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Spectrum analysis of bio-chemical thin films on the surface of a semiconductor by terahertz surface plasmon polaritons

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Abstract: A new method for measuring terahertz (THz) transmission spectra of bio-chemical thin films placed on the surface of a semiconductor by terahertz surface plasmon polaritons (SPPs) was proposed. A strongly confined surface plasmon in the terahertz frequency range on the surface of semiconductor was theoretically proved, which can be used to enhance the light-matter interaction with bio-chemical layers above the semiconductor surface. By employing a free-space THz time-domain spectroscopy (TDS) system, the transmission power spectra of onion epidermis films with free space transmission and SPP transmission, respectively, were experimentally obtained. Experimental results show that SPP transmission spectra represent many characteristic absorption peaks for a layer of onion epidermis film, which are more than 100 times thinner than the free space wavelength of the terahertz wave.

Key words: bio-chemical thin film, terahertz wave, surface plasmon polaritons, spectroscopy
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基于太赫兹等离激元的半导体表面生化薄膜光谱检测

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摘要:提出了一种利用太赫兹表面等离激元对放置在半导体表面的生化薄膜进行光谱测量的新方法.从理论上证明了半导体材料对其传输的太赫兹表面等离子体波具有较强的表面束缚性,从而提高太赫兹波与半导体表面生化薄膜之间的相互作用.通过采用太赫兹时域光谱测量系统,从实验上分别得到了洋葱表皮的太赫兹表面等离子体波和自由空间太赫兹波透射波谱.实验结果表明,当测量对象是厚度仅为自由空间太赫兹波波长约1%的单层洋葱表皮时,表面等离子体波的透射波谱与自由空间太赫兹波透射波谱相比具有更加多的特征吸收峰.

关 键 词:生化薄膜;太赫兹波;等离激元;光谱

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Introduction

Terahertz (THz) waves are electromagnetic radiation in the frequency range of 0.1 to 10 THz. Because of its nonionizing nature and the distinctive absorption response of many molecules in the terahertz frequency range, there has been a lot of interest in using terahertz spectrum for chemical and biological analysis. However, since the wavelength of the THz region is several hundred microns, which is about three orders of magnitude larger than that of the visible and near infrared regions, it is very difficult to investigate sample films much thinner than the THz wavelength[1]. Artificial resonant structures provided a possible solution to this problem[2]. The sensitivity of the thin film detection could be significantly improved around the resonant frequency, so as to realize the THz spectrum analysis for a very thin film. However, the implementation of the THz spectrum analysis with those resonant structures is still challenging due to the difficulty of large area arrayed device fabrication[3].

By using terahertz surface plasmon polaritons (SPPs), which can exhibit subwavelength confinement of the terahertz electric field in the direction normal to a semiconductor interface[4-6], we realize both the high sensitivity and the large area spectrum analysis for a very thin bio-chemical film. In comparison with conventional THz spectroscopic method, the subwavelength field distribution of THz SPPs enhances interactions of light and matter with material in the region just above the semiconductor surface. Since the energy confinement of THz SPPs is related to the approaching degree between the frequency of SPPs and the semiconductor's plasma frequency, a high sensitivity in spectrum inspection is achieved by using a THz wave with frequencies slightly lower than the plasma frequency of the semiconductor. In this paper, we demonstrate spectrum analysis for biochemical films laid on the surface of an intrinsic indium antimonide (InSb) wafer with a carrier density of 1×10^{16} cm^{-3} and a plasma frequency of 2 THz at 300 K, thus showing that THz spectra in the range from 0.1 THz to 1.9 THz can be measured by the SPP spectrum analyzer with high sensitivity.

