Final report on NKFI project #125808, entitled

Terahertz-Driven Electron Acceleration

1. Introduction

Electron beams at various energy levels from keV to GeV are fundamental tools for science and technology. For example, ultrashort electron bunches with a narrow energy spread, high charge (for some cases), and low jitter are needed for electron spectroscopy, for resolving state transitions in atoms and ions, for ultrafast electron diffraction measurements, and as a seed, for x-ray free-electron lasers.

The achievable accelerating gradient determines the performance, cost, size, and availability of electron guns and accelerators. In an electron gun, the electrons need to be accelerated from rest to high energies as quickly as possible to avoid the beam- and pulse-degrading effects of space charge. Due to plasma breakdown mechanisms on the walls of metal cavities, microwave guns have surface field limitations of around 200 MV/m. Conventional high-power microwave accelerating structures operate with 30–50 MeV/m gradients. For this reason, microwave electron accelerators with tens-of-MeV energy or beyond are complex, large-scale facilities with extremely high construction and operation costs. A further drawback of microwave technology is the difficult synchronization.

Since the plasma breakdown threshold is proportional to the root of the frequency and is inversely proportional to the fourth root of the pulse duration, using higher frequencies and short driving pulse durations are advantageous. The acceleration of electrons in dielectric grating structures using laser pulses was proposed [Plettner-2006], and the working of such a miniature accelerator was demonstrated recently [Peralta-2013]. However, the short optical wavelength seriously limits the bunch charge and requires sub-micron electron beam sizes and sub-femtosecond timing precision. Furthermore, the period of the optical pulse is shorter than the typical duration of a (compressed) electron bunch of about 100 fs. We recognized ten years ago that the terahertz (THz) frequency range could be superior to radio-frequency and optical radiation for electron acceleration and manipulation [Hebling-2011]. THz radiation, with a wavelength of two orders of magnitude longer than in the optical range, is ideally suited for such applications, enabling the significant increase of both the interaction length and the bunch charge. Furthermore, much better synchronization can be achieved between the accelerating THz field and the particles due to the picosecond-long oscillation cycle.

The technology of optical rectification of femtosecond pulses in $LiNbO_3$ (LN) using tilted-pulse-front pumping (TPFP) has been proposed and developed by our group [Hebling-2002]. The development of such intense THz sources enabled the generation of THz pulses with 100 MV/m scale field strength in the low-frequency (0.1–2 THz) part of the THz spectral range [Fülöp-2014].

Our collaborating partners demonstrated THz-driven electron acceleration first in a proof-of-principle experiment (the group of Prof. Kärtner at DESY) [Nanni-2015]. Based on the experimental results achieved with a small scale LN THz source, GeV/m accelerating gradient and electron energies on the 10 MeV scale was predicted for energetic THz driving pulses.

In our project, two bunches of research directions were assigned: 1., Proposing and investigating new THz driven electron guns and accelerators. Furthermore, a feasibility study on THz-driven proton acceleration was also planned. 2., Development of high-field THz sources for driving electron guns and accelerators.

This report summarizes the research result according to these two bunches followed by a short sub-chapter on investigation the possibility of single-cycle attosecond pulse generation using high-field THz pulses, which theme was not planned originally.

2. Research results

2.1. THz-driven particle accelerators

2.1.1. THz-driven electron gun and accelerator

We have proposed and numerically investigated an setup for accelerating electrons to relativistic energies by counter-propagating focused THz pulses [Tibai-2018]. The schematic view of the arrangement is shown in Figure 1a. The two THz pulses have the same waveform and the same polarization direction. In this case, the magnetic fields of the two THz pulses have opposite directions, thereby minimizing magnetic deflection effects on electrons. Electrons are injected by a synchronized short laser pulse which ionizes the atoms in a desired small volume within a gas jet. The generated free electrons are then accelerated by the superposition of the electric fields of the two THz pulses.

