THE NEW IR FEL FACILITY AT THE FRITZ-HABER-INSTITUT IN BERLIN

Wieland Schöllkopf, Sandy Gewinner, Wolfgang Erlebach, Heinz Junkes, Andreas Liedke, Gerard Meijer, Alexander Paarmann, Gert von Helden, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany
Hans Bluem, David Dowell*, Ralph Lange*, John Rathke, Alan M.M. Todd, Lloyd M. Young*, Advanced Energy Systems, Inc., 27 Industrial Blvd., Medford, NY 11763, USA
Ulf Lehnert, Peter Michel, Wolfgang Seidel, Rudi Wünsch, Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany
Stephen C. Gottschalk, STI Optronics, Inc., 2755 Northup Way, Bellevue, WA 98004, USA

Abstract

A mid-infrared oscillator FEL has been commissioned at the Fritz-Haber-Institut. The accelerator consists of a thermionic gridded gun, a subharmonic buncher and two S-band standing-wave copper structures. It provides a final electron energy adjustable from 15 to 50 MeV, low longitudinal (<50 keV-ps) and transverse emittance (<20 π mm-mrad), at more than 200 pC bunch charge with a micro-pulse repetition rate of 1 GHz and a macro-pulse length of up to 15 µs. Pulsed radiation with up to 50 mJ macro-pulse energy at about 0.5% FWHM bandwidth is routinely produced in the wavelength range from 4 to 48 µm. Regular user operation started in Nov. 2013 with 6 user stations. The user experiments include nonlinear spectroscopy of solids, spectroscopy of bio-molecules (peptides and small proteins), which are conformer selected in the gas-phase or embedded in superfluid helium nano-droplets at 0.4 K, as well as vibrational spectroscopy of mass-selected metal-oxide clusters and protonated water clusters in the gas phase.

INTRODUCTION

In 2008 the Fritz-Haber-Institut (FHI) embarked on setting up an infrared FEL facility. The aim was to set up an FEL that is capable of providing intense, pulsed laser radiation, continuously tunable from a few micron in the near to mid-infrared (MIR) all the way to several hundred micron in the far-infrared (FIR), or Tera-Hertz (THz) regime. Installation of the FEL started in mid 2011 in a new, dedicated FEL building on the FHI campus in Berlin. We observed first lasing in 2012 [1–3] and started user operation in November 2013.

IR radiation in the spectral region from 3 to 100 µm is often referred to as the molecular fingerprint region, because it is the region in which the fundamental vibrational modes of molecules, clusters or solid materials are located. The vibrational IR spectrum is intimately connected to the molecular structure and dynamics, which is why IR spectroscopy is one of the basic methods for molecular structure characterization. The highest energy vibrations, involving stretching motions of light atoms, are found around a wavelength of 3 µm. Lower energy vibrations are found throughout the IR region, down to the FIR region beyond 100 µm. The vibrational frequencies in the FIR result from heavy atoms and/or weak bonds, or from soft modes that involve large amplitude motions and global geometry changes. Thus, FIR spectroscopy also allows to study the folding dynamics of (bio)molecules.

FIR radiation can also be used to directly probe surface-adsorbate vibrations. This allows to study the structure and dynamics of adsorbates on surfaces, to measure the properties of deposited or gas-phase cluster materials, or to investigate real-world catalysts in action. In addition, the intense FEL radiation can readily induce multiple photon excitation processes and is well suited for double-resonance experiments with other, table-top laser systems.

DESIGN OF THE FHI FEL

To cover the full wavelength range of interest from about 4 to 500 µm the basic design of the FHI FEL includes two different undulator lines as outlined in Fig. 1 [1–5]. The MIR branch works for wavelengths up to about 50 µm and the FIR branch covers the wavelength range from about 40 to 500 µm. A normal-conducting linear accelerator provides electrons of up to 50 MeV energy with a beam transport system feeding either of the FEL branches or the diagnostics beamline (Fig. 1).

