Terahertz-Pulse Emission Through Laser Excitation of Surface Plasmons in a Metal Grating

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The second-order processes of optical-rectification and photoconduction are well known and widely used to produce ultrafast electromagnetic pulses in the terahertz frequency domain. We present a new form of rectification that relies on the excitation of surface plasmons in metal films deposited on a shallow grating. Multiphoton ionization and ponderomotive acceleration of electrons in the enhanced evanescent field of the surface plasmons results in a femtosecond current surge and emission of terahertz electromagnetic radiation. Using gold, this rectification process is third or higher-order in the incident field.

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Rectification, producing electromagnetic radiation at terahertz frequencies from optical pulses is a well-known phenomenon with a long history [1] and a wide range of applications [2]. Recent technological developments have made it possible to use subpicosecond terahertz laser pulses [3] in numerous fundamental studies and practical applications. The oldest method of rectification of laser light is optical rectification, [1] which is a phase-matched second-order nonlinear optical process taking place in noncentrosymmetric media such as ZnTe and $LiNbO_3$. The alternative rectification process of photoconduction relies on the excitation of electrons into the conduction band of a semiconductor photoconductive switch. These real electrons are then accelerated in an external dc bias field resulting in Hertzian dipole emission of terahertz radiation [4]. In the latter case, each absorbed laser photon produces one conduction electron resulting in a process second-order in the incoming laser intensity. More recently, it has been shown that the demagnetization of ferromagnetic materials results in rectification [5]. Rectification in laser-induced plasmas is seen as a promising source of high-power terahertz radiation [6,7] and allows coherent control of the electric field shape [8]. Here, we will describe and explain a novel method for generating terahertz laser pulses that is not second order in the incident laser field and based on the excitation of surface plasmons.

It was recently reported [9] that gold and silver surfaces irradiated with 800 and 400-nm femtosecond laser pulses emit terahertz radiation. This could in principle lead to new compact sources of terahertz radiation while also opening a new window on the study of surface physics and dynamics. The presence of a surface breaks the translational symmetry thereby allowing $\chi^{(2)}$ processes to take place with isotropic materials such as amorphous bulk metals. It is well known that this accounts for second-harmonic generation on surfaces and could well explain optical rectification on surfaces too. However, optical rectification should result in a second-order dependence on input fluence inconsistent with the reported fifth-order dependence [9]. The metal surfaces used to produce terahertz radiation in these studies were by no means crystallographically flat. This leads one to consider ''bumpy'' or nanostructured surfaces. Such nanoscale structures can interact strongly with light and ''concentrate'' fields resulting in very high field strengths and enhanced nonlinear optical phenomena.

Here, we will describe for the first time the influence of surface-plasmon excitation on terahertz-pulse generation on a gold surface. Surface plasmons can facilitate multiphoton excitation of electrons in metals by femtosecond laser pulses resulting in the emission of photoelectrons under vacuum conditions [10,11]. Under the right conditions, an evanescent field associated with the surface plasmons is created causing a ponderomotive force pushing photoelectrons out of and away from the surface [12]. A number of nanostructured surfaces have been studied using rectification of amplified femtosecond laser pulses. The rectification results will be compared with ⟨110⟩-cut ZnTe, which is typically used to produce broad bandwidth short pulse-duration free-space propagating terahertz pulses. Comparison with ultrafast electron-emission studies [12] will allow us to explain the paradoxical rectification results on ''flat'' metal surfaces [9].

