Raja Ramanna Centre for Advanced Technology
Indian Society for Particle Accelerators

Second User Interaction Meeting on

Science with IR/THz Free Electron Lasers



BOOK OF ABSTRACTS

Organised by

Free Electron Utilization Section, RRCAT, Indore

Sponsored by

Indian Society for Particle Accelerators (ISPA)

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Coherent dynamics and imaging of nanoscale quantum systems using x-ray free-electron laser pulses

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X-ray free-electron lasers (XFELs) provide an unprecedented capability to capture the ultrafast structural dynamics and electronic evolution of nanoscale quantum matter in real time [1][2]. Superfluid helium nanodroplets (HNDs) represent ideal quantum matrices for investigating multi-center dopant aggregation, quantum vortex dynamics, and laser-driven nanoplasma evolution [1–4]. Here, we report on recent experimental and theoretical advances combining single-shot soft X-ray Coherent Diffractive Imaging (CDI), two-color diffractive nanomoviemaking ("Dichography"), and simultaneous photoelectron Velocity Map Imaging (VMI) across multiple XFEL facilities (FLASH, European XFEL, and SwissFEL) [1, 2]. Using single-shot soft X-ray CDI (250 eV), we resolve distinct spatial dopant aggregation regimes inside superfluid HNDs [1]. Heavy xenon (Xe) atoms submerge into the droplet core or cluster along quantum vortices, giving rise to asymmetric, leaf-like diffraction patterns when located near the boundary [1, 4]. Conversely, alkali metal potassium (K) clusters remain weakly bound in surface dimples, staying invisible in static X-ray scattering due to low optical contrast [1]. However, upon near-infrared (NIR) laser ignition of a nanoplasma, localized ionization shells (He^{2+}/He^{+}) form around surface K clusters [1]. Time-resolved CDI reveals extended surface dimples and distinct ray-like diffraction patterns emerging at pump-probe delays ≥ 9 ps [1]. Simultaneous photoelectron VMI establishes fundamental nanoplasma scaling relations, confirming that scattered X-ray intensity scales with emitted electron yield as $N_{ph} \propto N_e^{1/2}$, while maximum electron kinetic energy follows Coulomb explosion kinetics $E_{max} \propto N_e^{2/3}$ [1]. To bypass detector speed limitations and track structural evolution in real time, we demonstrate "Dichography"—an imaging framework that disentangles two-color, single-shot diffractive patterns produced by collinear X-ray pulse pairs (1.0 keV and 1.2 keV; Δt = 750 fs) [2]. Combined with Droplet-CDI (DCDI) phase retrieval, Dichography successfully retrieves two-frame nanomovies of Xe-doped droplets, proving the structural integrity of the superfluid helium matrix up to 750 fs post-excitation and tracking filamentary Xe vortex structures prior to cluster explosion [2]. These findings establish a powerful multidimensional paradigm for probing real-time quantum fluid dynamics, surface roaming, and transient plasma states in nanoscale matter [1–4].

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A Compact Free Electron Laser based THz facility at IUAC, New Delhi

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A THz facility based on Free Electron Laser source has been commissioned at Inter University Accelerator Centre (IUAC), New Delhi in 2025. The facility is made compact using the concept of pre-bunched FEL where a train of electron micro-bunches with a maximum energy up to 8 MeV are being injected into a compact Undulator to produce the THz radiation in the range of 0.18 - 3.0 THz. Presently the ultrashort electron beam bunches having energies 1- 6.5 MeV are produced on routine basis using the copper photocathode of the normal conducting RF Gun and user experiments were conducted as per the requests received from the user. The electron beam having energy of 3Mev was interacted with thin Aluminium foil to produce Coherent Transition Radiation (CTR) consisting of broadband electromagnetic radiations up to the range 0.8 THz. The same beam was also injected in to the Undulator consisting of arrays of permanent magnets to produce narrowband electromagnetic radiations in the range of 0.3 THz. The produced THz radiations were detected using a Schottky Barrier Diode detector and Michelson Interferometer set up is being developed for characterization of the produced THz for user experiments. First THz imaging has been carried out using the produced narrowband THz from Copper photocathode. New semiconductor photocathode materials are being developed to increase the electron beam current thereby THz intensity for experimentalists. The transport of THz radiation is being worked out for the setting up of Experimental stations with the available detectors. Efforts are also going on for the development of the experimental facilities with help of active groups from various institutes to perform experiments using the electron beam and the THz radiation. Many publications are reported on the development of various subsystems of FEL and also by users using the electron beam. The details of the facility and operational details will be presented along with possible experimental set up for user experiments.