1 Principles of operation

SPPs can only propagate at the interface between two media with permittivities of opposite signs[7-8]. THz SPPs propagating along the surfaces of semiconductors resemble SPPs for metals at visible frequencies. However, SPPs with visible, near infrared, or even some THz frequencies cannot propagate along the surface of a semiconductor. This is because the real part of the semiconductor's permittivity is positive for SPPs with frequencies higher than the plasma frequency of the semiconductor. According to the Drude model[9], the permittivity of a semiconductor is related to the plasma angular frequency which is given by

$$\tilde{\varepsilon}_2 = \varepsilon + i\varepsilon' = \varepsilon_{\text{static}}\left[1 - \frac{\omega_p^2\tau^2}{1+\omega^2\tau^2} + i\frac{\omega_p^2\tau}{\omega(1+\omega^2\tau^2)}\right], \quad (1)$$

where $\varepsilon_{\text{static}}$ is the static permittivity; τ is the average collision time of the charge carriers; ω is the angular frequency of THz SPPs; ω_p is the plasma angular frequency, which can be expressed as

$$\omega_p = 2\pi f_p = \sqrt{\frac{Ne^2}{\varepsilon_0\varepsilon_{\text{static}}m^*}}, \quad (2)$$

where f_p is the plasma frequency; N is the free carrier density; e is the fundamental charge; ε_0 is the vacuum permittivity and m^* is the carrier effective mass.

Therefore, in order to support the THz SPPs propagation, the permittivity of the semiconductor should be negative and the frequencies of the THz wave should be smaller than the plasma frequency of the semiconductor. If the plasma frequency of the semiconductor is not high enough, we need to increase the carrier density of the semiconductor, which can be increased by chemical doping.

However, it does not mean that the plasma frequency should be as large as possible. This is because when the plasma frequency is increased, the decay length in the dielectric is also increased. Consequently, the energy is not confined on the interface, which may affect the sensitivity of the detection. The following calculations prove the estimation. According to the dispersion relation of SPPs[10], the propagation constant component with a direction normal to the semiconductor-dielectric interface (Z-axis) are given by

$$k_z = \frac{\omega}{c}\sqrt{\frac{\tilde{\varepsilon}_1^2}{\tilde{\varepsilon}_1 + \tilde{\varepsilon}_2}} = P + Di, \quad (3)$$

where $\tilde{\varepsilon}_1$ is the permittivity of the dielectric, i.e. the effective permittivity of thin film and air, $\tilde{\varepsilon}_2$ is the permittivity of the semiconductor, c is the speed of light in vacuum, P is the phase constant along the direction normal to the interface, D is the decay constant along the direction normal to the interface.

Therefore, we can calculate the wavelength and decay length in the dielectric along the direction normal to the interface, which are given by

$$\lambda_z = \frac{2\pi}{|P|}, \quad (4)$$

$$\delta_z = \frac{1}{|D|}. \quad (5)$$

According to Eqs. 1 ~ 5, we obtain the results as shown in Figs. 1 and 2. Figure 1 shows the relation between decay lengths of some THz SPP frequency components and plasma frequency of the semiconductor. We find that when the plasma frequency is increased, the decay length in dielectric is also increased. In order to achieve high energy confinement on the interface, we need to choose a semiconductor with a plasma frequency a little bit larger than the frequency range of the THz SPPs for spectrum measurement. Considering plasma frequencies of metals are much larger than the frequency of THz SPPs, the terahertz surface electromagnetic mode is only weakly bound on metal surface, thus metals are not suitable materials for supporting THz SPP propagation in our design. Figure 2 shows the wavelength of THz wave in free space and the wavelength of THz SPPs in dielectric along the direction normal to the interface. We found that

