Measurements of THz and X-ray generation during metal foil ablation by TW, sub-relativistic laser pulses

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Abstract. We discovered that intensity of \( \sim 10^{16} \text{ W/cm}^2 \) and 20 mJ energy in femtosecond laser pulse is enough to observe THz emission from the rear side of metal foil in a vacuum. In the same experiment for 2-10 mbar air pressure and two-color pump similar energy of THz pulse form gas plasma was detected. Comparable amplitude and spectra of THz emission are also observed in reflection from the metal foil front side. X-ray emission is also studied as a criterion of intensity optimization. For much lower \( (10^{14} \text{ W/cm}^2) \) intensity reflected THz emission was detected as a result of optical rectification in a thin metal film with 10\(^{-6}\) efficiency. While for sub-relativistic intensities the observed efficiency 10\(^{-5}\) of THz generation from metal is higher than predicted by known theories. The main benefit of THz generation in metal is the absence of yield saturation at TW level of laser energy and above.

1. Introduction

In recent years, there have been increasing interest in powerful terahertz (THz) pulses [1–3], they can be used to excite such low-energy excitations as phonons, excitons, etc. [2]. There are several ways to obtain powerful THz pulses, for example, optical rectification in nonlinear crystals with a large aperture [4] or linear accelerators (in particular, FEL) of electron bunches [5]. In the last two decades, methods for generating THz pulses by the interaction of intense laser radiation and plasma were developed. The plasma source is usually either a gaseous medium [6–8] or a thin metal target. In this paper, we study both sources to obtain powerful THz pulses and also x-ray pulses of fs duration.

In the gaseous medium; the generation of THz pulses is most effective in a two-color laser field, which is a coherent mixture of the fundamental and second harmonic fields. In this case, the THz generation mechanism is the motion of an electron cloud in an asymmetric field [7], and the generation efficiency may increase with decreasing gas pressure [8, 9] but maximal available THz energy (\( \sim 1 \mu\text{J} \)) is limited by THz yield saturation at intensities above \( 10^{15} \text{ W/cm}^2 \).

To obtain more powerful THz pulses (\( \sim 100 \mu\text{J} \)) one cane use thin metal targets in a vacuum, but in this case, relativistic (\( > 10^{17} \text{ W/cm}^2 \)) intensities are required. In this case, the predicted conversion efficiency into the THz range can be up to 10\(^{-3}\) [1, 10–13]. Powerful THz radiation in the metal layer can be generated in several ways: 1) due to the currents in the metal in the presence of pre plasma [14], 2) due to thermal and surface effects [15] (at an oblique incidence, a superluminal spot runs over the surface, it initiates a surface current, which emits THz according to the Cherenkov mechanism) or 3) due to the emission of electrons from the back surface of the metal (mechanisms of coherent
transition (CTR) and “sheath field” (SR) radiation, which act simultaneously [1]. More details about these mechanisms can be found in the review [3].

The authors of [16] used similar parameters to ours, the intensity was about $10^{17}$ W/cm², 3 TW, 35 fs, 5 µm titanium foil in 45 deg. reflection. The mechanism of THz generation was attributed to a bunch of relativistic electrons and its dipole moment. Most of the known works are devoted to intensities of $10^{17}-10^{18}$ W/cm², while in the lower (and more accessible) subrelativistic range of intensities [15], only a few of either experimental [17] or theoretical studies [13] are known. At an even lower, not destructive intensity <10$^{14}$ W/cm², the dominant process for THz generation is considered to be optical rectification (nonlinear response of the second and third-order of free electrons), but the efficiency of this process is rather low (10⁻⁶⁻⁻⁵) [18, 19].

In addition to THz radiation, X-ray radiation is also observed when exposed by intense laser pulses on the metal [20]. Moreover, part of the radiation in the form of a characteristic Kα line can have high spectral brightness and femtosecond duration, which is important for X-ray fs diagnostics of nonstationary states of matter.