Electron Accelerator and Beamline

The electron accelerator and beamline system, designed, built, and installed by Advanced Energy Systems, Inc. (AES), has been described before [4–7]. In brief, the accelerator system is comprised of a gridded thermionic gun, a subharmonic buncher cavity and two standing-wave, $\pi/2$ copper linacs. The first of the two S-band (2.99 GHz) linacs accelerates the electron bunches to a nominal energy of 20 MeV, while the second one accelerates or decelerates the electrons to deliver any final energy between 15 and 50 MeV. A chicane between the linacs allows for adjustment of the electron bunch length as required.

Key performance parameters of the accelerator are low longitudinal (<50 keV-psec) and transverse emittance (<20 π mm-mrad) at more than 200 pC bunch charge with a micro-bunch repetition rate of 1 GHz. The maximum

* Consultants to Advanced Energy Systems, Inc.

ISBN 978-3-95450-133-5

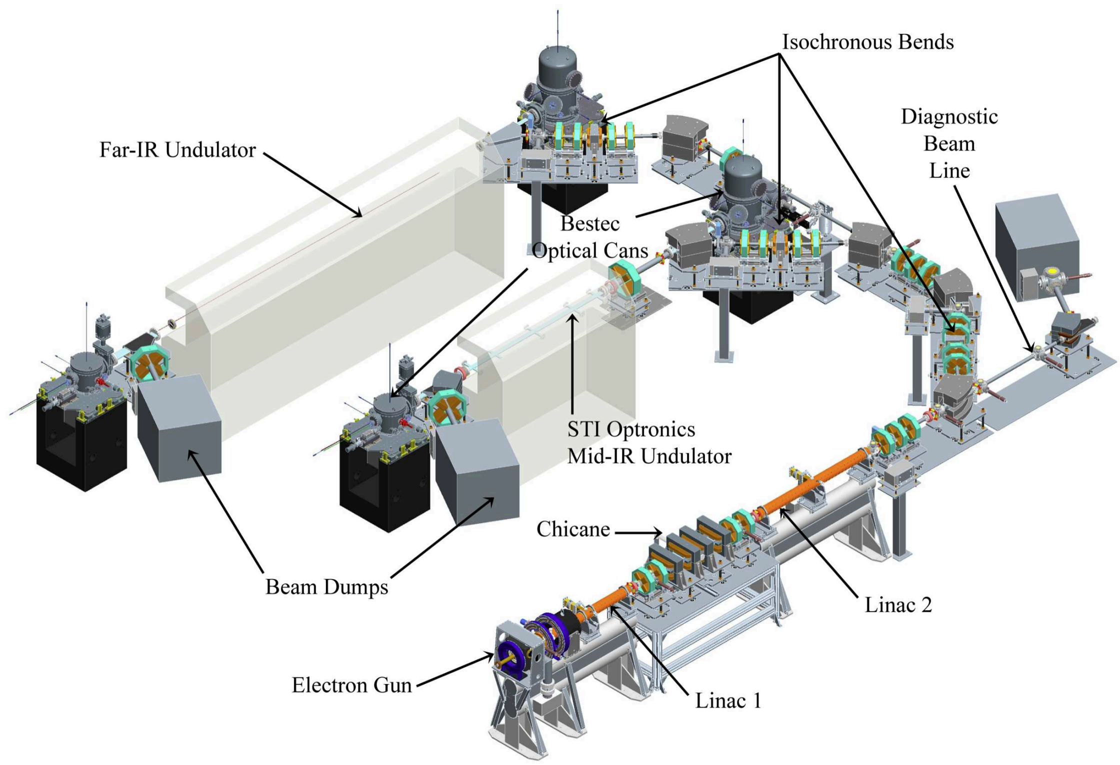

Figure 1: Overview of the FHI FEL installation showing the electron accelerator system and the MIR FEL (operational) and the FIR FEL (to be installed in the future).

