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Multi-Pass Free Electron Laser Assisted Spectral and Imaging Applications in the Terahertz/Far-IR Range Using the Future Superconducting Electron Source BriXSinO

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Free-Electron Lasers are a rapidly growing field for advanced science and applications, and worldwide facilities for intense field generation, characterization and usage are becoming increasingly popular due to their peculiarities, including extremely bright, coherent, wide band tunable ultra-short pulses which are not achievable with other techniques up to now. In this review we give a thorough survey of the latest advances in the Free-Electron Laser-based field generation and detection methodologies and then present the main characteristics of a future THz/IR source, named TerRa@BriXSinO, based on a superconducting linear accelerator. The foreseen source is strongly monochromatic, with a bandwidth of 1% or smaller, highly coherent both transversally and longitudinally, with extreme versatility and high frequency tunability. After introducing the most recent and novel FEL-assisted scientific investigations, including fundamental explorations into complex systems and time-dependent interactions and material dynamics, we present our vision on the potential use of the TerRa facility and analyze some possible applications, ranging from non-linear physics under extreme conditions to polarization sensitive imaging and metamaterial-based sensing.

Keywords: Terahertz (THz) radiation, infrared - IR, spectroscopy, accelerators, imaging, free electron laser (FEL)

INTRODUCTION

Free-Electron Lasers (FELs) physics and related technology is a rapidly growing field for advanced science and applications and currently represents a major subject of research worldwide. In the last 20 years FELs have demonstrated a great potential for tackling global issues in energy, transport, medicine, and bio-matter areas. Their widespread use in a desirably near future will be creating a range of novel opportunities achieving significant advances and extending the boundaries of different fields, from life sciences to novel investigations into condensed matter. FELs in fact combine unique capabilities and peculiarities with short pulse length, high brightness, monochromaticity and spatial coherence. The radiation produced by FELs show properties similar to optical laser light together with an ultra-broad frequency tunability, covering from X-rays to the microwave regime. A major advantage is that FEL emission consists of extremely intense, ultra-short, monochromatic radiation pulses allowing atomic-level detection of structural and functional properties with exceptional time resolution. Therefore, radiation produced by FELs in the X-ray, Ultra-violet (UV), visible (VIS), infrared (IR) and Terahertz (THz) region presents characteristics inherently different from the mainly used techniques such as synchrotron sources, Fourier Transform InfraRed (FTIR) systems and laser-based THz emission sources. As such, the development of FEL-assisted experiments, and the possible breakthrough in many scientific challenges of multi-disciplinary interest, requires a combined expertise in accelerator and laser science and related technologies to cover the various zones of the electromagnetic spectrum. In the X-ray range the pulse length can reach the limiting value of about 10^{-14} s and less, which is more than 10^3 shorter than the synchrotron generated radiation, while achieving a peak intensity (brightness) 10^9 greater. Due to the extremely high brightness and the ultra-short pulse durations, the measurements are unaffected by atomic motion, allowing a non-invasive material investigation. Data acquisition in fact can be completed in fs periods of irradiation, eliminating the radiation damages even under high irradiances. This becomes an important advantage for spectroscopic and imaging analysis of radiation (invasive) sensitive protein structures, soft tissues, and biological material. Apart from the ultra-high brightness, the capability of coherent detection and wide tunability obtained with FEL sources gives incomparable advantages upon well studied FTIR spectroscopy. For example, changes in atomic structure during protein function occurs at nano to femto-second timescale which is far short to achieve with conventional methods. Similarly, terahertz science, which keeps the peculiar properties of many materials in the spectral band of interest, benefits from both the high intensity electric field generation and the ultra-wide spectral tunability achieved by using FEL based generation methodologies. In standard THz spectroscopy based on photoconductive antenna (PCA) or nonlinear crystal emission, the most common methods used potentially covers the range 0.1–10 THz, but indeed the bandwidth is upper limited to six THz for the most part of optical instrumentation. Whereas, using FEL assisted techniques, the peak THz wavelength can be tuned over a range larger than 40 THz, allowing a stronger bridge between the photonic and electronic approaches. An extremely important feature of THz FELs is due to the operation with superconducting (SC) linear accelerators (LINACs) which can deliver high quality beams during all the acceleration time. The temporal structure of electron beams is characterized by a continuous train of pulses, and a repetition rate that can reach values of few hundreds of MHz. FEL generated beams are ideal to conduct diverse types of 'pump-probe' experiments, including THz pump/X-ray probe (or vice versa X-ray pump/THz probe), when operated together with an independent X-ray source as, for instance, an Inverse Compton Scattering (ICS) source alimented by the same electron beam. Moreover, the given electron beam manipulation techniques make it possible to tailor the THz band characteristics (narrow-band or broad-band) to be used for specific THz-pump/THz-probe measurements. The flexibility of combining diverse radiation sources is extremely beneficial for a multitude of experimental scenarios where an element-specific external (X-ray, IR and THz) excitation is needed combined with a corresponding surface/interface-sensitive suitable detection. In combination with the given radiation types, it is also possible to conduct time resolved pump-probe experiments with atomic level spatial resolution and femtosecond time resolution. As such, FEL assisted radiation exhibits superb spatial coherence, opening new perspectives for imaging techniques. For example, high-resolution 3D imaging of bio-systems and even single-molecule imaging by using polariton chemistry is a challenging objective which is prone to an enormous scientific impact. The study of exotic states of matter, characterization of materials under extreme conditions (deformation mechanisms, phase change dynamics, crystallization and polymorphic transitions) and observation of their evolution at ultra-short time scales, mapping the internal details of viruses and bacteria, decoding the molecular composition of cells and bio-materials, real time monitoring of chemical reactions, are just a few other examples to name in order to show the ground-breaking potential of FEL science.

In this paper, we aim to show some examples of free electron laser-assisted spectral and imaging applications in the THz/IR range that can be pursued using the future superconducting BriXSinO source. After this brief introduction, we summarize in **Section 2** the possible THz radiation (with extension in the Far Infrared Region) mechanisms putting a special emphasis on the undulator based techniques. In **Section 3**, the FEL source Terahertz Radiator (TerRa) in the framework of the BriXSinO superconducting accelerator, TerRa@BriXSinO, is described. In **Section 4** several possible detection systems are analyzed. Then, in **Sections 5, 6, 7** and **8** a selected list of interesting and novel applications using the TerRa radiation is presented. A last session with conclusions closes the paper.

RADIATION GENERATION

The THz frequency range lies between the radio and infrared bands, combining the photonic and electronic approaches within the broad electromagnetic spectrum. With the growing technological advances in THz science, this frequency window has been extended to include part of the IR spectrum. The recent "Terahertz Science and Technology Roadmap" broaden a prior definition of THz band (0.1–10 THz) setting its frequency range up to 30 THz [1].

THz pulses with ultra-strong electromagnetic field are of increasing interest, due to their growing potential for probing and controlling a variety of complex phenomena ranging from fundamental processes of molecules and nanostructures to phase transitions in solids and dynamical interactions in biomaterials.

TABLE 1 | THz/IR FEL sources worldwide. Parameters taken from [4].

