

Luo *et al.* Reply In the Comment [1], Schaich calculated the mode dispersion of surface plasmons supported by a planar metal-dielectric-metal (MIM) structure, and concluded that our model [2] fails to mimic the effect of nonlocality at high frequencies. Here, we shall clarify the difference between our calculations and that in Schaich's Comment, and highlight the validity of our model for a general class of plasmonic structures.

First, the difference between Schaich's calculations and ours in the lossless case results from the implementation of Q , which characterizes the decaying of longitudinal plasmons. Figure 2 of our original Letter [2] shows the maximum thickness needed to recover the exact hydrodynamic calculations. Therefore, we set

$$Q = \sqrt{k^2 + [\omega_p^2/\epsilon_\infty - \omega(\omega + i\gamma)]/\beta^2}. \quad (1)$$

Here, the permittivity of the dielectric layer [as calculated by Eq. (1) of Ref. [2]] is k dependent. However, in contrast to the hydrodynamic model where the transverse and longitudinal fields need to be treated differently, our model applies to both the transverse and longitudinal fields, and the k dependence only appears in the thin dielectric layer.

The k -dependence property can be neglected when

$$k \ll \left| \sqrt{\omega_p^2/\epsilon_\infty - \omega(\omega + i\gamma)}/\beta \right|. \quad (2)$$

Since β is normally a small quantity (e.g., for noble metals $\beta < 0.01c$, where c is the free space light velocity), Eq. (2) holds true for most of the cases. We also note that the condition given by Eq. (2) breaks down if the frequency ω approaches ω_p and the wave vector k is relatively large. For instance, the validity of Eq. (2) at $\omega = \omega_p$ requires $k \ll \sqrt{\omega_p \gamma}/\beta$. For a Drude metal permittivity (e.g., $\omega_p = 3.3$ eV, $\gamma = 0.165$ eV, and $\beta = 0.0036c$), this gives rise to $k \ll 1.04$ nm⁻¹ $\sim 62k_0$ (where k_0 is the free space wave vector). When the surface plasmon wave vector is larger than or comparable to this value ($62k_0$), Eq. (2) breaks down and the k dependence is no longer negligible. However, such a large wave vector is not easily accessible. It has never been observed for noble metals, to the best of our knowledge. Hence, for most of the realistic cases where the nonlocal optical responses of a plasmonic system are dominant by the contribution from relatively small k (e.g., $k \leq 20k_0$), our local approximation can be safely applied. In our original Letter [2], we checked the validity of this approximation by studying different nanoparticle geometries (see Fig. 3 and Fig 4 in Ref. [2]). Reference [1] also confirms that when applied to a MIM system, our model yields excellent agreement with the hydrodynamic theory at $k \leq 1$ nm⁻¹ (or $k \leq 60k_0$) below the surface plasmon frequency. This point is nontrivial, because most of the useful plasmonic resonances studied experimentally occur in this frequency range [3]. Moreover, if realistic

metallic losses are considered, the surface plasmon wave vector will be truncated at a finite value [4]. In this case, the validity of our theory can be extended to even higher frequencies (see the Supplemental Material [5] for detailed discussions).

We remark that any semiclassical model has its scope of applicability. Our local mode is valid whenever the screening length of surface charges is independent of k . In this sense, it is even more general than the hydrodynamic model. For instance, if the screening length of surface charges is obtained from full quantum approaches [9], the local model can be extended to treat other quantum effects in plasmonics. A similar concept was recently implemented in Ref. [10] to study the quantum corrections to the hybridization at subnanometer gaps.

We draw attention to recent experimental observations where the SERS enhancement from rough metal surfaces as a function of the surface roughness is probed and found to agree well with our theoretical model [11].

To conclude, we concede that for the case of a lossless metal in the worst case scenario of flat surfaces, Schaich's comments [1] have some validity, but for realistic parameters and in the complex geometries where our original Letter [2] thoroughly tested our model, the errors we found in the resonance positions were less than 1% in the analytical calculations (e.g., Fig. 3) and less than 1.2% in the numerical simulations (e.g., Fig. 4). The disagreement in the magnitude is less than 5% in the analytical results and less than 9% in the numerical ones below the surface plasmon frequency. Hence, we assert that by adopting realistic parameters and applying our model to challenging complex systems we gave a fair test of its accuracy.

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