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Novosibirsk free electron laser as a user facility

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Abstract

The Novosibirsk free electron laser is the first multi-turn energy-recovery linear accelerator with three separate laser systems (the terahertz, far-infrared and mid-infrared ones). The facility is well equipped with optical elements and instrumentation available to Russian and foreign users. In this paper, we describe in brief the workstations of the facility and survey selected recent experiments using intense monochromatic terahertz laser radiation, which can be tuned from 90 to 240 μm .

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1. Introduction

The Novosibirsk free electron laser (NovoFEL), a user facility [Kulipanov et al. (2015)] consisting of three laser systems (the terahertz, far-infrared and mid-infrared ones), is part of the Siberian Synchrotron and Terahertz Radiation Center. It emits monochromatic high-power radiation in spectral ranges from 5 to 240 μm . The terahertz laser system has been in operation since 2003, and most experiments have been performed using terahertz radiation. This system emits radiation in the spectral range of 90 μm to 240 μm as a continuous stream of 100-ps pulses with a repetition rate of 5.6 MHz and a line width of less than 1%. In a routine regime, the average power of radiation at the user stations is 50-150 W at $\lambda = 130 \mu\text{m}$. The high-power radiation, relatively narrow linewidth and tunability of the radiation wavelength enable a wide variety of experiments. Terahertz methods and techniques are rapidly developing fields of science, the potential of which is obviously far from being exhausted. The unique characteristics of the NovoFEL radiation and the techniques developed at the facility during past 10 years enabled experiments most of which would be impossible with conventional terahertz sources. The NovoFEL opened new possibilities for original experiments and novel methods and techniques in the field. Twenty eight research groups from Russia (Novosibirsk, Tomsk, Krasnoyarsk, Samara, Nizhny Novgorod, and Moscow) and abroad (South Korea and Germany) have worked at the facility. In this paper, we survey selected recent experiments on the terahertz line of the NovoFEL.

2. Beamline and workstations

Characteristics of the Novosibirsk free electron laser facility achieved by the summer of 2016 are described in the paper [Shevchenko et al. (2016)]. The accelerator and the laser resonators are situated underground in the radiation protected hall. The beam emitted from a separate laser resonator passes through the primary beamline and then enters the main beamline, in which the mirror system directs the beam to a selected workstation. A sketch of the beamlines and workstation positions is shown in Fig. 1 [Kubarev (2016)]. Stations 1 – 7 are situated on the first floor, whereas stations 8 – 13 are on the second floor. The beamline sections shown in the figure in the left side have been already assembled, and workstations 1 – 7 and 12 – 13 are available to users. Outputs 8 – 11 can be used for construction of new workstations. The beamline sections shown in the right will be assembled in near future.

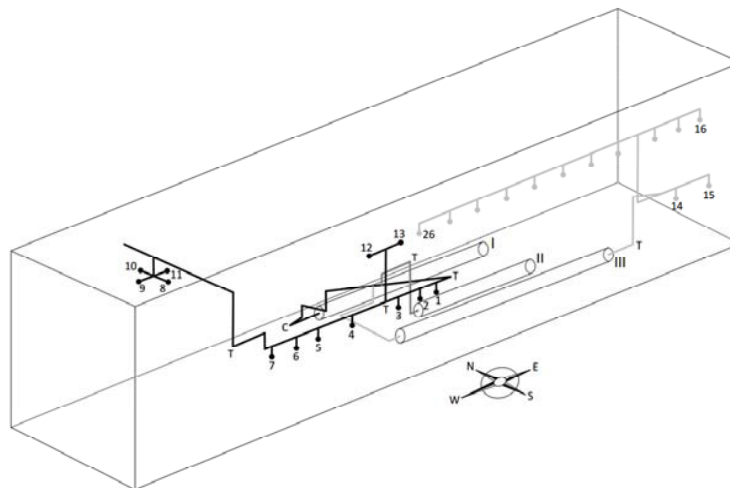


Fig. 1. Beamline system at NovoFEL. T – toroidal mirrors, C – spherical mirror, 1, 2, 3, ... – workstations. I – terahertz FEL, II - far infrared FEL, III - infrared FEL.

