Study of terahertz radiation generated by optical rectification on thin gold films

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Emission of terahertz (THz) radiation as a result of optical rectification of intense femtosecond laser pulses on thin gold films has been studied by time-domain THz spectroscopy. The THz amplitude was measured as a function of film thickness and incidence angle. The experiments reveal that the emitted THz field is suppressed for a thickness below 100 nm, which gives evidence of the nonlocal character of the response. The variation of incidence angle allows us to estimate the components of susceptibility tensor $\chi^{(2)}_{ijk}$. For thicker films and near grazing incidence, the emitted THz field attains a peak value of 4 kV/cm.

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In the past few years the effort to develop compact and efficient sources of terahertz (THz) radiation has stimulated a number of achievements in methods of generation of electromagnetic waves in the THz region. One possible method of optical rectification (OR), on metals, emerged recently.[1,2] It is based on a nonlinear optical response of free electrons at the metal surface where the translational symmetry is broken; one well-known implication of this nonlinearity is second-harmonic generation (SHG), which has been studied extensively in the past few decades.[3] The potential advantage of measuring THz radiation in the time domain over SHG is the possibility of detecting the emitted electric field with a phase sensitivity and on a time scale close to that of the dynamics of free electrons. Therefore information about the processes triggered by the incident light can be richer than that provided by time-integrated light intensity in SHG experiments.

In Ref. 2 we showed, for the case of a Au film deposited by plasma sputtering on a glass substrate, that the amplitude of the emitted electric field is proportional, as expected, to the intensity of the incident ultrashort laser pulse; this dependence is quite different for bulk Ag. This difference may be explained partly by chemical contamination of the metal surface, which is expected to be substantially higher for Ag. Consequently, Au films appear to be more convenient for further studies unless a higher purity of the metal surface can be ensured.

The thickness of the film, $d=100$ nm, used in Ref. 2 was much greater than the penetration depth, $1/\alpha$, at the incident wavelength used, $\lambda_0=810$ nm, which amounts[4] to $1/\alpha\approx 12$ nm. It is also known that the intensity of SHG radiation on Ag films is essentially constant[5] for thickness $d>50$ nm. We then incorrectly supposed that the OR on the 100-nm Au film was close to that on bulk material. In this Letter we show that the amplitude of an optically rectified field in Au dramatically increases as the layer thickness exceeds the value of 100 nm. We also study THz signal variation as a function of incidence angle.

The experimental setup was essentially the same as that described in Ref. 2; briefly, 0.7-mJ pulses with a mean wavelength of $\lambda_0=810$ nm and a duration of $\Delta t=50$ fs were used to excite the sample, and electro-optic sampling with a ZnTe crystal was employed for detection of the p-polarized component of the THz field. All the experiments were performed at reduced pressure (5×10^3 Pa) to avoid water-vapor absorption. As in the study in Ref. 2, the Au films were deposited on cleaned glass substrates by Ar-ion plasma sputtering (with a Baltec SCD-050 sputter coater). Thickness d of the films was determined with a precision of ±5 nm by surface profile measuring; the surface roughness of the films was ~0.7 nm rms. To determine the optical constants of the films used, we also measured the THz transmittance of a $d=100$ nm film; for the complex refractive index of the Au film, this yielded a value of $N(400\ \mathrm{GHz})\approx 500-500i$.

In a first series of experiments we measured the THz signal $E(t)$ coming from OR on six films with d ranging from 100 to 280 nm. The integrated THz pulse amplitude $A=\int|E(\omega)|\mathrm{d}\omega$ measured as a function of d is shown in Fig. 1; we see that at $d=100$ nm the amplitude of emitted THz radiation is the lowest among all the samples. As in Ref. 2, we can measure the peak value of the electric field by calibration of the compensator shift. For $d=100$ nm we obtain $E=50$ V/cm. Note that this value is four times lower than that obtained in Ref. 2 for a sample with the same nominal thickness. This is because the function $A(d)$ is steep near $d=100$ nm, so variations in d as small as a few nanometers (within the uncertainty) lead to significant changes in A. An increase of $A(d)$ is observed up to $d=160$ nm; the signals for samples $d=100$ nm and $d=160$ nm differ by approximately 2 orders of magnitude. When d is further raised to 300 nm, the amplitude drops by ~25%. The spectra for the individual samples scale well linearly with A;

