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A THz spectrometer combining the free electron laser FLARE with 33 T magnetic fields

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The free electron laser Free electron Laser for Advanced spectroscopy and high Resolution Experiments (FLARE) at the FELIX Laboratory generates powerful radiation in the frequency range of 0.3–3 THz. This light, in combination with 33 T Bitter magnets at the High Field Magnet Laboratory, provides the unique opportunity to perform THz magneto spectroscopy with light intensities many orders of magnitude higher than provided by conventional sources. The performance of the THz spectrometer is measured via high-field electron spin resonance (ESR) in the paramagnetic benchmark system 2,2-diphenyl-1-picrylhydrazyl (DPPH). The narrow ESR linewidth of DPPH allows us to resolve a fine structure with 3 GHz spacing, demonstrating a considerable coherence of the individual THz micropulses of FLARE. The spectral resolution $\Delta\nu/\nu$ is better than 0.1%, which is an order of magnitude higher than typical values for a rf-linac based free electron laser. The observed coherence of the high power THz micropulses is a prerequisite for resonant control of matter, such as THz electron spin echo spectroscopy.

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The frequency range between 0.3 and 3 THz is important for fundamental scientific research because remarkable physical phenomena, such as the pseudogap in superconductors, spin and cyclotron resonances of electrons, and vibronic and magnetic excitations in solids, are observed in this region. Technologically, this frequency range is known as the THz gap, since available THz sources (backward-wave oscillators, far-infrared lasers), as well as developments coming from microwave (gyrotrons, Schottky diode multipliers) and optical (quantum cascade lasers) techniques, do not have the same performance in power and spectral qualities as narrowband sources at microwave and optical frequencies.

Free electron lasers (FELs) produce very powerful narrowband radiation and for many applications fill the THz gap. For instance,¹ the 0.09–0.15 THz range is covered by a FEL at ENEA, the 0.12–4.75 THz range by a FEL at UCSB; 2.5–100 THz at CLIO; 1.2–70 THz at FELBE; and 0.3–100 THz at FELIX with its most recent extension, Free electron Laser for Advanced spectroscopy and high Resolution Experiments (FLARE). FELs have a very high peak power, particularly relevant for nonlinear spectroscopy, and a widely tunable spectral range compared to conventional sources, and usually a pulsed time structure. The spectral width of the FEL generated THz radiation is typically of the order $\Delta\nu/\nu \approx 1\%$ for rf-linac based FELs, which is relatively large for high-resolution THz spectroscopic studies, often requiring a width as low as 10 MHz.²

A THz FEL may serve as a source for high-power pulsed electron spin resonance (ESR) spectroscopy^{3–5} in high magnetic fields. High magnetic fields result in a strong spin polarization, thereby enhancing both the sensitivity and the resolution. Furthermore, in a spin echo experiment an intense FEL generated THz radiation pulse may induce spin

rotation by $\pi/2$ in a few nanoseconds, which is an order of magnitude faster than achievable with low-frequency solid-state sources.⁶ These improvements may allow the detection of weak or poorly resolved signals from samples that exist only in a limited quantity, as is quite common for biological systems. However, such an application requires a high-power THz pulse with a narrow bandwidth.

Details of FLARE (Free electron Laser for Advanced spectroscopy and high Resolution Experiments) are described elsewhere.^{7–9} Because of the long wavelength of FLARE radiation, a waveguide is incorporated into the optical cavity to avoid excessive diffraction losses in the undulator. This waveguide consists of two parallel metallic plates at a distance of 10 mm, extending over the entire resonator from mirror to mirror. A slit in the downstream mirror is used for outcoupling. Over its tuning range from 0.3 to 3 THz, FLARE produces intense narrowband lines but exhibits spectral gaps shown in Fig. 1. The reason for the occurrence of these spectral gaps is as yet unclear. However, in combination with the tunable magnetic field, fixed frequency operation, as it is available now, provides unique experimental opportunities for spectroscopic applications.