Two stations (1 – Radiation characteristics control and 7 – Fourier spectrometer) are devoted to beam diagnostics. Other stations are multi-purpose, and their names reflect their primary areas of research: 2 – Electron paramagnetic resonance, 3 – Biology and materials science, 4 – Metrology, 5 – Molecular spectroscopy, 6 – Medical, 12 – Spectroscopy and imaging, 13 – Evacuated chamber.

3. Manipulation of high-power terahertz radiation

The terahertz beam of the NovoFEL entering the workstations is a good-quality Gaussian beam the diameter and curvature radius of which differ at different stations. Since the users may have unlike requirements to the beam shape, one of the main tasks was the development of optical elements for transformation of terahertz beams. Optical elements for the terahertz range quite differ from the classical optics. A number of silicon diffractive optical elements, which enabled transforming the NovoFEL Gaussian beam into the Laguerre-Gaussian and Hermite-Gaussian ones, have been designed and fabricated [Agafonov et al. (2015a)]. Other elements, transforming the NovoFEL beam into pre-determined volumes like a pencil-like beam [Agafonov et al. (2015b)] or areas like a uniformly illuminated square are shown in Fig. 2. The problem of strong Fresnel reflection was solved with anti-reflection Parylene C film coating [Agafonov et al. (2013)]. A silicon multilevel kinoform lens [Komlenok et al. (2015)] was fabricated by the laser ablation technique for focusing high-power THz and IR radiation.

Using binary phase spiral axicons [Volodkin et al. (2016)], non-diffractive Bessel beams (Fig. 3) with an angular orbital momentum (vortex beams) of different topological charges were formed [Knyazev et al. (2015)]. Since such beams have great potential for data transmission and remote sensing, we investigated both numerically and experimentally the techniques which allow increasing the distance of beam propagation without beam divergence. That can be realized by reduction in the wavelength or expansion of beam with a telescopic system [Choporova et al. (2016a)]. Another experimentally verified feature of the Bessel beams useful for beam transport was the self-healing ability of these beams after passing a randomly non-uniform media or obstacles blocking several central Bessel rings.

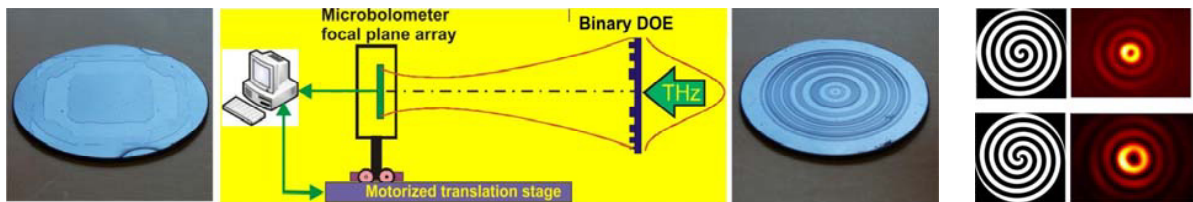


Fig. 2 Diffractive optical elements transforming Gaussian beam with wavelength $\lambda_0=141 \mu\text{m}$ into uniformly illuminated square (left) and pencil-like beam (right). Beam shape were studied using uncooled microbolometer matrix [Dem'yanenko et al. (2008) and Dem'yanenko et al. (2011)] moved by motorized translation stage.

Fig. 3. Silicon binary diffractive axicons, forming Bessel beams with angular orbital momentum of the first kind, first and second orders.

4. Terahertz imaging

Many applications require imaging in the THz range. Many devices can record images in the visible range, whereas only few kinds of THz radiation imagers are available because of the low energy of “terahertz photons”. Both direct imaging devices (uncooled microbolometer matrices and a pyroelectric array) and devices sensing thermal fingerprints of THz radiation are used at the NovoFEL workstations. Some imaging devices developed at the facility were described in detail in the paper [Knyazev et al. (2011)]. The microbolometer matrix and a thermal sensitive phosphor plate were recently applied [Choporova et al. (2015)] to recording holograms of objects illuminated with the NovoFEL radiation.

