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Effect of inhomogeneity and plasmons on terahertz radiation from GaAs (1 0 0) surface coated with rough Au film

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ABSTRACT

We measured terahertz (THz) radiation from GaAs (1 0 0) surface coated with rough Au film in the thickness ranging from 5 to 21 nm under the incident angle from 0° to 50°. Anomalous THz emission was observed with inhomogeneous crack structures at normal incidence, which originates dominantly from the lateral photo-Dember current. Meanwhile, enhanced THz radiation from Au/GaAs was investigated with the variation of the Au morphology, which confirmed that localized surface plasmons play an important role in the THz radiation. The results indicate the prospect of harnessing surface plasmons for efficient THz emission with controllable morphology of Au on semiconductors.

1. Introduction

Terahertz (THz) wave, bridging electronics and photonics in the electromagnetic spectrum, features many exotic properties and promising applications [1]. However, because of low THz emission efficiency [2], less sensitive detectors [3], and few manipulating devices [4], THz wave is still on the horizon for practical applications since 1980s. Thus, how to raise the radiation efficiency of THz emitters possess one of the most urgent problems in THz science and technology [2]. As frequently used THz emitters, GaAs has been widely exploited in photoconductive antennas [3,5]. The dominant THz radiation mechanism for GaAs emitters depends on the crystal orientation, exciting laser intensity, surface condition, intrinsic or extrinsic electric field supplied, and other features [6]. For example, due to the crystal orientation of GaAs (1 0 0), both optical rectification and photocurrent surge mechanisms are forbidden when the exciting laser impinges normal to surface, but both mechanisms will involve into the competition when the excitation is under oblique incidence [6]. In the case of oblique incidence, the drift current caused by intrinsic electric field in the depletion layer dominates the THz radiation when the excitation intensity is relatively low. With the increasing of excitation intensity, however, the photo-Dember current, which arises from the different diffusion coefficients of electrons and holes, will become the primary THz emission mechanism [7].

Emission efficiency may be effectively raised when the GaAs surface is changed by a properly designed coating [8,9]. For example, the replacement of the pristine surface with either inorganic molecule [8], organic molecule [10], or nano-structures [11] directly onto the surface can promote THz radiation from semiconductor surfaces. In 1990s, Au film coating was found to enhance THz emission at oblique incidences [12]. Our further studies on the Au/GaAs system have revealed ultrafast carriers dynamics in Au/GaAs interface [13], while the mechanism for the enhanced THz emission remains unclarified. Although THz emission from either metal nanoparticles [14] or Au films [15] was realized with huge single pulse energy (in the order of mJ/pulse), THz emission contribution from metals is negligible under relatively lower laser field (in the order of nJ/pulse) and can not explain the enhanced THz radiation from Au coated GaAs.

In this work, we report the THz emission from Au coated GaAs (1 0 0) surface excited by femtosecond laser pulses (nJ/pulse) at incident angles ranging from 0° to 50°. At oblique incidence of 50°, THz emission from Au/GaAs was found to be four times as efficient as the pristine GaAs. More importantly, THz emission with a

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considerable intensity was also measured at normal incidence, which demands explanation other than optical rectification or transverse surface transient current. To figure out the responsible mechanism, the dependence of THz radiation on the thickness of Au coating was investigated, and compared with that from Cr/GaAs, as well as numerical simulations of electric field distribution on surface.

2. Experimental

THz emission measurement was performed on the (1 0 0) surface of semi-insulating GaAs (0.5 mm thick, double-side polished) coated with Au film of various thicknesses (3–21 nm) via magnetron-sputtering (FJLX500, Shenyang Scientific Research Center). The thickness of Au film was controlled by choosing appropriate depositing current and time. In our sample preparation, the current for depositing was 2 mA and the depositing time was on the scale of several minutes. The longer the depositing time was, the thicker the Au film would be. For reference, the emission from a GaAs with pristine surface was measured under the same conditions. Also a sample coated with Cr (~8 nm) was investigated for comparison. Control samples are Au films of 5 nm, 10 nm and 15 nm in thickness grown on silicon wafer and quartz glass, respectively, which are expected to rule out the effect of accelerated free photoelectrons induced THz emission. The morphology of the metal coatings was characterized by scanning electron microscope (SEM, Raith) and atomic force microscope (AFM, Bruker). A custom-designed THz time-domain emission spectrometer in transmission geometry was employed for the THz radiation measurement. A p-polarized femtosecond laser with an average power of ~390 mW (Maitai Spectra-Physics, repetition rate 80 MHz, pulse width 70 fs, central wavelength 800 nm) was focused onto the samples with a maximum energy density of ~16 μJ/cm^2. The forward THz radiation was collected by a parabolic mirror and then detected by ZnTe via electro-optic sampling. As depicted in the literature [6], the contribution from optical rectification generated via oblique incidence, was first minimized by rotating the azimuthal angle of the samples to a proper position. The propagation path of THz waves was sealed in a space filled with dry nitrogen in order to eliminate the influence from water vapor.