The time structure of the THz radiation is defined by the radio-frequency linear accelerator and has two time scales: a long optical pulse (or macropulse) with a duration of 4–8 μ s and a repetition rate up to 10 Hz, which consists of a train of short (~ 20 –200 ps) THz pulses (or micropulses) with a repetition rate of 3 GHz.¹⁰ The round-trip time of the 7.5 m long resonator is 50 ns, which means that there are 150 independent micropulses circulating in the resonator. The typical average power of FLARE shown in Fig. 1 corresponds to a very high peak power during a single micropulse: 100 mW average power in Fig. 1 corresponds to ~ 3 –30 kW of peak

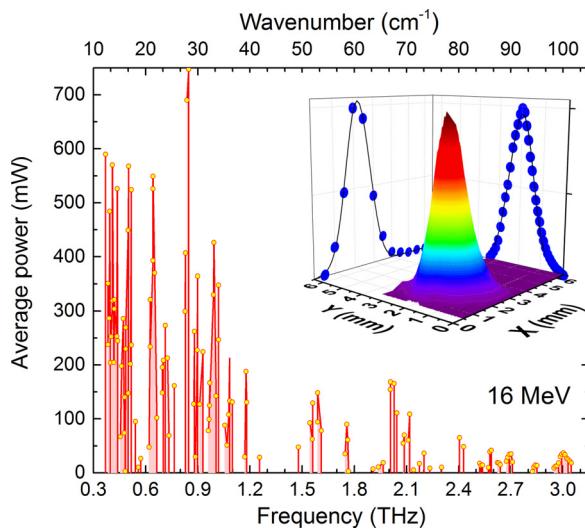


FIG. 1. THz spectrum of the FLARE laser at 16 MeV energy of the electron beam. The average power was measured at the diagnostic table. The repetition rate of macropulses is 5 Hz and pulse length is about 5–7 μ s. The inset shows the intensity profile of the 600 GHz beam focused by a 152 mm off-axis parabolic mirror.

power of the micropulse. This high peak power is especially relevant for systems with a sub-ns relaxation time. The average power in the macropulse is more than twice the highest power (1.6 kW) used before for pulsed ESR at high magnetic fields.^{4,5} Using parabolic mirrors, the THz radiation can be focused to a nearly diffraction limited, almost Gaussian spot (Fig. 1, insert), resulting in a strength of the terahertz electric and magnetic fields of 7–70 kV/cm and 2–20 mT for 100 mW of average power, respectively. These high values open perspectives for THz nonlinear spectroscopy, i.e., for pulsed electron spin resonance promising a $\pi/2$ spin rotation within a few nanoseconds. The high intensities correspond to an enormously high photon flux beneficial for nonlinear spectroscopy. For a typical THz photon energy of a few meV, the number of photons in a micropulse is 10^{16} and 10^{20} for a 3 μ s macropulse.

The spectrum of FLARE is routinely measured by nonlinear mixing of the THz macropulses and near-infrared laser radiation inside a nonlinear ZnTe crystal.¹¹ The 2%–5% of power split off from the THz beam is converted into sidebands of the main line. These sidebands are resolved with a 1.5 m focal length grating spectrometer with the option to extend to double pass configuration. The THz side peaks are very close to the main near-IR laser peak and it is important to effectively suppress the main line by a combination of optical polarization components. In the single pass setup, the detection limit is about 1 mW of average THz power for the applied ~ 400 mW power of 780 nm radiation supplied by a continuous-wave fiber laser. This set-up can conveniently be used during experiments and allows a fast detection of each macropulse spectrum with an absolute accuracy of ~ 1 GHz. Employing the double pass spectrometer, higher THz power is needed and a 1.5 GHz spectral resolution is obtained, which is just enough to measure the fine structure of the FLARE radiation.