5. Applications requiring high-power beams

The high power of the NovoFEL terahertz radiation enables experiments that cannot be made using conventional THz sources. The high pulsed power of NovoFEL radiation was used [Kubarev (2015)] for demonstration of ignition of continuous optical discharge (COD) in gases at the atmospheric pressure (Fig. 4). It was found that a sequence of 66-ps terahertz pulses struck a COD in Air, Ar, He, N₂ and CO₂ at a specific power density of about 1 GW/cm². Other applications requiring a high-power radiation are the non-destructive ablation of biological substances and production of nanoparticles and hydrosols, see, e. g., the paper [Kozlov et al. (2016)]. Because of the very low efficiency of the acousto-optical effect for a long-wave radiation, experiments on the deflection of terahertz radiation performed in [Nikitin et al. (2015)] would also be impossible using sources with an average power less than that of the NovoFEL. Furthermore, next section experiments with surface plasmon polaritons travelling along a cylindrical surface also require terahertz radiation of high average power.

6. Study of surface plasmon polaritons

Surface plasmon polaritons (SPPs) are a subject of special interest in the integrated optics, but SPPs in the far-infrared and terahertz ranges have been investigated insufficiently. Previous experiments [Gerasimov et al. (2013), Kotelnikov et al. (2013)] with SPPs launched using NovoFEL radiation showed that the propagation length of terahertz SPPs was about 10 cm, while the Drude model predicts a propagation length of several meters. In new experiments at the NovoFEL, characteristics of THz SPPs were thoroughly studied at a wavelength of 130 μm using the experimental schematics shown in Fig. 5. The devices shown in Fig. 5 (upper drawing) enabled studying characteristics of both plasmons and the radiation decoupled off the surface (radiative losses). It was found that the propagation length at the gold-ZnS-air interface had a maximum when the ZnS thickness was about several hundreds of nanometers, depending on the surface quality. Such dependence has not been reported for the visible range and may be of special interest to integrated optics systems. These results will be published elsewhere.

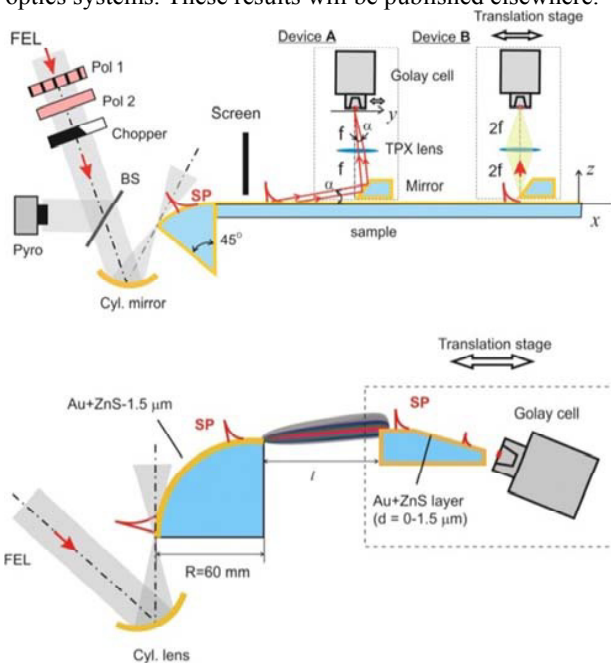
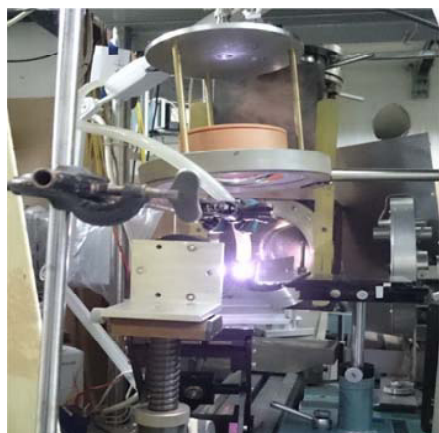


Fig. 4 Quasi-continuous optical discharge ignited by NovoFEL radiation in the focus of high-NA parabolic mirror; $\lambda=130 \mu\text{m}$.

Fig. 5. Experimental setups for investigation of surface plasmon polaritons in the terahertz spectral range. Upper schematic: study of SPP propagation length, decay length and radiative loss; lower schematic: study of plasmon jumps between two metal-ZnS-air interfaces.

Another phenomenon important for integrated optics was investigated with the device drawn in Fig. 5 below. It was found [Gerasimov et al. (2015)] that THz SPPs can “jump” from one metal-dielectric interface to another over air gaps up to 100 nm wide. The phenomenon can be exploited for splitting an SP beam into two new ones, guided by their own individual plane-surface substrates. A novel effect, the dependence of SPP generation efficiency on the direction of vortex beam rotation, was discovered [Knyazev et al. (2015)] when the “end-fire coupling” technique was applied to SPP generation. This effect can be used for the development of a new-type plasmonic switch.