Table 1: Summary of Electron Beam Parameters of the S-band Linear Accelerator

Parameter	Unit	Spec.	Target
Electron energy	MeV	20 - 50	15 - 50
Energy spread	keV	50	< 50
Energy drift per hour	%	0.1	< 0.1
Bunch charge	pC	200	> 200
Micro-bunch length	ps	1 - 5	1 - 10
Micro-bunch rep. rate	GHz	1	1
Micro-bunch jitter	ps	0.5	0.1
Macro-bunch length	µs	1 - 8	1 - 15
Macro-bunch rep. rate	Hz	10	20
Normalized rms transverse emittance	π µm rad	20	20

macro-bunch repetition rate is 20 Hz. Table 1 summarizes the top-level electron beam performance.

The beam is delivered from the accelerator to either the MIR or the FIR FEL by 90°-isochronous achromats that minimize the effect of the micro-bunch and macro-bunch jitter to ensure radiation wavelength stability and timing consistency for pump-probe experiments. A quadrupole triplet downstream of the second linac leads to the first dipole where the diagnostic beamline goes straight through. There is another quadrupole triplet in the back leg before the second 90° isochronous achromat delivers the beam to the MIR FEL. Downstream of the undulator the electrons are bent to the beam dump by a 60°-dipole.

MIR Oscillator FEL

Both the MIR and FIR FEL consist of an undulator placed within an IR cavity as summarized in Table 2. The MIR FEL includes a planar, radiation-resistant, high-field-strength, wedged-pole hybrid undulator, manufactured by STI Optronics, Inc., which has been described in detail elsewhere [8]. It is 2 m long containing 50 periods with a period length of 40 mm. At a minimum undulator gap of nominally 16.5 mm, a maximum root-mean-square undulator parameter K_{rms} of more than 1.6 is reached [8]. This, in combination with the minimum electron energy of 15 MeV corresponds to a theoretical maximum wavelength of more than 50 µm for the MIR system.

The 5.4 m long FEL cavity is formed by a cavity end mirror and the outcoupling mirror. These are gold-plated copper mirrors of spherical concave shapes with radius of curvature of 2.65 m and 3.51 m, respectively. As a result, the Rayleigh length of the cavity mode is 2 m, thereby matching the undulator length, and the location of the mode's waist is shifted by 50 cm away from the cavity center to coincide with the undulator center [3]. The cavity-end mirror is mounted on a precision translation stage to enable cavity length adjustment with a nominal precision of 1 µm. The

ISBN 978-3-95450-133-5

Table 2: Summary of Essential Parameters of the MIR (commissioned) and FIR (prospected) FEL Setups

Undulator	MIR	FIR
Type	Planar hybrid	Planar hybrid or PPM
Material	NdFeB	NdFeB or SmCo
Period (mm)	40	110
No. of periods	50	40
Length (m)	2.0	4.4
K_{rms}	0.5 - 1.6	1.0 - 3.0
IR-cavity	**MIR**	**FIR**
Length (m)	5.4	7.2

outcoupling mirror has a hole in its center. Each one of a set of mirrors with hole diameters of 0.75, 1.0, 1.5, 2.5, and 3.5 mm can be precisely positioned at the cavity end by an in-vacuum mirror changer. This allows to choose the out coupling-hole diameter best suited for the wavelength range of interest.

The FIR FEL has not yet been installed, but its design has been outlined. The right column of Table 2 gives the essential design parameters for the FIR undulator and cavity. The electron branch-off beamline has been included in the current system, thereby allowing for implementation of the FIR system at a later point of time.