Location	Abbreviation	Frequency [THz]	Maximum micro pulse energy [μJ]	Minimum pulse width [ps]	Micro pulse repetition rate
Netherlands	FELIX	6.7–100	50	0.25	25 MHz/1 GHz
	FELICE	3–60	1,000	0.3	16 MHz/1 GHz
France	CLIO	2–60	100	1	62.5 MHz
Russia	NOVO-FEL	20–60	30	10	3.76 MHz
Germany	FELBE	14.2–75	1.5	0.8	13 MHz
		1.2–16.5	5	1	
		7.5–60	3	0.8	
United Kingdom	ALICE	33.3–60	0.6	1	16.25 MHz
China	BFEL	12–60	-	4	2.86 GHz
		6–15	-	4	
Japan	FEL-TUS	21.4–60	6	1	2.9 GHz
	KU-FEL	13–83	13	0.7	29.75 MHz
	OSAKA-FEL	13.6–60	-	5	22.3 MHz
		5–15	-	5	22.3 MHz

Laboratory scale laser-based sources with moderate frequency band (<10 THz) and mid-range pulse energies (<μJ) are widely used in the broad scientific community due to the ease of use, cost effectiveness and commercial availability. Nevertheless, new scientific attention is given to large-scale accelerator-based sources with ultra-high pulse power (MW) coupled with a wide tuning range (<60 THz) and extremely fast repetition rates. Being prone to many novel scientific explorations through photon-matter interactions, extremely bright, coherent, widely tunable ultra-short, monochromatic pulses open new possibilities to fundamental physics of complex systems and to material structural analysis, including time-dependent interactions, nonlinear effects, and dynamics of different compounds. In this context, the THz science becomes a valid tool including the infrared and Raman spectral analysis but with superior properties due to the high intensity and coherency properties.

There is a fast-growing demand for FEL facilities due to their tunability and high spatial and temporal resolution. There are more than 30 FEL facilities actively used worldwide for scientific research, many of them including various user stations operating in different frequency regimes ranging between X-rays and THz waves. A detailed list of FELs and their characteristics are summarized by Neyman *et al.* [2]. A more recent status with the technical specifications of these facilities can be found at the Virtual Library web link [3] maintained by Ramian *et al.* from the University of California, Santa Barbara. Moreover, a frequently updated list is edited by Klopf *et al.* from the Helmholtz-Zentrum Dresden-Rossendorf [4].

Regarding specifically the THz band regime, an up-to-date list of operational facilities covering THz/IR band and main characteristics of interest (repetition rate, maximum micro-pulse energies, minimum pulse width) are given in **Table 1**. From the table, one can deduce that FELs operating in the high THz frequency band (>6 THz) are limited. In the following subsections, among all current THz sources, we will concentrate on high-field THz sources briefly summarizing the most common generation methodologies and comparing their scientific and technological capabilities with undulator based THz facilities.

Conventional Terahertz Sources

Since the very early stages of THz technologies, femtosecond laser-based techniques have been widely used to generate coherent pulses, enabling Time Domain Spectroscopy (TDS). Photoconductive switches [5–7] and optical rectification [8, 9] are the most widely used techniques due to commercial availability, cost effectiveness and easy adaptation to different experimental schemes. On the other hand, to satisfy the demand on wide band, high-intensity THz pulse generation, a large variety of techniques has been developed following the technological advancements in femtosecond laser systems. Some major sources to name are Schottky diode mixers [10, 11], backward wave oscillators [12–14], quantum cascade lasers [15, 16], nanosecond gas lasers [17] and gas plasmas assisted THz generation [18–20]. The main limitations of these laser driven THz sources stem from the emission intensities (<μJ) and/or the reduced frequency bandwidth (<10 THz) related to the intrinsic absorption characteristics of the nonlinear conversion media used. In this review the laser driven generation/detection methodologies and applications based on the mid-band THz radiation are not covered. The interested readers can find a detailed review on the metrology techniques in [19, 21] and on the broad mid-band applications in [1, 22–25].

Accelerator-based sources generate coherent high-field THz radiation from ultra-short relativistic electron bunches. Linear accelerators are used to generate electron bunches that can radiate a fully coherent (super-radiant) THz radiation in proportion with the number of electrons emitting radiation in phase. The super-radiant and the high field THz emission in these sources require ultrashort electron bunches or density-modulation at THz frequencies [26]. Some major examples of this type of

TABLE 2 | Beam characteristics comparison for various sources (modified from [42]).

	FEL undulator radiation	FEL dipole radiation	FEL transition radiation	Laser optical rectification	Laser photo-switch	Laser plasma
Maximum Pulse Energy [μJ]	>100	>100	>500	1–100	<1	<5
Frequency Band [THz]	1–30	1–3	0.3–3	0.1–10	0.1–5	1–30
Repetition Rate [Hz]	10^1–10^7	10^1–10^7	10^1–10^7	10^2–10^6	10–10^8	10–10^3

interactions are radiation produced by undulator [27, 28], magnetic dipole [29], transition [30, 31], diffraction [32], Cerenkov [33, 34] and Smith–Purcell [35, 36] effect. One of the greatest benefits for novel experimentation using electron bunch triggered radiation is that the intense THz emission becomes self-synchronized with all radiation types (from IR to X-ray) emitted with the same electron bunch [37]. This also allows the use of coherent THz pulses for electron beam diagnostics and to reconstruct the charge distribution of the electron bunch [45]. Generating short electron bunches is the most essential point for achieving a high peak THz power. Coherent Transition Radiation (CTR) with high pulse energies reaching mJ levels [31, 38] can be generated via colliding the ultra-relativistic electron bunches with a metal screen [39–41]. Electric field magnitudes exceeding 10 MV/cm level and covering a very broad spectral windows can be reached with this technique. TELBE, FLUTE, FLASH, FERMI, LCLS, and SPARC are some examples of large facilities using this type of CTR modules [42]. A similar mechanism for high field THz emission is the edge radiation [43, 44] generated by directing electrons through a longitudinal magnetic field gradient. Edge radiation-based THz generation attains energies up to 10 μJ, giving a peak electric field up to 3 MV/cm. Even if the edge radiation keeps similar field properties, with the CTR based generation mechanism the THz signal can be completely synchronized and made phase stable with an accelerator-based X-ray source, as both sources are generated by the same electron bunch. This allows a convenient combination of THz/X-ray pump/probe experiments. A similar technique has been implemented at FLASH, reaching pulse energies around 1 μJ with electric field intensities up to 900 kV/cm [42].

Free-Electron Laser Based Terahertz Sources

Another mechanism for THz generation is the FEL emission by an electron beam driven through an undulator. With this technique pulse energies up to mJ range can be obtained. The first demonstration was held at FLASH [45] by using a nine-pole electromagnetic THz undulator. Fully synchronized and phase stable (fs levels) THz radiation is emitted reaching pulse energies up to 100 μJ with peak electric fields up to 1 MV/cm [37]. The THz FELs can operate in various kind of configurations: seeded THz FEL amplifiers, Self-Amplified Spontaneous Emission (SASE) THz FELs, short-pulse super-radiant THz facilities and multi-pass oscillators.

Mainly, IR, Far-IR (FIR) and THz FELs are designed to operate as oscillators. Examples of IR and FIR oscillators are FELIX, ELBE FELs, FLARE, ALICE and Novo-FEL. The advantages of this operational mode are the compactness, relaxed requirements for the quality of electron bunches, and the fact that oscillators are suitable for SC-LINACs, enabling the generation of powerful quasi-Continuous Wave (CW) light. Differently to other schemes, FEL oscillators require a stable repetition rate of electron bunches with very low jitter.

A comparison of the main beam characteristics grouped for different type of sources is given in **Table 2**.

TABLE 3 | BriXSinO electron beam parameters.