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Room-Temperature Experiments at the RRCAT IR-FEL: Strong-Field Nonlinear Optics, Degenerate Pump–Probe Spectroscopy, and Resonant Condensed Matter Physics Across 12.5 to 50 Micrometers.

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The Infrared Free Electron Laser (IR-FEL) at the Raja Ramanna Centre for Advanced Technology (RRCAT), Indore, is India's first operational free-electron laser user facility, delivering monochromatic, continuously tunable coherent radiation across the 12.5 to 50 micrometer (24 to 6 THz; 800 to 200 cm^{-1}; $\hbar\omega$=99.2 to 24.8 meV) spectral band. Operating with ~10 ps micropulses at 29.75 or 59.5 MHz within 4 to 6 microsecond macropulses (1 to 4 Hz), the facility delivers micropulse energies exceeding 35 microjoules (~3.5 MW peak power at 17.7 micrometers) to the user stations via a >55 m nitrogen-flushed optical transport line. When focused using standard F/4 parabolic optics, the delivered beam achieves focal intensities of 3.4 to 55.0 GW/cm^2 and peak optical electric fields of 1.61 to 6.44 MV/cm.
This talk presents some experiments at User Ports P-1 and P-4 designed specifically to operate at room temperature (300 K), zero external magnetic field (0 T), and without external synchronized tabletop lasers.

We detail two targeted materials physics experiments executable under these conditions at RRCAT:

(1) Dirac and Topological Semimetals (Graphene, Bi_2Te_3, Bi_2Se_3, $PtSe_2$): Pumping graphene below its 200 meV optical phonon energy (λ=13 to 45 micrometers) to quench optical-phonon emission and resolve picosecond relativistic collinear versus non-collinear Coulomb scattering bottlenecks, macroscopic momentum anisotropy decay, and the interband Pauli-blocking to intraband Drude crossover, alongside sub-bandgap surface-state saturation in topological chalcogenides.

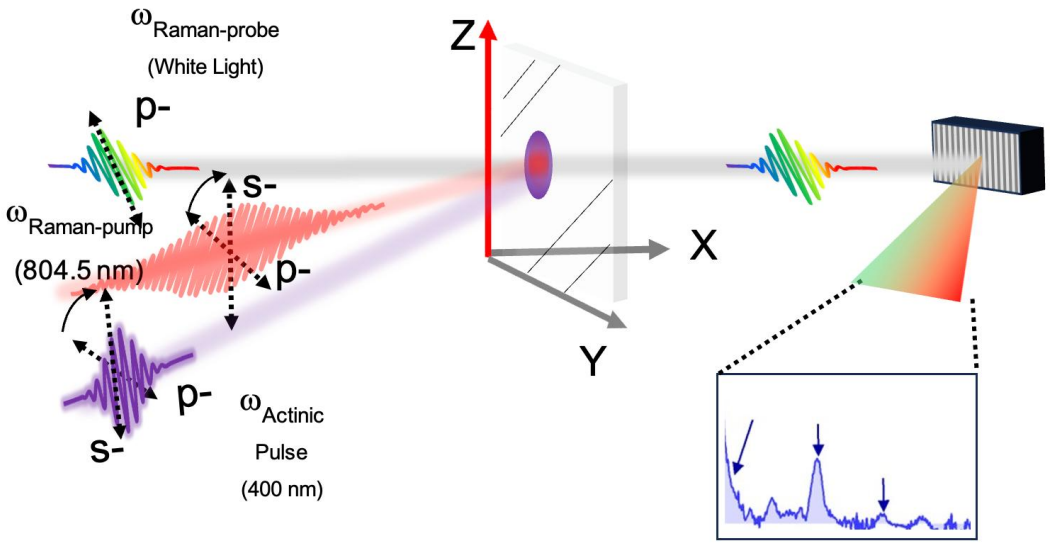
(2) Correlated Oxides and Ferrites (VO_2, Fe_3O_4/Si(100), $Bi_2Fe_4O_9$, $Ba_{0.8}Sr_{0.2}TiO_3$): Driving the non-thermal insulator-to-metal transition in VO_2 via resonant V–O phonon pumping (16.7 and 29.8 micrometers), and probing transient small-polaron delocalization and T_{1u} optical phonon lifetimes at 17.5 and 27.8 micrometers in ferrimagnetic Fe_3O_4 thin films.