Investigations were performed first for a single electron at different THz frequencies (0.14-3.0 THz) based on experimentally achieved or numerically calculated THz pulse parameters. The acceleration rate was optimized in each case by varying the phase of THz pulses and the θ angle between the propagation direction of the electron and the THz pulses. We showed that the optimal THz propagation angle is higher at higher electron velocity. Simulations were carried out with realistic electron bunches as well, with different bunch sizes. It was supposed that a THz pulse pair is focused to a gas jet, where the electrons are generated, and a high power fourth harmonic beam of a 1 μm wavelength laser is focused to the gas jet, where it ionizes the krypton atoms, resulting in electron bunch with 0.5 eV average energy.

Post-acceleration of accelerated electrons is also feasible by focusing further, properly synchronized THz pulses along the path of the electrons. In the double-stage acceleration setup (in an early version of the setup shown in Fig. 2a), the electrons accelerated by a first THz pulse pair are further accelerated by a second pulse pair comprising the 2nd acceleration stage. According to the calculation, the final electron energies of 142 keV can be achieved in the case of electron bunches with a 20 µm radius, using two THz pulses with 1 mJ energy per pulse.

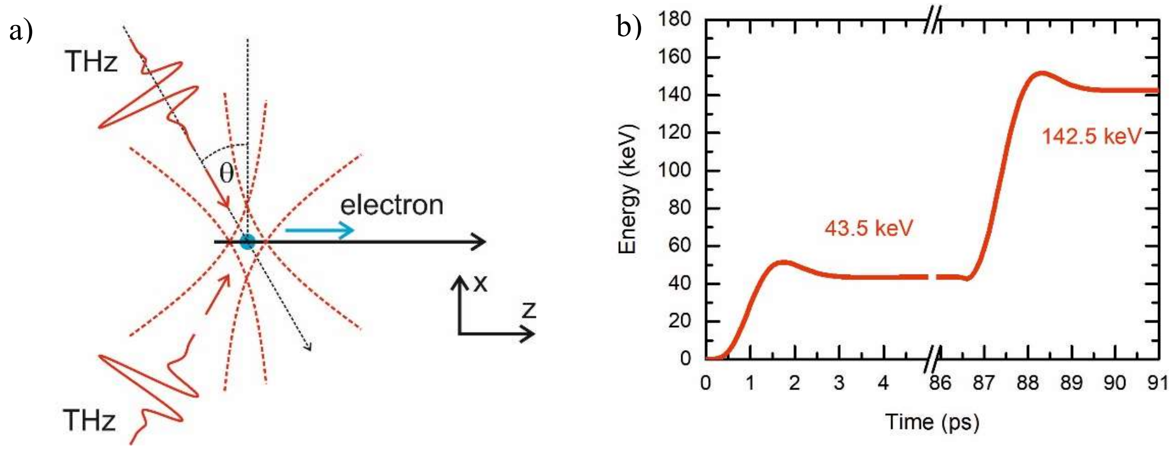

Figure 1: (a) Electron acceleration setup with two single-cycle THz pulses propagating in close to opposite directions at a beam tilt angle θ. (b) the energy evolution of the electrons in the double-staged THz driven accelerator.

We improved our previously mentioned setup by proposing the Compact THz-based Electron Acceleration (C-TEA) setup driven by 0.5 mJ THz energy per pulse. This setup, which is shown in Figure 2a, consists of 4 main parts (two acceleration stages (see red painted parts) and two bunch shaping stages (see yellow-painted parts), and the size of the whole system is 150 mm [Turnár-2021]. In the numerical investigation, four-four THz pulses with 0.3 THz mean frequency and 3.7 MV/cm peak field each were supposed in both acceleration stages. In the shaping stages, much weaker THz pulses (with 50 kV/cm peak field) were supposed. The electrons generated in a gas jet can be accelerated from rest up to 80 keV in the 1st acceleration stage, and they are post-accelerated to 350 keV in the 2nd acceleration stage. The extended transversal and longitudinal bunch size caused by space charge was reduced as much as possible using solenoids and bunch compression technique. The dependence of the energy spectrum of the bunch on the bunch charge has been investigated. The simulations predict the production of electron bunches with 20 fC charge, 80 keV energy, 49 nm rad transverse emittance, 1.0% relative energy spread from the electron gun (first stage). Utilizing electron beam spatial and temporal focusing by a magnet and bunch compression by THz pulses, post-acceleration boosts the electron energy to 346 keV, and the electron bunch has 600 nm rad transverse emittance, 2.0% relative energy spread, 200 fs duration, and 8 fC charge.