Energy [MeV]	22–45
Bunch charge [pC]	50–200
Repetition rate [MHz]	50–100
Average current [mA]	5
Peak current [A]	8–12
Rms electron emittance [mm-mrad]	1.4–2
Rms energy spread [%]	0.1
Energy beam fluctuation [%]	<0.05

TABLE 4 | TerRa@BriXSinO parameters of the undulators with permanent magnets, variable gaps, and linear polarization. First module is the Terahertz Undulator (TU). Second module is the Infrared Oscillator (IO). λ_w undulator period, B peak on-axis magnetic field, λ emission wavelength, L_w length.

Undulator	TU	IO
λ_w [cm]	3.5	3
B [T]	0.5–1	0.5–1
λ [μm]	5–50	3.5–35
L_w [m]	1.75	1.75

THE BriXSinO PROJECT

The BriXSinO project is developed by INFN (Istituto Nazionale di Fisica Nucleare), Section of Milan in collaboration with INFN, Section of Naples and Italian National Institutions and Universities, and it will be located at LASA (INFN and University of Milan, Italy). The nominal characteristics foreseen at BriXSinO for the electron beam and for the undulator are given in **Tables 3** and **4**.

The BriXSinO project was primarily intended as a demonstrator for the physics of the two-pass two-way

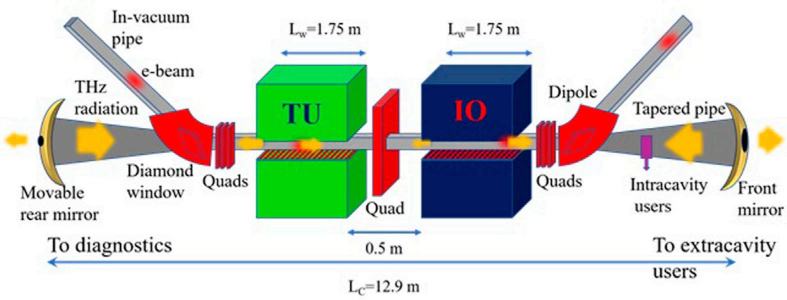

FIGURE 1 | Schematic of the TerRA@BriXSinO source, with details of the segmented undulators TU&IO. The radiation produced by the electrons in the undulators is recirculated inside a cavity of length L_c = 12.9 m. The undulator modules are 1.75 m long and separated by 0.5 m where a quadrupole, phase shifters, diagnostics and correcters are located. The elements are not in scale.

acceleration scheme using SC cavities [46, 47]. After being accelerated up to 22–40 MeV, the 50–200 pC electron beam is sent into an arc, whereby it retraces the accelerator in the opposite direction. Depending on the entrance phase, the electron beam can give the energy back to the SC as an Energy Recovery Linac (ERL) or being further accelerated. In this way, the energy deposited by the electron beam to the dump can be limited and controlled. In the zero dispersion zones of the arc, the electron beam finds on one side a Fabry-Peròt assisted Inverse Compton source and on the other one a THz FEL oscillator. The two radiation sources can be alimented either independently or in sequence, thus generating two synchronized radiation pulses in two different ranges of frequencies, namely X-rays and THz. The combination of limited space and low peak current together with a high repetition rate - up to 100 MHz - and high stability in beam energy has led to design the THz source as a FEL oscillator: i.e., the FEL undulator is placed inside a cavity whereby the radiation is re-circulated and high average THz power is realized.

Due to the wide versatility of BriXSinO's beam lines, it exhibits strong potentiality as user facility for both X-rays and THz radiation.

The scientific case of the FEL source TerRa indicates strong interest in radiation of wavelengths between 10 and 50 μm (3–17 THz). The BriXSinO accelerator line delivers a 50–200 pC electron beam with energy between 22 and 40–45 MeV, as shown in **Table 3**. Taking the highest energy values and assuming an undulator with a period λ_w equal to 3.5 cm, from the resonance relation:

$\lambda = \lambda_w(1 + a_w^2)/(2\gamma^2)$ Eq. 1

(where $a_w = 0.657\, B(T)\cdot\lambda_w$ (cm), γ the Lorentz factor of the electron beam).

radiation having wavelength λ ranging from 10 to 60 μm (3–20 THz) can be generated with a peak efficiency (evaluated as the ratio between the Extra-Cavity radiation energy and the energy of the electron beam) of $4.4\cdot10^{-3}$ around 20 μm.

The FEL Oscillator TerRa@BriXSinO source has been designed as the sequence of two undulator modules, separated by a drift where a quadrupole, phase shifters, diagnostics and correctors are allocated, and embedded into an optical cavity equipped with mirrors suitable to the considered frequency range. The segmented undulator TU&IO, whose pictorial view is represented in **Figure 1**, is made of two different sections that could be also characterized by different periods: TU (Terahertz Undulator) and IO (Infrared Oscillator). They can work either tuned at the same wavelength or separately, delivering different wavelengths.

TABLE 5 | Properties of the intra-cavity (IC) and extra-cavity (EC) radiation at different frequencies for TerRa@BriXSinO, assuming beam peak current of 10 A (a), 10 A (b), 20 A (c), 25 A (d).

	(a)	(b)	(c)	(d)
Frequency [THz]	15	10	8.5	6
Beam energy [MeV]	40	33.2	30	26
IC Energy [μJ]	355	300	952	560
EC energy [μJ]	17.7	15	46.6	16.8
EC average power [kW]	1.77	1.5	4.66	1.68
Peak power [MW]	250	100	250	100
Bandwidth (%)	0.6	1.6	2.7	1.3
Rms waist [mm]	2.6	3.5	4.5	5

The numerical modeling of the TerRa source has been carried on by using the three-dimensional, time-dependent FEL code Genesis 1.3 [46]. Starting from the electron beam parameters listed in **Table 3**, we have injected into the undulator a sequence of randomly prepared electron beams different one from each other both microscopically and macroscopically, to simulate the fluctuations of a bunch train. The study has been performed for radiation wavelengths between 10 and 50 μm, with an undulator having λ_w = 3.5 cm. **Table 5** reports the main properties of the radiation at different wavelengths. As shown in **Figure 2**, single shot power in time (**Figure 2A**) and spectrum vs wavelength (**Figure 2B**) appear to be single spiked. Intra-cavity (IC) single shot radiation energy of 355 μJ (meaning 17.7 μJ of extra-cavity (EC) energy and 1.77 kW of average output power) at λ = 20 μm has been estimated. The option constituted by two undulator modules of different period allows therefore to obtain two different wavelengths with the same electron beam [47].

Regarding the stability and coherence properties, the THz radiation is almost fully coherent and widely stable both in space

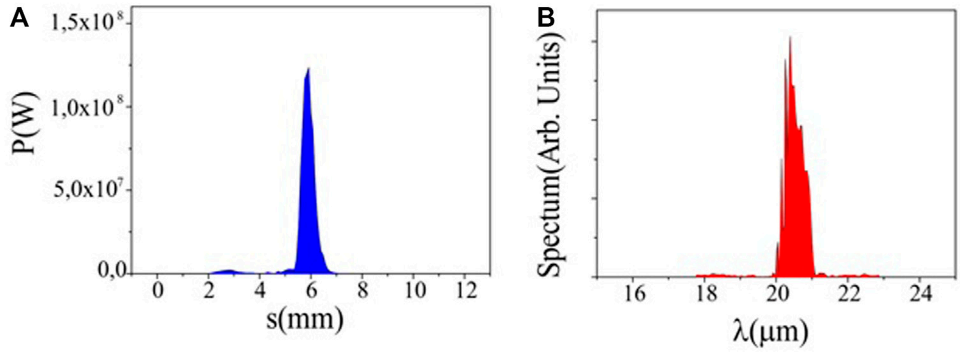

FIGURE 2 | Radiation properties of the TerRA@BriXSinO source. **(A)** The Intra-cavity longitudinal profile of the power for a single shot of 355 mJ and **(B)** the spectral profile, when the two undulators (TU&IO) are in tune for an emission wavelength of λ = 20 μm at the saturation point of the oscillator.

and in time. The X rays produced by the Compton back-scattering in the other beam line of the device is temporally incoherent but can acquire a partial transverse coherence length of about few microns after meters of propagation.