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Probing coupling between charge carriers, spontaneous polarization, and polar phonons in prototypical ferroelectric $BaTiO_3$

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Coupling of charge, orbital, spin, and lattice degrees of freedom is essential for realizing various functional properties, such as ferroelectricity, thermoelectricity, the magnetoelectric effect, the metal-insulator transition, superconductivity, and charge-density waves. However, the relative contributions of the competing orders in controlling the desired behavior, and how and at what time scale they interact, are difficult to discern. A detailed experimental mapping of various competing order parameters is necessary to decipher the governing mechanism(s). In this talk, I will describe our recent efforts to experimentally map the ultrafast response of charge carriers, polarization, strain, and lattice vibrations in the prototypical ferroelectric $BaTiO_3$ using transient absorption, time-resolved second-harmonic generation, and femtosecond stimulated Raman scattering.



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Photothermal spectroscopy using micro-sensors for sensitive and selective chemical sensing

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Real-time monitoring of trace hazardous chemicals is essential for environmental remediation and public health protection. Integrating micro/nano-sensors with mid-IR spectroscopy enables the sensitive and selective detection of trace contaminants. This talk introduces photothermal cantilever deflection spectroscopy (PCDS), which integrates the high thermo-mechanical sensitivity of bi-material microcantilevers with the molecular selectivity of IR light. We demonstrate the label-free detection of trace perfluorooctanoic acid (PFOA)—an emerging environmental plastic contaminant—and dimethyl methylphosphonate (DMMP), a sarin simulant, without pre-concentrators. Furthermore, we show how low-cost, commercial quartz tuning forks paired with mid-IR radiation achieve rapid standoff classification of plastic samples. The talk will conclude with prospective photothermal sensing experiments using the IR-FEL source available at RRCAT.

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Indigenously Developed Pyroelectric Detectors for IR-THz FEL Beam Detection and Profiling Applications

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The development of indigenous pyroelectric detectors for infrared (IR) and terahertz (THz) radiation is essential for advanced IR-FEL beam diagnostics and characterization. This talk will present the fundamental principles of pyroelectric detection and the technology development of indigenously developed single-channel and multi-channel detector structures, incorporating silicon chip carrier technology and specially synthesized IR/THz absorbing materials, along with their characterization using IR-FEL facilities at RRCAT, Indore. Potential applications in IR-THz beam detection, profiling, and non-dispersive infrared (NDIR) gas sensing will also be highlighted. Furthermore, ongoing exploration of CMOS-compatible pyroelectric materials, particularly aluminium nitride (AlN) and hafnium oxide (HfO_2), for next-generation monolithically integrated detector development will be presented. The talk will conclude with future prospects about detector array development for IR-FEL beam profiling along with our vision for establishing indigenous capabilities in advanced IR-THz detector technology for various societal and strategic applications.

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UGC–DAE CSR User Station at RRCAT: Ultrafast Spectroscopy Prospects Enabled by the IR Free-Electron Laser

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This talk highlights the scientific opportunities of a proposed UGC–DAE CSR user experimental station at the Infrared Free-Electron Laser (IR-FEL) facility at RRCAT, Indore. Envisioned as a national user facility, the station aims to advance research in ultrafast and vibrational spectroscopy by providing access to intense, wavelength-tunable ultrashort IR-FEL pulses spanning the 12.5–50 µm range. This spectral window enables selective resonant excitation of low-energy vibrational modes and lattice phonons, opening new avenues for investigating ultrafast lattice dynamics, nonlinear phononics, resonant phonon control, and light-induced nonequilibrium structural transformations through IR-FEL pump-optical probe measurements. The station will also facilitate advanced nonlinear spectroscopic techniques such as time-resolved sum-frequency generation (SFG), alongside synchronized optical pump–probe measurements. By integrating these complementary capabilities, the proposed facility will provide a versatile platform for user-driven research into the coupled electronic, vibrational, and structural dynamics of quantum, strongly correlated, and low-dimensional materials. Ultimately, it will enable new frontiers in light–matter interaction and pave the way for ultrafast control of material properties through resonant vibrational excitation.

