http://dx.doi.org/10.5012/bkcs.2014.35.3.953

Real-Space Mapping of Hole Plasmons†

Deok-Soo Kim and Zee Hwan Kim*

*Department of Chemistry, Seoul National University, Seoul 151-747, Korea. *E-mail: zhkim@snu.ac.kr*
Received September 25, 2013, Accepted October 8, 2013

Key Words : Nanoparticle, Microscopy, Raman scattering, Localized plasmon

The nano-voids,[1] nano-holes,[2] and nano-slits[3] have shapes that are complementary to the nanospheres, discs and nanorods, respectively. According to the Babinet principle,[4] the plasmon modes and resonances of such nano-objects are fairly similar to (the Babinet principle is exact only when the constituting metal is a perfect conductor) the original objects. As such, the plasmons of such complementary nanostructures can be equally good, or even better (in terms of ease of fabrication), structures for plasmonic sensing, surface-enhanced Raman scattering (SERS)[5] and plasmonic devices. Therefore, it is important to fully characterize the plasmon modes of such objects.

In this Note, we report on the direct spatial mapping of dipolar plasmons of a gold nano-hole, which provides information on the spatial distribution of enhanced local fields. We employ the nano-holes instead of nano-slits because the former can be easily fabricated by low-cost nanosphere lithography.[6] The operating principle and the detailed setup of the scattering-type scanning near-field optical microscopy (sSNOM) can be found in ref. 7, and only the key concept is outlined here. Linearly polarized light excites plasmonic nanostructure (in our case nano-hole) and the plasmonic field induces an oscillating dipole of the tip-end (see Figure 1(a)). Therefore, the far-field scattering from the tip-end contains the information on the intensity, phase, and direction of the tip-dipole, and hence the local field information. In particular, we detect z-component (parallel to the tip-axis and perpendicular to the sample plane) of the local field, E_z.

Figure 1 shows the atomic force microscopy (AFM) topography, sSNOM intensity and phase images of an isolated nano-hole. Also shown in comparison are the simulated (*via* the finite-domain time-dependent, FDTD, method) E_z-field intensity and phase images. Close agreement between the experiment and simulation is evident. First, we observe enhanced field intensity at the rim of nano-holes (compare Figures 1(c) and (e)). Because of the topographic convolution effect of the tip-end, the apparent boundary of nano-holes in AFM image does not exactly match the physical boundary. The dashed circles in the images are the estimated positions and sizes of the boundary.

Secondly, we observe a 180 degree phase shift across the center of the nano-hole (compare Figures 1(d) and (f)). This corresponds to the E_z-field pointing opposite directions (upward and downward) on either side of the nano-hole. This, within the Gauss law, reflects the dipolar charge distribution at the rim of the nano-hole. This phase signature proves that the enhanced field does not arise from random hotspots associated with surface roughness. Rather, this arises from the overall circular geometry of the nano-hole. With an excitation of nano-hole with p-polarized light, these two features disappear (Figures 1(g) and (h)), which further substantiate our interpretation. The agreement between the