A 1-kHz repetition rate regeneratively amplified laser providing 1-mJ pulses with a 100-fs pulse duration centered at 800 nm was used for all experiments. Figure 1 shows a schematic diagram of the experimental setup used. The laser beam is split into an intense pump beam containing 98% of the power and a weak probe beam. The pump beam is passed through an optical-delay stage followed by a telescope producing a collimated beam of approximately 5-mm diameter on the sample [13]. The power density was varied between 0.25 and 4 mJ/cm^2. The sample was positioned for p-polarized excitation (grooves perpendicular to the table) with a variable angle of incidence and a half wave plate used to allow for excitation with s polarization. Terahertz radiation produced by the samples is detected by standard electro-optic sampling techniques [3] in a 1-mm-pathlength ⟨110⟩ ZnTe detection crystal followed by a quarter wave plate, Wollaston prism, and balanced photodiodes. The electro-

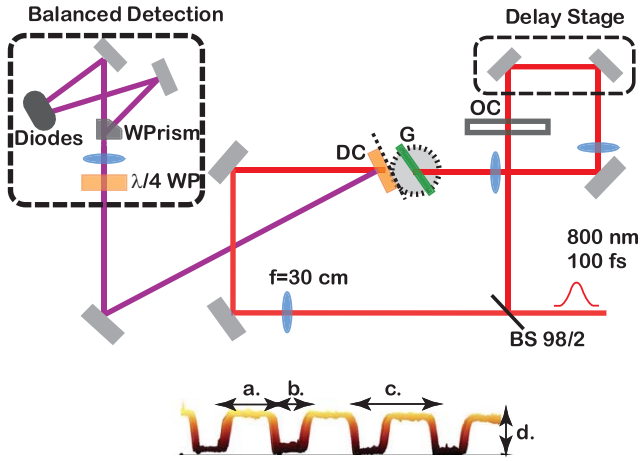

FIG. 1 (color online). (top) Schematic of the experimental setup. BS, Beam Splitter; OC, Optical Chopper; G, Grating on rotation mount; DC, 1-mm thick ZnTe Detection Crystal with black card to block pump light; WP, Wave Plate; WPrism, Wollaston Prism. (bottom), Grating profile obtained by AFM; $a = 340$ nm, $b = 160$ nm, $c = 500$ nm, and $d = 40$ nm etch depth.

optic sampling setup employs a reflection geometry allowing for easy sample changes but reducing the signal-to-noise ratio in the experiments considerably. It was confirmed experimentally that the 5 cm distance between the generator and detector provides a far-field signal [3].

Terahertz-pulse emission has been obtained in our studies from a number of nanostructured surfaces. In initial experiments, self-assembled ordered monolayers of latex spheres [14] were used as metal-deposition masks to make nanostructured metal surfaces. Roughened surfaces have also been used successfully. Nanostructured surfaces made using these techniques gave varying and somewhat irreproducible results as far as terahertz-pulse generation is concerned. Consistent terahertz-pulse results could be obtained using a custom made shallow grating (Ibsen Photonics, Denmark). The $10 \times 10\text{-mm}^2$ grating is produced in UV-grade fused silica and has a 500-nm grating period and a 40-nm etch depth. The grating was coated with 30 nm of gold using a standard vacuum evaporator. The thickness was selected for maximum absorption of the pump laser by modeling a multilayered flat surface with standard Fresnel equations. After gold deposition, the grating properties were measured using a Witec Alpha AFM (Atomic Force Microscope) operating in tapping mode. The measured cross section (see Fig. 1) shows a 40-nm etch depth and a high section of 340 nm in every 500-nm grating period.

The grating was then mounted in air on a rotation stage and excited from the rear in transmission with the grating surface facing away from the incident laser beam as shown in Fig. 1. A half wave plate and polarizing beam cube were used allowing the power to be continuously varied while keeping the pump-probe time delay constant. The emitted terahertz-pulse electric field is measured using electro-optic sampling in 7-ps scans. The terahertz-pulse fluence (power) was determined by squaring the measured time-domain data and integrating over delay time.

Figure 2 shows the dependence of the terahertz-pulse fluence on the (external) angle of incidence of the excitation beam onto the grating for both p and s polarizations. Also, shown in Fig. 2 is the amount of absorption experienced by the 800-nm pump beam as a function of angle of incidence. The absorption is relatively constant as a function of angle except for a sharp peak at $\sim 40°$ due to the excitation of surface plasmons in the 30-nm gold film.