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Probing Geometric and Electronic Effects in Doped Metal Clusters: Opportunities for IR-FEL-Based Vibrational Spectroscopy

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Clusters occupy a unique size regime in which their geometric and electronic structures can differ markedly from those of bulk materials, providing an ideal platform for investigating the effects of finite size on chemical bonding, stability, and composition. In this talk, the role of single-atom doping in metal and metal–oxide clusters will be discussed as a versatile approach to exploring the interplay between geometric and electronic effects in nanoscale alloys. Particular emphasis will be placed on how the incorporation of a single foreign atom can induce structural transformations, modify electronic properties, and influence the stability and reactivity of these ultrasmall systems. In this context, infrared free-electron laser (IR-FEL)-based vibrational spectroscopy offers exciting opportunities for obtaining characteristic vibrational fingerprints of size-selected clusters. Such measurements, in combination with theoretical calculations, could provide definitive structural assignments and deeper molecular-level insights into their bonding characteristics and reaction mechanisms.

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Role of Hydration in Biomolecular Condensation: Insight from THz Spectroscopy and the Potential Effect of High Intensity Radiation

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Biomolecular condensation has emerged as a pertaining phenomenon in the field of soft-matter physics. The process involves formation of protein self-aggregates in the form of liquid droplets or amyloid fibrils, which are often cytotoxic and could develop detrimental health conditions including neuro-degenerative diseases. The process of biomolecular condensation has mostly been understood from the protein's perspective whereas the pivotal role of water as a matrix of the cellular environment has been shunned. Our group has been using THz spectroscopic technique to determine the role of water in various biophysical processes including biomolecular condensation. THz spectroscopy (1 THz = 1012 Hz) is a rather non-conventional but highly potential tool to determine the network structure of water at biomolecular interfaces. In this talk I will discuss some results from our group on the pivotal role of hydration towards biomolecular condensation. We will also discuss the plausible effects of intense THz radiation on these biomolecular aggregates.

IR-FEL RRCAT Facility for Three Avenues of Metamaterial Research: Near-zero Refractive Index Platform, Bul-Dirac Semi-Metal-Based Ultrafast THz Optics and Chiral-Fingerprint Detection

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The infrared free-electron laser (IR-FEL) facility at RRCAT can support three avenues of metamaterial research: Near-zero-refractive-index (NZRI) photonics, bulk Dirac semimetal (BDS)-based ultrafast THz metamaterial devices, and chiral-fingerprint detection via extreme light-matter interactions. A metamaterial with a near-zero refractive index is an excellent platform for achieving phase matching, high intensities, and time-varying nonlinear optical (NLO) effects. Recently, the time-varying NLO effect was experimentally reported in an ITO thin film using an optical pump at an epsilon-near-zero wavelength [1]. However, unlike the composite thin film, the metamaterial architecture readily realizes near-zero values in the 6-24 THz IR regime for extreme light-matter interactions, enabled by slow-wave light and tight confinement. Secondly, the picosecond pulses and high fluence of the IR-FEL are attractive for exploring ultrafast THz optics in a bulk Dirac semimetal (BDS). An ultrafast THz meta-device based on BDS using an optical pump-THz probe was experimentally demonstrated with a 40 GHz switching speed [2]. Time-modulating the optical and electronic properties of BDS and graphene is promising for unravelling plasmonic effects in the 6-24 THz range. Moreover, this frequency window is an excellent tool to detect the chiral fingerprint of proteins and biomolecules. If we realize a chiral metamaterial IR sensor, one can investigate the chiral fingerprint of biological samples under extreme light-matter interactions. In this talk, I'll highlight a few results and present the scope of IR FEL for these research avenues of metamaterials.