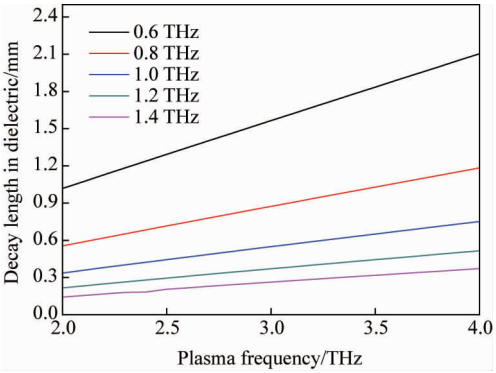

Fig. 1　The relationship between decay length in the dielectric and plasma frequency of the semiconductor
图 1　不同频率太赫兹表面等离子体波在介质中的衰减长度与半导体的等离子体频率之间的关系

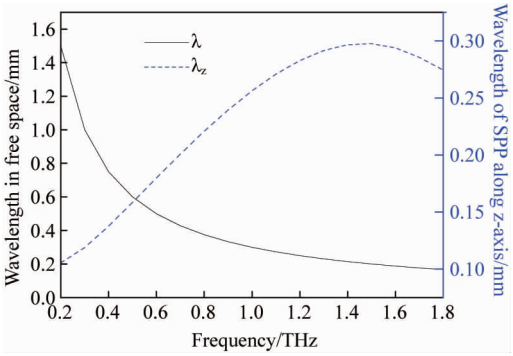

Fig. 2　The solid line shows the wavelength of THz wave in free space and the dashed line shows the wavelength of THz SPPs in dielectric along the direction normal to the interface
图 2　图中实线为太赫兹波在自由空间中的波长,虚线为相同频率的太赫兹表面等离子体波沿着垂直于半导体表面方向在介质中的波长

the wavelength of SPPs along the direction normal to the interface is much smaller than the wavelength of free space THz wave with the same frequency particularly in low frequency band, which is especially favorable for spectrum analysis of thin film.

2　Experimental results

In order to show that the THz SPP on a semiconductor is significantly more sensitive to a thin film than free space THz wave, we modify a traditional THz time-domain spectroscopy (THz-TDS) system[11] to obtain the transmission THz SPP power spectra. In the setup as shown in Fig. 3, a train of pulses from a Ti:sapphire laser is split into two beams. One of these beams constitutes the excitation beam while the other is the probe beam. The excitation pulses are employed to optically generate coherent broadband THz radiation from a low temperature grown gallium arsenide (LT-GaAs) surface field emitter and the THz radiation is coupled into THz SPPs propagating in the surface of a InSb wafer for optical modulation[12-13]. By varying the time delay between the two pulse trains, the coupled out THz pulse amplitude can be detected as a function of time with subpicosecond resolution. The InSb wafer used in the experiment is 0.5 mm thick with 2 inches in diameter. The size of the razor apertures is 300 μm and the distance between the two razor blades is 3.5 mm. The thin film used in the experiment is a layer of onion epidermis with a thickness of 8 nm. The measured THz pulses are Fourier transformed to obtain the transmission power spectra. The transmission spectra were measured with and without the thin film, respectively, and the ratio of these two spectra show the absorption peaks of the thin onion epidermis in THz band. Figures 4 (a) ~ (c) shows the time-domain, frequency domain and transmission absorption spectra. From Fig. 4(c), we found the absorption peaks are located at the frequencies of 0.234 THz, 0.468 THz, 0.82 THz, 1.05 THz, 1.28 THz, 1.52 THz and 1.76 THz, respectively.

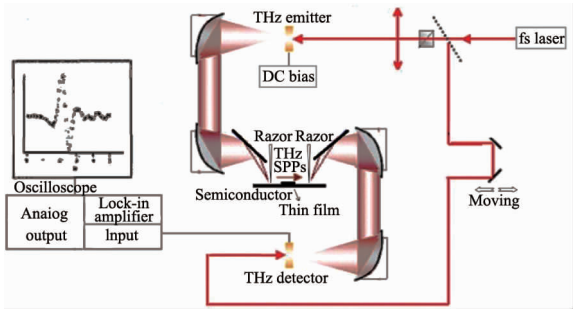

Fig. 3　Schematic of the THz-TDS to excite and to detect THz SPP pulses
图 3　太赫兹时域波谱系统产生与探测太赫兹表面等离子体波