Surface plasmons (SPs) represent electromagnetic surface waves traveling on the boundary between a metal and air. The electric fields associated with the SP decay away exponentially from the boundary. SPs do not normally couple to free-space propagating electromagnetic waves except in special cases such as corrugated surfaces [15]. One such corrugated surface is a grating in which SPs can be excited at the appropriate phasematching condition. Phasematching is obtained when the wave vector of the external field projected onto the grating combined with multiples of the grating wave vector matches the wave vector of the SP. This can be expressed as [15]

$$\sin(\theta) + N\lambda/\Lambda = n_{\mathrm{SP}}, \tag{1}$$

where $\sin(\theta)$ is the projection of the wave vector of the excitation beam onto the grating with angle of incidence θ and wavelength λ, N is the diffraction order, and Λ the grating period. The SP refractive index is given by $n_{\mathrm{SP}} = \varepsilon^{1/2}/(\varepsilon + 1)^{1/2}$, where ε is the dielectric function of the metal. Using Eq. (1) and a value for the dielectric function of gold at 800 nm of $-26.3 + 1.86i$, [15] a SP resonance

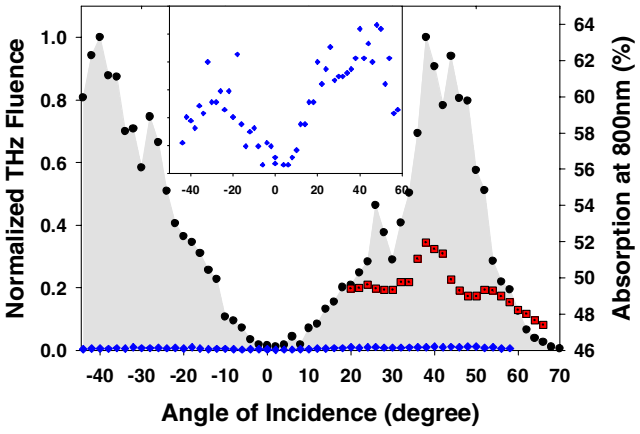

FIG. 2 (color online). The circles (diamonds) show the normalized terahertz-pulse fluence vs. the angle of incidence of the pump laser obtained by rectification on a gold-covered grating excited by p-polarized (s-polarized) radiation. The fluence incident on the grating at 800 nm was $3.0\ \mathrm{mJ/cm^2}$. The squares show the absorption of the 800-nm p-polarized pump beam. The inset shows the terahertz fluence for excitation with s-polarized radiation multiplied 150 times.

angle of 35° is calculated. The measured angle of maximum absorption is 40° as can be seen in Fig. 2.

It can be seen in Fig. 2 that maximum terahertz-pulse fluence is observed around the angle where surface plasmons are excited for p-polarized light. Significantly, the terahertz-pulse fluence drops by a factor of ~200 when the grating is excited with s-polarized light. The distribution of angles appears to be somewhat wider than the surface-plasmon absorption resonance. One may expect higher-order ($N > 1$) excitation of surface plasmons widening the distribution. Further, complications arise as Eq. (1) is only strictly valid for gratings with vanishing modulation depth. The finite modulation depth (40 nm) may cause a slight shift of the resonance angle [16]. It can also be seen in Fig. 2 that the terahertz-pulse fluence is symmetric and tends to zero at zero angle of incidence. This is a result of the boundary conditions that require the (terahertz) electric field component parallel to the metal surface to be zero. We observe that the terahertz electric field flips sign as a function of the angle of incidence as explained later.

