Physica B 474 (2015) 64–69

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Broadband THz pulse emission and transmission properties of nanostructured Pt thin films

Mingzhe Hu [a,d,*], Kaijun Mu [b], Cunlin Zhang [b], Haoshuang Gu [c], Zhao Ding [d]

[a] Department of Physics and Electronics, Liupanshui Normal University, Liupanshui, Guizhou 553004, China
[b] Department of Physics, Capital Normal University, Yuquan Road 100082, Beijing, China
[c] Department of Electronic Sci&Tech, Hubei University, Xueyuan Road 430062, Wuhan, Hubei, China
[d] College of Electronics and Information, Guizhou University, Huaxi 550025, Guiyang, Guizhou, China

ARTICLE INFO

Article history:
Received 13 April 2015
Received in revised form
11 May 2015
Accepted 12 May 2015
Available online 14 May 2015

Keywords:
THz radiation
Ag nanowire
Impedance matching
THz transmission enhancement

ABSTRACT

The THz transmission and emitting properties of a composite metallic nanostructure, composed of Ag nanowires electrodeposited in an anodic aluminum oxide (AAO) template and a Pt thin film, were investigated by using a femtosecond pulse laser irradiation. The microstructure of the above sub-wavelength nanostructure was investigated by XRD, SEM, AFM and TEM. The results indicated that the thickness of the Pt thin film was about 200 nm and the Ag nanowire array had a sparse and random distribution inside the AAO template, with a length distribution in the range of 10–25 μm. The THz radiation properties of above sub-wavelength nanostructure indicated that the generated THz fluence from the Pt film was a magnitude of μW scale with a broadband frequency range and its subsequent transmission could be significantly improved by the better impedance matching property of the Ag nanowire embedded AAO film compared with that of the empty AAO film.

1. Introduction

High power and broadband THz sources have recently become an international focus due to their extremely important applications in space wireless communication, security imaging and drug detecting [1–3]. However, they have always become a hindrance because they cannot be simply generated by extending from neither side of microwave/millimeter region nor mid-infrared (IR) region. It was not until mid-1980s have several methods been developed for generating strong THz pulses by using femtosecond pulsed laser techniques [4–5].

The most typical method is the second-order nonlinear optical rectification (OR) in the non-centrosymmetric crystals, such as ZnTe, $LiNbO_3$ and InAs [6–10]. The laser field interacts with the second-order susceptibility tensor to produce a nonlinear polarization field, which radiates ultra-short THz pulse extending from 0.1 to 2.5 THz in a proper phase matching condition. The $LiNbO_3$ emitters have been able to generate terahertz pulses that have at least 1 μJ of energy. Another typical well known rectification process is the Auston switch antenna, where the photoconductive carriers, generated by incident laser field, are accelerated in a bias DC field, resulting in the emission of THz radiation [11–12]. Besides these semiconductor THz emitters, Hilton et al. [13–14] also found that THz pulse could be produced through OR in ferromagnetic films due to the demagnetization effect. More recently Gregor et al. and Filip et al. [15–18] revealed a novel coherent OR effect in metals, such as in gold or silver films, which could generate high power and large band width THz emission and gave a new possibility to produce compact THz sources. However, during the process, the generated THz power in metal surface was third to fourth order dependence on incident laser power in some certain domain, inconsistent with the traditional second order relationship in coherent OR effect. Gregor et al. [17–18] ascribed this to the surface plasmon effect in metals, which produces an evanescent field away from metals surfaces under the irradiation of femtosecond laser pulses. The involved free photoelectrons could be accelerated by the ponderomotive force in this evanescent field, resulting in the enhanced nonlinear emission of THz pulse.

Additionally, high efficiency THz sources require optical components such as beam splitters, dispersive optical substrates and transparent windows in broadband THz region. However, these components usually cause reflections at interfaces due to the mismatched refractive index, therefore, an impedance matching layer, operating at broadband terahertz region, is highly advantageous for high power and compact THz sources. Recently, Thoman et al. [19] demonstrated that randomly nanostructured gold films were suitable to work as efficient impedance-matching layers to

* Corresponding author at: Department of Physics and Electronics, Liupanshui Normal University, Liupanshui, Guizhou 553004, China.
E-mail address: guhs@hubu.edu.cn (H. Gu).

http://dx.doi.org/10.1016/j.physb.2015.05.013
0921-4526/

suppress the internal reflection at a silicon–air interface to less than 1%. And owing to their non-Drude behavior, these randomly nanostructured gold films hardly showed frequency-dependent sheet conductivity, which could be allowed to match the impedance over 0–3 THz broadband performance. However, high efficiency impedance matching layers with extended THz frequency range and more flexible controllable nanostructure are still highly desired.

In the present paper, we report a new composite metallic nanostructure for generating and transmitting THz pulses with powerful and broadband properties. The THz pulses were initially generated through OR process in the Pt metal film, and subsequently transmitted through the AAO template with randomly filled Ag nanowires. This sub-wavelength nanostructure possessed an efficient wave-impedance matching property and could enhance the THz transmission coefficient in a very broadband frequency range of 0.2–20 THz. This novel composite nanostructure provided a meaningful and flexible method for the development of high power and broadband THz sources in the future.

