

# FIRST OPERATION OF A TWO-COLOR MODE IN A DUAL-OSCILLATOR INFRARED FREE-ELECTRON LASER

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## Abstract

Since 2013 the infrared FEL at the Fritz Haber Institute (FHI FEL) has been providing intense, pulsed mid-infrared (MIR) radiation continuously tunable from  $<3\ \mu\text{m}$  to  $>50\ \mu\text{m}$  for in-house users. In 2023 an additional short-Rayleigh-range far-infrared (FIR) FEL was commissioned lasing from  $<5\ \mu\text{m}$  to  $>170\ \mu\text{m}$ . In addition, a 500 MHz kicker cavity has been installed downstream of the electron accelerator. It deflects the electron bunches alternately left and right by an angle of  $\pm 2^\circ$  thereby splitting the high-repetition-rate (1 GHz) electron bunch train into two bunch trains of half the repetition rate each; one is steered to the MIR FEL and the other one to the FIR FEL. The wavelengths in both FEL's can be tuned independently over wide ranges of up to a factor of four by undulator gap variation. Furthermore, two additional small dipole magnets upfront and behind the kicker cavity permit conventional single-color operation of either the MIR or the FIR FEL when the 500 MHz kicker field is off. Regular user operation in two-color mode is scheduled to start in 2024.

## THE FHI FEL FACILITY

The FHI started operation of its IR FEL facility in 2013 [1]. The system is based on a thermionic DC electron gun and a normal-conducting accelerator including two standing-wave S-band linac stages delivering 200 pC bunches at energies from 15 to 50 MeV. The temporal structure is characterized by a bunch repetition rate of 1 GHz (with optional reduced rates) with a burst (macro-bunch) length of typically 12  $\mu\text{s}$  at 10 Hz macro-bunch repetition rate. The electron beam is delivered by two  $90^\circ$  isochronous achromats to the MIR FEL as can be seen in Fig. 1.

The MIR pulses are generated by the electrons passing through a planar hybrid-magnet undulator (2 m long, 50 periods) located within a 5.4 m long IR cavity. Measurements of the FEL macro-pulse energy as a function of wavelength are shown below in figure in section "FIR and two-color performance". Each trace in the figure corre-

sponds to an individual accelerator tune-up at the indicated electron energy. The typical energy of  $\sim 10\ \mu\text{J}$  per micro pulse corresponds to  $\sim 100\ \text{mJ}$  per macro pulse. The central wavelength of the MIR FEL can be set to any value between 2.9 and 60 micron, thereby covering the full mid infrared regime [1]. The spectral width of the radiation, and hence the micro-pulse length, can be varied by fine adjustment of the FEL cavity length. For most spectroscopic applications it is set to less than 0.5% (full width at half maximum) of the central wavelength, corresponding to a Fourier-limited micro-pulse length of a few ps. Ultrashort pulses as short as 0.5 ps of high peak power, as sometimes needed for non-linear spectroscopy or time-resolved measurements, can also be generated at a correspondingly broader spectrum of several percent.

The FHI FEL has been serving as an MIR in-house user facility producing, to date,  $\sim 100$  peer-reviewed journal articles in physical chemistry basic research. An FIR/two-color upgrade to the existing facility was approved in 2019. It comprises a second oscillator FEL as well as installation of a 500 MHz kicker cavity (see Fig. 1). As will be described below, the MIR and FIR FEL's can be operated individually or simultaneously in two-color mode. The latter will open up a wealth of novel user applications such as, for instance, MIR-FIR pump-probe or nonlinear experiments. First lasing of the new FIR FEL was achieved on June 8, 2023. Subsequently, the first separately tunable, two-color lasing from the MIR and FIR beamlines was achieved at 18  $\mu\text{m}$  and 55  $\mu\text{m}$  on December 8, 2023. To date, the FIR FEL has generated radiation from 4.7 to 176  $\mu\text{m}$ , the latter being achieved at 16.75 MeV electron energy.

## THE TWO-COLOR FHI FEL UPGRADE

The design of the two-color FHI FEL upgrade [2, 3] presented here was based on three aims: (i) extend the wavelength range of the facility into the FIR/THz regime out to at least 150  $\mu\text{m}$  (2 THz) by setting up a second oscillator FEL; (ii) make it possible to operate both MIR and FIR FEL's simultaneously, thereby offering users two-color capabilities; and (iii) design the FIR FEL such that the wavelength ranges (undulator-gap tuning ranges) of both FEL's overlap at any given electron energy.