Figure. 3 shows the terahertz electric field measured by electro-optic sampling emitted by the grating and a reference sample consisting of a 0.5-mm length ⟨110⟩-cut ZnTe crystal. At an angle of incidence of 40°, corresponding to maximum terahertz-pulse emission, the peak electric field measured from the grating is half that of the 0.5-mm ZnTe crystal. The inset of Fig. 3 shows the power spectrum obtained by taking the absolute value of the Fourier transform of the time-domain traces. Although the detection bandwidth in electro-optic sampling is limited to about 2.5 THz due to the use of a 1-mm pathlength ZnTe crystal, it appears that the terahertz pulses produced in both types of generator are of about equal pulse length (~1 ps).

By varying the excitation (pump) power, it is possible to obtain a power dependence of the terahertz emission from the grating vs ZnTe. Figure 4 shows the measured input power dependence of the terahertz-pulse fluence with least-squares fits to power laws. The ZnTe data fits a power of 1.8, which is sufficiently close to 2 to be consistent with the expected $\chi^{(2)}$ optical-rectification process. The grating data fits a power of 3.2 or—when fitted to a double power law—to a power of 3.5 at low incident intensity leveling off to a power of 2.0 at the highest incident intensities. At power densities above 3.6 mJ/cm^2, gold is ablated from the surface of the grating due to heating. However, at power densities above 3.0 mJ/cm^2, second-harmonic generation effects occur in ZnTe resulting in crystal damage.

The third-to-fourth-order power dependence of the generation of terahertz radiation in the grating is inconsistent with standard optical rectification or photoconduction. The work function of gold is approximately 5.3 eV, [17] suggesting that rectification may be associated with the absorption of three to four 800-nm (1.5 eV) photons and the emission of a photoelectron through the multiphoton photoelectric effect. Previously, we have shown that the excitation of surface plasmons with high-power 800-nm femtosecond laser pulses causes the emission of electron bunches and acceleration [12]. The excitation of surface plasmons is associated with an evanescent electromagnetic field extending into the dielectric medium (air in this case). Electrons placed in this evanescent field experience a ponderomotive force directed away from the metal surface resulting in measured electron energies approaching 1 keV [10–12]. We surmise that the effect takes place in the grating studied here resulting in ultrafast electric currents and emission of terahertz pulses.

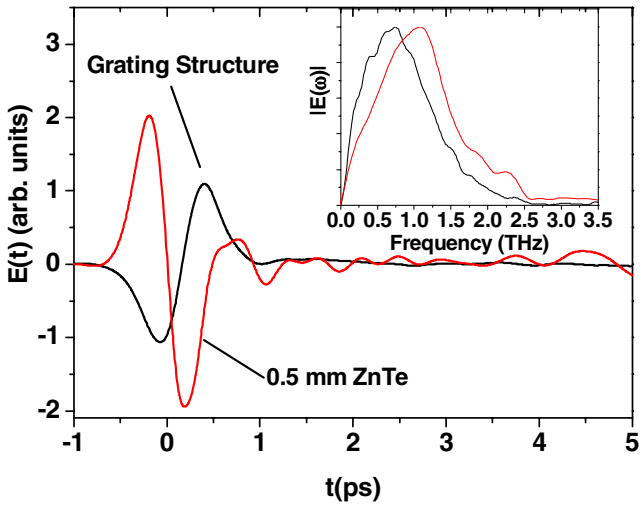

FIG. 3 (color online). Comparison of terahertz electric field generated in a 0.5-mm ZnTe crystal with that generated in the grating for an incident 800-nm laser intensity of 3.0 mJ/cm^2. (inset) The absolute value of the Fourier transform of the time-domain traces.