CHARACTERIZATION OF THE MIR FEL RADIATION

To characterize the IR radiation different commercial IR detectors including a liquid-nitrogen cooled MCT (HgCdTe) detector (Judson J15D24) and a large area (46 mm diameter) pyroelectric detector (Ophir PE50BB) are used for IR power and pulse energy measurements. In addition, a vacuum Czerny-Turner grating spectrometer (Acton VM-504) equipped with a 0.5”-wide pyroelectric linear array detector (DIAS 128LT) allows online monitoring of the FEL spectrum (see Fig. 2). Further commercial diagnostics equipment includes a 5 stage IR beam attenuator (LASNIX Model 102). It is planned to set up additional equipment for characterizing and manipulating the IR beam in the future including a photo-acoustic cell and a polarization rotator.

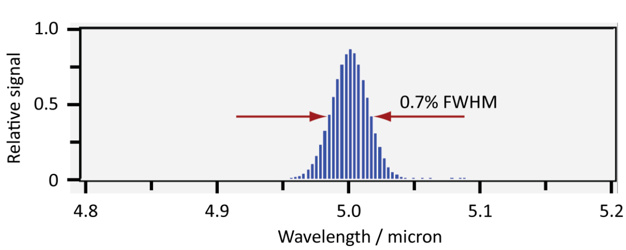

Figure 2: Spectrum of the FEL laser line centered at 5 µm measured with the grating spectrometer.

Figure 2 shows a spectrum of the FEL line centered at a wavelength of 5 µm obtained for an electron energy of 38 MeV. The spectral line FWHM (full width at half maximum) in this measurement is about 33 nm corresponding to a relative line width of less than 0.7%. As discussed in detail in the next section, the line width can be varied by adjusting the FEL cavity length. Most user experiments at the FHI FEL facility benefit from narrow line width, even at the expense of pulse energy. Therefore, the FEL is usually tuned to exhibit a narrow line width of typically 0.35% to 0.6% FWHM, when lasing at 10 µm or less, increasing to 1.0% to 1.5% FWHM at the longest wavelengths close to 50 µm. This corresponds to absolute FWHM bandwidths of 3.6 to 6 cm^{-1} and 2 to 3 cm^{-1} at 10 and 50 µm central wavelength, respectively.

As of summer 2014 the accelerator is operated at either one of three electron energies of 38 MeV (regime I), 26.5 MeV (regime II), and 18.5 MeV (regime III). With these three energies lasing over the entire MIR wavelength range from 3.5 to 48 µm is achieved. Figure 3 shows measurements of the macro-pulse energy for the three regimes, when the FEL is operated at narrow line width conditions. Largest macro-pulse energies of about 50 mJ are found at highest electron energy (regime I), decreasing to less than 10 mJ in regime III at wavelengths larger than 25 µm. We expect lasing at even shorter or longer wavelengths once the accelerator is operated at its maximum and minimum electron energies of 50 and 15 MeV (see Table 1), respectively. So far, a maximum IR pulse energy of 130 mJ was observed with a broad spectrum centered around 9 µm.

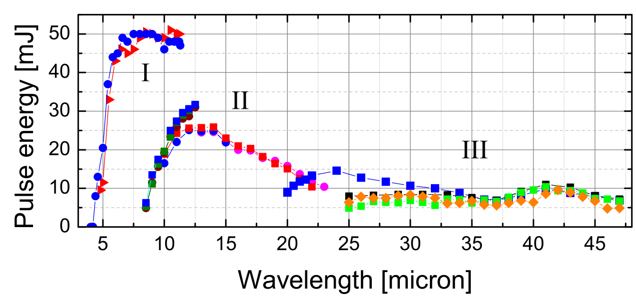

Figure 3: Macro-pulse energies measured at narrow bandwidth conditions for electron energies of 38 MeV (regime I), 26.5 MeV (regime II) and 18.5 MeV (regime III).