The upgraded FHI FEL layout fulfilling these three

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aims is shown in Fig. 1, where the new FIR FEL beamline is visible on the left. In addition, a 500 MHz side-deflecting-cavity (kicker cavity) was installed downstream of the accelerator. It deflects the electron bunches alternately left and right into the MIR and FIR beamlines, respectively. Using a side-deflecting RF cavity that operates at one-half of the bunch repetition rate to split a high-repetition-rate bunch train in two was first proposed by Schwettman and Smith in 1989 [4]. To the best of our knowledge this scheme was not implemented anywhere in the context of an infrared FEL until now.

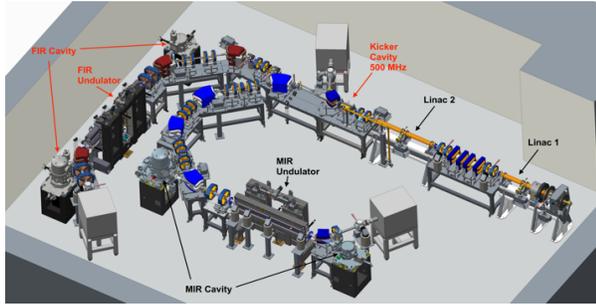


Figure 1: Layout of the FHI FEL. Red labels indicate the new components installed as part of the two-color upgrade and commissioned in 2023, whereas black labels indicate the parts that have been in operation since 2013.

The layout of the kicker cavity and the beamline are shown in Fig. 2. A strong 500 MHz RF field generates a transverse horizontal electric field (peak field of up to 11.5 MV/m) leading to side deflection of 50 MeV electrons by  $\pm 2^\circ$ . This allows every second electron micro-bunch to be sent to the MIR FEL and every other second bunch to the FIR FEL branch after a relatively short drift region behind the cavity, as shown in Fig. 2(a). The kicker-cavity setup includes two small dipole magnets before and behind the cavity. This configuration makes three different modes of operation possible: (i) 100% of the electron beam goes to the MIR FEL when the dipoles are off; (ii) 100% of the beam goes to the FIR FEL when each dipole deflects

by  $+2^\circ$ . In modes (i) and (ii) the RF to the kicker cavity is off. Finally, in mode (iii) the 1 GHz repetition-rate bunch train is split into two 500 MHz repetition rate beams feeding both MIR and FIR FEL, when the side deflecting cavity is energized. In this two-color mode each of the two dipole magnets is set to deflect the beam by  $+1^\circ$ . The diagnostic dipole visible in Fig. 2 (a) is not energized in any of these modes.

Downstream of the separation region an isochronous achromat deflects the beam by a total of  $94^\circ$  delivering the bunches to the FIR FEL which makes an angle of  $90^\circ$  with respect to the accelerator axis. The IR wavelength range of the FIR FEL is defined by its undulator period of 68 mm. This period was chosen such that at any electron energy set in the accelerator, the MIR FEL (at close to minimum undulator gap) and the FIR FEL (at close to maximum undulator gap) generate IR pulses at identical wavelength (3<sup>rd</sup> aim described above). By opening up the MIR undulator gap and/or closing the FIR undulator gap the wavelengths of the FEL's can be independently reduced and increased, respectively, over a wide tuning

range (typically more than a factor of 2.5 in each FEL, see below). Furthermore, for the given range of electron energies accessible with the accelerator from 15 to 50 MeV, the FIR FEL allows lasing from  $< 5 \mu\text{m}$  to  $\sim 175 \mu\text{m}$ , compliant with the users' request (1<sup>st</sup> aim described above).