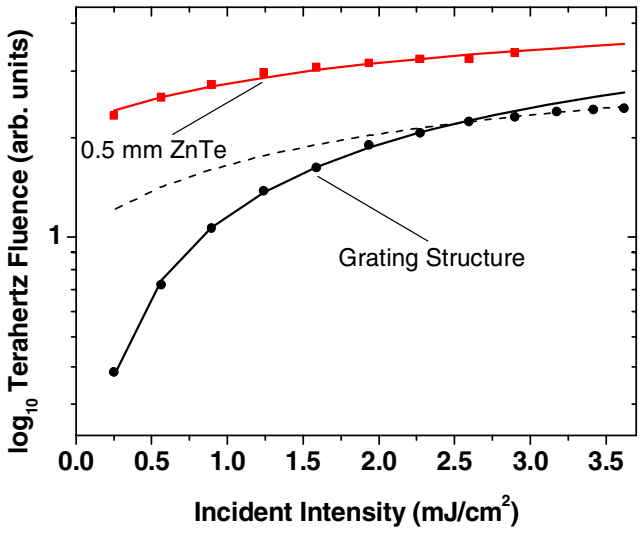

FIG. 4 (color online). Input laser-power dependence of the terahertz-pulse fluence for ⟨110⟩ ZnTe (squares) and the grating structure (circles). The solid lines are least-squares fits to power laws with the upper solid line corresponding to a power of 1.8, the lower solid line a power of 3.5, and the dashed line 2.0.

Acceleration of charge causes emission of electromagnetic radiation. In the far field, the electric field is given by $\vec{E}_{FF}(t) \propto d\vec{j}(t)/dt$, where $\vec{j}(t)$ is the (photoinduced) current density. In the case of the grating, three to four 800-nm photons are absorbed producing a photoelectron near the surface of the metal film. The evanescent field associated with the surface plasmon decays away from the surface over a distance of approximately a wavelength (800 nm). Therefore, the photoelectrons will be accelerated in the ponderomotive potential associated with the evanescent field over a distance no longer than ~800 nm and for a period no longer than the laser-pulse duration (~100 fs). This explains the relatively short terahertz pulse observed (see Fig. 3). The grating has a rectangular profile leading to a complicated shape for the evanescent field. However, as the grating period (500 nm) is less than the excitation wavelength (800 nm), the evanescent field is perpendicular to the plane of the grating to a good approximation. That implies that $\vec{j}(t)$ and hence the terahertz field is perpendicular to the plane of the grating explaining the flip of the sign of the terahertz field observed as a function of angle of incidence. The magnitude of the current density will scale with the third to fourth power of the incident laser intensity except at high power where tunnel ionization reduces the apparent order.

We have been unable to reproduce terahertz emission from flat gold surfaces reported previously [9]. In our experiments, 150–250 nm layers of gold were vacuum evaporated directly onto glass slides. It is likely that the terahertz-pulse signal is simply too weak to measure in our experimental setup. This suggests that terahertz-pulse emission from ''flat'' surfaces may be due to surface roughness and the (inefficient) excitation of surface plasmons. The involvement of surface plasmons in our experiment is demonstrated by the strong dependence of the terahertz fluence on excitation angle and polarization.

Our experiments have been performed in air, which may seem inconsistent with the emission of photoelectrons. However, in the evanescent field acceleration model, the photoelectrons are only accelerated over a very short distance for a very short period. The probability of a collision with a molecule in air is therefore vanishingly small. On longer time scales, one does expect the electrons to collide with and ionize molecules in the air. On a time scale of nanoseconds, the charge will recombine with the holes left in the metal.

The terahertz-pulse generation process described here, naturally leads to a possible extension: acceleration by an external dc bias field. In that case, the distance over which acceleration takes place would be strongly dependent on the morphology of the nanostructures. The time dependence would still be mainly determined by the duration of the laser pulse producing the photoelectrons. At very large dc potentials, one would expect field emission and a lowering of the order of the process. This would be similar to a recent report inferring terahertz-pulse emission by optical rectification at a metal tip [18] except over a much larger area.

In conclusion, the results presented here show a new process of rectification and terahertz-pulse generation from nanostructured surfaces. We have found a third to fourth-order power dependence ruling out second-order optical rectification or photoconduction but consistent with photoelectron production and evanescent wave acceleration. This suggests the possibility of dc biasing in order to produce short and high-power terahertz pulses.

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