7. Spectroscopy, biology, medicine

The tunability of the NovoFEL radiation enabled a number of experiments on absorption spectroscopy of molecular gases and flames. In the paper [Chesnokov et al. (2014)], OH radicals and NO molecules were detected in flames. In this case, the width of the laser generation line was practically the same as that of the molecule absorption line, and laser radiation may be assumed to be monochromatic. But, in fact, in some cases several vibrational-rotational transitions of a molecule can lie inside the laser line bandwidth. This feature was used in [Chesnokov et al. (2013)] for fast one-pulse spectroscopy of HBr molecule in the gas phase. Excitation of the ($J = 4$) \leftarrow ($J = 3$) lines of $H^{79}Br$ and $H^{81}Br$ (66.70 μm and 66.72 μm) with a laser pulse is followed by a complicated free induction decay signal (Fig. 6). The signal was measured in real time using an ultrafast Schottky diode detector. Such a signal corresponds in the frequency domain to a spectral resolution of about 10 MHz, which allows resolution of the isotopic and quadrupole structure of the absorption line. The molecular spectrum can be reconstructed using the Fourier transform, but the induction decay signal for any molecular transition has a unique pattern, which can be used directly for detection of molecule.

The possibility of measuring the concentration of H_2O molecules in flames, based on the absorption of terahertz free-electron laser radiation, was studied in [Chesnokov et al. (2012)]. These measurements were performed using the 77.32 cm^{-1} absorption line in the rotational spectrum of H_2O . This line has a low intensity at the room temperature, whereas at about 1000 K its intensity is comparable with that of the strongest lines. The temperature dependence of the radiation absorption coefficient at a frequency of 77.32 cm^{-1} was studied theoretically and experimentally. It was shown that the method can be used for measurements in a sooty $C_2H_4/O_2/Ar$ flame, which strongly scatters the visible and UV radiation.

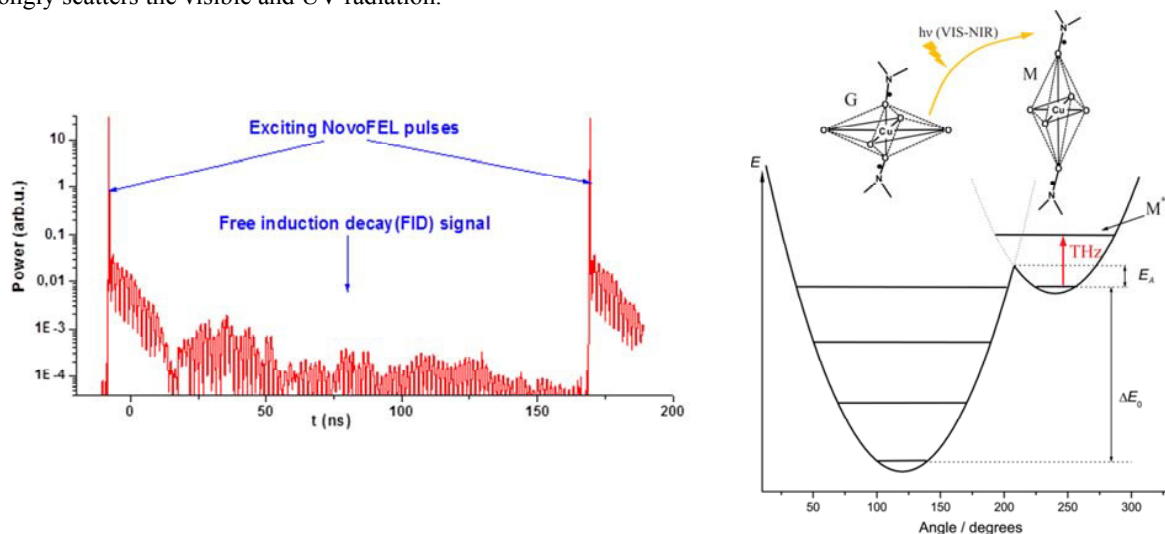


Fig. 6. Free induction decay emission of molecules of isotopic gas mixtures $H^{79}Br + H^{81}Br$ (pressure of 0.04 Torr, pipe 1 meter long) between two exciting NovoFEL pulses, following with frequency of 5.6 MHz.