Micro-pulse Characterization by SHG Autocorrelation Measurements

To determine the micro-pulse time structure we implemented an auto-correlation measurement setup shown schematically in Fig. 4. The FEL beam is split in equal parts by a Ge-coated ZnSe beam splitter. Both partial beams are focused onto a 1 mm thick CdTe single crystal. The optical path-length difference of the beams is accurately controlled by a commercial precision translation. Non-linear interaction in the CdTe sample leads to second harmonic generation (SHG). Due to the angle between the incident beams, the generated SHG beam is separated from the reflected FEL beams, allowing to block the latter by an aperture. As a re-

ISBN 978-3-95450-133-5

sult, the relatively weak SHG signal can be detected free of the second harmonic background signal inherently present in the FEL beams.

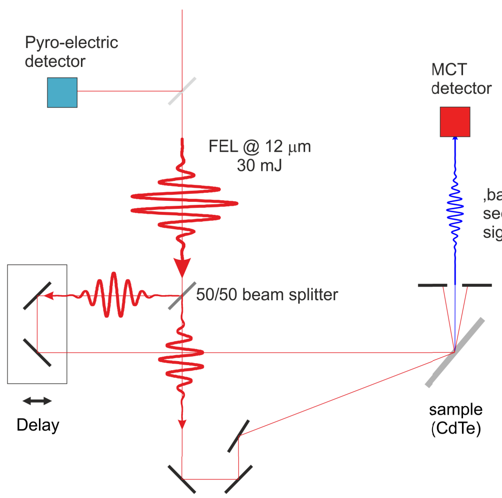

Figure 4: Setup to measure auto-correlation functions by second harmonic generation in a CdTe sample.

Auto-correlation data measured at a wavelength of 12 μm is shown in Fig. 5 for three different cavity length detunings of $\Delta L = -3$, -16, and -50 μm, where $\Delta L = L - L_0$ with L and $L_0 = 5.4$ m standing for the actual and nominal cavity length, respectively. The 2-D plots on the left of Fig. 5 show the SHG signal as a function of the delay time and real time. The former describes the micro-pulse arrival-time difference due to the optical path-length difference, while the latter refers to the time referenced to the beginning of the electron macro bunch. Thus, the plots show the evolution of the micro-pulse auto-correlation over the macro-pulse time window.

For the smallest cavity detuning of -3 μm (Fig. 5A) micro-pulses occur rather late in the macro-pulse indicating a relatively small FEL gain. These pulses show very sharp auto-correlation functions with FWHM values down to 0.6 ps. The auto-correlation time structure changes significantly when the cavity detuning is increased to -16 μm (Fig. 5B). Pulses now appear earlier within the macro-pulse indicating a larger FEL gain. In addition, the auto-correlation function exhibits multiple peaks separated by about 1 ps. Finally, at even larger cavity detuning of -50 μm (Fig. 5C) the multiple-peak structure has evolved into a single broad peak with a relatively large FWHM increasing from 3 ps to about 8 ps over the course of the macro-pulse.

A multiple-pulse time structure of the micro-pulses in an oscillator FEL was observed at FELIX before and explained in terms of limit-cycle oscillations [9, 10]. Our observations agree well with those findings. The large variation of the micro-pulse time structure allows us to tune the temporal and spectral properties of the FEL radiation by changing the cavity length. In fact, in most FHI FEL user experiments, which will be described in the next section, a rather narrow spectral bandwidth is advantageous. Time resolved experi-