The BriXSinO project is going to open new research opportunities for the run of high field two-color THz spectroscopy measurements or X-ray pump/THz probe experiments.

As a further option, the THz source could be coupled to a laser synchronized with the oscillator. A pump laser delivery station with photon energies in the three main bands, NIR, VIS and UV, could be installed in a dedicated area. The possibility to investigate the temporal recovery scale of different excited states of the sample in time resolved pump-probe experiments is directly related to the performance of the pump laser in terms of photon energy, time duration and jitter as well as pulse energy and repetition rate. The pump lasers will be distributed at the end-stations and carefully synchronized with the FEL beams by means of a dedicated synchronization unit. A 1 MHz repetition rate or higher will be obtained. A possible way to accomplish this option should be to operate with a laser system delivering 800 nm, 15 fs pulses aiming at a pulse energy of 0.2 mJ or larger, in burst mode (4÷5 MHz). An optical parametric amplifier and a second/third harmonic generation box would convert this wavelength into the desired pump wavelength throughout the UV-VIS-IR ranges (ca. 200—2000 nm).

DETECTION

In the framework of FEL applications, the detection and characterization of high field, large bandwidth electromagnetic signals having ultrashort time duration are of extreme importance and represent a challenge. The commonly used electronic devices and/or detectors do not satisfy the need for such advanced signal responses, due to the technical low capacity in terms of the rise times (ps level) and relatively low response times (ranging from ns to μs). The possibility to simultaneously measure pulse duration and intensity profile temporal response, and to monitor electric field evolution, is extremely crucial for phase and amplitude sensitive spectral measurements.

The electro-optic (EO) sampling is one of the most intensively used and proven candidates for high intensity THz/IR emission characterization. This technique also combines the advantage of the possible characterization of electron bunches. Optical pulses in the visible and NIR laser sources can reach ultra-high pulse durations (few tens of fs), that are much smaller than the oscillation period of the THz electric field (few hundreds of fs). Thus, THz sampling with such sources can be achieved with high resolution. EO techniques are based on the Pockels effect, based on the refractive index change of crystal media used in the presence of the THz electric field. The THz electric field induces a transient birefringence and therefore a phase shift between polarization components in these optically active crystals. THz and laser beams are superimposed in the EO crystal, and the transmitted electric field detected by using balanced photodiodes in conjunction with a Wollaston prism, as shown in the schematics of **Figure 3**.

In detecting high field THz beams using EO sampling there are a few technical considerations that one should consider. First, lattice symmetry [48] and thickness [49] of the EO crystal are important parameters affecting the sensing performance and the achievable bandwidth of the impinging electric field. The intrinsic optical phonon resonances of the crystal are also important and should be taken into account since THz frequencies in proximity are prone to strong absorptions and reflections. For example, resonances are found at 5.3 THz in ZnTe and 11 THz in GaP. Moreover, they correlate also with an anisotropic index of refraction profile within the media, producing a strong variation in between phase and group velocities of the probe pulse inside the crystal and limiting the temporal resolution [50]. Similarly, the dispersion of the probe pulse would significantly distort the THz time domain profile. These effects can be minimized by using a thin crystal media, at the expense of reduced intensity profile, or compensated by model combined EO detection methodology [51].

The TELBE THz facility consists of two super radiant THz sources where a sequential EO sampling is implemented for ultrafast benchmark experiments for pulse- and field-resolved

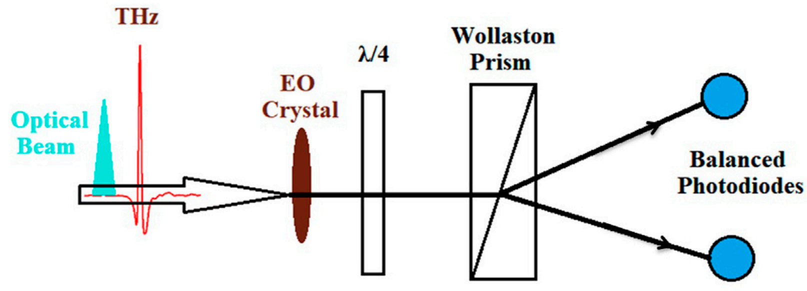

FIGURE 3 | The basic electro-optic sampling scheme with a balanced detection method.

THz diagnostics. ZnTe and GaP are the most commonly used crystals for EO sampling of broadband high THz fields. The interested readers may find a comparative work authored by Wu *et al.* [52] on the properties and the EO detection performances of commonly used nonlinear crystals for FEL assisted sources, and a thorough investigation into terahertz pulse EO sampling abilities of various crystals (including CdTe, DAST, GaAs, GaP, and ZnTe) in [53].

It is very important to note that the EO sampling technique can be also used for measuring the longitudinal electric field distribution of electron bunches with a sub-ps time resolution [54]. The given approach is realized at HUST (Huazhong University of Science and Technology) THz-FEL facility for sampling both the coherent THz electric field and the field distribution of the electron bunches [51]. The system uses a mode-locked 10 fs laser at 800 nm with 79.33 MHz repetition rate as a probe pulse, actively synchronized to the accelerator RF clock (2,856 MHz) and achieving a jitter around 100 fs [55]. Analogously to the EO sampling of transient electric fields, phase mismatch, dispersive propagation and absorption affects the detection of the temporal profile of fs electron bunches [56]. The emission and focusing properties of the original electron bunch profile differ with respect to the transition and diffraction radiation [51, 57].

Another well-established technique is field-streaking, that is based on the photoelectron momenta change created in the presence of an impinging electric field [58]. Mainly streak cameras are used for this purpose, yet their resolution (limited to a few tenths of picoseconds) is not sufficient for FELs pulse durations (few fs). On the other hand, for longer streaking field periods needed for FEL sources correspond with THz and FIR frequencies. The use of THz field-streaking has been demonstrated at FLASH [45] where researchers have used THz pulses from an undulator for measuring the single shot X-UV pulse durations.

THz Air Breakdown Coherent Detection (ABCD) is a recent technique removing the bandwidth limitations [59]. This approach uses the third-order nonlinear frequency mixing in air or selected gases as the sensing media and is based on the same physical phenomena of field-induced second harmonic generation under two-color excitation [60]. The greatly reduced absorption and dispersion allows detection up to 30 THz. This technique is favorable in terms of synchronization for gas plasma generated THz field detection. The interested readers can find in two thorough reviews authored by Dai *et al.* a detailed introduction to the mechanisms [19] and potential applications [61] of the THz air photonics technique.