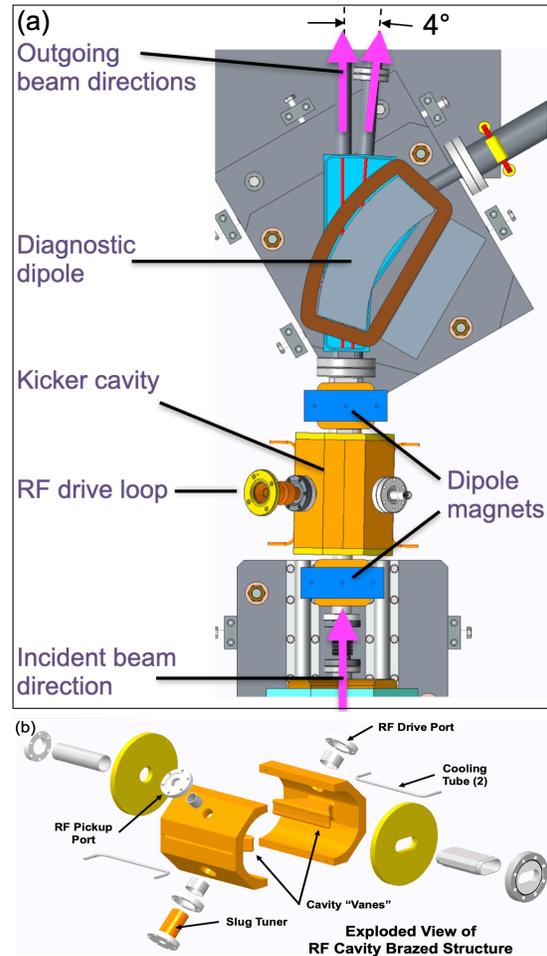


Figure 2: (a) Schematic of the kicker-cavity location, where the bunches coming from the accelerator are alternately separated by  $4^\circ$  into the MIR and FIR beamlines. Two dipole magnets before and after the cavity are indicated. (b) Internal shape of the kicker cavity where a 500 MHz transverse RF electric field builds up between two 158-mm-long vanes separated by 20 mm. The cavity is powered by a solid-state pulsed RF amplifier delivering up to 56 kW of pulse power.

To avoid a waveguided optical cavity, which is known to interfere with lasing at various wavelengths in the FIR regime (spectral gaps) [5-7], we implemented a large-clearance undulator vacuum chamber, thereby allowing for free-space propagation of the optical mode even at the longest wavelengths. The vacuum chamber, shown in Fig. 3, has an inner height at the central section of 23 mm, which is further linearly increasing towards both ends of the FIR undulator in two regions 15 and 56 cm from each end as indicated in Fig. 3(a). This shape mimics the optical mode size variation along the undulator axis with its mode waist located at the undulator center. This design

has allowed us to choose a short Rayleigh range of the optical cavity of just 68 cm corresponding to 1/3 of the undulator length. Finally, the nominal cavity length of 5.4 m is identical to the MIR cavity length, which makes synchronization of MIR and FIR radiation pulses straightforward. Just as for the MIR cavity we use hole-outcoupling in the FIR cavity. Outcoupling-mirrors with hole diameters of 2.5, 4.0, and 6.0 mm were used in the present work.

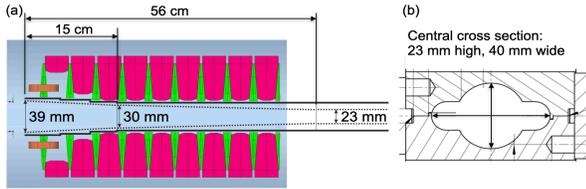


Figure 3: Cross sectional views of the undulator vacuum chamber. The increasing inner height and the recessed poles and magnets at both ends of the undulator can be seen in the longitudinal cross section in (a). The transverse cross section at the undulator center is shown in (b).

As a consequence of the relatively large height of the FIR undulator chamber, the 68-mm-period (U-68, Fig. 4) undulator's minimum gap is 32 mm. To achieve the high magnetic-flux density needed to get the undulator parameter required for the long wavelengths, we implemented an optimized wedged-pole undulator design, schematically visible in Fig. 3(a). It also allowed us to choose a radiation-hard grade of NdFeB for the wedged-shape magnets. The peak on-axis magnetic-flux density  $B_0 = 0.49$  T at minimum gap corresponds to a maximum root-mean-square undulator parameter  $K_{rms} = 2.2$ .



Figure 4: Completed FIR U-68 undulator during magnetic mapping at FHI.

The undulator was fabricated at FHI with remote direction from STI Magnetics. Magnetic field measurements were performed at the FHI. To this end, dedicated equipment, including a 7 m monolithic-granite magnet scanner (see Fig. 4) was set up in the FEL building at the FHI. Magnetic center line vertical field trajectories versus undulator gap, are shown in Fig 5. The spikes are at the entry and exit of the undulator. The main effect is a steadily increasing offset with gap caused by end fields. The completed undulator is shown in Fig. 4.

