

## Comment on “Self-Similar Chain of Metal Nanospheres as an Efficient Nanolens”

In a recent Letter, Li *et al.* [1] reported a self-similar chain of Ag spheres as an efficient nanolens, which induced a huge enhanced electromagnetic field by a factor of  $g \geq 1000$  at the nanofocus in the cavity between two smallest metal particles in the chain calculated by the quasistatic approximation. However, there are two problems in their model simulations. First, all the sizes of the chains composed with three, five, and six self-similar Ag particles in their study can be comparable to the favorite incident wavelength  $\lambda$  for a reasonable radius of the smallest Ag particle  $r = 5$  nm. Obviously, the quasistatic approximation in their model simulations is not valid anymore. Second, the smallest particles in the chains are too small where the intrinsic size effects, i.e., the broadening effects, damp the plasmon resonance to diminish the field enhancement [2]. The ignorance of the broadening effects in their model simulations in Ref. [1] leads to the overestimation of the field enhancement again. Although the basic qualitative predictions in Ref. [1] are not ques-

tioned here, based on the above reasons, the potential enhancements and the validity of quantitative results deserve serious attention.

We use both the orders of scattering method of the generalized Mie theory [3] and the three-dimension finite difference time domain (3D-FDTD) method [4] to calculate the field enhancement in the nanofocus. As shown in Fig. 1, with the consideration of full retardation by the generalized Mie theory, the maximum field enhancements (the dashed lines) for three configurations are all smaller than half of the corresponding maximum values in Ref. [1]. If the size-dependent dielectric function of the smallest Ag sphere is considered [5], the enhancement will become even smaller (the solid lines). As shown in Fig. 1(a), the result by the 3D-FDTD method (asterisks) is comparable to the generalized Mie theory. Without consideration of both the retardation and the broadening effect, the maximum field enhancements in Ref. [1] were overestimated at least 4 times. For surface-enhanced Raman scattering ( $\sim g^4$ ), the overestimation will become more than 2 orders larger.

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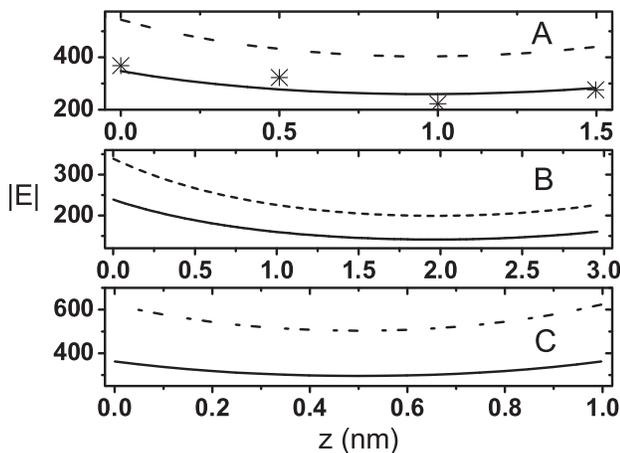


FIG. 1. The distribution of field enhancement in the symmetric axis of the self-similar chain between the two smallest spheres (start from the smallest sphere) calculated by the generalized Mie theory (lines) and the 3D-FDTD method (asterisks) with the incident polarization parallel to the symmetric axis. A, B, and C correspond to the cases of Figs. 2(b), 3(b), and 4(a) in Ref. [1], respectively. The solid and dashed lines correspond to with and without consideration of the size-dependent dielectric function of the smallest Ag sphere, respectively. The asterisks in A are calculated by 3D-FDTD. The radius of the smallest Ag sphere  $r = 5$  nm. The order of the multipoles used in the generalized Mie theory was 30 to ensure the convergence. The Yee cell size used in the 3D-FDTD calculation was set to be  $0.5 \times 0.5 \times 0.5$  nm, and the total number of time steps was set to be 21 000 to ensure the convergence.

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- [5] The dielectric function of small Ag nanoparticles was calculated here according to Eq. 2.53 in Ref. [2] by modifying the bulk dielectric function of Ag [P. B. Johanson and R. W. Christy, Phys. Rev. B **6**, 4370 (1972)], where the damping constant  $\Gamma_\infty = \nu_F/l_\infty$ , and  $\nu_F = 1.39$  m/s is the Fermi velocity,  $\Gamma_\infty = 52$  nm is the electron mean free path of the bulk Ag at the static case, and the corresponding plasmon resonance frequency in the Drude free electron model  $\omega_p = 9.2$  eV.