ADVANCED APPLICATIONS

As already mentioned above, with the growing technological advances in THz science the current definition of THz band (0.1–30 THz) is merging with the IR radiation. In this context, the THz science becomes a valid tool including the infrared and Raman spectral analysis but with superior properties due to the high intensity and coherence characteristics. The FEL based THz/IR methods have superiority among the other types of IR emission and detection techniques [70, 71]. The output characteristics are MW range pulse power, pulse durations in the order of picoseconds (10–1,000 optical cycles), up to few GHz of micro pulse repetition rates, and the continuous and fast tuning over a wide range of wavelengths combined with coherent radiation properties. Infrared and THz radiation is of great interest for various applications and for studying fundamental mechanisms [62–66]. Just to name, some examples are: strongly correlating quantum systems, magnetic states in solids, complex fluids, bio-fluids, nanoscopic structures, membrane proteins, bio-molecules, functioning dynamics in bio-systems, materials chemistry, electronic transitions in semiconductors, including interband dynamics, phonon scattering and inter-sub band dynamics in quantum wells, inter-sublevel spectroscopy and polaron dynamics in quantum dots, polaron dynamics, coherent dynamics, spin dynamics, impurities in semiconductors, vibrations in crystalline solids and amorphous materials covering the local modes in insulators, vibrational modes of hydrogen-like atoms and molecules.

FEL assisted radiation sources are gaining extreme importance for novel exploration and fundamental understanding of material structures through photon-matter interactions. Tunable wavelength and coherent radiation are generated by using relativistic electron bunches directed through undulators. The beam packages produced by FELs are compressible to few fs pulses allowing to study the minute interactions and ultra-fast dynamics of different materials. A number of significant works across a diverse range of subject areas have been conducted within the last few years using pump-probe experiments at X-ray FEL facilities. The combination of IR/THz and X-ray pulses allows to study the temporal evolution of different complex atomic and molecular systems. THz/FIR region of spectrum is of extreme importance for structural biology, condensed matter, and molecular dynamics, yet it cannot be separated away from X-ray, extreme UV and optical spectrum studies.

In this section devoted to application we will first give to the readers an overall picture in terms of the fundamental and material characterization studies done by FEL-assisted emission sources, covering the subject presented above. In the second part we will present a brief review on low band THz applications covering the biomedical and life science. Lastly, as a recent and novel approach, we will give a glance to the polariton chemistry, which combines the bio-medical and chemical structures of materials of biological interest with the fundamental physics and chemistry. We will focus on the mechanisms and possible future applications of this promising technique.

Fundamental Studies and Condensed Matter Applications

The inter-band processes occurring within the conduction band are more sensitive in the FIR frequency region. While the inter-sub-band transitions above Longitudinal Optical (LO) phonon frequencies have their sub-picosecond relaxation in the Mid-Infrared (MIR), relaxation mechanisms below this frequency can be observed in the THz range. The relaxation lifetime was first time measured based on the saturation detection utilizing a FEL source [67]. The transition selection rules are polarization sensitive. To this aspect the polarization sensitiveness of coherent THz radiation becomes an important factor for identification of the electronic transitions in semiconductors and quantum dots. Inter-sublevel transitions within confined states, transitions between singlet and triplet states, and composite states transitions (polarons) all occur in the MIR/FIR regime of the spectrum. The high intensity and tunability for FIR/VIS wavelength double resonance capability of FEL assisted IR/THz measurements of inter-sub-level transitions allow to characterize the electronic structure of the materials. Spin polarized electron dynamics in semiconductor spin and spin-split electronic structures are often studied at MIR/FIR regime of the spectrum. For this reason, the pump and probe measurements with circular polarization at MIR/FIR wavelengths are prone to many applications and fundamental studies. Many of the major FEL facilities such as FELIX, JFEL, FELBE, CLIO provide FIR output with simultaneous coverage of MIR (around 3–250 μm in wavelength) [68]. The wide wavelength tuning makes it possible to achieve resonant pumping of discrete transitions and multi-wavelength spectroscopy. There are many intrinsic properties and phenomena occurring in this frequency regime and enabling time-resolved spectroscopy of weak absorptions and non-linear optical processes. Combined pump-probe experimental stations at the European XFEL are another good example for the powerful, coherent IR/THz radiation related research tool where the time structure is synchronized with X-ray FEL. The undulator generated radiation can reach up to 30 MeV of energies. The SASE-FEL mechanism provides a sensitive polarization control with wide band tunability (0.3 THz - 30 THz) with few hundreds of μJ pulse energies and up to 100 MW peak power. The future generation of FELs is prone to combine THz with VIS-UV and X-rays, together with complementary techniques such as strong magnetic fields and near-field techniques. The rapid tunability together with the high photon flux character of FEL beams allows to study the weak absorption by using action spectroscopy methodologies based on the detection of different chemical and physical processes occurring, such as the change in particle mass through fragmentation and/or disassociation, change in ionization state through photoconduction, and photoemission. Some examples of this type of specialized experimentation techniques are the ion-trap system at FELIX, the sum-frequency generation set-up at CLIO and the near-field optical microscopy at FELBE. Moreover, the spin dynamics can be extracted by using a circularly polarized variant of pump-probe methods.

The very high intensity beam characteristics of FELs made it possible to experimentally investigate advanced nonlinear optical dynamics such as the characterization into relaxation rates of electronic transitions in semiconductors by bleaching, exploring the heavy ion formations on surfaces by the frequency-sum generation and multi-photon absorption and the Franz–Keldysh type of strong-field effects [69, 70]. Having ultra-short pulse duration, FEL generated radiation enables to observe very fast decays like the electronic lifetimes of low energy excitations. Infrared-active vibrational transitions are of extreme importance to investigate energy distribution of phonon modes within the material. The excited density starting from the bleaching condition can be probed when the ground and excited state populations reach the near saturation by ultrafast high field pump-probe experiments.

Low energy transitions in semiconductors in relation with the electron dynamics are of great interest not only for fundamental physics but also for the development of novel optoelectronic devices. Notable phenomena include band-to-band transitions and transport confinement in semiconductors for quantum cascading, impurities, and spin polarized excitations in solids for the manipulation of the polarization states of matter. Understanding the electronic structures under extreme magnetic fields by using the cyclotron resonances are important for a deeper investigation into new materials, such as semiconductor nitrides and magnetic semiconductors for spintronic applications.

There have been many reports investigating the relaxation rate properties of various advanced materials such as HgCdTe [71, 72], PbSe [73, 74], InSbN and hetero-structures of InAs/GaSb [75] in pump–probe experiments. The phonon dynamics and electron-phonon interactions are studied by monitoring the absorption and decay processes [76]. High intensity pump-probe measurements on the GaAs/AlGaAs quantum wells at low temperature and on Si/SiGe super-lattices have shown the fundamentals behind the inter-sub-band relaxation mechanisms and their dynamics under temperature changes. There have been successful attempts of lifetime measurements by using novel active device structures which combines quantum well infrared photodetector technique within a waveguide structure to determine the transition energy by voltage modulated waveguide transmission [77]. The FEL source at CLIO is one of the pioneers in the field. Some cornerstone studies have been conducted on GaAs/AlGaAs coupled quantum wells, with four bound state sub-bands observed under two color excitations and the gain measurement performed with time-resolved pump–probe experiments by the FEL [78].

Very complex dynamics such as Franz–Keldysh effect between electron and hole pairs under a strong driving field, electron tunneling through the electrostatic confining potential at low frequencies, ionization by multiphoton absorption, high harmonic generation at mid-range frequencies, resonance enhancement and second order susceptibility dispersion leading to second harmonic generation at THz frequencies [79] are observed in the presence of intense FEL assisted THz electric fields [69, 80, 81]. Moreover, side-bands of the quantum confined states [82–85] can be triggered under strong THz electric fields revealing new virtual states [86–88], frequency doubling and tripling [89, 90], characterization of depolarization shift [91]. Using synchronized pulses with a two-color scheme, time-resolved spectroscopy of internal dynamics of excitons is allowed by tuning the THz frequencies at the transition frequencies.

