

The Postech Times

No. 99

WED., Nov. 9, 2016

<http://times.postech.ac.kr>

First Edition SEP. 2, 2009

Publisher: Doh-Yeon Kim | Professor Editor: Jinhee Kim | Editor-in-chief: Choi Jong-hyeok | eng-reporter@postech.ac.kr | 77 Cheongam-Ro, Pohang, Gyeongbuk, KOREA | Tel 82-054-279-3718

Academics Understanding and control of surface electron dynamics in metal by optical frequency comb

Surface Plasmon (SP) is a collective electron oscillation at metal dielectric interface excited by an incoming light. The resonant interaction between the charge oscillation and the electromagnetic field of the light at surface constitutes the SP and gives rise to its unique properties. Interest in SPs has been renewed because of the recent advances in nanotechnology that allow one to fabricate and characterize metals on the nanometer scale.

One of the aspects of SPs that has attracted researchers in the field of optics and science is the possibility in which one can use SPs to concentrate and channel light in a sub-wavelength scale. This might lead to miniaturized photonic devices with length scales much smaller than those currently achieved. An appealing feature is that, when embedded in dielectric materials, the nano-structures used to propagate SPs can also be used to carry electrical signals. Developments such as this open the prospect of a new branch of photonics using SPs, called plasmonics.

Concentrating light in this way leads to an electric field enhancement that can be used to manipulate light-matter interactions and boost non-linear phenomena. The ability to dynamically control the plasmonic properties of materials is a key to develop novel nanoscale photonic electronics. So far, many studies on characterizing the plasmonic properties have been reported. For example, localized electric field enhancement, enabling dramatic enhancement of Raman signal, which is also known as Surface Enhanced Raman scattering, has been investigated using various measurement techniques so that the enhanced field associated with SPs makes them suitable

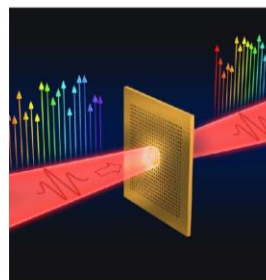
for use as sensors. Commercial systems have already been developed for sensing biomolecules. SP-based sensing applications

SPs are hence of interest to scientists, ranging from physicists, chemists, and materials scientists to biologists. This in turn has enabled us to control SP properties to reveal new aspects of their underlying science and to tailor them for specific applications. For instance, SPs are being explored for their potential in many fields of science and applications such as bio/chemical spectroscopy, optical communication, and nanoscale photonics.

Since SP is the electron oscillation in a metal (condensed matter), the electrons' motion can physically be interrupted by the Ohmic loss or dispersion by the material that possibly causes the difference/distortion between electron oscillation frequency and excited electromagnetic (light) frequency. Unfortunately, this study has not been investigated and yet it has been assumed that the collective electron oscillation can precisely follow the frequency of excited light frequency. In this work, we have carried out a series of experiments to clarify this hypothesis. We have used a well defined or very exact light frequency to excite the SP and measure the frequency difference.

As an exact frequency tool, we have used the optical frequency comb, millions of optical modes whose frequencies are equally spaced to prove our hypothesis. Because the optical frequency comb can be locked (referenced) by an atomic clock (which is known as precision frequency standard), each optical frequency comb can be used as the reference frequency to evaluate the oscillation frequency characteristics of SP. Among many ways of

exciting SPs to evaluate the frequency characterization, in our experiment, we especially exploited sub-wavelength-sized nano-hole array fabricated on a thin gold film (~100 nm thickness). According to classical optical theory, if the diameter of a nano-hole is smaller than the wavelength of light, the transmitted light suffers from huge loss, resulting



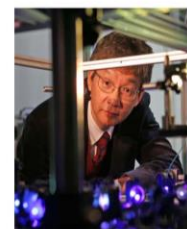
▲ Transmission of light through Nano-hole array

in no light transmission. However, thanks to SP, the light energy is converted into the SP on the metallic surface. These SP will be in resonance with the pitch of the nano-hole and funnel through the nano-hole. The transmitted SPs re-radiate in the form of electromagnetic wave. This phenomenon is well known and called "Extraordinary optical transmission of light," which was firstly observed by Thomas Ebbesen in 1998. This transmitted electromagnetic wave has the information during the transformation from light wave into SP. By measuring this frequency of light, we can understand the quality of SP frequency.

Interestingly, we experimentally found that

the frequency stability of the transmitted light is almost the same as the stabilized optical frequency comb. This result was reported in *Nature Communications* in 2016. This implies that the SP does not degrade the quality of optical frequency comb. In other words, the SP can oscillate in the same way as a light wave.

In a practical sense, we can exploit of the optical frequency combs in plasmonics in various advantageous ways. The optical frequency comb can be used in ultra-precision spectroscopy, quantum information delivery, or optical rulers. All these intriguing applications can be integrated in plasmonic device, enabling the system size down to nanoscale. Our finding indicates that a plasmonic device can work in optical frequency to full degree, but further investigation in other plasmonic system or demonstration of novel plasmonic devices is still to come.



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