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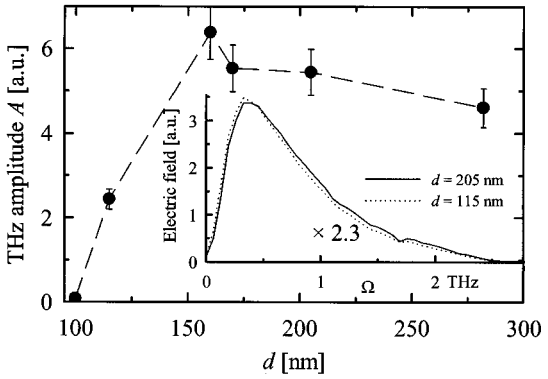

Fig. 1. Integrated THz pulse amplitude as a function of d with both the excitation and the THz beam p polarized; angle of incidence 45°. Inset, THz spectrum for $d=205$ nm and $d=115$ nm (linearly scaled).

as an example, two selected spectra are shown in the inset of Fig. 1.

This behavior can be tentatively explained by depolarization effects owing to the electron current normal to the surface (other components can be neglected[2]). At a frequency of 400 GHz, the wavelength in free space is $\lambda=0.75$ mm and the THz penetration depth $1/\alpha_{\mathrm{THz}}\approx 120\pm 40$ nm is comparable with d. This means that the film can contribute to the OR signal throughout its whole thickness. On the one hand, the absence of signal for $d<100$ nm cannot be explained simply by a destructive interference; this would require a much greater thickness and would significantly influence the shape of the spectra depending on d, which was not observed. On the other hand, we know that the nonlinear polarization induces a current perpendicular to the surface. This leads us to the idea that the charges accumulate near the metal–glass interface, and the resulting field is opposite the initial current at the origin of the emitted THz field. Indeed, the measured $A(d)$ dependence strongly indicates the nonlocal character of the thin-film response. If this hypothesis is correct, this means that the charge carriers arrive at the metal–glass interface within the incoming pulse duration. Supposing that the free electrons propagate with the Fermi velocity[6] 1.4×10^6 m/s, they pass through the $d=100$ nm film within ~ 70 fs, which is close to Δt. This would imply a ballistic character of the electron transport, in agreement with earlier observations.[7]

In a second series of experiments we measured the p-polarized THz field $E(t)$ emitted by the film with $d=205$ nm as a function of incidence angle θ. To this purpose we used a parallelogram-shaped device that enables one to vary θ, while the direction of both of the input and output (reflected) beams and the optical path length[8] are preserved. The values of integrated THz amplitude A for p- and s-polarized incident pulses are shown by the data points in Fig. 2. Note the similarity between the data corresponding to the p polarization and those observed in SHG experiments[5,9] with Ag; in both cases the signal increases toward higher θ. With this sample at $\theta=76°$, we measured an electric field of 4-kV/cm peak value, which is comparable with the typical field generated by OR in a ZnTe nonlinear crystal. From waveforms $E_p(t)$ and $E_s(t)$ it is also possible to find the relative phase shift η_{s-p} between the two signals for each θ; this shift appears to be frequency independent, and its measured values are shown in the inset of Fig. 2.

In a first approximation the nonlinear polarization of the metal surface can be described[10] by the susceptibility tensor $\chi^{(2)}_{ijk}$. By solving Maxwell's equations, we may write, for the p-polarized electric field intensity at THz frequency Ω in analogy with Eq. (15) in Ref. 10,

$$E_p^{\Omega,p}=B[\chi_{xzx}F_c(f_c^{*}f_s+f_cf_s^{*})|t_p|^2+\chi_{zxx}F_s|f_ct_p|^2+\chi_{zzz}F_s|f_st_p|^2] \quad (1)$$

for incident p polarization and, in analogy with Eq. (16) in Ref. 10,

$$E_s^{\Omega,p}=B\chi_{zxx}F_s|t_s|^2, \quad (2)$$

for incident s polarization. Here we introduce

$$B=\frac{\varepsilon_0}{2}\frac{|E_0^{(\omega)}|^2}{NF_c+nf_c}\frac{1}{N\cos\theta+F_c}, \quad (3)$$

and we use the notation from Ref. 10, $F_s=\sin\theta/N$, $F_c=(1-F_s^2)^{1/2}$, $f_s=\sin\theta/n$, $f_c=(1-f_s^2)^{1/2}$, $t_s=2\cos\theta/(\cos\theta+nf_c)$, $t_p=2\cos\theta/(n\cos\theta+f_c)$; n and N are the complex refractive indices of the metal at incident and THz frequencies ω and Ω, respectively.