Double resonance spectroscopy correlates the inter-band and inter-sublevel transition energies [92]. One can use the mapping of the double resonance FIR modulated photoluminescence spectroscopy for distinguishing the mechanisms from dot to dot in correlation with the size and composition distributions [93, 94]. The non-linear optics of intra-band transitions including the third-harmonic generation [95, 96] is associated with resonant intra-valence band transitions, second harmonic generation [97, 98] and two-photon absorption [99, 100]. Polaron dynamics [101, 102] has been studied with the absorption saturation technique [103] and pump–probe spectroscopy [104–107]. Spin mixing effects and spin-lattice coupling have been studied [108] using a THz probe to improve the fundamental understanding of magneto-transport mechanisms in metals [109].

Since optical transitions are in the FIR region, the very short coherent pulses attained using FELs are required for experiments. The high power and coherent pulsed characteristics make it possible to investigate the coherent dynamics of matter such as Rabi oscillations and more complex time-resolved, coherent, degenerate four wave mixing effects. In such a way damped optical Rabi oscillations of polarons have been observed [110]. Spin polarized electron dynamics in semiconductors spin and spin-split electronic structures are often studied at MIR/FIR regime of the spectrum. The inter-band non-radiative and spin lifetimes and the inter-band transmission and circularly polarized intra-band transitions dynamics can be probed by FIR FEL pump-and-probe experiments [111].

Mechanisms driven by intense THz electromagnetic fields in spin systems recently gained great interest in modern spintronics as a novel way to obtain ultrafast control in macroscopic magnetization. Observation on the reconfiguration dynamics of magnetic domain structures under high field THz FEL pulses by Kurihara *et al.* [112] has proven the possible use of realizing thermal spin effects at this frequency range. To irradiate the ferromagnetic domains of single crystal $ErFeO_3$, authors use intense (10 mJ/pulse) THz pulses generated by the FEL. Findings are that the domain mechanisms near the boundaries can be locally reconfigured under the THz–FEL pulse irradiation.

In another experiment, Gauthier *et al.* [113] used a chirped pulse amplification technique for the compression of free electron laser pulses in the extreme UV range. This work has proven the importance of FEL techniques to explore fast electron dynamics. Usenko *et al.* [114] demonstrated the use of attosecond interferometric autocorrelation technique by using the self-amplified spontaneous emission of FEL to gain insights of phase-controlled pulses. De Ninno *et al.* [115] implemented an interferometric method in order to measure and control the spectro-temporal characters of single shot ultrashort pulses seeded by a femtosecond laser.

A novel technique to achieve high intensity two-color FEL pulses and probe ultrafast dynamics by using twin electron bunches has been proposed in the optical range at SPARC [116, 117] and then implemented in the X-ray range at LCLS [118]. A similar electron beam with two beamlets at relatively low energy (50–80 MeV) can be used in an Inverse Compton experiment to generate two color X-rays [119]. At BriXSinO, two color X-rays radiation can be obtained by passing the electron beam in a two Fabry-Peròt cavity Inverse Compton source [120]. An all-optical synchronization of the soft X-ray FEL pulses with ultra-fast rates less than 30 fs was achieved by Schulz *et al.* at FLASH [121]. Ferrari *et al.* [122] has demonstrated a novel configuration technique to achieve elemental selectivity by use of a two-color seeded FEL source in an independently tunable resonant-pump resonant-probe to specific electronic excitations. The origin of incipient ferroelectricity in lead telluride, correlated with coupling of band-edge electrons and phonons revealed by ultrafast X-ray scattering, was reported by Jiang *et al.* [123]. Gerber *et al.* directly studied the photoinduced lattice dynamics in $BaFe_2As_2$ by measuring the rapid lattice oscillations which can alter the electronic and magnetic properties of the material [124]. Symmetry breakdown of electron emission in photoionization of argon atoms and ions was demonstrated by Ilchen *et al.* [125].

MOLECULAR DYNAMICS, STRUCTURAL BIOLOGY AND BIO-MEDICAL APPLICATIONS

During the years, THz spectroscopy proved to be a very useful and successful tool for the investigation of many processes in pharmaceuticals, biology, and bio-medicine, since it probes low frequency vibrational modes [126–129]. These vibrations define large scale conformational changes occurring along the torsional degrees of freedom in complex biopolymers [130–132]. Moreover, the fingerprints of many materials of biological interest lie in the THz frequency band. THz related energies correspond to intermolecular hydrogen bonds and are therefore an ideal probe for the investigation of the binding of water and hydration levels in large biomolecules. THz spectroscopy techniques are also widely used for the real-time detection of protein–water dynamics upon protein folding and for the study of the water surrounding biomolecules, including carbohydrates [133–136], small peptides [137–140] and DNA oligomers [141–144].

There has been an intensive work done in the literature for pharmaceutical material characterization [145–149], for monitoring intermolecular interactions in pharmaceutical formulation [150–152], and for investigating drug delivery mechanisms. The interested reader may find a detailed list of terahertz spectral signatures of crystalline pharmaceuticals of major interest summarized by Shen [149].

THz spectroscopy in both frequency and time domain have been also applied for tracing gases. THz spectral signatures for example in hydrogen cyanide (HCN) [153, 154], carbon monoxide (CO) [155, 156], formaldehyde (H_2CO) [157, 158], and water (H_2O) [159, 160] allow to identify and quantify the individual gases from the pure rotational transitions.

Since THz waves are non-ionizing, they represent a suitable option for noninvasive spectroscopy, imaging [161, 162] and *in-situ* monitoring [163, 164]. Since THz radiation is highly sensitive to polar molecules, it has been successfully used to determine the hydration levels in skin tissue [159], both *in vitro* and *in-vivo* [161, 165], in order to discern between tumoral and healthy tissues for cancer diagnosis [166–168]. In many bio-applications, a key factor is that the THz radiation is strongly reflected by conductive materials and strongly absorbed by polar liquids, on the contrary the THz waves can penetrate a large number of organic or inorganic materials which are opaque in visible or IR ranges. For this reason, THz spectroscopy and imaging techniques are ideal candidate for the detection of concealed (multilayered) materials and tissues.

Even if the given examples on imaging and spectroscopy show the great potential of the investigation in the THz range, the major part refers to works done in the low frequency range (<6 THz) and at low energy level (μJ) [169]. Many of the complex molecular investigations were complemented with the integrated analysis of THz time domain response, yielding structural information together with classical IR spectroscopy [170, 171]. In this context, the FEL-assisted THz sources covering both bands of the spectrum will throw aside the technical difficulties faced with infrared and Raman spectroscopy. Moreover, IR/THz pump-probe experiments satisfying wide range tunability with high peak powers, high spectral and temporal resolution, polarization control and the possibility of synchronization with the X-ray pulses will allow to study complex materials on atomic and molecular time scales [172–174].

Spectral Analysis of Biomolecular Aggregates

Self-assembly of peptides and proteins have important role in bio-function. Similarly, the dissolution method of peptide fibril is potentially useful in medical fields, as a modification technique of cell structure and as a fabrication tool of biomaterials. Using a novel approach Kawasaki *et al.* [175] recently proposed a physical engineering technology based on a THz FEL. Using microscopy analysis, authors observed a remarkable reduction in the β-sheet with increasing α-helix of calcitonin peptide fibril irradiated by the FIR-FEL, with respect to the MIR-FEL. Their findings show that intense terahertz waves can dissociate fibrous conformation of peptide with little influence of thermal effects, proving that high field FIR-FEL irradiation can be used as an innovative processing tool in medical research.