Combining the THz FEL with high-field magnets at the high field magnet laboratory (HFML),¹² high-field and multi-

frequency ESR experiments can be performed. The THz beam propagates from the exit slit of the FEL resonator to the top of a 33 T Bitter magnet through a 90 m-long evacuated, quasi-optical transport line with 41 gold-coated mirrors with an aperture of 250 mm. The total loss from the diagnostic station (which is at the half-way of the beam line) to the output waist above the magnet is about 40%. The entrance of the ESR probe is in the last waist of the Gaussian beam,¹³ having the same size and position for a given slit width over the entire frequency range of FLARE. The ESR probe has a resonator-free transmission configuration, allowing for broadband ESR experiments. Inside the probe a single oversized cylindrical waveguide, with an inner diameter of 13 mm and a collimating end-cone, guides the light to the sample with a typical loss of 50%. After the sample a second inverted cone couples the light into a similar piece of the waveguide. The whole assembly is placed inside a tube filled with He exchange gas (to cool the sample) and sealed at the bottom by a polypropylene window. This tube is placed in a bath cryostat with cold z-cut quartz and warm polypropylene windows at the tail. After the cryostat, a brass tube guides the radiation over 3 m to a He-cooled InSb hot electron bolometer. The ESR spectrometer is completed with an amplifier, a temperature controller, and a fast data-acquisition system. A reference signal to account for the FLARE intensity fluctuations is measured with a pyroelectric detector and is used for normalization of the transmitted intensity (Fig. 2). A digital boxcar averager was used to measure both signals from the detectors.

We used this set-up to perform ESR measurements detecting the THz absorption of a sample as a function of the applied magnetic field. Sweeping the magnetic field, a peak-shaped signal appears in the transmitted intensity with its center at the resonance field determined for paramagnetic materials by $h\nu = g\mu_B B$, where ν and B are the radiation frequency and magnetic field, respectively, and h and μ_B are the Planck constant and Bohr magneton. Therefore, the field dependence of the transmitted intensity is directly proportional to the frequency spectrum of FLARE with the resolution defined by the linewidth inherent to the sample and the magnetic field homogeneity.

Solid polycrystalline DPPH (2,2-diphenyl-1-picrylhydrazyl)¹⁴ is chemically one of the most stable free radicals. This radical was extensively characterized at low fields^{15,16} and the g -factor is 2.0036 ± 0.0002 . The small anisotropy

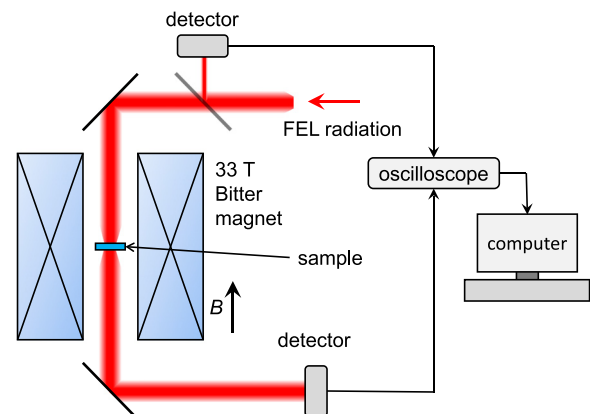


FIG. 2. Schematic experimental set-up.