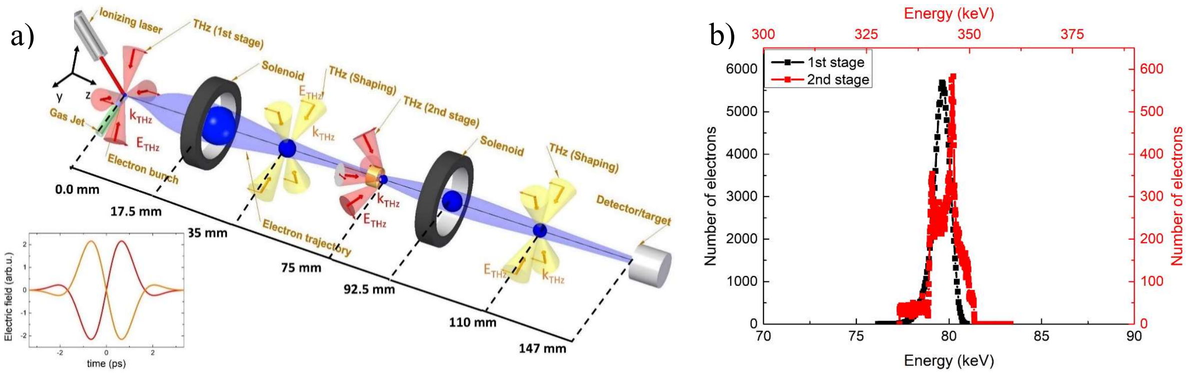

Figure 2: (a) Scheme of the compact THz-driven electron accelerator setup. (b) The energy spectra after the first (black curve) and after the second (red curve) acceleration stage.

According to very recent calculation, using 10 MV/cm level THz pulses in the first stage of the setup shown in Fig. 2, it is possible to generate relativistic electron bunches. Furthermore, when deuterium gas is ionized, the Coulomb explosion of the remaining ion bunch results deuterons with 100 keV level average energy. These could generate ultrashort neutron bunches (patent application 2021033001 WIPO (PCT)).

2.1.2. THz-driven proton post-accelerator

Hadron therapy motivates research on production proton beams with 70÷100 MeV energy and relative energy spread of about 1% by non-conventional accelerators. Laser-driven plasma accelerators produce proton beams with only a few tens-of-MeV energy and extremely broad spectra, hindering direct applications for hadron therapy [Haberberger-2012].

Recently, we proposed a method for post-acceleration and monochromatization of laser-produced proton bunches [Pálfalvi-2014], which utilizes the evanescent field of THz pulses in the vacuum gap between a pair of dielectric prisms (Fig. 3a). To improve the predictive power of our model, in this project, we performed numerical simulations taking into account the finite size of the original proton bunch. Supposing THz driving pulses with 1 MV/cm field strength, our simulations predict a monochromatization of the proton bunch with a relative energy spread of only 0.5% at 42 MeV average proton energy.

We also investigated the acceleration efficiency at higher electric fields. The predicted dependence of the energy gain on the peak electric field strength, together with a square-root fit, is shown in Fig 3b. According to our calculations with 10 MV/cm electric field strength, 7 MeV acceleration can be achieved over less than 2-cm distance [Tibai-2017].