2. Experimental process

A ~200 nm thickness Pt thin film was magnetically sputtered on one side of a commercial AAO template with 200 nm pore diameter and 390 nm interpore distance. After a careful sealing process, the Ag nanowires were deposited into the above Pt film coated AAO template by using an electrodepositon method. The electrolyte was composed of 0.2 g/L $AgNO_3$ and 0.1 g/L citric acid. The electrodepositon voltage was −1.2 V and the electrolyte was stirred by magnetic field during the whole 40 min electrodepositon process. The AAO template was pre-annealed at 800 °C for 10 min before magnetic sputtering and electrodeposition process in order to resist the etch of electrolyte.

The phase assemblage was detected by an X-ray diffraction method using CuKα radiation (Bruker D8 Discover). The microstructure of the composite nanostructure was observed by a scanning electron microscope (SEM JEOL 6490) and a transmission electron microscope (TEM, Technai G20). The composition of the Ag nanowire was confirmed by TEM with an EDS energy spectrum from Oxford equipment. The surface microstructure of the Pt thin film was observed by using an atom force microscope (MicroNano AFM-III). The THz emission properties were investigated by the experimental setup illustrated in Fig. 1. A 1 kHz, 800 nm, 2.6 mJ and 50 fs Ti-sapphire laser beam (Spitfire Pro of Spectra-Physics) was focused by 15 cm focal length planoconvex lens. The THz radiation generated from the Pt film was collected by a parabolic mirror, and then interfered by a typical Michelson interferometer.

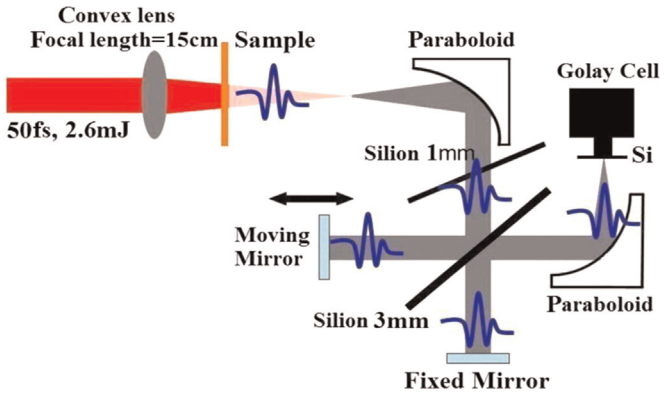

Fig. 1. Schematic setup for measuring the THz emission properties of the composite nanostructure.

The final THz signal was detected by a Golay Cell (Microtech Company), whose effective detecting frequency range is 0.2–20 THz with a polyethylene window. The whole experiment environment was in ambient pressure and N_2 atmosphere to avoid the THz absorption of water in air.

3. Results and discussion

3.1. Microstructure analysis

Fig. 2(a) illustrates the atomic force microscope (AFM) image of the Pt film magnetically sputtered on an AAO template with 200 nm pore diameter. As can be seen, the Pt film had a rather small grain size of about 150 nm with the root-mean-square (RMS) surface roughness of about 70 nm. These bumps on the surface of the Pt film could help focus the incident laser into a very strong field and enhance the nonlinear optical process, which was beneficial for both lowering the work function of the Pt film and the emission of surface plasmon [20–22]. Photoelectrons would be subsequently emitted by the surface plasmon through multiphoton process under the irradiation of femtosecond laser pulses, resulting in the emission of THz pulses. Both the TEM picture and the EDS spectrum clearly indicated the Ag nanowires with 200 nm diameter which were electrodeposited in the AAO template, as illustrated in Fig. 2(b) and (c) respectively. However, due to the high power of transmission electrons in TEM, the Ag nanowires were easily melted under long time irradiation, therefore, it was failed to obtain high resolution TEM pictures for the Ag nanowires. Through multiple SEM cross section pictures of the above sample, one could see that the thickness of the Pt film was around 200 nm and the Ag nanowires were randomly distributed in AAO template with a very low filling rate of about 10%. The length distribution of the Ag nanowires was uneven and in the range of 10–25 μm, which indicated that the electrodeposition speed under the present condition was inhomogeneous due to the bending and branching of the AAO pore channels. The growth rate of the Ag nanowires under this circumstance was about 240–600 nm/min.

The phase assemblage of the Ag nanowire decorated Pt thin film was detected by X-ray diffraction, as shown in Fig. 3. The Pt thin film coated AAO template was symbolized as sample A hereafter and the composite nanostructure sample, which was composed of the Ag nanowire array electrodeposited in sample A was symbolized as sample B hereafter. The Ag (111) diffraction peak could be clearly observed in the pattern (c), which corresponded well with JCPDF card 040783. However, the peak intensity was rather weak, which indicated that the distribution of Ag nanowires was sparse and the filling rate was quite low. The Pt thin film was poly-crystallized with (111) and (200) two planes, which corresponded with JCPDF card 040802 (see pattern b). Besides the diffraction peaks of Ag and Pt phases, the diffraction peaks of Al_2O_3 phase from the AAO template could also be seen (JCPDF card 88-0107, see pattern a), indicating that the AAO template was partially crystallized during the 800 °C annealing process, which was beneficial for the resistance of the etching of the electrolyte during electrodeposition.

3.2. THz radiation property

Fig. 4(a) shows the time domain spectra of THz radiation properties of samples A and B with the DC component being extracted. Fig. 4(b) illustrates the corresponding frequency domain THz spectra through Fourier transformation from Fig. 4(a). Both of the samples were irradiated normally on the Pt film side using 800 nm laser with 50 fs pulse and 2.6 mJ power in N_2 atmosphere. The pump beam was weakly focused onto sample surface with a