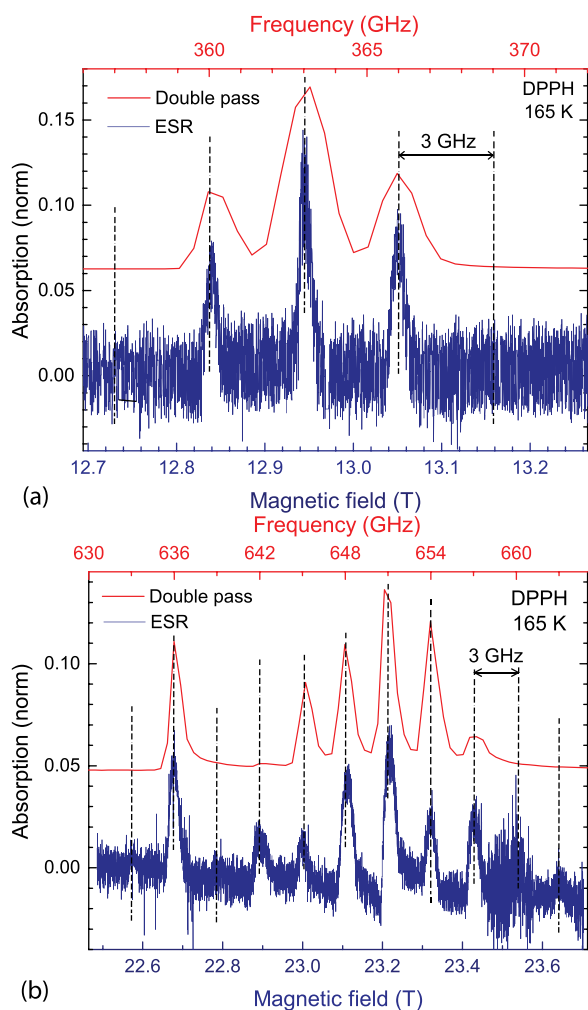


FIG. 3. FLARE spectra measured for two different frequencies using a grating spectrometer in a double pass configuration (red lines, top axes) and the ESR spectrum of DPPH in high magnetic field (blue lines, bottom axes). Dashed lines indicate harmonics of the RF frequency (3 GHz) of the accelerator.

$(g_{\parallel} - g_{\perp})/g \approx 4 \cdot 10^{-4}$ makes DPPH widely used for evaluating the sensitivity of ESR spectrometers and as a marker for high-field ESR spectroscopy.^{17,18} The room temperature powder spectrum consists of a narrow line with a linewidth

increasing from 0.2 mT at 109 GHz to 0.85 mT at 465 GHz. This broadening is caused by a combination of the g -factor anisotropy and the frequency dependent relaxation time T_2 .¹⁸

The measured ESR spectra (blue line) are shown in Fig. 3 together with the spectra obtained via optical up-conversion (red line) for two different frequencies. A fine structure is well resolved with both methods, but the ESR spectra show a much better resolution for the low frequency case (upper panel). Focusing on the ESR spectra, we see that the peaks are well separated with a spacing of 3 GHz. Since the ESR linewidth of DPPH is smaller than 0.5 GHz, the observation of this 3 GHz fine structure implies that the 150 independent micropulses exhibit a considerable coherence; otherwise, the spectrum would consist of a random distribution of 20 MHz-spaced resonator modes underneath the envelope. A similar fine structure, but with a poorer resolution, can also be seen in the grating spectrometer in the double pass configuration. This set-up has a better signal-to-noise ratio but a lower resolution than the ESR approach. The inherent peak broadening is probably substantially smaller than the value found experimentally $\Delta\nu/\nu \approx 0.1\%$ (Fig. 3), which is solely limited by the intrinsic resolution of the experimental setup.¹⁹ Therefore, the FLARE linewidth is at least an order of magnitude smaller than the natural width of a FEL, $\Delta\nu/\nu \approx 1/2N$, which for $N=40$, the number of undulator periods of FLARE, gives a width of 1%.

Using the optical up-conversion approach, the same 3 GHz structure was also observed for higher frequencies up to 2.7 THz. The fact that a series of 3 GHz-spaced peaks is generated by FLARE instead of a single spectrally narrow line is not necessarily a severe disadvantage for simple spectra, like DPPH, and in fact allows relatively high resolution ESR at frequencies and fields far beyond the range of any standard spectrometer. Of course, when spectra are more complicated and consist of several closely spaced lines, the FLARE spectrum with several 3 GHz spaced modes adds complexity. In the future, we envisage introducing external filtering,²⁰ to obtain a truly single frequency operation. The number and amplitude of the neighboring peaks reflect the mode pattern of the laser pulse, depending very much on the setting of the FEL. Close to the synchronous condition, where the cavity length is an exact multiple of the 3 GHz repetition rate, the optical pulse will be rather short and the spectral envelope therefore broad. Occasionally, the spectrum also shows an extra frequency component indicative of another waveguide mode being excited as well.

In particular cases, one of which is shown in Fig. 4(a), it is possible to obtain a quasi-single THz line by optimization of electron beam properties and cavity length detuning. The narrow line shape with only one dominant peak means that the micropulses overlap to form a continuous THz pulse with a duration equal to that of the macropulse (Fig. 4(b)). The small spectral sub peaks are caused by some residual amplitude modulation going from one micropulse to the next. The phase noise of the carrier is determined by the coherence of the 150 micropulses circulating in the resonator. As the linewidth of the peak is at most 0.5 GHz (or $\approx 0.1\%$ of 495 GHz), the corresponding coherence length of 6 (or 3 GHz/0.5 GHz) micropulses can be considered a lower limit.