In our studies with TW pulses, we investigated the generation of THz and X-ray radiation. First from a gaseous medium and two-color pump and then from a metal target. We adapted metal ablation and X-ray emission methods to study filament properties for low-pressure nitrogen case, optimal for THz generation from TW laser pulse. We observed THz generation from the metal itself with efficiency higher than expected. Our research can help to understand what happens in a metal under such sub-relativistic intensity.

2. TW-laser system and measurement scheme
We use the laser system “Pulsar 200 TW” in NRC “Kurchatov Institute”, capable of generating 800 nm, 25 fs pulses with the energy of up to 7 J. With f=2.5 m focusing mirror (or f=1;0.8;0.3 m lenses or parabolic mirror) and 70 mm beam diameter we double laser radiation in thin KDP crystal, and launch a 5-20 cm long two-color filament in nitrogen and air that emits THz radiation [6, 8]. When the metal foil is inserted in the beam waist region, X-ray radiation is also registered, see Figure 1 (a). The entrance thick window of the experimental camera limits the available energy of a laser pulse due to phase self-modulation in the window material, therefore in this work, we are limited to a laser pulse energy of 80 mJ.

![Figure 1](image)

**Figure 1** (a) Measurement scheme (b) Reconstructed spectrum of transmitted THz radiation from metal and from gas plasma.

THz energy was detected by a Golay cell and pyroelectric two detectors, X-ray energy was characterized by x-ray photomultiplier and its spectra by Si-PIN X-ray detector. We also measured laser-induced breakdown spectra (LIBS) in the visible spectra range.
For gas pressure below 3 mbar, THz emission from metal target becomes more efficient than from gas plasma. Further for metal experiments we removed KDP crystal and use a single color pump at 0.5 mbar air pressure, 0 and 45 deg incidence angle, p-polarization. The target was continuously moved by stepper motors in two directions, fast enough to provide new area (100*100 µm) for each laser short. One measurement with metal required about 1*1 cm target surface and 5 minutes of signal optimization and averaging, all with 10 Hz laser repetition rate. We estimated THz spectral bandwidth obtained in two-color filament and from metal surface (<10 TH and < 0.5 THz respectively), see Figure 1 (b) by the usage of several THz filters: “high pass” ones from doped Si and “low pass” ones from PMMA polymer [8]. In the case of the metal target, the observed spectrum is as low-frequency as in CTR and SR mechanisms. We also used KHz rep.rate, 5 mJ laser system for non destructive THz generation on 200 nm gold films and electro-optical sampling for detection of THz pulses with sub-nJ energy.

3. THz generation in TW filament at low pressure and loose focusing.
We first study THz generation from gas plasma [8] and x-ray generation from metal to characterize TW monofilament. The pressure decreasing from ambient to tens of mbar avoids multiple filamentation and excessive ionization and provides a 100-times gain in the THz generation efficiency [6, 8] inside filament. The optimization of the second harmonic crystal position, the gas pressure, and the pulse duration allowed us to obtain a THz pulse of about 2 µJ, with 400 fs duration for 40 mJ, 30 fs, 800 nm pulse at 20 mbar of N₂ pressure and 2.5 m focusing length. We observe saturation of THz yield at energies above 40 mJ in all modifications of that experiment, while x-ray emission yield didn’t saturate with energy [21]. X-ray efficiency (flux about 10³ ph/(sec*sr)), spectra, and metal foil ablation rate allowed us to estimate laser radiation parameters in the beam waist or filament region. Maximal THz power in 20 mbar gas was obtained at a fluence of 15 J/cm², with an intensity of 2.5*10⁹W/cm². Here, with laser power P=1 TW; we obtained maximal THz energy of 2 µJ, which corresponds to efficiency ~10⁻⁴. Main limitations for THz yield are: THz absorption in long plasma channel and ionization losses of the pump. So we proceed with vacuum configuration and short focus case.

4. THz generation during thin foil ablation in vacuum and short focusing.
We decrease focal length from 250 to 30 cm, decrease pressure below 0.5 mbar, that increased X-ray yield two-four orders for Kα line (Figure 1 a)), while bremsstrahlung radiation background extended to hundreds of keV, transmitting through 1 cm of iron.