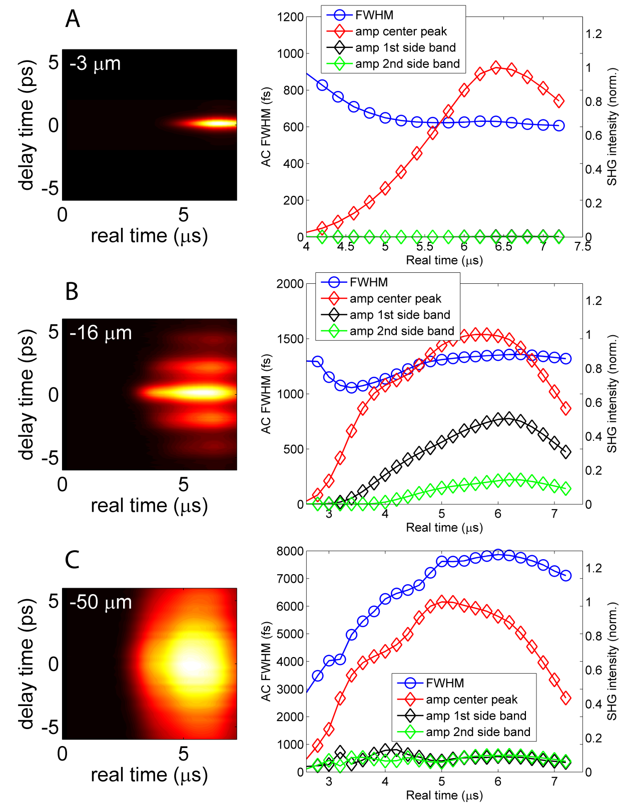

Figure 5: Auto-correlation measurements of FEL pulses at 12 μm wavelength for 3 different cavity length detunings of $\Delta L = -3$ μm (A), -16 μm (B), and -50 μm (C), measured with the setup shown in Fig. 4. The plots on the left show the observed SHG signal as a function of delay time and real time within the electron macro-bunch time window with zero set by the beginning of the macro-bunch. The data analysis results shown on the right indicate the evolution of the micro-pulse structure over the macro-pulse.

ments, however, will benefit from short FEL pulses that can be achieved at small cavity length detuning. The FWHM of 0.6 ps observed in the auto-correlation function shown in Fig. 5A corresponds, assuming a Gaussian pulse shape, to a deconvoluted micro-pulse length of less than 0.43 ps FWHM.

FACILITY AND USER STATIONS

The IR pulses, extracted from the MIR FEL, enter the IR beamline through a CVD diamond window under Brewster angle. The window separates the ultra-high vacuum of less than 1 x 10^{-8} mbar in the undulator and cavity-mirror chambers from the high vacuum (10^{-5} mbar) in the IR beamline. The IR beamline consists of 10 cm inner-diameter stainless steel pipes interconnected at right angle by vacuum chambers housing 90°-deflection broadband IR mirrors. The mirrors are either flat or toroidal (focusing) and are made out of copper with a gold coating. The nominal mirror reflectivity is 99.2% over the entire IR range. A total of 6 such mirrors steer the IR beam from the FEL cavity in the vault to the

ISBN 978-3-95450-133-5

IR diagnostic station, located in the neighboring building, over a total length of 18 m (see Fig. 6). Another IR beamline system (user beamlines) brings the FEL beam from the diagnostic station to either one of six experimental stations located in the basement and the ground floor as indicated in Fig. 6.

As of summer 2014 five experiments from the area of gas-phase spectroscopy of clusters and bio-molecules as well as one experiment from the field of solid-state physics have been using the MIR FEL radiation.

Station I: Chemistry of transition metal clusters The group of André Fielicke (FHI & TU Berlin) has been using the FHI FEL radiation to investigate transition-metal clusters in the gas phase. For instance, vibrational spectra of CO attached to small cationic ruthenium and cobalt-manganese clusters have been measured. These studies allow to probe the effects of alloying on the d-band center that is controlling the strength of the p-backbonding to the CO. Further systems studied so far by FHI FEL radiation include: (i) neutral iron oxide clusters studied by IR-UV two color ionization; (ii) neutral gold clusters in search for O_2 activation in the presence of deuterated water; (iii) positively charged $Rh_n^+CH_4$ clusters; and (iv) neutral Si_nB clusters.

Station II: Vibrational spectroscopy of gas phase clusters: catalysis, astrochemistry and new materials The group of Knut Asmis (FHI & U Leipzig) has been using the FHI FEL radiation to study negatively charged aluminum oxide and iron oxide clusters in the gas phase. The small negatively charged metal oxide clusters are prepared in an ion trap, cooled to temperatures on the order of 20 K, and tagged with a few D_2 molecules per cluster. Mass-selective vibrational spectra are measured by way of IR photodissociation of the messenger-tagged complexes in order to probe (close to) the linear absorption regime.