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Photoinduced effects in Ferroic Materials

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The most natural way to induce, manipulate, and probe the dynamics of fundamental excitations in condensed matter, such as electronic excitations, phonons, and magnons, is through their interaction with electromagnetic radiation. When the excitation energy is resonant with a lattice vibration, a specific phonon mode can be coherently driven. At sufficiently high field strengths, where the electric field approaches the scale of the interatomic potential, the driven lattice motion can induce significant atomic displacements and thereby modify the material's functional properties. In this context, free-electron laser (FEL) sources provide a unique platform for selectively exciting lattice vibrations using intense, tunable mid-infrared and terahertz pulses. By driving specific phonon modes, ferroic order can be manipulated on ultrafast timescales while largely avoiding direct electronic heating.

In this presentation, I will discuss our recent FEL-based experiments on a variety of ferroic materials, including ferroelectric, magnetic, and ferroelastic systems, demonstrating how resonant phonon excitation can induce both transient and long-lived changes in polarization, magnetization, strain, and domain configurations. Using time-resolved optical imaging techniques, we directly visualize the spatiotemporal evolution of these photoinduced states and reveal the interplay between lattice, charge, spin, and strain degrees of freedom. Finally, I will propose future experimental directions for the phononics control of ferroic materials.

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FTIR Investigation of Thermally Evaporated Sb and Bi_2Se_3 Thin Films Using Free Electron Laser Radiation

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Thin films of antimony (Sb) and bismuth selenide (Bi_2Se_3) are promising materials for applications in infrared optoelectronics, photonics, spintronics, and next-generation electronic devices. The optical response of these materials in the infrared spectral region provides important information about their electronic structure, lattice dynamics, carrier response, and surface/interface-related phenomena. In the present study, we propose to investigate thermally evaporated Sb and Bi_2Se_3 thin films using Fourier Transform Infrared (FTIR) spectroscopy coupled with Free Electron Laser (FEL) radiation at RRCAT, Indore. Thin films will be deposited by thermal vaporation under controlled vacuum conditions on suitable substrates. Particular emphasis will be placed on the mid- and far-infrared spectral regions, where conventional laboratory FTIR sources may have limitations in terms of spectral intensity and tunability. The high-brightness and tunable FEL source can provide enhanced sensitivity for investigating weak infrared absorption and transmission features of the deposited films. To complement the optical measurements, lowtemperature resistance–temperature (R–T) measurements will be performed to investigate the temperature-dependent charge transport and identify the dominant conduction mechanisms in the thermally evaporated films. The evolution of resistance at low temperatures will provide information about carrier localization, impurity/defect-assisted transport, and possible surface- or bulk-dominated conduction in Bi_2Se_3. Magnetoresistance (MR) measurements at low temperatures will further probe the field-dependent electronic transport and carrier dynamics. Particular attention will be given to the magnitude and field/temperature dependence of MR, which may provide insights into multichannel conduction and surface-state-related transport in Bi_2Se_3.

IR FEL–Biomacromolecule Interactions: From Fundamental Mechanisms to Biological Applications

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Infrared free-electron lasers (IR-FELs) offer a unique and powerful approach for investigating and modulating biomacromolecular systems through tunable wavelengths and high-intensity, monochromatic infrared radiation. By precisely matching laser wavelengths to the characteristic vibrational modes of biomolecules, IR-FEL irradiation enables the selective excitation of specific molecular bonds, providing opportunities to investigate molecular interactions and structural dynamics. This wavelength-selective excitation can induce localized, non-thermal conformational changes in biological macromolecules, offering valuable insights into the relationship between molecular structure and biological function. In addition IR-FELs have emerged as versatile molecular cutters, enabling selective cleavage and modification of biomolecular structures under ambient-temperature and solvent-free conditions. These capabilities open new avenues for analyzing and manipulating complex biological systems. The talk will highlight recent advances and emerging applications of IR-FEL technology in biological sciences, including the study of molecular structure and function, modulation of biological activity, and potential approaches for targeted biomedical applications.