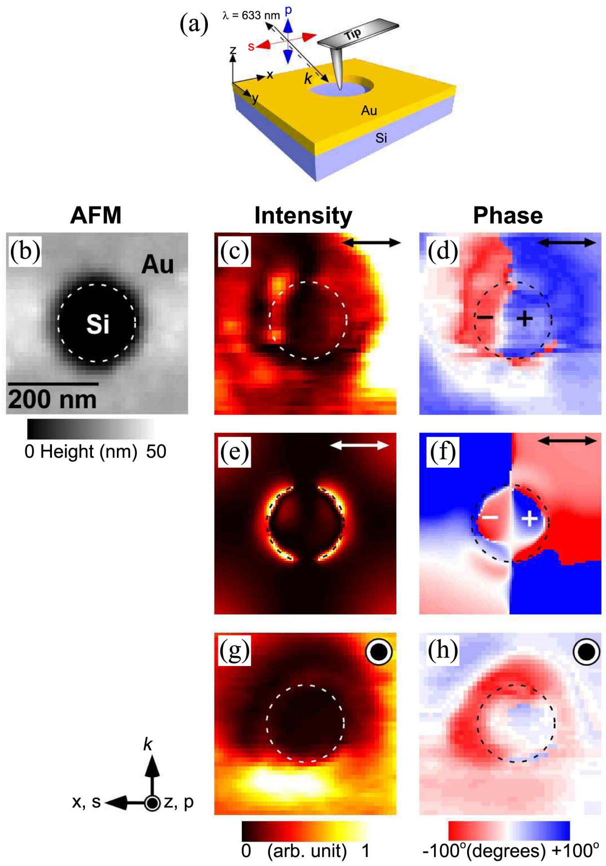

Figure 1. (a) Schematic of sSNOM measurement; (b) AFM topography of the nanohole; (c) sSNOM intensity (in-plane excitation); (d) sSNOM phase (in-plane excitation); (e) FDTD-simulated intensity; (f) FDTD-simulated phase; (g) sSNOM intensity (out-of-plane polarized excitation); (h) sSNOM phase (out-of-plane polarized excitation). The dashed circles represent the estimated position of Au/Si boundary. The double-sided arrows in (c), (d), (e), and (f) and concentric circles in (g) and (h) are the excitation polarization direction.

†This paper is to commemorate Professor Myung Soo Kim's honourable retirement.

experiment and theory is not exact. Part of the deviation is caused by the sSNOM experimental artifact. For example, the intensity profile shows slight asymmetry, which is caused by the asymmetry of the tip-end. Some other part of the deviation is likely to arise from the non-ideal shape (especially edges of holes) of the nano-holes fabricated. Nevertheless, the result clearly shows the detailed spatial profiles of the dipolar plasmons of nano-holes. The study of near-field interaction of the two or more Au nano-holes and their possible application to SERS are currently under way.

Experimental

The Au-hole with a diameter of 200 nm is made by the simple nanosphere-lithography: polystyrene beads of a corresponding diameter are dispersed onto the Si-wafer, and is coated with an Au film of thickness of 50 nm by thermal evaporation coating, and the bead is removed by sonicating in dichloromethane (CH_2Cl_2). This procedure yields randomly distributed nano-holes with a well-defined diameter. To minimize the tip-induced perturbation (shift in resonance frequency and distortion of the recorded field) in sSNOM,[5] we use weakly scattering Si-tip (tapping mode), and use the cross-polarization geometry in which excitation light is polarized orthogonal to the tip-axis (*s*-polarized) and only the scattered light that is polarized near-parallel to the tip-axis (*p*-polarized) is detected.

Acknowledgments. The authors acknowledge the financial support (Grant Nos. 2013-028021, 2012-0008686, and 2013-004512) from the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology (MEST). This work was also supported by Research Resettlement Fund for the new faculty of SNU. This paper is dedicated to Professor Myung Soo Kim on the occasion of his honorable retirement.

References

1. Cole, R. M.; Baumberg, J. J.; Garcia de Abajo, F. J.; Mahajan, S.; Abdelsalam, M.; Bartlett, P. N. *Nano Lett.* **2007**, *7*, 2094.
2. Gao, H.; Henzie, J.; Odom, T. W. *Nano Lett.* **2006**, *6*, 2104.
3. Seo, M. A.; Park., H. R.; Koo, S. M.; Park, D. J.; Kang, J. H.; Suwal, O. K.; Choi, S. S.; Planken, P. C. M.; Park, G. S.; Park, N. K.; Park, Q. H.; Kim, D. S. *Nature Photon*. **2009**, *3*, 152.
4. Bonn, M.; Wolf, E. *Principles of Optics*; Cambridge University Press: Cambridge, 1999.
5. Wei, H.; Håkanson, U.; Yang, Z.; Höök, F.; Xu, H. *Small* **2008**, *4*, 1296.
6. Hulteen, J. C.; Van Duyne, R. P. *J. Vac. Sci. Technol. A* **1995**, *13*, 1553.
7. Kim, D. S.; Kim, Z. H. *Opt. Express* **2012**, *20*, 8689.