As described above, the standard mode of operation of the accelerator system is a 1 GHz bunch repetition-rate, which, in the two-color mode, results in a pulse repetition rate of 500 MHz in each FEL. In addition to this mode the

bunch repetition rate of the accelerator can be scaled down from 1 GHz by a factor of 36, 18, 12, 9 or 6, resulting, respectively, in 1, 2, 3, 4, or 6 optical pulses in the optical cavity during single-color operation. For instance, at an electron bunch repetition rate of 55.6 MHz two optical pulses are generated in either the MIR or FIR FEL. This mode is needed for some user experiments which use existing 55.6 MHz tabletop lasers. Operating the electron gun at 111.1 MHz will then deliver 55.6 MHz radiation in both FEL's in two-color lasing mode (two optical pulses in each FEL cavity).

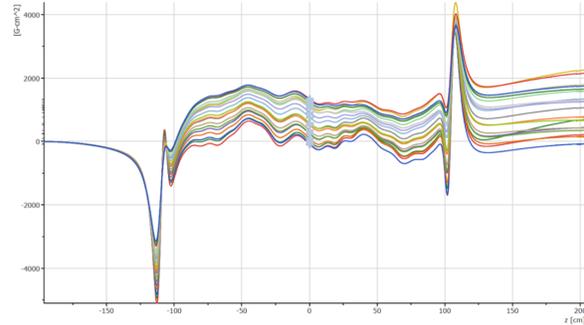


Figure 5: U-68 vertical field trajectories at the magnetic center line versus undulator gap in 2 mm steps from 32 mm (red top) to 68 mm (blue bottom).

## FIR AND TWO-COLOR PERFORMANCE

On June 8, 2023, first light was achieved at 8  $\mu\text{m}$  from the FIR FEL beamline. To characterize the FIR FEL we subsequently measured power curves at different electron energies between 16.7 and 45 MeV. The plots in Fig. 6 show the macro-pulse energy as a function of wavelength. In each curve the wavelength was tuned by changing the undulator gap at the given electron energy.

The five curves of the FIR FEL plotted in Fig. 6 indicate continuous lasing from 4.7  $\mu\text{m}$  to 176  $\mu\text{m}$ . For each curve the tuning range is at least a factor of 3, almost a factor of 5 for some curves. This large continuous tunability of the FIR FEL is particularly advantageous for many user experiments. In addition, the maximum pulse energy of the curves ranging from 50 mJ at low electron energy to more than 200 mJ at 45 MeV appears to be more than the corresponding values observed with the MIR FEL. This might reflect the short-Rayleigh-range design of the FIR FEL leading to more energy in the optical pulses despite the fact that the smaller number of undulator periods results in a smaller gain. This observation is in agreement with simulations.

Finally, it is noteworthy to mention that dips in the power curves are visible at wavelengths of 23, 33, 51, 93 and 133  $\mu\text{m}$ . In particular, the ones appearing at 33 and 51  $\mu\text{m}$  are significant. The origin of the dips is not yet understood. More experiments and/or simulations will be needed to understand the cause of the dips and, if possible, to get rid of them.

On December 8, 2023, after only  $\frac{1}{2}$  a day of commissioning, first light was obtained simultaneously at 18 and 55  $\mu\text{m}$ , respectively, thereby achieving the project goal of separately tunable, two-color lasing. Fig. 7 shows the

lasing traces from that milestone.

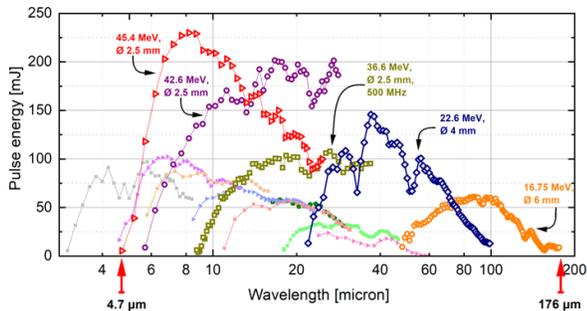


Figure 6: Spectral range measured in the FHI FEL’s to date ranges from 3 to 176  $\mu\text{m}$  as shown by these gap scans. The lower faded-color curves, replotted from [2], show results of the MIR FEL at various electron energies for comparison. The upper bright-color traces indicate the pulse energy measurements of the new FIR FEL at the indicated electron energies. Except for the data at 36.6 MeV, the measurements were done at 1 GHz repetition rate. Different outcoupling-hole diameters of 2.5, 4, and 6 mm were used as indicated.