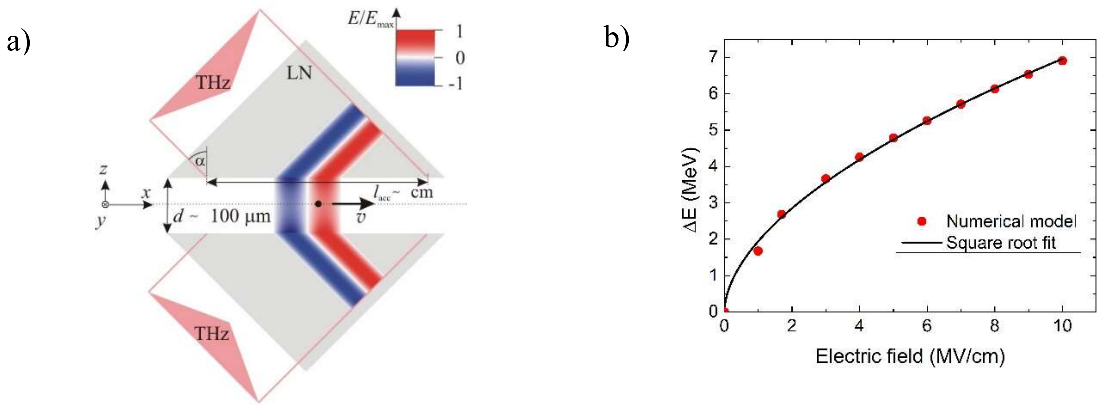

Figure 3: (a) Scheme of the THz evanescent-wave accelerator. (b) Dependence of the energy gain on the electric field.

Furthermore, increasing the proton bunch energy from 40 to 70 MeV can be expected in five such post-acceleration stages with 8 MV/cm field strength in each. The 70—MeV final proton energy can already be sufficient for applications in hadron therapy.

2.2. Development of high-field THz pulse sources

2.2.1. Development of single-cycle high-field THz pulse sources

Although the TPFP LN THz source [Hebling-2002] has become very popular and worldwide used, more than one decade ago, we recognized a few properties of the original (from now conventional) TPFP LN source, which limit its scalability in THz pulse energy or peak electric field strength. The limitation originates in the needed large pulse-front tilt and the needed large angular dispersion of the pump beam. This results in a large group-velocity-dispersion [Hebling-1996], and with it a short effective THz generation length in the LN. Another consequence of the large needed tilt angle is that a prism-shaped LN has to be used, resulting in inhomogeneous generated THz beams for large pump spot sizes.

In 2016 we proposed a hybrid-contact-grating setup, where the tilt angle needed for velocity matching is introduced in two steps [Pálfalvi-2016]. First, an optical grating – imaging system introduces a pre-tilt, about half of the required tilt. Second, the remaining tilt needed is introduced by diffraction of the pump beam on the contact grating (a transmission grating prepared on the entrance surface of the LN crystal. An important advantageous result of the hybrid solution is that the wedge angle of the LN prism can be about two times smaller than in the conventional TPFP setup. The hybrid-contact-grating setup and the "echelon" setup [Ofori-Okai-2016] inspired us to develop the nonlinear echelon slab (NLES) setup [Pálfalvi-2017].

This one is also a hybrid type setup: a transmission grating – imaging part for introducing a pre-tilt, and an NLES (see Fig. 4), a plane-parallel LN crystal with an echelon structure on its input surface. The NLES can be parallel if all three angles indicated in Fig. 4 are equal: $\gamma_0 = \gamma_{slab} = \gamma$, where γ is the needed pulse-front-tilt angle inside the LN to achieve velocity matching between the pump pulse and the generated THz radiation [Hebling-2002, Pálfalvi-2017]. Please notice that in the conventional TPFP setup, it is necessary to introduce a significantly larger pulse-front-tilt angle by the grating-imaging system! ($\tan(\gamma_{gi}) = n_{gr}\tan(\gamma)$, where $n_{gr} = 2.25$ is the group index of refraction of the LN crystal and γ_{gi} is the tilt-angle introduced by the grating – imaging system in the air just before the LN crystal.). Please notice that the NLES setup is the very first one that uses plan-parallel LN crystal! This is advantageous since this, together with the small needed pre-tilt angle, makes it possible to use as large as 40 mm diameter pump beams and (according to numerical simulations) generate THz pulses having a few mJ energy.