3. Results and discussion

Fig. 1 illustrates the morphology of the Au films on GaAs (1 0 0), as revealed by SEM and AFM. In the case of thin coatings (such as 3 nm and 5 nm in thickness), Au films are composited with Au clusters and some cracks are shown in the SEM. With the film thickness increasing to 8 nm and 21 nm, the density of Au deposit and the cracks also increase and the whole rough Au films form percolating patterns. This is a disaster for some applications, however, a good fortune for THz emitters. Firstly, high electro-magnetic field is locally enhanced, which is associated with the gaps in the cracks [12]. Secondly, the scattering near the edges of cracks affords the momentum matching to excite surface plasmon resonance without the total reflection prisms and gratings.

As is well-known, no matter THz radiation from optical rectification or the photocurrent surge effect [6], no forward or backward THz radiations from GaAs (1 0 0) are detectable at normal incidence without an external magnetic field. This is because that the THz radiation dipoles (polarization) oriented perpendicular to the GaAs (1 0 0) surface. However, we observed abnormal THz radiation from GaAs (1 0 0) coated with Au, as demonstrated by the results obtained from the sample with 8 nm Au film (Fig. 2). Even at normal incidence of the exciting laser, an obvious single-cycle THz electromagnetic signal (red circle, Fig. 2a) was detected,

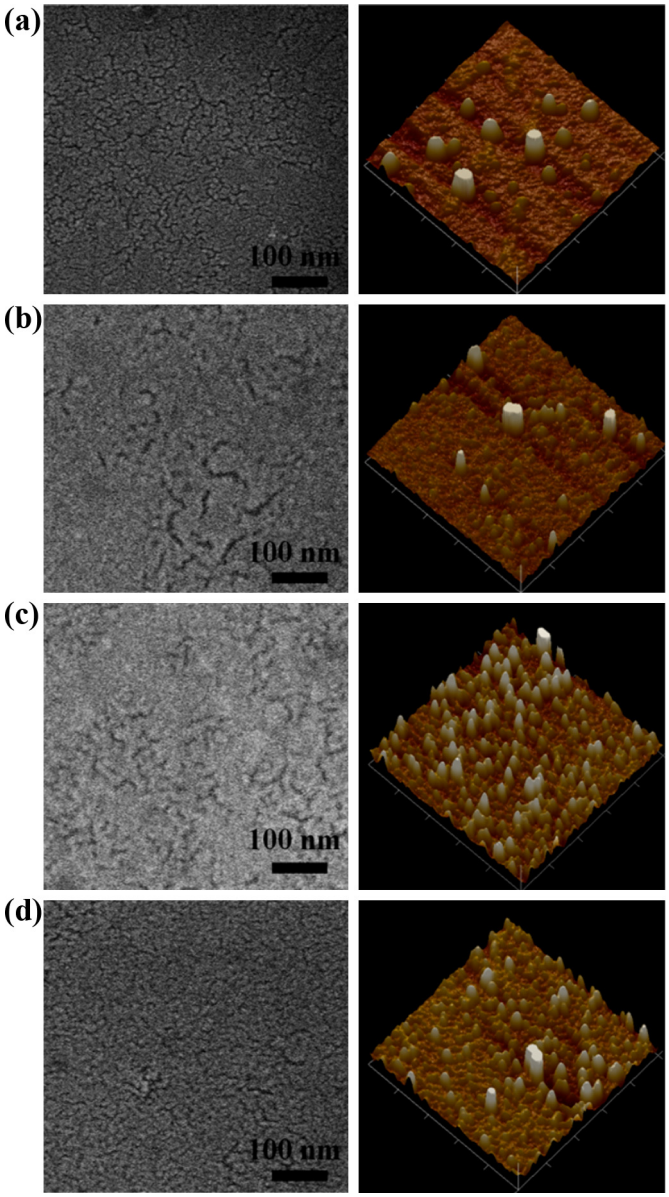

Fig. 1. Left panel: scanning electron microscopic images of SI-GaAs surface coated with Au film of various thicknesses: (a) 3 nm, (b) 5 nm, (c) 8 nm, and (d) 21 nm. Right panel: the corresponding AFM images scanned in 1 μm × 1 μm. The maximum brightness corresponds to a height of 23.2 nm, 9.2 nm, 13.1 nm, and 16.7 nm, respectively.