Equations (1) and (2) allow us to calculate for given complex values of χ_{xzx}, χ_{zxx}, and χ_{zzz} functions $|E_p^{\Omega,p}(\theta)|$, $|E_s^{\Omega,p}(\theta)|$, and $\eta_{s-p}(\theta)$ to describe our experimental data. For the refractive index of gold, we used the value[4] $n=0.2-5.2i$. Although we found no set of χ_{ijk} that would unambiguously describe the data, there are possible values leading to a satisfactory agreement with the experimental values for both $A(\theta)$ and $\eta_{s-p}(\theta)$; the curves in Fig. 2 show one possible solution. The fitting procedure led us to the con-

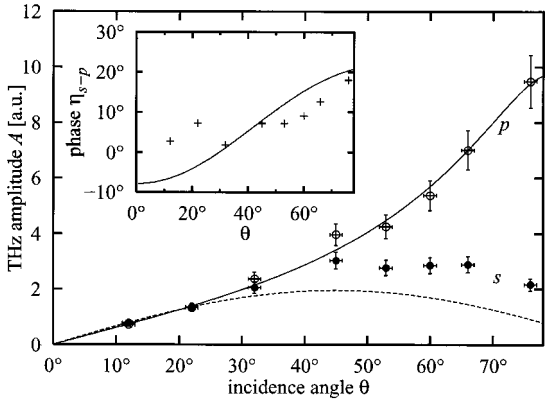

Fig. 2. Integrated THz pulse amplitude as a function of θ for p and s polarization of excitation beam $d=205$ nm. Inset, relative phase shift between the THz pulses in the two polarizations. Symbols, experimental data; curves, best fit using Eq. (1) for p polarization and Eq. (2) for s polarization.

clusion that the χ_{zzz} term has the highest amplitude, whereas that of χ_{zxx} is approximately ten times smaller and the χ_{xzx} term is more than 2 orders of magnitude smaller than χ_{zzz}. Concerning the phases of the terms, the fit suggests that the absolute difference between the phases of χ_{zzz} and χ_{zxx} is $\pi/2$.

Although the curves clearly reproduce the salient features of our experimental results, the overall agreement is not excellent. In particular, for any set of χ_{ijk}, the maximum of the $|E_s^{\Omega,p}(\theta)|$ curve is at $\theta = 45°$, and its peak in our experimental data is shifted toward higher values. As a matter of fact the above solution of Maxwell's equations supposes that susceptibility tensor $\chi_{ijk}^{(2)}$ does not depend on the **k** vector. In reality there are most probably two different sources of $\chi_{ijk}^{(2)}$, one corresponding to the current density normal to the surface and localized in the interface (selvedge) region, i.e., related to the surface electron states, and another due to the bulk states modified by the presence of the surface.[11] The interplay of these contributions, together with those of the electric quadrupole and magnetic dipole moments,[12] will lead to a **k** dependence of the effective nonlinear response. Should all these contributions be taken into account in the fit, the number of free parameters allowed by the symmetry would be too high to permit their unambiguous determination. Thus, to obtain better agreement between the calculated curves $E_p^{\Omega,p}(\theta)$, $E_s^{\Omega,p}(\theta)$, and $\eta_{s-p}(\theta)$ and the experimental data, it would be necessary to develop a microscopic model taking into account these effects and also including the sample thickness d.

In conclusion, we have shown that the THz field produced by OR at metallic surfaces probes a surface region extending a few hundreds of nanometers below the surface, an order of magnitude more than in case of SHG.[13,14] The suppression of the THz signal for $d < 100$ nm is a clear sign of a nonlocal nonlinear response.

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