Figure 7: Oscilloscope traces showing the first simultaneous two-color lasing from the MIR and FIR beamlines, each one operating at 500 MHz micro-pulse repetition rate. The yellow and orange traces show the 10- $\mu\text{s}$ -long macro-bunches in the dumps (micro-bunches are not resolved). The green and violet traces show the optical signals of the MIR and FIR FEL, respectively, recorded with pyro detectors. Here, too, the micro-pulses are not resolved. The opposite sign of the radiation traces is a technical artefact.

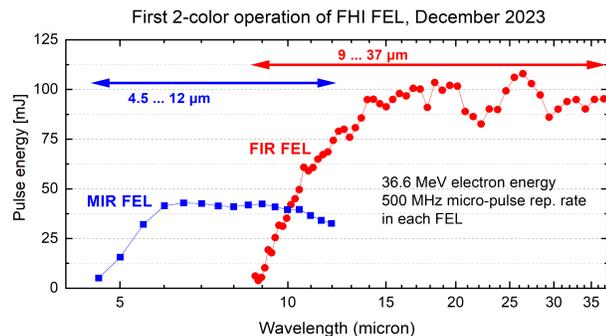


Figure 8: First two-color, separately tunable, simultaneous lasing gap scans observed at an electron energy of 36.6 MeV. The plot indicates the overlap of the wave-

length ranges at 9 to 12  $\mu\text{m}$  (fulfilling user request no. 3, see text) and the wide, continuous tuning capabilities of both FEL’s.

Subsequently, the first separately tunable, two-color lasing gap scans were obtained at 36.6 MeV electron beam energy, as shown in Fig. 8. A factor of four in the wavelength tuning range can be seen for the FIR radiation spectrum (9 to 37  $\mu\text{m}$ ) and nearly a factor of three for the MIR spectrum (4.5 to 12  $\mu\text{m}$ ). This corresponds to a tuning range of almost a factor of 10 for the ratio of FIR to MIR wavelength; it can be continuously varied from 0.75 to 7.0. The FIR pulse energy is a substantial 100 mJ, which would deliver 200 mJ in 1 GHz FIR-only lasing mode.

## CONCLUSIONS

To date, the newly installed FHI FIR FEL has delivered radiation from 4.7 to 176  $\mu\text{m}$  as, shown in Fig. 6, using electron beam energies ranging from 17 to 45 MeV. We expect to further extend the wavelength range by tuning the accelerator to its full nominal energy range from 15 to 50 MeV. The pulse energies of the FIR are larger than those we get with the MIR FEL. We attribute this behavior, which was predicted by simulations, to the short-Rayleigh-range design of the FIR FEL.

In addition, we have demonstrated operation of 2-color lasing by running both MIR and FIR FEL’s simultaneously. In the two-color mode it is possible to tune the FIR-to-MIR wavelength ratio from 0.75 to 7.0, i.e. almost by a factor of 10. The 2-color mode is made possible by a 500 MHz side-deflecting cavity deflecting every second bunch to the MIR and every other second to the FIR FEL. This way each of the two FEL’s generates radiation pulses at a rate of 500 MHz repetition. We expect the MIR and FIR pulses to be highly synchronized, because they are generated by electron bunches picked alternately from a single bunch train of the accelerator. We intend to run cross-correlation measurements to investigate the remaining MIR-FIR pulse-to-pulse timing jitter.

The first two-color operation of an IR FEL was demonstrated in pioneering work by Ortega and coworkers at the CLIO facility in Paris Orsay in the 1990’s [8-10]. In those experiments two identical undulator segments within the CLIO cavity where gap tuned independently, referred to as step-tapered undulator. Based on the strong gain of CLIO it was possible to observe two-color operation with a single cavity IR FEL. However, lasing in the first undulator segment interferes with lasing in the second one leading to power fluctuations when gap tuning. In addition, the wavelength ratio was limited to a factor of  $\sim 1.5$ . The two-color mode of a dual-cavity IR FEL presented in this work overcomes those limitations and makes it possible to provide users with the wide and continuous tunability of both wavelengths that is needed for many applications. We expect to see first results of two-color experiments from FHI FEL user groups later in 2024, with potential research over the next few years performing FEL pulse structure characterization by balanced optical cross-correlation, pump-probe experiments in molecules and doped semiconductors, and four-wave-mixing microscopy, among other topics.

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