while there was no obviously detectable signal from the sample with the pristine surface (black square). This suggested that rough Au films lead to considerable radiation dipoles parallel to the GaAs (1 0 0) surface. Klatt et al. studied THz edged emission from metal-semiconductor edge on GaAs and they concluded that strong density gradient of photo-induced electrons and holes results in a photo-Dember polarization parallel to the surface where the exciting laser impinges at the edge of metalized stripe [16,17]. Our rough Au films show associated gaps in the cracks, which induced the intrinsic carrier gradient for the laser excitation with the polarization parallel to the surface.

Fig. 2b displays the peak-to-peak amplitude of the emitted THz electric field as a function of the excitation fluence. The radiation was observed to first increase with the incident fluence (till ~9 μJ/cm^2), then experienced a relative flat saturation process, and decreased when the power of the exciting laser increased further. With the increasing of incident fluence, the number of the carriers

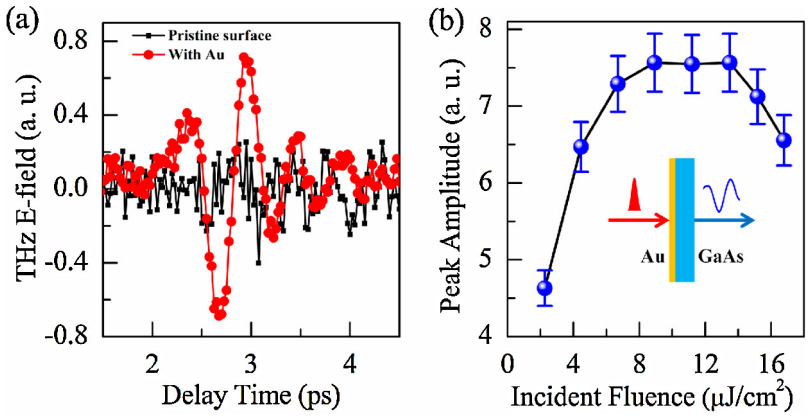

Fig. 2. (a) Time-domain waveform of THz radiation from the (1 0 0) surface of SI-GaAs without (black square) and with (red circle) Au coating (~8 nm) obtained at normal incidence of the exciting light; (b) peak-to-peak amplitude of the THz emission from the coated sample as a function of the excitation fluence. Inset in (b) shows the measurement geometry. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

will increase, resulting in the electromagnetic screening effect of photocarriers [18] and decreasing of the carrier gradient. This in turn brings the saturation and decreasing process to THz radiation.

Thickness dependent peak-to-peak amplitude of the THz emission under different incident angles was summarized in Fig. 3. For all the samples concerned, with the thickness of Au film varying from 3 nm to 21 nm, the oblique incidence at an incident angle up to 50° (maximum angle measurable by current THz system) always results in a steadily increasing emission. The increasing of THz radiation with incident angles can be explained by the dipole approximation radiation patterns as proposed by Zhang et al. [19]. Maximum THz radiation appears near the Brewster angle, which is ~75° for GaAs. In Fig. 3, for all the incident angles, the radiated THz wave first increases and then decreases with the Au film thickness, with the maximum appearing at ~8 nm. This is in accordance with previous reports, while cannot be explained by the models of optical rectification and photoconductition [12,14]. Noticeable, however, the THz emission from the sample with 20 nm Au coating is only about 5% less than that obtained in the case of 8 nm Au coating.

