16a-D5-1

# Nonlinear Plasmonics in Metal-Insulator-Metal Structure

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## 1. Introduction

Surface plasmons (SPs) in a metal-insulator-metal (MIM) structure couple with light without using attenuated total reflection (ATR) geometry or a periodically corrugating surface. At the resonance condition, enhanced electric fields are produced adjacent to the metallic surfaces, similar to the SPs excited in the ATR geometry. Because of the simple optical geometry, the MIM is useful for practical applications of SPs. So far, the MIM structure has been used for optical filters [1] and biosensors [2]. However little is known about the nonlinear optical (NLO) properties of the MIM structures. In this paper, I will talk about two NLO phenomena in MIM structures: one optical second-harmonic generation (SHG) and the other optical bistability.

## 2. Second-Harmonic Generation (SHG)

SHG is one of the second-order NLO phenomena, which are inhibited in a system with inversion symmetry under the electric dipole approximation. Since the metallic surfaces face each other in the MIM structure at which the enhanced electric fields are induced, the second-harmonic (SH) fields from the nonlinear electric dipoles produced at the metallic surfaces are canceled because they are out of phase. In other words, the MIM structure has local inversion symmetry. However, enhanced SH light was actually observed at the resonance angle of SP.

The SH intensity profile as a function of the angle of incidence leads to conclude that the SH light observed in the MIM structure is mainly from bulk nonlinear polarization (quadrupoles or magnetic dipoles) in the gold layer rather than from electric dipoles at the gold surface, where inversion symmetry is broken. The nonlinear bulk polarization $\boldsymbol{P}^b$ produced by incident light $\boldsymbol{E}$ is described as

$$P_i^b = \gamma\nabla_i(\boldsymbol{E}\cdot\boldsymbol{E}) + \zeta(E_i\nabla_i E_i)$$

where $i = x$, $y$ or $z$. Coefficients $\gamma$ and $\zeta$ are certain phenomenological constants. The variation of the electric field in the metal is the origin of the second-order nonlinear polarization. Although the surface nonlinear polarization is also generated, the SH fields from both metal surfaces in contact with the insulator layer is canceled and SH light from bulk nonlinear polarization is dominantly observed. The comparison of the SH intensity from a quartz crystal used as a standard, the effective susceptibility for the angular-independent nonlinear bulk susceptibility is found be a few hundreds pm/V.

## 3. Optical Bistability

Optical bistability is a nonlinear optical (NLO) phenomenon that is applicable to optical signal processing and optical memories. A number of all-optical bistable devices that are free from electrical circuits have been reported. These are based on the Fabry-Perot cavity, in which an NLO medium layer is sandwiched between a pair of half mirrors. Such materials show fast response, but strong illumination (more than 1 kW/mm$^2$) is necessary because the optical nonlinearity of the NLO materials is usually low.

A combination of MIM structure and liquid crystals (LCs), a low-power all-optical bistable device can be realized [3]. LCs are materials with large anisotropy, and their refractive indexes can be controlled by adjusting the temperature, which induces changes in their order parameter or the phase transition from the nematic to isotropic phase. This feature leads to a high optical nonlinearity based on the thermooptic effect. The lowest threshold switching illumination achieved was 0.3 mW/mm$^2$, which is extremely low. The threshold illumination is lower at higher temperature up to the phase transition of LC. The LC device is promising for two-dimensional optical memories or spatial light modulators, since the structure is simple and free from electronic circuits.

**Acknowledgements**

This work has been performed with Dr. R. Fujimura, Mr. D. Teshima and Mr. Pham Tien Thanh of Tokyo Institute of Technology.

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