Figure 2 (a) X-ray spectra and (b) LIBS visible spectra for laser ablation of Al, Cu, and Ti foil of 10 µm thickness at 40 mJ, 30 fs laser pulse.

LIBS spectra show at least up to the 3rd degree of ionization [22], this confirms high intensity on the target, see Figure 2 (b). Simultaneously with the presented x-ray and LIBS we detected THz radiation from metal both in reflection and in transmission directions – Fig. 3. We observed weak dependence on incidence angle, film thickness, and metal type.
Next step we studied dependence of transmitted and reflected THz energy (and of X-ray energy) on intensity and on energy of laser radiation. We varied laser intensity by pulse duration and chirp increasing with translation of diffraction gratings in the optical compressor. We discovered that transmitted THz signal is more sensitive to pulse duration (see Figure 3 (a)), than both reflected THz and x-ray signals. For energy dependence we observe some sublinear behavior for reflected THz signal and almost linear for transmitted one. Thus transmission configuration of THz generation should be more perspective for higher laser energy. In those experiments x-ray signal is a criterion of actual intensity on the target, that could be reduced by plasma screening, residual gas pressure, or bad intersection of target surface and focus point.

In last experiment we compare metal types (Figure 4 (a)) and thickness influence (Figure 4 (b)) on THz yield. For variety of conditions (thickness, pulse duration, energy) we always obtained that Al provides the strongest THz yield and Ti – the worst.

On the example of cupper foils we studied film thickness influence, below 10 μm thickness THz yield decreases for both transmission and reflection directions. Similar tendency was observed for Ti foils with maximal efficiency around 10 μm thickness. For thickness more than 25 μm single pulse energy was not enough to ablate a through hole, in that case transmitted THz were not detected.
5. Discussion
Some of observed THz yield behavior is opposite in comparison with known publications for higher laser intensities. For the relativistic intensity, a sharp increase in the THz yield is known at foil thicknesses less than 2–5 μm [11]; in our case, for thicknesses less than 10 μm, the THz yield decreased for both detectors. We did not observe quadratic or exponential increase of THz yield with pulse energy as predicted for higher intensities. Normal and 45 deg. incidence angles did not influence considerable on THz yield in our case.

Other features agree with similar studies. Spectra bandwidth is about or below 1 THz in our case and in CTR and SR processes. The dependence on the type of metal is given in [10], aluminum gives the best yield, as in our study, but copper is worse than titanium in [10].

In our experiment with another, kilohertz laser system, for an intensity less than the ablation threshold (and pulse energy up to 4 mJ) we observed THz generation from 200 nm of the gold film on quartz substrate in reflection geometry with an efficiency of about 10^{-6}, for two-color and for one-color pumping. The breakdown threshold of the gold film (and the optimal intensity for generation from metal) was about 1 mJ per 1 mm². In this case, the mechanism of optical rectification on nonlinearities of the second and third orders prevailed [12], the spectrum reached 2 THz, the optimal metal thickness was 200 nm. With an increase in intensity, the optimal thickness was 10 μm, the THz spectrum was already less than 0.5 THz, the other mechanism of THz generation became dominant. At the same time, the efficiency increased by an order of magnitude.

6. Conclusions
For laser pulse intensities above metal damage threshold but below relativistic level, that is 10^{14} - 10^{17} W/cm², there is no detailed and proven explanation of dominated mechanism of THz generation. Our study provides additional experimental data to clear out this question. Up to know only reflected THz where described for intensity below 10^{17} W/cm². We discovered that 10^{16} W/cm² and 20 mJ energy is enough to observe THz emission from the rear side of metal foil. In transmission and reflection directions, comparable THz signal values and spectra are observed, but for shorter pulse duration and for higher laser energy the ration between transmitted and reflected THz signals increases. The dependence of film thickness and on pulse energy does not confirm dominated contribution from the known relativistic processes of CTR or SR. The advantage of metal over a gas as a conversion media– is the absence of saturation at energy increase above TW level. We expect to obtain 10 μJ of THz pulse at 10 TW pump in the suggested scheme of moving metal foil.

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