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Highly-Enhanced Plasmonic Biosensors based on Atomically Thin Two-Dimensional Chalcogenide Phase-change Materials

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Abstract: We designed an enhanced plasmonic sensing device based on 2D Ge₂Sb₂Te₅ phase change nanomaterials. The sensing capability has been experimentally demonstrated to be 7000,000 μm/RIU with a detection limit of 10 fM for BSA molecules. © 2020 The Author(s)

1. Introduction

Surface plasmon resonance (SPR) sensors have been exploited as an effective tool for real-time and label-free biochemical detection over these years [1]. They are used to detect the optical signal change in the reflected light beam at the sensing interface where surface plasmon polaritons are excited by the incident light. This signal change is strongly dependent on the evanescent field perturbation induced by the molecular binding at the plasmonic interface. However, the sensitivity of those sensors is not able to detect target analytes with low molecular weight (less than 400 Dalton) by the conventional angular scanning method. To overcome this challenge, we proposed an enhanced plasmonic sensing technique through the phase singularity-related Goos-Hänchen (GH) interrogation method with an enhanced zero-reflection performance by the atomically thin phase change nanomaterials [2-4]. We constructed an optimized multi-layered metallic sensing substrate based on GST and gold thin film. Experimental results show that the sensitivity in Goos-Hänchen (GH) shift has been greatly enhanced compared to pure gold substrate by more than one order of magnitude. Bovine serum albumin (BSA) molecules with low concentrations ranging from 10 fM to 10 μM have been successfully detected. Thus, we believe that this device has great potential in detecting chemical and biological reactions with ultra-high sensitivity especially for the clinical diagnostic usages.

2. Results and discussion

In this paper, we designed a novel configuration of multi-layered SPR sensor based on the Kretschmann scheme. To achieve a significant phase singularity through zero-reflection effects, a thin layer of GST materials with the thickness of 2 nm is deposited on the top of the gold thin film (40 nm). These atomically thin phase change materials are known to have a high absorption rate for the light waves in visible and near-infrared region. The dielectric constant of GST material is measured to be 13.00+11.10i by spectroscopic ellipsometry (SE), which indicates its high absorbance. It is worth mentioning that the thicknesses of both GST and gold layer are optimized to obtain the maximum sensitivity. We utilized transfer matrix method to simulate the reflectivity of the substrate based on this metastructure. GH shift [5], which is the lateral position shift between the reflected TM-polarized and TE-polarized light from the sensing substrate, is used as the sensing signal to extract the information of analytes in the microfluidic chamber. According to the stationary phase approach, GH shift is defined by $GH = -\frac{1}{k} \frac{\partial \phi}{\partial \theta}$. Therefore, the largest GH shift is achieved when the sharpest phase change occurs, which corresponds to the minimum reflectance achieved at surface plasmon resonance angle.

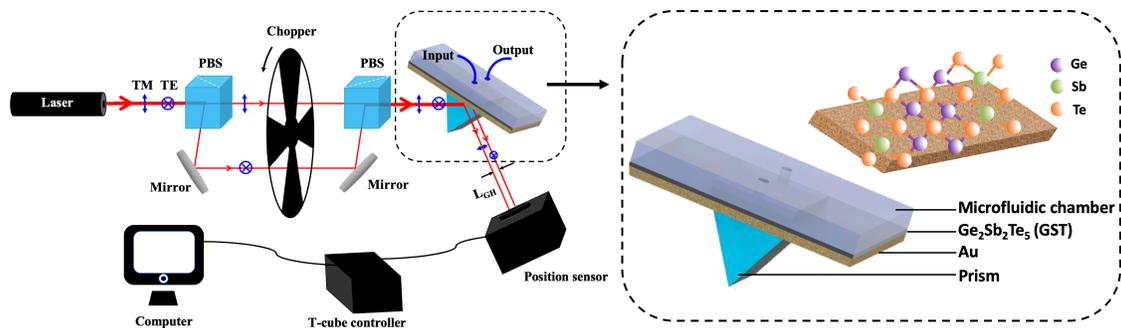


Fig. 1. Optical setup for measuring differential GH-SPR shift between TM- and TE-polarized light based on GST-gold metastructures.