For comparison purpose, we have also measured free space THz wave transmission absorption spectra. Figures 5 (a) ~ (c) show the absorption spectra of free space THz wave transmitting through one layer, two layers and three layers of onion epidermis. There are four obvious absorption peaks located at frequencies of 0.82 THz, 1.05 THz, 1.29 THz and 1.52 THz, respectively, as shown in Figs. 5(a) ~ (b). However, there are only two evident absorption peaks located at frequencies of 0.468 THz and 0.82 THz as shown in Fig. 5(c). The free space THz absorption spectra for one or two layers of onion epidermis represent the absorption peaks at high frequencies. This is because that high frequency THz wave components normally have relative small wavelengths, which are comparable to the thickness of the onion epidermis films. In contrast, low frequency THz components have relative large wavelengths, which are much larger than the thickness of the sample. Therefore, the low frequency THz wave cannot fully interact with the onion epidermis films and the distinctive absorption peaks in low frequency band cannot be represented. However, if we increase the thickness of the sample by adding the layers of onion epidermis film, we would find the distinctive absorption peaks in low frequency band as shown in Fig. 5(c). The THz signal in high frequency band is lost because that high frequency waves normally suffer

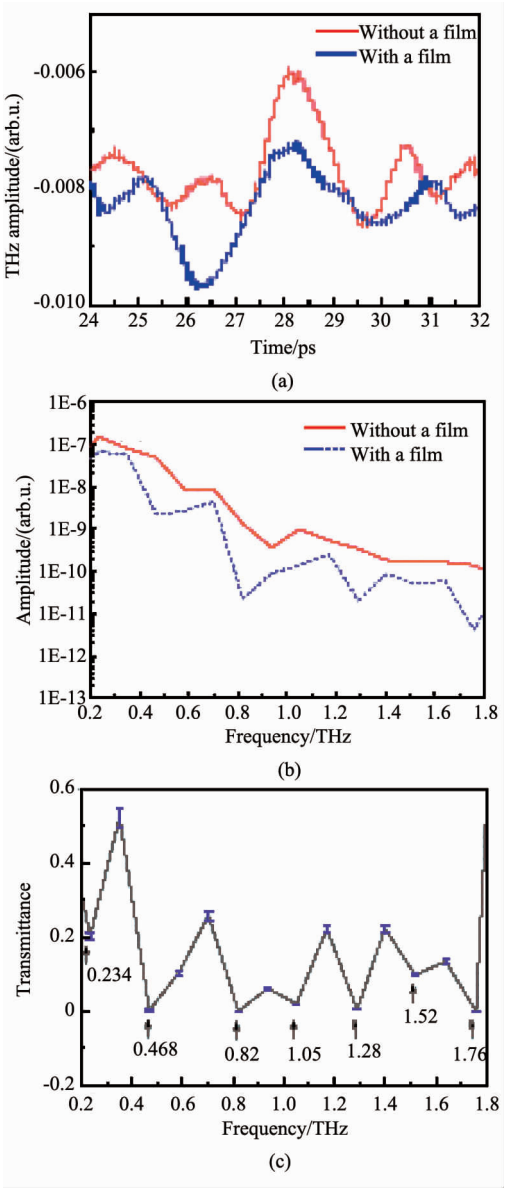

Fig. 4 (a) The time domain spectrum of the transmitted THz SPPs (solid red line) and the time domain spectrum of the transmitted THz SPPs passing through the thin onion epidermis (dashed blue line), (b) The frequency domain spectrum of the transmitted THz SPPs (solid red line) and the frequency domain spectrum of the transmitted THz SPPs passing through the thin onion epidermis (dashed blue line), (c) Absorption spectrum of the transmitted THz SPPs passing through the thin onion epidermis