From Probe to Process: Tunable IR-FEL Light for Functional Thin Films & Devices

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The tunable mid- to far-infrared radiation (12–50 µm) generated by the Infrared Free-Electron Laser (IR-FEL) at RRCAT, Indore, serves as a unique photonic workbench: the IR light from the FEL can be utilized both as a non-destructive diagnostic probe to qualify optoelectronic devices and also as a selective processing tool to engineer functional thin films. The talk showcases this dual capability across the characterization of the superconducting far-infrared detectors and low thermal-budget processing of device grade Group-IV semiconductor thin films. In the first part, we present the sputter-growth optimization and superconducting transport of disordered $Ti_{40}V_{60}$ alloy thin films and their fabrication into 30-pixel lumped-element kinetic inductance detector (LEKID) meander arrays for the 12–50 µm wavelength band. By matching the meander impedance to SiO_2/Si and diamond substrates, electromagnetic simulations and simultaneous IR-FEL reflection–transmission measurements demonstrate peak optical absorption up to 95 % (narrowband) and > 50 % (broadband).

In the second part, the FEL becomes a processing tool. Germanium and silicon–germanium alloy films are easy to deposit as disordered (amorphous) layers but work best as crystals, and the usual route to crystallization needs hours of heating or metal additives. Infrared FEL light (λ = 14.0–24.89 µm; 10 ps pulses at 29.75 MHz, contained in 4 µs bursts repeated at 2 Hz) passes through a thin film and heats it throughout its volume, while the silicon wafer beneath stays cool: a tool for achieving crystalline Ge-based films on heat sensitive or polymer-based substrates. The IR light from FEL having sub-gap photon energies converted pulsed-DC magnetron-sputtered amorphous Ge, $Ge_{0.7}Si_{0.3}$, $Ge_{0.3}Si_{0.7}$ and Si films on silicon substrate into nanocrystalline films without any metal additives. At 17.11 µm, crystallization starts at an average power of 1.35 mW for Ge and 1.70 mW for $Ge_{0.7}Si_{0.3}$, and tuning the wavelength changes the power needed: Ge crystallized at 2.2, 1.7, 1.15 and 0.85 mW at 14.0, 19.54, 23.49 and 24.89 µm, even though the photons carry less energy at longer wavelengths (from 88.6 meV down to 49.8 meV). The film therefore absorbs the light 1.6–3.4 times more effectively at the long-wavelength end (23.49–24.89 µm), where it crystallizes and remains free of ablation or damage. X-ray diffraction and Raman spectroscopy confirm crystallization through the full film thickness across the Ge–Si series. Finally, we followed the film's electrical resistance while it was being irradiated: a 600 µm disc of $Ge_{0.7}Si_{0.3}$ film with four contacts carried a steady current while the IR pulses irradiated its center, and the voltage was recorded with every laser burst triggering a 17-msec. recording cycle. The resistance was found to reduce to a permanently stable low value (−20 %) within four IR macro-pulse bursts, i.e. only 16 µs of light in total, and then held stable within ±0.27 % through more than 1,400 further bursts. Changes in the film's resistance on millisecond time scales after each burst show that the crystallized region keeps growing, sustained by the latent heat the film releases as it crystallizes. The crystallization front propagates outward at speeds slowing from at least 0.3 m/s to 0.04 mm/s; finally stopping at a radius of about 60 µm, several times larger than the ~15-20 µm core heated directly by the laser, and the film then stays in its new crystalline state.

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IR-FEL User Facility: The Machine & its Operational Scenario

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The IR-FEL facility has been operated successfully from Oct-25 to Oct-26 with beam delivered to Users for over 900 hrs for experiments. The wavelength was varied from 12.5 to 28 microns with up to 5 mW CW power at user table. The FEL shows good power & wavelength stability over 8 hrs in a day. The peak pulse optical power is from 2 to 5 MW in about 10ps micro pulses. For the tuned r.f. accelerators and beam transport magnets delivering reliable daily operation, the electron beam was characterized to have a low emittance. The FEL optical beam size and position were characterized inside the user building for full operating wavelength scan with undulator gap change. The optical beam size variation was small and the beam centroid position was also with in half mm. The M-square value of the FEL in beam receiving area in user building 55 m away was also estimated to be around 2.1. The FEL can be tuned for high micro pulse power as well as high average power as per the need of user experiments. After a correction to beam energy at an estimated wavelength, the wavelength can then be changed with better than 0.1micron accuracy.