# Extreme THz Radiation from Lithium Niobite Materials

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When discussing extreme terahertz (THz) radiation, we always mean that its electric peak field is in the megavolt per centimeter with a peak magnetic field at the Tesla level. Therefore, strong-field THz can accelerate electrons, stimulate spin procession, excite lattice vibration, align and rearrange molecules, and has many fantastic potential applications.[1–3] However, all the applications finally rely on strong-field THz sources. For some cases, millijoule level or even tens of millijoule level THz radiation is required. There are many approaches to THz radiation generation, but generating high-energy THz radiation is difficult. Thanks to the rapid progress of ultrafast laser technology these years, at present, the mainstream to generate strong-field THz radiation is employing femtosecond laser pulses to interact with matters, such as nonlinear crystals, liquids, gas, or plasma. Practical users always want THz sources with extremely high efficiency, high beam quality, and high stability. Lithium niobate materials have a very high nonlinear coefficient, and the crystal size can be huge. Its damage threshold after doping is also very high. Therefore, the lithium niobate THz source has been recognized as one of the best candidates for obtaining high-energy THz radiation.

However, it is not so easy to efficiently generate THz radiation from lithium niobite. There are at least the major difficulties and challenges. Since most of the high-power femtosecond lasers are based on Ti:sapphire technologies and have very short pulse durations, in particular, not custom-built for THz generation, if Ti:sapphire femtosecond lasers are employed to efficiently generate THz radiation, there will be at least three challenges: (1) phase mismatching due to huge refractive indices gap in optical and THz frequencies, (2) intrinsic low efficiency due to ultrashort pulse duration, and (3) nonlinear effects under extremely high pump intensity.

For the first obstacle, the tilted pulse front technique of the pump laser pulses enables making the group velocity of the pump match with the phase velocity of the generated THz radiation inside the crystals,[4] resulting in longer effective interaction length, and can coherently boosting the 800 nm-to-THz energy conversion efficiency.[5] At the beginning stage, various femtosecond laser systems with different wavelengths, repetition rates, pulse durations, and pump energies have been applied to radiate THz radiation, and pushed the THz single pulse energy from several microjoules to hundred microjoule level. With these microjoule-level THz sources, sub-keV all-optical THz gun as well as THz electron acceleration and manipulation have been successfully demonstrated.[6,7] Moreover, THz field induced optical Kerr effect was also observed in liquid water.[8]

Noticeably, millijoule-level THz generation needs higher pump energies which can be provided by Ti:sapphire femtosecond lasers. Up to date, plasma from solids stimulated by femtosecond laser pulses has been demonstrated to be able to generation more than millijoule-level THz radiation. However, although most Ti:sapphire lasers can deliver more than joule-level single pulse energies, most of them have extremely short pulse durations and not built for THz radiation generation. Nevertheless, theoretical results predicted that the best optimal pulse duration for 800-nm Ti:sapphire pumped THz sources should be around four hundreds of femtoseconds. Therefore, chirping the pump pulses to a longer pulse duration is the best way to circumvent this difficult.[9] Under this application, 1.4-mJ THz radiation was achieved from lithium niobate crystals driven by 214-mJ Ti:sapphire laser pulses.[10]

When further scaling up the pump energy at the entrance facet of the crystal, the THz efficiency is already very high, but the residual pump spectrum will be destroyed, and the residual pump energy cannot efficiently be employed for further THz generation. In this case, the effective interaction length will become shorter under a strong pumping region. Furthermore, other nonlinear effects, such as self-phase modulation (SPM), will also occur inside the crystals in this circumstance. Due to these side effects, the generated THz energy as well as its properties, such as beam center and size, will also vary along with increasing the pump energy. To obtain tens of millijoule THz, we must consider all the above factors, and prepare the pump beam to be elliptical, chirp the pump pulses to longer pulse duration, tailor the pump spectrum, stack several lithium niobate crystals together, and cryogenically cooling them. The key point is all these factors have to be optimized and synchronized, and there may be the opportunity of generating $>10$-mJ THz radiation.

In the work recently published on Advanced Materials, we collaborated with Shanghai Institute of Optics and Mechanics, Chinese Academy of Sciences, which has a Ti:sapphire laser system producing 6-J, 30-fs laser pulses at a 1-Hz repetition rate. At the beginning, when the

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crystals were pumped at room temperature, we obtained more than 1-mJ THz at 400-mJ pump energy. This result demonstrates the feasibility of generating high-energy THz radiation with this high-power laser. However, THz energy saturated, and what's even worse was the reduced efficiency along with scaling up the pump energy. According to the broadened residual pump spectra, the SPM effect of the pump energy inside the crystals may be the primary mechanism. To verify this suspicion, the crystals were cryogenically cooled, and the THz efficiency was enhanced, but the saturation phenomenon was still there. Since the SPM is not so sensitive to the crystal temperature, but the THz linear absorption can be avoided by cooling, the THz energy saturation could originate from the SPM effect. Moreover, numerical simulation was also conducted, and under higher pump fluence, the SPM shows the destruction of the pump spectrum, and reduction of the effective interaction length, leading to decreased THz efficiency under a strong pumping case.

After understanding the saturation mechanisms, we safely scaled up the pump energy up to 1.2 J, optimized the system and tuned the pulse width by stretching the distance between the grating pair in the compression chamber. We finally obtained 13.9-mJ THz from lithium niobates and 1.2% 800 nm-to-THz energy conversion efficiency.[11] The peak electric field at the focused spot is $\sim 7.5$ MV/cm. This is the first lithium niobate-based THz source with a single pulse energy larger than 10 mJ in the world. This THz source is very friendly for high-field THz users due to the very good stability of the Ti:sapphire laser. Furthermore, as a function of the pump fluence, the THz energy curve did not show saturation behavior, enabling the opportunity to further enhance THz radiation output by scaling up the pump energies.

Along with the rapid progress of strong-field THz sources, extreme THz sources based on lithium niobate materials delivering more than several tens of millijoule or even hundred-millijoule single pulse energy are expected. With extreme THz science and applications in interdisciplinary research fields such as condensed matter physics, electron acceleration and manipulation, and biomedical sciences, undoubtedly, there will be another wave of extreme THz research upsurge worldwide. Moreover, with the further improvement of processing technology for lithium niobate materials and the development of bulk crystal to the thin film, THz devices, under the assistance of phonon, will also achieve great attention. More on-chip integrated lithium niobate THz devices will also emerge, and even nonlinear THz optics and phononics in devices may become very hot research topics. Soon, we can foresee extremely strong-field THz light sources that will be employed to observe more new discoveries and new physics in physics, chemistry, materials, biology, accelerators, and so on. On-chip integrated lithium niobate THz devices and systems will gradually move from the laboratory toward more real applications.

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