ mJ of energy. Timing of the switching is controlled within $\pm500$ ps accuracy by precisely triggering the Q-switch and flash lamp of the Nd:YAG laser with FEL advanced trigger pulses using a Stanford research systems delay generator DG645. The 532 nm green radiation illuminates the Si plate through an anti-reflection coated window. The CDC works in conjunction with the recently installed injection-locking system\textsuperscript{7} for single mode operation. The injection-source is coupled to the FEL cavity through the Si coupler. Injection-locking was achieved even with the minimum Si coupling at Brewster’s angle.

A CDC has been previously demonstrated in the UCSB FEL\textsuperscript{13}. Although the experiment clearly showed the switching of the FEL with a Si plate, the performance of the CDC was much lower than what is anticipated because of inadequate drive laser energy. The current CDC system has been completely upgraded with a proper drive laser and Si wafer. In addition, a combination with injection locking makes the FEL a suitable source for high-power pulsed ESR experiments.

Fig. 2(a) and (b) show the recorded 240 GHz FEL output signals without and with CDC respectively. A $\sim4.7$ $\mu$s pulse, in the normal FEL mode, was observed in the FEL user lab as shown in Fig. 2(a). The signal shows no evidence of mode-beating effects because of the operation of the injection-locking system. The peak power of the FEL pulse, measured using a Thomas-Keating energy meter\textsuperscript{14}, was $\sim190$ W in the user lab. When the CDC is operated,
FIG. 2: (a) 240 GHz FEL output without the CDC operation as a function of time. The signal was measured using a video-mode detector from VDI. The FEL signal was attenuated down to tens of microwatts in order to protect and not to saturate the detector. Digitization noise was filtered without affecting rise and fall times using a 6th order Savitsky–Golay smoothing filter. (b) 240 GHz FEL output with CDC operation. The inset shows a detailed shape of the CDC FEL signal. Rise and fall time ($\tau_r$ and $\tau_f$) of the CDC FEL $\tau \sim 10$ ns is given by a time difference between 90 % and 10 % of the signal. (c) Power spectral density of a CDC FEL pulse with the injection-locking technique. The width of the CDC FEL spectrum is approximately 22 MHz.

The FEL output trace is drastically different, as shown in Fig. 2(b). The FEL radiation rises at the time= $-3 \mu$s point, and the CDC drive laser is triggered later at time=0. In this case, the measured peak power of the CDC FEL is 1.5 kW which is approximately 8 times more than the normal FEL power. As shown in the inset of Fig. 2(b), the radiation lasts $\sim 40$ ns.
FIG. 3: Spin echo signals by Hahn echo sequence ($\pi/2-\tau-\pi-\tau$-echo) with the normal FEL (a) and CDC FEL (b). Data was taken at 1.5 Kelvin and the sample is Fe8 single molecule magnets. Details of this experiment will be presented elsewhere.

which corresponds to the $L=6.5\text{m}$ length of the FEL cavity ($t = c/2L$, where $c =$ speed of light). The rise and fall times of the CDC FEL pulse corresponds to the switching time of the Si plate. In the present case, we found the duration of the both rising and falling edge to be $\sim10 \text{ ns}$, which is similar to the FWHM of the excitation laser pulse. As explained later, this short decay time is very important to reduce background noise after the pulse. The spectrum of the CDC FEL radiation was measured by a heterodyne spectrometer. As shown in Fig. 2(c), the CDC pulse spectrum remains single mode as enforced by the injection-locking system. The observed spectral line-width is $\sim 22 \text{ MHz}$, consistent with the Fourier transform limit.

