Optical rectification in dielectrically modulated metallic thin film

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Introduction

Optical rectification(OR) is one of 2nd order nonlinear effects which generates DC polarization in a material without space inversion symmetry. When a material with free carrier is irradiated by light, voltage is generated in the material due to the DC polarization induced by OR effect. In terms of momentum conservation law, the voltage is attributed to the momentum transfer from light to free carrier in the material. Historically such effect is investigated in semiconductors and known as photon drag effect. Vengurlekar et al. extended the photon drag effect to metallic thin film with the use of field enhancement effect of surface plasmon polariton (SPP) excited with a prism coupler[1]. Recently Hatano et al. investigated the photon drag effect in metallic grating slabs and evaluated the voltage numerically in terms of momentum conservation law[2]. Although numerical calculation in Ref[2] obtained qualitatively good agreement with experiment, detailed features such as magnitude of the voltage and sharp resonance structure were not reproduced. This indicates that in some cases momentum conservation used in Ref[2] is quite insufficient to evaluate photo-induced voltage(PIV).

The purpose of this study is to demonstrate the anomaly of momentum conservation approach to the evaluation of PIV experimentally and to construct a method for the numerical calculation of PIV from the microscopic point of view.

Samples and Experimental setup

Using electron beam lithography, we fabricated periodic structure composed of 210 nm-thick electron beam resist (ZEP520A, ZEON Corporation) with the periodicity 700 nm on 50 nm-thick Au film as seen in the schematic in Fig.1(a).

The experimental setup is shown schematically in Fig. 1(b). Idler light from an optical parametric oscillator (OPO) pumped by a tripled Nd:YAG laser was sent to the sample with p-polarization, for which surface plasmon polariton is excited. We measured photo-induced voltage across the sample with an oscilloscope (Tektronix TDS3012B) through a high-speed amplifier with a gain of 125.

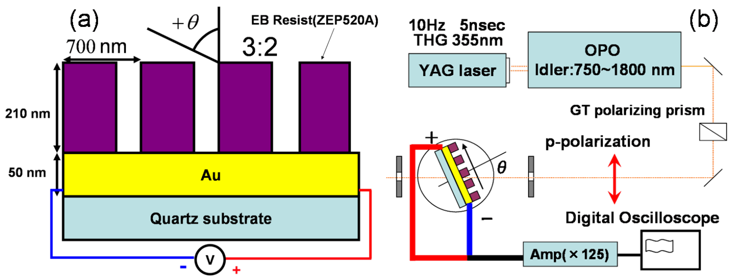

Fig. 1: (a):Schematic of the sample, (b):Experimental setup.

Results and Discussion

In Fig.2(a), angle resolved reflection spectra are shown as a function of wavelength. There are some dips that shift in association with the change of incident angle, which corresponds to the SPP excitation. The characteristic feature of PIV spectra shown in Fig. 2(b) also shifts in association with the SPP excitation. Thus SPP excitation dependence of PIV is clearly shown in Fig.2(b) and it is considered that the generation mechanism of PIV is attributed to the SPP resonance. In Fig.2(c) we numerically calculated the PIV spectra by the method used in Ref[2] and the result is shown. Although same feature in Fig.2

(b), the SPP dependence, was well reproduced, the polarity of PIV is reversed compared to the experimental result. Thus momentum conservation approach to the evaluation of PIV fails to reproduce the experimental result in the sample. Approach used in Ref[2] utilizes optical spectra such as transmission and reflection, which is evaluated *far away* from the sample. However SPP strongly enhances *near field*. So the previous approach cannot include near field enhancement effect properly. Moreover when using the momentum conservation approach we cannot distinguish the amount of momentum transfered from light to free carrier in the metal and to the bound electron in the dielectric , while only the metallic thin film contribute to PIV. These are the possible reason why the previous approach cannot explain the experimental result in this abstract.

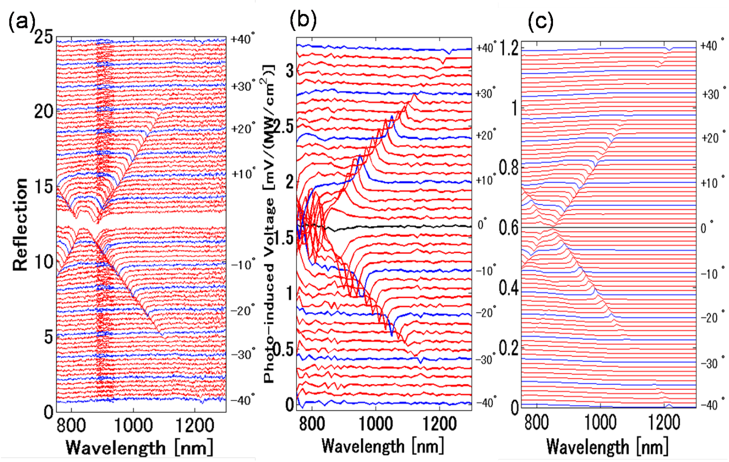

Fig. 2: (a):Numerical calculation of reflection Spectra. (b): Experimental result of PIV measurement. (c): Numerical calculation of PIV spectra evaluated by momentum conservation.

Here we take another approach to include near field effect correctly. Using complex polarizability $\alpha = \alpha_R + i\alpha_I$, electromagnetic DC force acting on free carrier is generally given as [3]

$$\boldsymbol{F}_{DC} = \frac{\alpha_R}{4}\nabla|\boldsymbol{E}|^2 + \frac{\alpha_I}{2}\sum E_j \nabla E_j^*.$$

The first term in the equation above, which is called as gradient force, vanishes when integrating it on the whole volume of metallic plane film due to the periodic boundary condition. So the 2nd term, which is called as scattering force, is the origin of the PIV here. The primary difference between previous approach and this one is that in the momentum conservation approach far field electromagnetic field is used to calculate momentum transfer from light to the material system, while in this approach microscopic electromagnetic field is used to calculate the force acting on electron. Figure 3 shows the microscopic calculation result of PIV spectrum with the experimental result. Numerical calculation readily reproduced the experimental result. So near field effect, the SPP field enhancement, is correctly included in the calculation and scattering force is the origin of the PIV here.

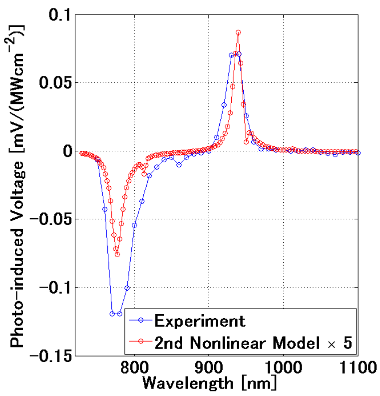

Fig. 3: PIV evaluated by microscopic alculation(blue) and experimental data (red). Incident angle : 8 degree

Conclusion

We have experimentally and numerically investigated PIV in dielectrically modulated metallic thin film. While momentum conservation approach used in Ref[2] gives wrong sign for PIV at the SPP excitation, microscopic calculation used in this paper correctly reproduced it. It is also clarified that scattering force is responsible for the generation of PIV in this structure.

References

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