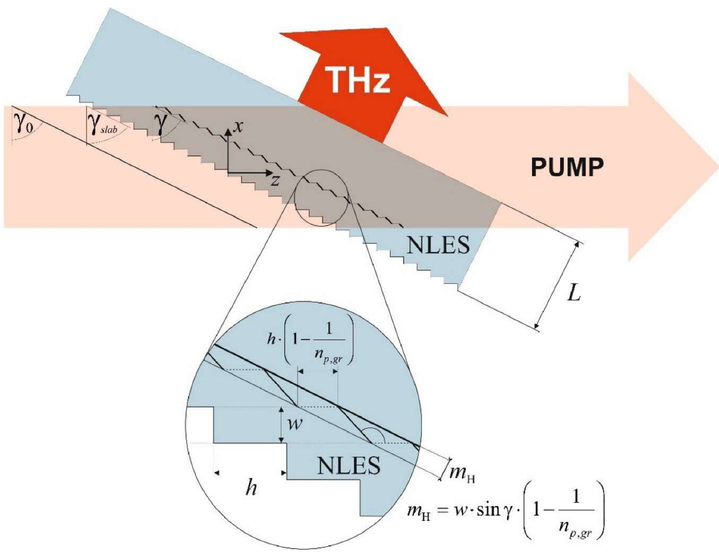

Figure 4: The scheme of the NLES based hybrid THz source.

We also proposed and numerically investigated a modified NLES setup, which does not contain imaging [Tóth-2019a]. In this case, if the transmission grating is used in Littrow arrangement to achieve high diffraction efficiency, the NLES has to be wedged, but the needed wedge angle (<10º) is much smaller than the required 63 º angle in the conventional TPFP setup. We have recently shown [Krizsán-2021a] that it is possible to use volume-phase-holographic-grating (VPHG) having appropriate slant angle, illuminating it in normal incidence so that the diffraction efficiency remains high and the NLES can be plan-parallel. In this case, a "straightforward" arrangement can be used.
Finally, we have also proposed and numerically investigated a reflective nonlinear slab (RNLS) THz source [Tóth-2019b]. It is made from a plane-parallel nonlinear crystal slab (e.g., LN) (see Fig. 5). The front (top) surface of the slab remains plane, but in its back (bottom) surface, a periodic structure is created (e.g., by diamond milling) to form a blazed reflective grating. The pump beam incident perpendicular to the top surface of the slab (see Fig. 5) does not generate practically any THz radiation when it propagates inside the LN towards the back surface since the velocity matching condition is not fulfilled. The period of the structured back surface and the slope of the surface pieces are chosen so that after reflection/diffraction, two beams with $\pm\gamma$ propagation angles will propagate. For these beams, the velocity matching is fulfilled, and THz radiation will be generated efficiently. The THz radiation propagates upward. Please notice that for the RNLS setup, there is no limitation for the usable beam size, and so on the pump and generated THz pulse energy. Numerical simulations predict the generation of THz pulses with 10 mJ energy when a 50 mm diameter LN RNLS is pumped by laser pulses having 1 J energy and 500 fs duration. The predicted temporal shape of the generated THz pulses is close to a single cycle. By focusing the THz beam, it is possible to achieve a 40 MV/cm electric field. Such pulses could be very attractive for THz driven electron acceleration using the setup described in [Turnár-2021].

