Origin of Shape Resonance in Second-Harmonic Generation from Metallic Nanohole Arrays

The physics of a system with broken symmetry is rich and interesting. Second harmonic generation (SHG) is one of the most useful methods for investigating surfaces, since it is only sensitive to the few layer surface atoms with broken spatial symmetry. Ultrafast surface SHG is especially useful for investigating nonlinear optics in nanophotonics. Although nano-optics is seen as a natural extension of conventional optics, the nonlinear optics aspect is non-trivial. Recently some groups have found that geometric shape greatly affects the nonlinear signal intensity in a nano-system. With identical area, the one with optimized shape can lead to two orders of magnitude enhancement in the SHG intensity. However, so far there is no clear understanding about the physical origin of this shape resonance effect.

Associate Professor ZHAO Jimin from State Key Lab for Surface Physics, Institute of Physics, Chinese Academy of Sciences has developed techniques of detecting weak SHG signal and 2fs resolution ultrafast spectroscopy. He and Professor LI ZhiYuan from Lab of Optical Physics, Institute of Physics, Chinese Academy of Sciences collaborated together to investigate this problem experimentally and theoretically. They conclude that the by-default modal spatial overlap that is fulfilled in conventional nonlinear optics may not fulfill automatically at nanoscale. It is greatly modified by the shape.

ZHAO Jimin and PhD student WANG Rui prepared gold film on glass substrate and milled rectangular-hole array on it, with different shapes (marked by the aspect ratio, AR, Figure 1). They used the technique developed in ultrafast spectroscopy to detect the SHG signal (10^{-10} of the fundamental wave signal) and found consistent results with those reported by other groups. Then they used the 2fs sub-pulse precision technique to measure the effect of light slow down. They found there is no strong correlation between the light slowdown and the shape. Thus slow light effect might

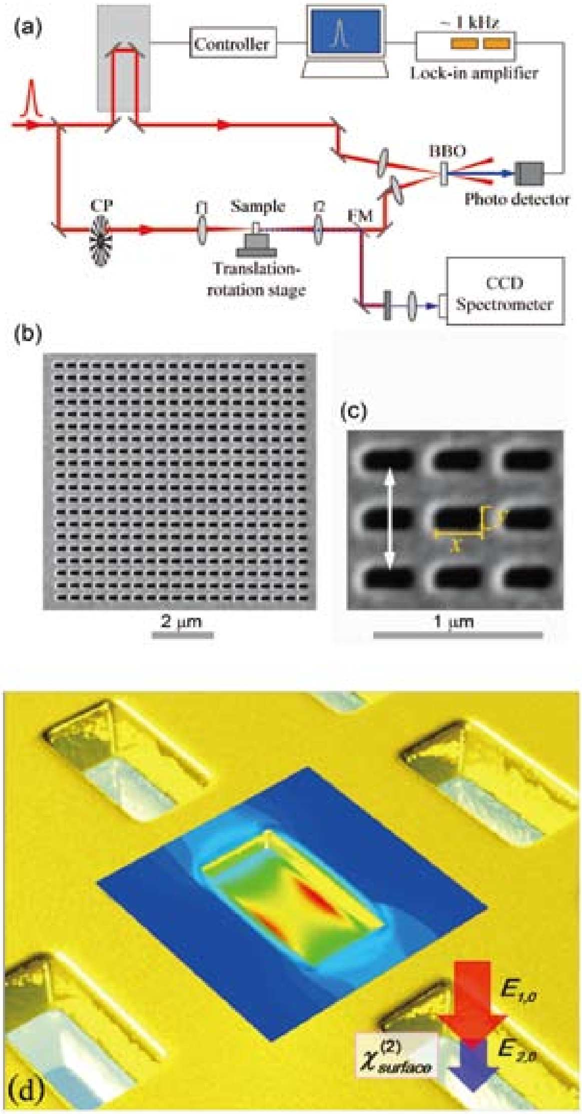

Figure 1: (a) Experimental setup; (b) and (c) Metallic nanohole array samples; (d) SHG due to nonlinear interaction of FW and SHW modes within each metallic nanohole. (Image by IOP)

not be the major reason leading to the shape resonance effect. This refreshes the previous understanding held by the community.

To further clarify the physical mechanism underlying the observed SHG shape resonance phenomena in nanoscale nonlinear optics, LI Zhiyuan and PhD student WANG Benli developed a nonlinear coupled-mode theory to solve energy conversion from fundamental wave (FW) mode to second-harmonic wave (SHW) mode within the nanoscale air hole. They started from nonlinear coupled wave equations well-established in nonlinear optics and solved the optical properties, including the dispersion relation, modal profile, and attenuation, of the transmission mode within each metallic nanohole for both fundamental wave (FW) and second-harmonic wave (SHW). Under the excellent approximation of single transmission mode, a nonlinear coupled mode theory was set up and analytically solved, leading to a very simple explicit solution to SHG.