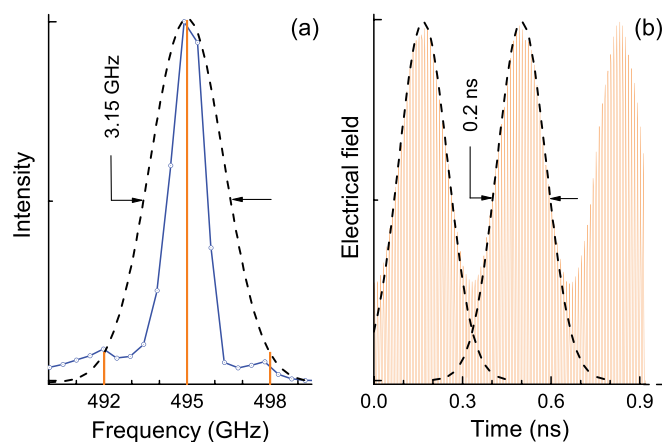


FIG. 4. (a) The experimental FLARE spectrum exhibiting one dominating mode (blue line). The orange lines correspond to the spectral positions resulting from (b) a series of coherent THz micropulses. The dashed black line shows a Gaussian lineshape of single micropulse in (a) frequency and (b) time domains.

The observed coherence between the independent micropulses is not completely surprising, even though FELs are usually said to start up from noise. Coherence has previously been observed²¹ especially for FELs employing a combination of short electron pulses and long wavelengths.^{22–24} In general, the spontaneous radiation emitted by the electron bunch consists of an incoherent part scaling linear with N_e and a coherent part, scaling as N_e^2 , with N_e the number of electrons in the bunch. Because N_e is about 10^9 , even a small structure in the density profile at the scale of the wavelength can easily dominate the spontaneous radiation and provide a seed for the phase. This way the very high stability of the bunch repetition rate, typical for RF accelerators, and hence the small jitter in the arrival times in the undulator, can be imprinted on the phase of the optical pulses. As the structure on the e-bunch depends on very subtle changes in the settings of the accelerator and the resulting amount of coherent spontaneous radiation can vary by orders of magnitude, it is difficult to predict the degree of coherence of the optical pulses at a certain wavelength. Moreover, the coherent spontaneous emission will depend on the exact cavity length, as this determines whether the circulating micropulse will remain in phase with the e-bunch structure until the FEL gain takes over.

The observed coherence of the micropulses is highly relevant for future applications in high magnetic fields, as for instance pulsed ESR and spin-echo experiments. In a frame rotating at the Zeeman frequency in a given DC magnetic field, the THz radiation is seen as a constant transverse magnetic field that makes the electron spins precess. The amplitude of this transverse field is given by the radiation intensity and determines the frequency of the spin precession, which together with the micropulse duration determines the amount of spin rotation. If all micropulses are coherent, the first pulse rotates the spins with a certain amount, and after a while the second pulse rotates them a bit further. Therefore, on a time scale that is short compared to the relaxation/dephasing time of a sample, a coherent set of intense micropulses is equivalent to a (lower) continuous field. Given the FLARE intensity, a rotation angle of π should be achievable. Therefore, our observations indicate that FLARE can be used for ESR spin-echo experiments at the high magnetic fields available at HFML.

In summary, we have determined the spectral characteristics of the spectrometer consisting of FLARE, which produces a μs -long pulse train of micropulses with a repetition rate of 3 GHz, and one of the 33 T Bitter magnets of HFML using DPPH as benchmark system and compared the results to spectral measurements of FLARE using optical upconversion. The width $\Delta\nu/\nu \leq 0.1\%$ of the observed ESR peaks allows for high-resolution spectroscopic studies. Furthermore, the frequency spacing is confirmed to be exactly the repetition

rate of the electron bunches, meaning that the origin of these well-separated peaks is a considerable, electron beam induced coherence between the micropulses. The latter finding shows the potential of this spectrometer for ESR spin-echo experiments.

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¹See www.fels-of-europe.eu for detailed information about other free-electron laser facilities.

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