Station III: Vibrational spectroscopy of gas phase clusters: atmospheric chemistry and ion solvation Protonated gas-phase water clusters $H_3O^+(H_2O)_n$ and $D_3O^+(D_2O)_n$ as well as $Cs^+(H_2O)_n$ and $Cs^+(D_2O)_n$ have been investigated by the Asmis group to illucidate their structural properties. Further systems studied at this beamline using the FHI FEL radiation include anionic $ClMgCO_2^-$ molecules as well as $NO_3^-(HNO_3)_n(H_2O)_m$ and $NO_3^-(HNO_3)_n$ clusters tagged with D_2.

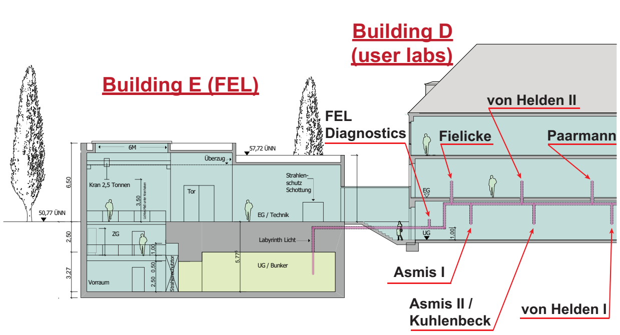

Figure 6: Cross sectional view of the new FEL building (left) and the spectroscopy-lab building (right) indicating the IR transport beamline system connecting the FEL with currently six user stations in the basement and ground floor, which are labelled by the principle investigator's name.

Station IV: Spectroscopy of conformer selected proteins The group of Gert von Helden (FHI) has been using the FHI FEL to study the structure of bio-molecules in the gas phase. IR spectra of the protein Ubiquitin (76 amino acids) in different charge states $n+$ and crown-ether complexes thereof have been measured. Prior to the interaction with the FEL beam, the molecules are not only selected according to their mass-to-charge ratio but they are also conformer selected using gas-phase IMS (ion mobility spectrometry) methods. Surprisingly narrow amide-I bands near 1650 cm^{-1} and amide-II bands near 1500 cm^{-1} have been found. Further molecules studied by IR spectroscopy combined with IMS include the penta-peptide leucine-enkephalin.

Station V: Spectroscopy of bio-molecules embedded in superfluid helium nano-droplets Helium nano-droplet isolation spectroscopy has been established as a method to record optical spectra of molecules at ultracold conditions. Charged bio-molecules, brought into the gas phase via electrospray ionization, are mass-to-charge selected in a mass spectrometer and stored in an ion trap. Suprafluid helium nano-droplets traversing the ion trap pick up the molecules. The IR spectra of the ions at 0.4 K in the helium droplet, measured with the FHI FEL radiation, reveal much narrower lines than those measured using conventional gas-phase methods. This is why the results are expected to improve the understanding of the peptide and protein structures. The von Helden group has so far observed spectra of leucine-enkephalin and of the proteins ubiquitin and angiotensine.

Station VI: Nonlinear spectroscopy of solids The group of Alexander Paarmann (FHI) has embarked on a project to investigate ultrafast dynamics in solid state systems under MIR excitation with the FHI FEL. These experiments on the one hand allow micro-pulse temporal characterization using second harmonic generation in MIR nonlinear optical crystals like CdTe and GaSe. On the other hand, ultrafast time-resolved experiments using the FEL as pump pulse and applying various probes, such as visible reflectivity or sample magnetization allow detailed study of fundamental interactions among low-energy excitations. As part of the project, sub-ps time synchronization of a near-IR femtosecond laser with the FEL micro-pulses will be implemented.