A FEL-powered pulsed ESR spectrometer is currently being developed with the injection-locked UCSB FEL. The system includes a new pulse slicer optimized at 240 GHz, a home-built detector, and protection for the detection system. Details of this setup will be presented elsewhere. As a novel application of the CDC FEL in pulsed ESR, Fig. 3 shows the first spin echo signal taken by the FEL-powered pulsed ESR spectrometer. In the case of FEL-powered pulsed ESR, the frequency of the applied pulses and detected signals is the same, therefore isolation of the applied pulses is very critical. However, because the detected ESR
signals are often less than nano-watts, while the excitation pulses are hundreds of watts, the isolation of the signal from the excitation pulse is extremely difficult. Fig. 3(a) shows a spin echo taken without cavity dumping normal FEL. Two Excitation pulses ($\pi/2$ and $\pi$) were applied before time=50 ns and the detection system was switched ON at 100 ns. After time=100 ns, as shown in Fig. 3(a), spin echo signals are hidden and distorted by a large background and noise due to imperfect pulse slicing in the pulse slicer. This issue can be solved using the CDC FEL. Because the CDC shuts off the FEL radiation quickly and completely, background and noise after the CDC FEL pulse are extremely small. Fig. 3(b) shows spin echo measurement with the CDC FEL, where timing of the spin echo is set at 200 ns. As shown in the figure, background noise is negligible and the CDC FEL gives a clear and non-distorted spin echo signal.

In summary, we have demonstrated cavity dumping of an injection locked-FEL. The CDC FEL radiation shows close to an order of magnitude enhancement in the FEL output power. The CDC FEL also gives abrupt falling edge of the FEL radiation which is critical for background-free pulsed ESR measurement. The combination of cavity dumping and injection locking in FELs enables one to use FELs for pulsed ESR as well as nonlinear spectroscopy of other systems with extremely narrow linewidths, like dilute molecular gases and Rydberg atoms.

We thank David Enyeart, Devin Edwards and Louis-Claude Brunel for support of the FEL-powered ESR operation. This work was supported by research grants from NSF (DMR-0520481, DMR-0703925 and CHE-0821589) and the W. M. Keck Foundation.

* Electronic address: susumu@itst.ucsb.edu

1 J. H. Freed, Annu. Rev. Phys. Chem. 51, 655 (2000).
2 W. Hofbauer, K. A. Earle, C. R. Dunnam, J. K. Moscicki, and J. H. Freed, Rev. Sci. Instrum. 75, 1194 (2004).
3 University of St. Andrews, Scotland, UK, http://www.st-andrews.ac.uk/~mmwave/epr/hiper.html.
4 D. A. Hall, D. C. Maus, G. J. Gerfen, S. J. Inati, L. R. Becerra, F. W. Dahlquist, and R. G. Griffin, Science 276, 930 (1997).
5 Fukui university FIR center report, FU-94, http://fir.u-fukui.ac.jp/Eng_index.html.
6 The University of California Santa Barbara Free-Electron Lasers, [http://sbfel3.ucsb.edu/](http://sbfel3.ucsb.edu/).

7 S. Takahashi, G. Ramian, M. S. Sherwin, L.-C. Brunel, and J. van Tol, Appl. Phys. Lett. **91**, 174102 (2007).

8 A. J. Alcock, P. B. Corkum, and D. J. James, Appl. Phys. Lett. **27**, 680 (1975).

9 H. Salzmann, T. Vogel, and G. Dodel, Optics Communications **47**, 340 (1983).

10 F. A. Hegmann, J. B. Williams, B. Cole, M. S. Sherwin, J. W. Beeman, and E. E. Haller, Appl. Phys. Lett. **76**, 262 (2000).

11 M. F. Doty, B. E. Cole, B. T. King, and M. S. Sherwin, Rev. Sci. Instrum. **75**, 2921 (2004).

12 Big Sky Laser Technologies Inc., Bozeman MT, USA, [http://www.bigskylaser.com/](http://www.bigskylaser.com/).

13 J. P. Kaminski, J. S. Spector, C. L. Felix, D. P. Enyeart, D. T. White, and G. Ramian, Appl. Phys. Lett. **57**, 2770 (1990).

14 Thomas Keating Ltd. West Sussex, UK, [http://www.terahertz.co.uk](http://www.terahertz.co.uk).

15 This relatively low power (for a FEL) arises because the UCSB MM-FEL was optimized to run at significantly higher frequencies, where powers in excess of 10 kW are possible without cavity dumping.