图4 (a) 太赫兹表面等离子体波不经过(红色实线)与经过(蓝色虚线)单层洋葱表皮所测时域光谱图,(b) 太赫兹表面等离子体波不经过(红色实线)与经过(蓝色虚线)单层洋葱表皮所测频域光谱图,(c) 太赫兹表面等离子体波通过单层洋葱表皮所测吸收光谱

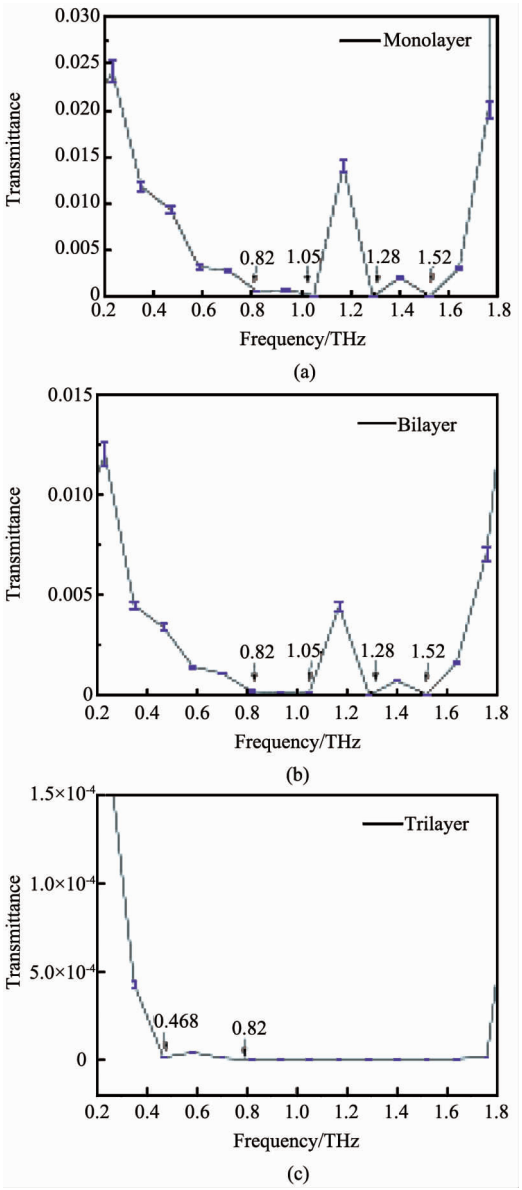

Fig. 5 Absorption spectra of free space THz wave transmitting through (a) one layer, (b) two layers and (c) three layers of onion epidermis

图5 自由空间太赫兹波通过(a)单层、(b)双层、(c)三层洋葱表皮的吸收光谱

more Rayleigh scattering[14] when we add scattering materials.

By comparing Fig. 4(c) with Figs. 5(a) ~ (c), we concluded that THz SPP transmission absorption spectra represent distinct absorption peaks with high sensitivity and precision in a wide frequency band. Transmission absorption spectra with free space THz wave for one bio-chemical film can represent absorption peaks in high frequency band rather than low frequency band. However, information of multilayer bio-chemical films is missing in the transmission absorption spectra at high frequency band due to Rayleigh scattering. Therefore, compared with traditional free space transmission method, the new method using THz SPPs is more sensitive for thin film THz spectrum analysis in a wide frequency range.

3 Conclusions

A new method to measure distinct absorption peaks of a thin film by using THz SPPs was proposed. Theoreti-

cal results prove that THz SPPs have a relative small wavelength in the direction of thickness dimension compared with free space THz wave in low frequency band, which is especially favorable for spectrum analysis of very thin film. Experimental results demonstrated that it is feasible to use semiconductor SPPs to interrogate the terahertz optical properties of thin bio-chemical analytes with high sensitivity in a wide frequency range. Experimental results show that direct transmission spectra cannot represent absorption peaks in a wide band with a single measurement.

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