As stated above, the rough Au films could support surface plasmon excitation. In order to further explore the possible role of surface plasmons in promoting THz radiation from GaAs, THz emissions from samples with Au and Cr coatings, which are 8 nm thick and prepared under equal conditions, are presented in Fig. 4 for comparison. Wherein the results from the pristine surface is also presented. The excitation fluence for all these measurements was set at 9.0 μJ/cm^2. Cr is not a good choice for surface plasmon excitation [20]. There was no detectable THz emission from Cr/GaAs at normal incidence of the exciting laser. With both metal coatings, the polarity of the radiated THz wave reversed with regard to that from the pristine surface (comparing Fig. 4a and b with Fig. 4c). Meanwhile, the spectra from pristine surface are much narrower than those samples with metal coatings. At oblique incidences, the THz radiation amplitude from both Au/GaAs and Cr/GaAs was much stronger than that from the pristine surface. With 50° incidence angle, the radiation from Au/GaAs was roughly four times stronger than that from the pristine surface, while that from Cr/GaAs is approximately doubled.

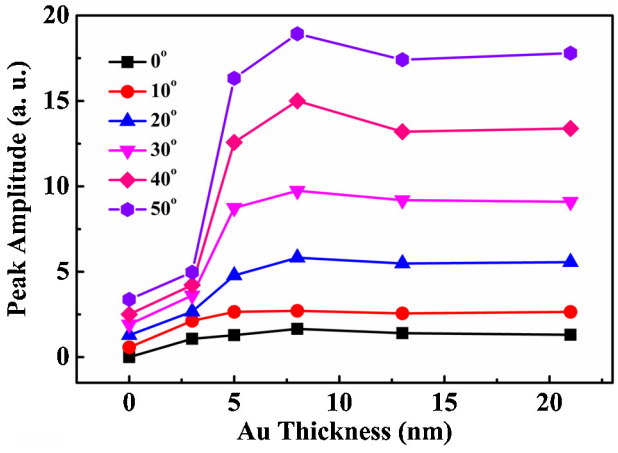

Fig. 3. Incident angle dependence of the peak-to-peak amplitude of THz emission from the (1 0 0) SI-GaAs surface with Au coating of various thicknesses.

Both the polarity reversal and enhancement of THz radiation at oblique angle from Au/GaAs have been observed previously [12,13]. The former was attributed to the opposite direction of the interface transient current while the latter has not yet been fully understood. To our best knowledge, the observation of THz radiation from Au/GaAs at normal incidence has not been reported. Recently, THz radiation was measured on Au films under high power excitation of femtosecond laser pulses [14], and it was reported that field emission of electrons could be generated by plasmons induced by the Au rough surface under the excitation of femtosecond laser of 80 MHz repetition frequency [21]. If the THz radiation from the Au/GaAs here concerned were due to either the high field induced optical rectification [15] or the free photoelectron acceleration [14], there should be detectable THz signals in the control samples, i.e., Au films (5 nm, 15 nm and 30 nm) deposited on silicon and quartz. However, no signals could be detected on the control samples, thus the free electron emission mechanism can be eliminated. It is quite reasonable that the free electron acceleration requires very high optical energy. Based on the aforementioned discussions, we postulate that the surface plasmons, which is related to the presence of the cracks contribution to the enhancement of THz radiation from the Au/GaAs surface.

Normally, prisms or gratings are used to bridge the momentum matching for surface plasmon excitation, while in our experiment, scattering from the Au cracks in the rough surface can work for the momentum matching automatically. Furthermore, the gaps in these cracks as revealed by SEM in Fig. 1 suggest high electromagnetic field in local region, which might lead to the enhanced THz emission. In principle, when gaps between Au cracks are in the sub-wavelength scale for the incident light (800 nm), the surface resonant enhancement of the local electromagnetic field was