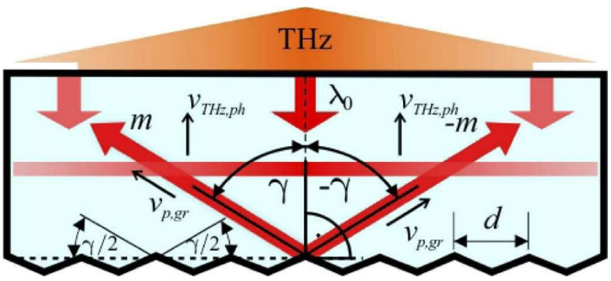

Figure 5: A schematic figure of the reflective nonlinear slab (RNLS) THz source. The red color indicates the pump pulse.

We have also proposed a practical RNLS setup [Krizsán-2020] which consists of a plane-parallel LN slab with smooth front- and back surfaces, a metal or dielectric grating, and an index-matching fluid between them. Furthermore, since the index of refraction of LN is large (2.15), we proposed to use a nanocomposite fluid consisting of GaP nanoparticles.

In proof-of-principle experiments [Nugraha-2019, Krizsán-2021b], we have demonstrated the working principle of the NLES and imaging-free NLES setups. The observed THz generation efficiencies were about an order of magnitude less than expected. For the imaging-free NLES setup, we proved that, in large part, the imperfect micromachining of the NLES (not smooth enough surface) is the reason for this [Krizsán-2021b]. We are working on the technical improvement of the micromachining and the accurate characterization of the NLES surfaces.

2.2.2. Development of multi-cycle high-field THz pulse sources

Just before the starting of this project, we successfully realized a monolithic (contact-grating) THz source in the semiconductor ZnTe [Fülöp-2016], which requires a much smaller tilt angle (than LN) when pumped at an infrared wavelength sufficiently long to suppress two- and three-photon pump absorption and the associated free-carrier absorption at THz frequencies. We demonstrated two orders of magnitude higher efficiency than any previous work with the same material. An alignment-free semiconductor source with outstanding beam quality can be advantageous for acceleration.

Both the semiconductor contact grating THz sources and the TPFP LN sources (including those that have been described in sub-chapter 2.2.1.) are suitable for multi-cycle pulse generation when pumped by laser pulse sequences or a pair of frequency-shifted pulses. We have shown [Tóth-2017] that using dual chirped optical parametric amplification (both the pump and the signal are chirped), periodic intensity-modulated pulses could be generated. Intensity-modulated pulses are achieved by the superposition of the amplified signal and idler (generated by the OPA-process) beams. Because both the signal and the idler are used, the efficiency is two times higher than the efficiency of a typical OPA. According to our calculation, using an efficient Yb pump laser with 1.03 µm central wavelength, an intensity-modulated pulse at ~ 2 µm central wavelength could be generated with near 50% energy conversion efficiency. The 2 µm wavelength is long enough to avoid three-photon absorption in ZnTe or GaP semiconductors, making a possible generation of multi-cycle terahertz pulses with high efficiency. Of course, it is also possible to pump LN THz source with such periodically modulated pulses to achieve energetic multi-cycle THz for particle acceleration.

2.3. Single-cycle attosecond pulse generation by Thomson scattering

We have numerically investigated the Thomson scattering of terahertz pulses on the ultrashort electron layer [Tóth-2018]. An electron beam with 34 MeV energy from a laser-plasma wakefield accelerator was assumed as an electron source. A laser-assisted bunching process generates ultrathin (~10 nm) electron layers are created in this electron beam. An extremely high energy (5 mJ) terahertz beam was focused on these electron layers in the numerical calculation. The scattered field, which goes opposite direction than the THz pulse, was calculated based on the Liénard-Wiechert potential. As a consequence of the process, the waveform of the THz pulse in the time domain is transferred to the waveform of the scattered radiation, as is shown in Fig. 6. As a result, approximately 1 nJ attosecond pulse energy was predicted.