Spectral Analysis at Strong Coupling Regime

In the past few years, strong coupling between electronic molecular transitions and photonic structures or in other words between organic molecules and confined light modes has been shown to modify the electronic landscape of the molecules, changing their physical and chemical properties, and affecting the chemical reaction rates, electronic and excitonic transport properties. The so called polaritonic chemistry, with its promising potential to tailor chemical structure and reactions through the hybrid light–matter states, has recently emerged as an exciting new research field. This phenomenon was observed in various types of materials, such as cold atoms [176–178], excitons in semiconductors [179–181], phonons in inorganic crystals [182, 183], organic molecules [184–186], nanocrystals [187].

Squibb *et al.* reported the acetylacetone photodynamics at femtosecond regime by using a seeded FEL source in a photoexcitation and photoemission scheme [188]. Their work has given a better understanding of the earlier stages of the photochemical processes involved on extremely short timescales. Perakis *et al.* revealed the highly complex water dynamics and the influence of cage effects on ultrafast phenomena by sub-100 fs measurements conducted by FEL sources [189]. The structures of peptide fragments formed during electron transfer dissociation was reported by Martens *et al.* by using FEL-based IR spectroscopy techniques [140]. The demonstrated technique was used for sequencing peptides and proteins by fragmentation. Levantino *et al.* showed that it is possible to track the motion of myoglobin, observing the breakage of a bond between the protein and a ligand occurring within picoseconds of time interval during the photoinduced ligand release [190]. Nogly *et al.* demonstrated a novel approach for the study of the complete photocycle dynamics of retinal proteins, reporting time-resolved experiments for the structural identification of the light-driven proton pump bacteriorhodopsin [191]. In a work by Grünbein *et al.*, the possibility of MHz rate high quality data acquisition was shown, allowing the noninvasive characterization of lysozyme crystals from proteins [192].

It is important to note that the strong coupling with inter-molecular vibrations occurring in organic materials is in the low terahertz region [193], whereas many of the fundamental excitations and collective modes in solids such as the free carrier Drude response, crystal lattice vibrations, charge density waves, transition gaps in superconductors, magnetic excitations, surface plasmon and phonon polaritons have characteristic energies in the FIR range. Analogously, FIR provides fingerprints of molecule specific vibrational modes such as macromolecules and polymer chains in many soft and biological molecular materials [203–206].

More recent works refer to different kinds of polymers, molecules in liquid phases, organic crystals and organic liquid crystals in various geometries, nanowires, nanographene, proteins, light-harvesting complexes, and photosynthetic bacteria hybridization and many more. Interested readers may find the cited works on strong coupling observed in exotic materials listed above in the short review given by Hertzog *et al.* [194]. Moreover, a thorough review on polaritonic devices, with a comparison between organic and inorganic materials, is given by Sanvitto *et al.* [195], that includes polariton lasing [196], bosonic condensation [197], Bose–Einstein condensation [198], nonlinear interactions [199] and optical response [200].

IMAGING APPLICATIONS

Accelerator-based radiation plays a major role in THz imaging applications, being capable to couple high spatial and temporal resolution with highly sensitive chemical state analyses, which are of major importance in bio-medical research on molecular dynamics in the living environment. Tunable, narrow band FEL sources allow the investigation of the sub-molecular aspects of biochemistry with superior analytical performance to investigate hard tissues even down to a single cell. In fact, most bio-processes are in the range of FEL pulse lengths, and time-resolved data on vibrational modes can be obtained [105, 201].

Ion beam microprobes such as particle-induced X-ray emission, Rutherford Backscattering Spectrometry (RBS), Particle-Induced Gamma-ray Emission (PIGE), or Nuclear Reaction Analysis (NRA), Scanning transmission ion microscopy, X-Ray Fluorescence (XRF) and X-ray Absorption Spectroscopy (XAS) at microscopic levels, X-ray diffraction microscopy, mid-infrared microscopy (FTIR) and THz radiation are used in a complementary manner to achieve the needs for imaging and spectroscopical investigations of bio-matter.

Single cell imaging of live cyanobacteria by using X-ray FEL source was demonstrated by van der Schot *et al.* The authors have recorded signals from free-flying cell samples with 4 nm resolution [202, 203]. Schneider *et al.* has successfully shown the possibility of *in situ* single-shot diffractive fluence mapping by using integrated gratings to reduce the uncertainty in the laser beam profile characterization [204]. Leshem *et al.* has reported the full image reconstruction by a direct single-shot phase retrieval acquired from the diffraction patterns [205]. This work proves that it is possible to obtain coherent diffractive imaging of non-crystalline objects such as single molecules.

Polarization Sensitive Imaging

Pakluea *et al.* [206] proved a new modality of THz imaging for polarization sensitive materials by using coherent THz transition radiation generated with short electron bunches of around 8 MeV from a 45° tilted aluminum target. Later, the radiation was used to create images with a reflection setup. The authors carried out a full characterization of backward transition radiation properties, including spatial distribution, polarization, and spectral radiation. Moreover, they made THz imaging of the flow channels plate of proton exchange membrane fuel by mapping separately the radiation of p- and s-polarization components. The result of their study is that polarized transition radiation can be used to improve the THz imaging contrast and realize a better THz image, with p-polarization providing the highest reflectance contrast.

Near-Field Nanoscopy and Scanning Near-Field Optical Microscopy

Nanoscopy and IR scattering-type Scanning Near-field Optical Microscopy (s-SNOM) provides imaging and spectroscopy at nanometer scales. These techniques can be adapted to cover a broad electromagnetic spectrum of NIR, MIR, FIR and THz frequencies. With recent advances in technology, they can be possibly implemented to low temperature and ultrafast regime studies.

As explained in detail previously, many molecular specific activities, electron, and lattice dynamics, and plasmonic and polaritonic effects in quantum matter lie within the THz and FIR regime of the spectrum. Up to date, s-SNOM detection spectral band was mainly limited in between the NIR and MIR regimes and/or low THz window, and it was not accessible through THz/FIR frequencies. Recently, a successful adaptation of an ultrabroadband FIR s-SNOM nanoimaging and spectroscopy has been demonstrated by Khatib *et al.* [207]. The authors have combined an IR synchrotron radiation with a novel low-noise fast modulating Cu-doped Ge photoconductor. Using this approach, they exceed conventional limits by achieving ultrahigh spatial resolution (30 nm) at wavelengths up to the THz/FIR spectral window.

PROGRAMMATIC USE OF THE BriXSinO TERAHERTZ/INFRARED BEAMLINE

In the previous sections we have given a comprehensive survey of the main FEL assisted terahertz sources and their applications. Even if up to date FEL sources have speed up the scientific exploration, the goal of more intense sources has not been achieved yet. The BriXSinO project is expected to fulfill this demand. In the following we present a perspective on the potential use of the Milan facility, including a vision on some possible applications that may come true soon in a programmatic manner.

Non-Linear Physics Using the BriXSinO Source

In the recent literature, the possibility of combining near-infrared optical pulses and intense, sub-ps, broadband terahertz pulses to generate a THz-optical four-wave mixing in the investigated material has been successfully demonstrated by Rubano *et al.* [208]. In this novel technique the frequency spectrum of the generated signal was observed to display two intense sidebands close to the optical second harmonic central frequency $2\omega_L$, where ω_L is the optical central frequency of the near-infrared fundamental beam. The two sidebands are peaked around the central frequencies $\omega_{s,a} = 2\omega_L \pm \omega_T$, where ω_T is the THz central frequency. These two frequencies are analogous to the Stokes and anti-Stokes components appearing in standard hyper-Raman scattering, leading to a coherent THz Hyper-Raman (THYR) effect.