Fig. 7. Schematic circular section of potential energy surface associated with two Jahn – Teller valleys in $Cu(hfac)_2L^{Pr}$ compound. Top: structures corresponding to ground (G) and metastable (M) geometries. The vibrational excitation by THz radiation which could induce the reverse $M^* \rightarrow G$

conversion is shown.

The station for the electron paramagnetic resonance (EPR) consists of the X-band (9 GHz) EPR spectrometer equipped with a He cryostat, a Nd:YAG laser system and a THz waveguide; thus, both the radiation sources (NovoFEL or Nd:YAG laser) can be used for sample irradiation during the EPR experiment. This station was used recently for exploration of the influence of vibrational excitation on the relaxation processes in the light-induced metastable state of $\text{Cu}(\text{hfac})_2\text{L}^{\text{Pr}}$ molecular magnet [Veber et al. (2013)]. The $\text{Cu}(\text{hfac})_2\text{L}^{\text{Pr}}$ molecular magnet undergoes thermo- and light-induced magnetostructural transitions and at temperatures of less than 20 K can be switched from a ground state (G) to a metastable state (M) by near-IR–vis light (Fig. 7). Far-IR and mid-IR spectra of $\text{Cu}(\text{hfac})_2\text{L}^{\text{Pr}}$ have intense absorption lines of different energy for G and M states (Veber et al. (2015), Barskaya et al.(2016)); therefore, these lines can be examined by high-power THz radiation generated by NovoFEL aiming to stimulate the conversion from metastable to the ground state. The experiments with THz irradiation at $1/\lambda = 200.4$, 217.8 (M and G characteristic bands) and 235.8 cm^{-1} (far-IR transparent region) did not confirm this assumption, probably because of the fast intermodal vibrational relaxation $\text{M}^* \rightarrow \text{M}$ as compared with the $\text{M}^* \rightarrow \text{G}$ conversion, but have demonstrated the robustness of the EPR system.

A one-color pump-probe system with a helium cryostat, recently commissioned at the facility, enables studies of relaxation processes in semiconductors. An original assembly combining precisely the pump and probe beams on the sample using an imaging microbolometer matrix is shown in Fig. 8. Two Golay cells are used as detectors of radiation passed through the sample. Results of the study of relaxation dynamics as a function of temperature (5 – 40 K) for bound-bound and bound-continuum transitions at 105, 141 and 150 μm in a Ge:Sb sample are presented in the paper [Choporova et al.(2016b)].



Fig. 8. ST-100 He flow cryostat and sample unit with precise focusing of pump and probe terahertz beams for study of transient absorption with 100-ps resolution.



Fig. 9. General view of the ellipsometer designed for study of surfaces and films using tunable radiation of NovoFEL.

Polarimetry and ellipsometry of materials and biological substances are important areas of activity at the NovoFEL. An ellipsometer developed and optimized for the terahertz range [Azarov et al. (2015)] is shown in Fig. 9. It was used for characterization of a number of biological and medical samples in the attenuated total reflection mode. This device proved itself to be the most sensitive ellipsometer in the THz range, especially in measurement of liquids.

The question whether the THz radiation affects biological objects and living organisms is the most important for biomedical applications, as well as for the development of systems for safety and non-destructive testing. Experiments to study the non-thermal effects of terahertz radiation on various living objects (from the archaea and bacteria, bluegreen algae, and crustaceans to the human stem cells) and different levels of their structural and functional organization (from DNA and RNA to changes in the proteome) are carried out at the NovoFEL. Some of the results are described in the paper [Demidova et al. (2015)].

8. Education

The NovoFEL, being a part of the Siberian Synchrotron and Terahertz Radiation Center (SSTRC), is not only a user facility, but also an educational center. Students of the Novosibirsk State University and the Novosibirsk State Technical University and PhD students from Novosibirsk Scientific Center, from Russia and abroad, use the facility to perform their student works and dissertations.

9. Summary

The Novosibirsk free electron laser is a user facility well equipped with instrumentation required for wide variety of experiments. It is available to Russian and foreign users. No payment is required from non-commercial users. Applications shall be sent to the NovoFEL executive board.

Acknowledgments

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