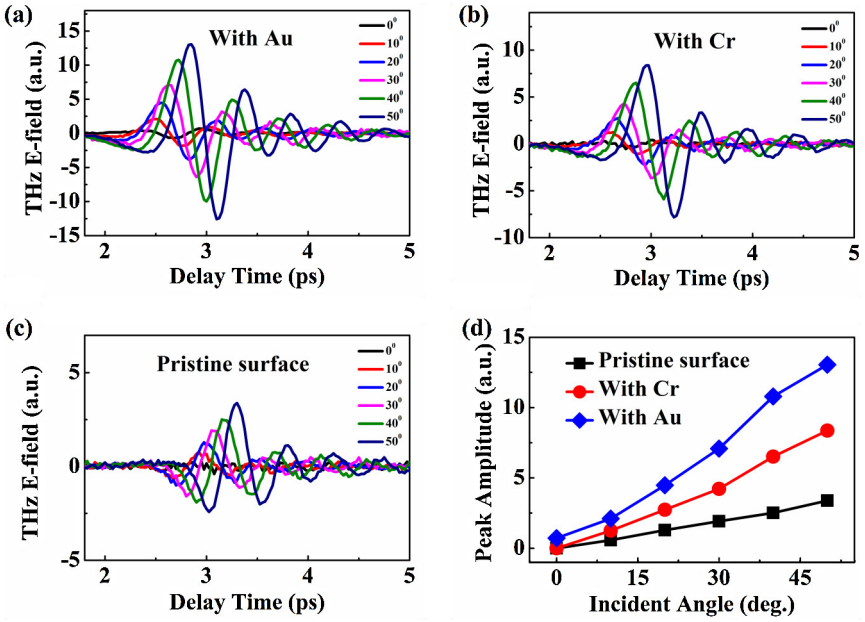

Fig. 4. Comparison of the time-domain waveform of THz radiation obtained at different incident angles for the samples with (a) Au coating (~8 nm), (b) Cr coating (~8 nm), and (c) a pristine surface. The peak-to-peak amplitudes for the three samples are summarized in (d).

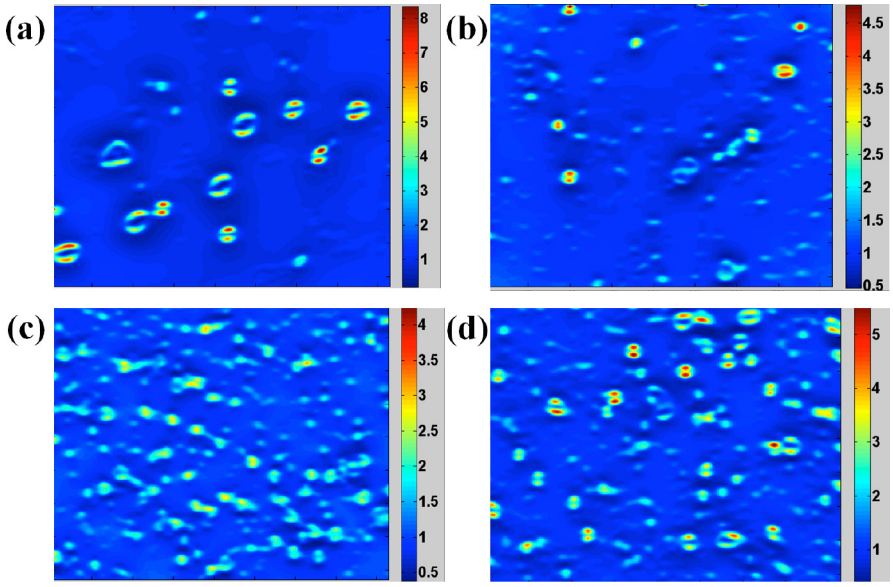

Fig. 5. Numerical simulation of the surface localized field based on the morphology of samples obtained with AFM referred in Fig. 1. Thickness of the Au coating: (a) 3 nm, (b) 5 nm, (c) 8 nm, and (d) 21 nm.

extremely huge [22,23]. We calculated the local field on the surface of Au/GaAs systems referred to Fig. 1 by using finite difference time domain method. It can be seen that there are a large number of hot spots on Au/GaAs surface (Fig. 5), where the electric field is huge that can induce quickly photo-carrier separation, which in turn causes enhanced THz emission. The calculation provides a qualitative picture for the possible mechanism governing the enhanced THz emission from Au/GaAs surface. This is different from the free photoelectron mechanism for THz emission from Au nanoparticles on glass. That the surface plasmon hugely enhanced the detection sensitivity of THz detector has also been demonstrated on the nanostructure-patterned GaAs antenna [3], which moreover verifies that the surface plamon plays a significant role in both THz emitters and detectors.

4. Conclusions

In summary, THz radiation was measured from (1 0 0)-GaAs surfaces covered by Au film of a thickness ranging from 5 to 21 nm, excited by femtosecond laser pulses at incident angles from 0° to 50°. The THz radiation is attributed to the lateral photo-Dember current due to the inhomogeneity of the cracks of the Au film. Enhanced THz radiation is also observed by the large local electromagnetic field by surface plasmons. The latter point is confirmed by the weak emission from the GaAs surface coated by Cr film of similar morphology. It is anticipated that the THz radiation efficiency can be further raised with specific design of nano-structures based on Au. Our results indicate the prospect of harnessing surface plasmons for the efficiency of THz emission with

controllable morphology of Au on semiconductor surface in THz photo-electronics.

Acknowledgments

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