The analytical theory reveals that several physical mechanisms, including the FW mode excitation amplitude, FW-SHW modal spatial overlap, FW-SHW mode phase mismatch, and SHW mode attenuation, all make contribution to SHG. They are all geometric shape sensitive and altogether act to induce the SHG shape resonance effect (Figure 3). In short, the SHG shape resonance can be ascribed to the strongly shape-dependent intrinsic phase and attenuation feature of the FW and SHW waveguide mode and the external excitation efficiency of the FW mode. The theory agrees well with experimental observations that the SHG signal intensity can be three orders of magnitude larger at a specific hole shape (namely, a specific value of AR) than at other shapes. Besides, the theory also indicates that the AR value where resonant SHG occurs is closely related to the nanohole area. This simple theory provides an accurate and complete explanation for the long-emphasized but elusive shape effect.

This study, in particular, the analytical nonlinear coupled-mode theory, may stimulate new angles and deeper insights to visualize general nonlinear nanophotonic processes and pave the way to engineering high-efficiency nonlinear nanophotonic structures. It is expected that their finding might inspire a very useful insight into other general nanoscale nonlinear optical problems that have been inconspicuous in conventional macroscopic nonlinear optics.

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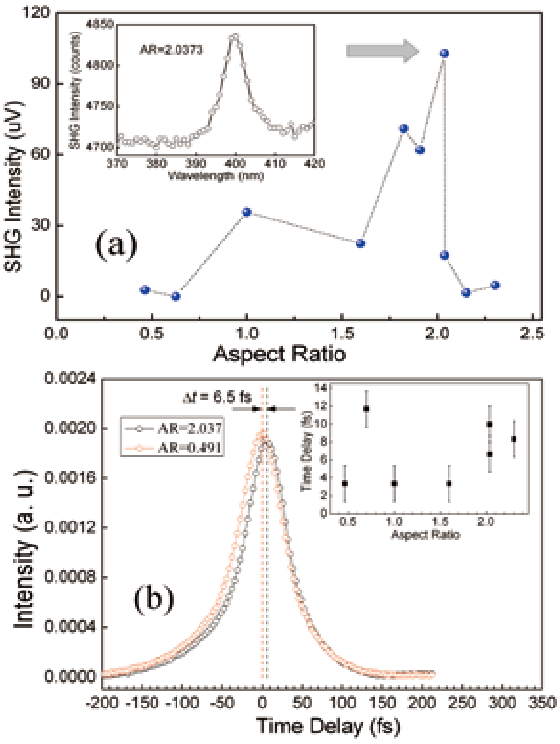

Figure 2: (a) Experimental result of SHG intensity depending on the aspect ratio of the rectangular air hole; (b) Measured time delay by comparing the convolution temporal profile of AR=2.037 and AR=0.491 samples, with a temporal accuracy of below 2fs. (Image by IOP)

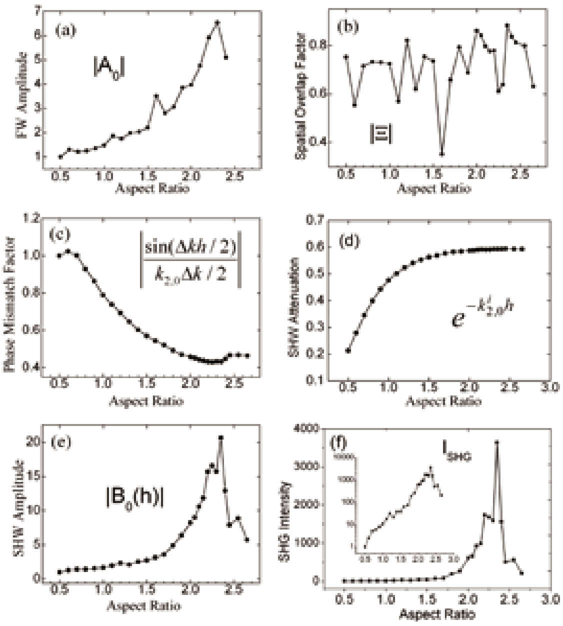

Figure 3: Theoretical results of SHG signals as a function of the air-hole aspect ratio and various mechanisms contributing to SHG shape resonance: (a) The amplitude of FW mode; (b) the spatial overlap factor; (c) the FW-SHW phase mismatch factor; and (d) the SHW attenuation factor. Panels (e) and (f) display the calculated amplitude of SHW mode and SHG intensity, which show a prominent resonant peak at AR=2.3 and a three orders of magnitude variation of the SHG. (Image by IOP)