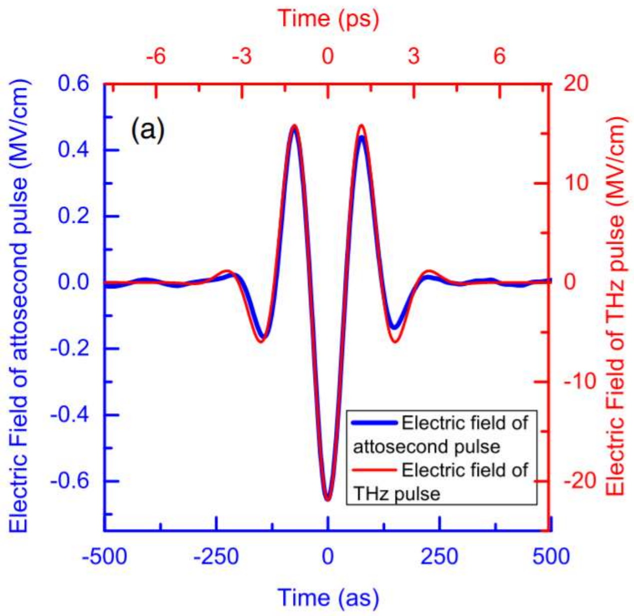

Figure 6. The electric field of the terahertz pulse (red) in the picosecond time scale and the scattered field (blue) in the attosecond time scale.

3. Summary of the research and publication results

In summary, we have achieved significant scientific results in both research directions: the development of THz pulse-driven particle accelerator concepts and development of high-field THz pulse sources devoted to driving particle accelerators. However, in both directions, we have performed less experimental work than we expected at the project's planning. In accelerator development, one reason for this was the more than expected difficulties of the experimental work. However, in both directions, the most significant troubles were caused by matters out of our control. The front-end laser of our laser system has broken down, and its reparation (because of the very long procurement procedure) took more than one year. Our other procurement suffered an even longer delay. We wanted to buy a high-energy laser and OPA for pumping the high-field LN and semiconductor THz sources, respectively, from a GINOP grant. The (first) procurement procedure started before starting the current project, but the contract was signed only this spring! The devices will be delivered to us in November, and after that, our experimental research can speed up.

Considering these awful circumstances, we regard our experimental achievement as more than expectable and the whole project very successful. The new generation (high-field) THz sources developed in this project, after technical development, very probably become worldwide used, similarly to our earlier invented conventional TPFP LN sources. We have published our achieved research results during the project in 13 refereed journal papers, and other three papers are in preparation (we have predicted 11-12 papers in our proposal). We have also filed five patents, from which two even have granted (see the list below). One important impact of the research result we achieved during the project is that we were invited by Christelle Bruni (CNRS Iréne Joliot-Curie Laboratory) to participate in an application for a Horizon-EIC-Pathfinder Open proposal: "THz Wave Accelerating Cavity for ultrafast science (TWAC), Proposal ID: 101046504.

4. Patent applications filed during the project period

Patent No.	Filed	Granted	Title	Inventors
10,481,468 USPTO	Jun. 25, 2017	Nov. 19, 2019	Method to generate terahertz radiation and terahertz radiation source	János HEBLING Gábor ALMÁSI László PÁLFALVI Levente TOKODI József A. FÜLÖP Csaba M. LOMBOSI
EP3493657A1	Apr. 06, 2018		Method and setup to produce relativistic electron bunches	János HEBLING Gábor ALMÁSI Zoltán TIBAI József A. FÜLÖP
10,747,086 USPTO	Aug. 6, 2018	Aug. 18, 2020	Method and setup to generate terahertz radiation scalable in energy	János HEBLING Gábor ALMÁSI László PÁLFALVI József A. FÜLÖP Gergő KRIZSÁN
2020188307 WIPO (PCT)	Oct. 31, 2018		Reflection- and/or diffraction-based method and setup to generate high-energy terahertz pulses	János HEBLING Gábor ALMÁSI László PÁLFALVI György TÓTH
2021033001 WIPO (PCT)	May 29, 2019		Efficient production of high-energy ultrashort ion-especially proton bunches	Gábor ALMÁSI János HEBLING László PÁLFALVI Zoltán TIBAI

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