Despite the resolution in the frequency domain being poor due to the large bandwidth of the initial pulses, yet it is possible to observe the strong oscillations following the time domain evolution of the THz signal. The latter are due to the resonant coupling of the THz pulses with the proper frequencies of the material. It was demonstrated in crystals that the THYR signal carries information on a large variety of low-energy excitations including polaritons and phonons far from the Γ-point.

This phenomenon was first time observed by means of a table-top laser-based THz sources, thus making the observation of this nonlinear optical effect challenging due to the limited intensity per pulse. The high intensity and wide band tunability offered by the THz BriXSinO source will allow to extend this novel technique for more advanced material investigation and reveal new concepts in the nonlinear physics. Moreover, recent studies predict new nonlinear hyper-Rayleigh and hyper-Raman scattering mechanisms, which are specific of the interaction with chiral molecules by use of twisted photons [209]. This can be done adding a helical phase to the beams participating to the Hyper-Raman process, making this new technique very appealing for bio-sensing and microscopy of chiral biological samples.

Imaging Using the BriXSinO Terahertz/Infrared Source

Imaging using intense THz sources can be applied in many processes involving the photoelastic effect. The effect can be essentially described in terms of a permanent or transient birefringence induced in the material by the action of some internal/external force. All dielectric media in principle can show photoelasticity, but besides the study of the fundamental physics of this effect, it was predominantly used as a tool for studying and experimentally mapping the forces/stresses applied into a given material. This is particularly important and useful around discontinuities in the material, where those forces are strong and the material can have weak points (critical stress points), and in general for irregular geometries, where calculations can be cumbersome and less reliable. Naturally, the material under investigation must be, at least partially, transparent to visible light, and indeed this technique has been applied successfully for many years on studying relaxations/deformations dynamics in glasses and plastics.

In the case of a material which is not transparent to visible wavelengths, one immediate solution can be found by changing the wavelength itself. This is not always easy or straightforward as it may seem, because one should match the very high technological level which has been reached on visible light (both in terms of sources, manipulation, and detectors). Anyway, modern technologies, developed enough to make this technique possible, are available today. THz waves seem particularly interesting in this context, especially when powerful sources such as BriXSinO will be developed. The photoelastic effect requires a crossed polarizers configuration. Only a small portion of light, whose polarization has been affected by the material birefringence, will be transmitted, and therefore strong sources (and sensitive detectors) are key to make the experiment practically possible. One intrinsic feature of the THz wave is its high transitivity through optically opaque materials, making it a suitable radiation range to investigate the internal properties of these materials. Today, the research field of THz photoelasticity is rapidly growing, but still relatively unexplored. The proven ability of THz radiation to distinguish between different kinds of human bone tissue could be extremely beneficial to study the osseointegration process in real conditions.

The research proposal is to implement in the BriXSinO facility a THz photoelastic setup, with the aim of studying dental implants in animal (and, in a future, human) bones. The implants can be studied at first on dead animals (no osseointegration), then in dead animals which have been implanted a given time before death (study as a function of the time of osseointegration process) and finally they can be measured *in-vivo*, which is a crucial step to achieve the possibility to study real human implants. This last step presents some difficulties, due to the large amount of soft tissue surrounding the bone, which could absorb the THz radiation to a level in which the experiment is not feasible anymore. In order to circumvent this problem, the only solution is to increase the source power (that is why the BriXSinO source is an inevitable choice), to increase the sensor sensitivity and to search for wavelengths which are less affected by absorption. Once the experimental procedure will be validated, it will be possible to study different clinical protocols: delayed or immediate load, with all possible variations among those two cases (different healing times/drugs in the former, different load conditions in the latter, the number of supports for each implant, and so on). This approach will be extremely beneficial for the whole community working in this area.

Hybrid Approaches and Metamaterial Research Using the BriXSinO Terahertz/Infrared Source

Small Angle X-ray Scattering (SAXS) using a THz transparent 3D printed microfluidic cell was first presented by Schewa *et al.* [210]. X-ray studies reveal THz-induced, nonthermal changes in the structure of crystallized proteins, where the excitations of large molecules are associated with THz and FIR radiations. Using a microfluidic cell for the experiments, authors combined SAXS with an external THz field to determine the collective vibrational modes. Even if the authors have successfully demonstrated the main concept, the idea halted because of the lack of high field and more broadband THz sources. TerRa@ BriXSinO can be the solution.

Photonic metamaterials are another concept of main interest, where tailored micro-structured subwavelength building blocks allow the light manipulation and the development of unconventional electro optical properties such as unnatural, metal level refractive index, zero reflection, perfect absorption and enhanced nonlinear phenomena. Main future advances are expected to focus on the possible applications into ultra-high-resolution imaging beyond the diffraction limit and in polarization based devices [211].

One of the major future perspectives for THz science and technology is focused on developing rapid, label-free, and cost-effective sensing applications for bacteriology and virology Research and analysis. THz spectroscopy gained extreme importance for the fast racing and qualification is needed including the morphological alteration monitoring of cells. All these innovative works show the importance of THz bio-matter interaction in the near future. The interested readers may find a full comparison of THz spectroscopy with common bacterial detection methods from the review by X. Yang *et al.* [212]. Main challenge however is the limited sensitivity, due to the THz wavelength being far larger than cells/viruses size (a typical bacterial cell is on the order of ~λ/100 with respect to THz range), however it can be resolved by engineering suitable metamaterials with an improved coupling in between the living matter and THz radiation [213, 214]. In any case of these scenarios the design structure and/or the organisms under investigation stems from the highly absorptive nature of water and or the construction materials of the meta-devices where the strong attenuation of THz radiation starts playing the main role in the detection precision. Given these limitations for standard TDS, the BriXSinO THz/IR Source with high intensity and tunability characteristics will be a complementary technique not only for research on bio-systems (where water absorbance becomes a major problem) but also for sensing applications where metamaterial-based THz devices are involved.

CONCLUSION

In this article, we gave a comprehensive survey of the main FEL-assisted terahertz sources worldwide and presented the main characteristics of a future THz/FIR source based on a superconducting linear accelerator. The source is constituted by a FEL oscillator, alimented by a 100 MHz electron beam of 22–40 MeV of energy and 50–200 pC of charge. The wavelength of the radiation is between 10 and 60 μm with a peak of efficiency around 20 μm. Intra-cavity single shot radiation energy of 355 mJ (meaning 17.7 μJ of extra-cavity energy at 100 MHz and 1.77 kW of average output power) at $\lambda = 20$ μm has been estimated. The source is strongly monochromatic, with a bandwidth of 1% or smaller, highly coherent both transversally and longitudinally, with extreme versatility and tunability.

After the schematic description of the TerRa@BriXSinO THz/FIR source, the paper is devoted to the analysis of the potential use of the Milan facility, including a vision on some possible applications that may come true soon in a programmatic manner. We present different scenarios, ranging from non-linear physics under extreme conditions to polarization sensitive imaging and metamaterial-based sensing, where the TerRa source can be successfully applied.

In a near future, FELs will speed up scientific explorations and open new perspectives in many scientific different fields, from condensed matter to biology and life science, with a need for more and more intense and highly monochromatic beams, and the envisaged facility can have a strong role in fulfilling this demand.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

Writing—original draft CK; Writing—review & editing AA and VP. All authors have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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