Station under preparation: Surface science experiment A surface-science beamline station is currently under construction by the group of Helmut Kuhlenbeck (FHI). It is scheduled for commissioning in 2015. It will be used

ISBN 978-3-95450-133-5

for vibrational spectroscopy of metal clusters deposited on flat ordered substrates, so-called model catalysts. Similar to the gas-phase experiments the clusters will be tagged with weakly bound rare-gas atoms. Upon vibrational excitation of the cluster by FEL radiation, the rare gas atoms will desorb due to the rapid resonant heating of the clusters, and vibrational spectra of the clusters can be measured by recording the desorption signal.

CONCLUSION

The IR FEL Facility at the Fritz-Haber Institute of the Max-Planck Society started regular user operation in November 2013. The MIR oscillator FEL, operated with 15 to 50 MeV electrons from an S-band copper accelerator made by AES, Inc., provides pulsed radiation from, currently, 4 µm to almost 50 µm. The temporal and spectral properties of the FEL radiation has been characterized using a grating spectrometer and by auto-correlation measurements. The width of the FEL's spectral line was determined to be as small as 0.35% (FWHM) for relatively large detuning of the FEL cavity length. For very small cavity detuning, IR pulses as short as 0.4 ps (FWHM) have been observed. As of now, 6 user stations have been applying the FHI FEL radiation for vibrational spectroscopy of charged and neutral cluster and bio-molecules in the gas-phase or embedded in helium-nanodroplets as well as for nonlinear spectroscopy of solids.

ACKNOWLEDGMENTS

We thank Johannes Bahrdt, Andreas Gaupp, Klaus Ott, and Ernst Weihreter (Helmholtz-Zentrum Berlin) as well as Lex van der Meer (Radboud University Nijmegen) for their valuable support to the planning and commissioning of the FHI FEL. Furthermore, we thank the members of the FHI FEL user groups listed in this paper for fruitful discussions.

REFERENCES

[1] W. Schöllkopf et al.. First Lasing of the IR FEL at the Fritz-Haber-Institut Berlin. In *Proceedings of FEL 2012, Nara, Japan*, MOOB01, 2013.

[2] H.P. Bluem et al. Accelerator Beamline Performance for the IR FEL at the Fritz-Haber-Institut, Berlin. In *Proceedings of FEL 2012, Nara, Japan*, WEOC04, 2013.

[3] W. Schöllkopf et al. The IR and THz Free-Electron Laser at the Fritz-Haber-Institut. In *Proceedings of FEL 2013, New York, NY, USA*, WEPSO62, 2013.

[4] W. Schöllkopf et al. Status of the Fritz Haber Institute THz FEL. In *Proceedings of FEL 2011, Shanghai, China*, TUPB30, 2012.

[5] A.M.M. Todd et al. Commissioning of the Fritz Haber Institute Mid-IR FEL. In *Proceedings of IPAC 2012, New Orleans, Louisiana, USA*, TUPPP087, 2012.

[6] A.M.M. Todd et al. Commissioning Status of the Fritz Haber Institute THz FEL. In *Proceedings of IPAC 2011, San Sebastián, Spain*, THPC106, 2011.

[7] H.P. Bluem et al. The Fritz Haber Institute THz FEL Status. In *Proceedings of FEL 2010, Malmo, Sweden*, MOPA09, 2010.

[8] S.C. Gottschalk et al. Design and Performance of the Wedged Pole Hybrid Undulator for the Fritz-Haber-Institut IR FEL. In *Proceedings of FEL 2012, Nara, Japan*, THPD13, 2013.

[9] D.A. Jaroszynski et al. Experimental observation of limit-cycle oscillations in a short-pulse free-electron laser. *Phys. Rev. Lett.* **70**, 3412 (1993).

[10] G.M.H. Knippels et al. Formation of multiple subpulses in a free-electron laser operating in the limit-cycle mode. *Phys. Rev. E* **53**, 2778 (1996).

ISBN 978-3-95450-133-5