

Li, Stockman, and Bergman Reply: In their Comment [1], Li, Yang, and Xu (LYX) contend that, within their electrodynamic computation, the maximum enhancement is about 2 times smaller than in our original Letter [2], and with the finite-size effects taken into account it is less by an additional factor of 2.

There were two main results reported in our original Letter [2]. The first is based on a qualitative idea of transfer of the excitation and field down the scale of sizes in self-similar systems, which leads to multiplicative enhancement of the local field. The maximum enhancement occurs in a small nanofocus (“the hottest spot”) in the gap between the smallest nanospheres. This is the main part of our Letter, and it is not contested in the Comment, which actually confirms the existence of this nanofocus and large field enhancement in it. The second part of Ref. [2] provides an illustration: some computations for three-, five-, and six-sphere nanolenses, which show that the fields are enhanced at the nanofocus, depending on the geometry, by a factor from 500 to 2200. These numbers are stated by LYX to be by a factor of 2–4 larger than their results.

In our original Letter, we clearly stated that we solve the problem in the quasistatic approximation, neglecting spatial dispersion and Landau damping. The parameters of our systems were deliberately chosen to be at the limits of applicability of that approximation. The Comment actually shows that these parameters are reasonable: An error by factor 2, when the total enhancement is 1200, is a reasonably good accuracy given that there are many other physical effects which were ignored in our model, and also in the Comment (see below). We do maintain that our model was solved accurately. A careful perusal of Fig. 1(a) shows that the field at the nanofocus, seen as the very narrow high peak just outside the smallest sphere in the 1.5 nm intersphere gap, changes by more than an order of magnitude in the tangential direction and in the radial direction as soon as that sphere is entered. This abrupt change occurs over a distance of less than 0.5 nm—the grid size used by LYX is too coarse.

The great enhancement of field strength arises due to the fact that the frequency is close to that of one or more localized scattering resonances of the system [2]. The generalized Mie theory used by LYX is a multiple scattering theory [3,4]. Such a procedure cannot be expected to be accurate at a frequency close to a scattering resonance: Precisely at the resonance, it will diverge.

For our part, we do not understand why the two different approaches, which were used by LYX, yield results that are so close to each other—perhaps this is fortuitous? Finally, LYX discuss numerical consequences for the surface-enhanced Raman scattering enhancement factor G^{RS} , ignoring the fact that $G^{RS} \neq |E|^4$, where $|E|$ is the enhancement factor of the local electric field. Figure 1(b) shows our original Green’s function computations [5] of G^{RS} along with $|E|^4$. Evidently, G^{RS} differs from $|E|^4$ by orders of magnitude.

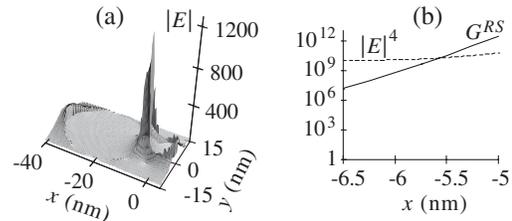


FIG. 1. (a) Distribution of local field enhancement $|E|$ shown for the two smaller spheres in the three-sphere nanolens with gap d equal to 0.3 of the radius of the minimum nanosphere [2]. (b) Distribution of Raman enhancement G^{RS} (solid line) and $|E|^4$ (dashed line) along the symmetry axis inside the gap.

The effects of the small size of nanoparticles on their optical responses cannot be taken into account by naively modifying the Drude relaxation constant as LYX have done, since the small size leads to spatial dispersion and Landau damping [6,7]. In particular, spectra of extremely thin nanoshells do not show any extra broadening [8]. Finally, not less important than retardation and spatial dispersion of electric permittivity are the atomistic roughness of the nanosphere surface in the minimum intersphere gap and the spillover of electrons and resultant underscreening of the fields at that surface [9]. These are the main outstanding problems which are faced in any attempt to use such nanosphere aggregates as nanolenses.

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