Based on our theoretical modeling, the maximum GH lateral shift could reach 2500 μm at the resonance angle, which is 100 times higher than the ones with standard 40 nm pure Au thin film. Fig 1 shows the schematic diagram of our experimental setup. The incident light beam from He-Ne laser is splitted into TM-polarized and TE-polarized light beam. The prism is placed at the translation stage and fixed at the surface plasmon resonance angle to achieve the largest GH shift change. The sensing substrate is integrated with a microfluidic chamber to realize convenient transportation of sample solutions, which also provides possibilities to realize multiplexed detection in a single chip. An optical chopper is placed to ensure that only one of the polarizations can reach the sensing substrate at a time. The position of reflected light beam is recorded by a position sensor.

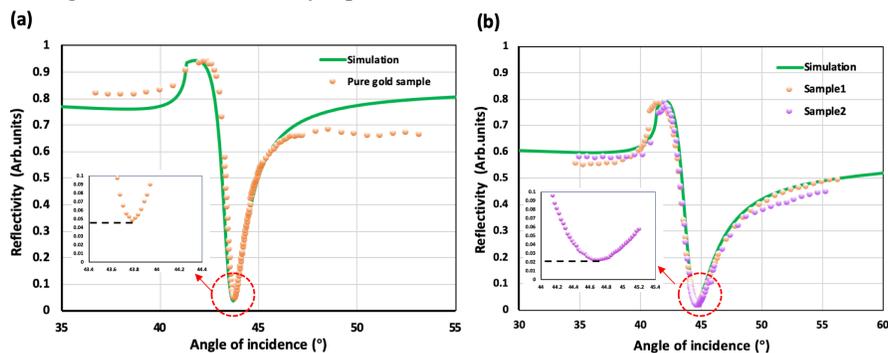


Fig. 2. Reflectivity spectra of (a) pure gold thin film substrate (b) multi-layered substrate with GST.

Fig. 2 illustrates the angular scanning reflectivity spectra of both pure gold thin film substrate and multi-layered substrate based on GST nanomaterial. The experimental measurements show good accordance with theoretical calculations. As clearly shown in the figure, the presence of GST material can lead to a deeper resonance dip (minimum intensity lowered by 50%), which led to a much higher GH shift.

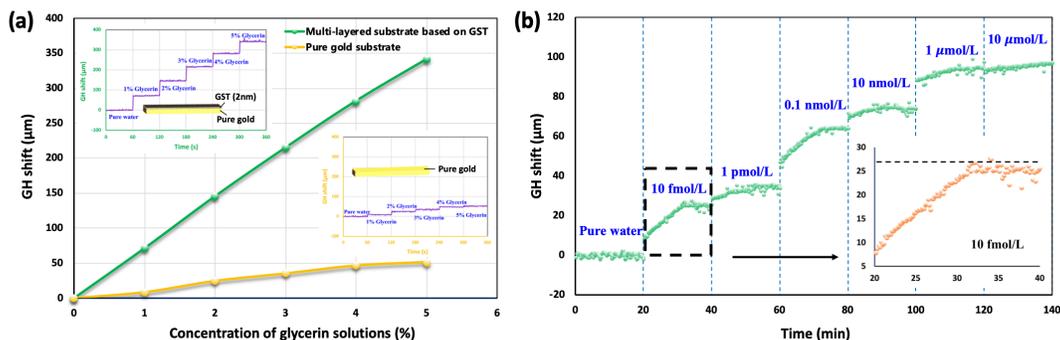


Fig. 3. (a) GH shift of sample solutions with different concentration. (b) Real-time GH shift of BSA solutions from 10 fM to 10 μM .

As shown in Fig 3(a), the GH shift has a good linear relationship with glycerin in different weight ratios. The GH shift for 1% glycerin (0.0012 RIU) was measure to be 70 μm . Thus, the sensitivity of our device can reach 7000,000 $\mu\text{m}/\text{RIU}$, which is more than 5 times higher than the pure gold SPR substrate (50 nm). In the biosensing experiment, we first injected (3-Aminopropyl) trimethoxysilane solutions into the microfluidic chamber to enhance the sensing signal by functionalizing the sensing substrate with alkoxysilane molecules. Then, BSA solutions with molarity ranging from 10 fM to 10 μM were injected to the chamber sequentially. In Fig 3(b), the gradually increasing GH signal indicates the binding process of BSA molecules onto the substrate, which demonstrated the capability of our device to realize real-time biosensing. The minimum detectable concentration proves to be lower than 10 fM. The GH shift has increase up to 25.49 μm after the binding process of 10 fM BSA molecules. We believe this device has great potential in sensing minute RI change, which is of great importance in biosensing applications for small cancer biomarkers and heavy metal ions in the human serum. In the near future, we will further fine tune the optical absorption of the GST materials through thermal heating due to the phase change properties of the metastructures.

3. References

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