Shalaev and Moskovits Reply: In their Comment [1] Stockman, Pandey, Muratov, and George (SPMG), on a basis of the numerical simulations, have concluded that there was no localization of dipolar eigenmodes in a cluster-cluster aggregate (CCA) projected into a plane. Although our previous simulations [2] for a 2D-projected CCA and for a 2D Viscek fractal did show localization at high-intensity zones ("hot spots"), we leave a discussion of the localization in 2D fractals for a future publication. Here we consider the localization in a CCA deposited in a plane (CCADP) which differs from a 2D-projected CCA and is certainly more relevant to our experiment [2]. The process of the deposition of a CCA was simulated so that a monomer with coordinates (x_i, y_i, z_i) reaches the plane of the deposition, $z = 0$, only if there are no other particles located underneath any given one, therefore, preventing the monomer from settling on the plane. Otherwise, the monomer remained on the top of the particle(s). Although, after such a deposition, a cluster is significantly reduced in z dimension, there are still a significant number of particles (~50%) located at the second level and higher. The average height of the CCADP with $N = 1000$ particles was $\bar{z} \approx 1.47a$, and the dispersion in the height was $\Delta\bar{z} \equiv [\bar{z}^2 - (\bar{z})^2]^{1/2} \approx 1.35a$ (a is the diameter of a particle). In Fig. 1 we plot the localization length L_X against the eigenfrequency a^3X (the simulations have been carried out by V. Markel). Despite the broad distribution of mode sizes, the results of the simulations clearly demonstrate the presence of the strongly localized eigenmodes in the CCADP. With increasing $a^3|X|$, L_X decreases down to a value $L_X \approx 0.1R_c \approx 2a$. Thus the most localized modes have a linear dimension which corresponds in size to only two touching particles.

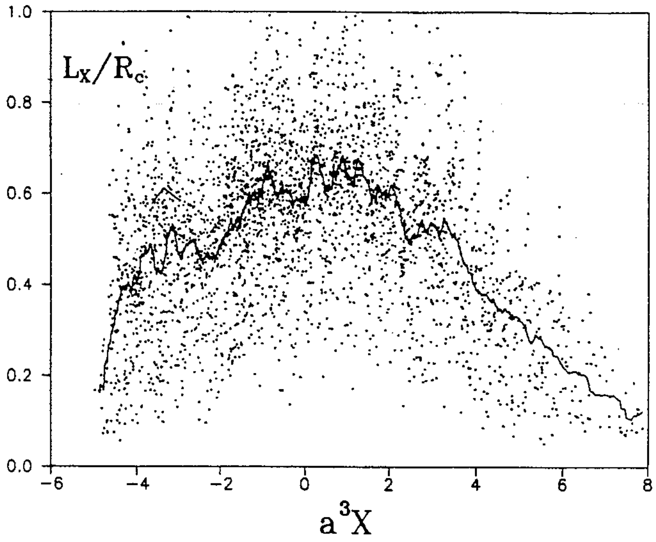

FIG. 1. Normalized localization length L_X of eigenmodes (R_c is the cluster radius) versus their eigenvalue a^3X for the CCADP with $N = 1000$ particles. The dependence L_X/R_c averaged over an interval of $a^3\Delta X = 0.04$ is shown by the solid line.

SPMG have also brought up the point regarding an anisotropic form of the hot spots. However, this is what one could expect, and it does not of course contradict the concept of the localization. Clearly, any particular dipolar model located on some fragment of a cluster is not supposed to be spherically symmetric. The local fields in a cluster are formed by the interaction between many particles with random (although correlated) spatial distribution and also by their interaction with the tip. The net result can be very complicated as seen in Fig. 3 of our Letter [2]. Some of the spots, like the one in Fig. 3(c), are elongated in the direction of **k**, while others, like the spots in the bottom of Fig. 3(a), are rather elongated in the perpendicular direction. The theory does not demand "circular" patterns. Likewise, the observation that some hot spots are longer and wider than they are high is not precluded by the theory. In fact, one rather expects it in view of the fact that we are dealing with a quasi-two-dimensional assembly. In this case the localized modes are asymmetric themselves, so that typically $L_\parallel \gg L_\perp$ ($L_\perp \equiv L_z$). The laser wavelengths (488 and 514 nm) used in our experiment are shifted to the red part of the spectrum with respect to the resonance frequency, $X = 0$, of a spherical silver particle (≈360 nm). For these wavelengths it is the longitudinal modes of a CCADP that are excited most and, accordingly, the dipole moments **d** associated with the excited modes lie predominantly in the (x, y) plane or close to it. The intensity of the local fields at a distance R from the modes associated with these dipoles is four times larger for the **d** direction, i.e., in the (x, y) plane, than at the same distance R but is normal to the plane. Thus the asymmetry of the mode images obtained with the PSTM operating at the constant preset intensity is the expected result and does not contradict the concept of the localized modes.

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Comment on "Photon Scanning Tunneling Microscopy Images of Optical Excitations of Fractal Metal Colloid Clusters"

In their recent paper [1], Tsai, Kovacs, Wang, Moskovits, Shalaev, and Botet (TKWMSB) have reported a direct observation of local optical fields in individual silver fractal clusters with a subwavelength resolution. These fields are concentrated in a few "hot spots" whose visible size is estimated in Ref. [1] as 300 to 700 nm. The position of these spots changes radically with the light polarization and considerably with frequency. TKWMSB argue that the local-field intensity is enhanced by the factor of $\sim Q^2(L_X/r)^6$, where L_X is the localization radius of excitations [2], and r is the distance from the photon scanning tunneling microscopy (PSTM) tip to the hot spot. From this, they obtain that the apparent size of a hot spot exceeds its true size, L_X, by a factor of $Q^{1/3} \sim 5$ and, consequently, $L_X \simeq 60 - 140$ nm. TKWMSB conclude that they have successfully observed the strong spatial localization of fractal eigenmodes predicted in [2].

We believe that the experiment [1] is of great importance for the optics of fractals and of disordered media in general. In this Comment, we augment TKWMSB's consideration and point out another possible interpretation of their experimental data. To simulate the response of a large cluster, we have generated a cluster-cluster aggregate (CCA) with the $N = 1024$ monomers, projected it into a plane, as is expected for the conditions of [1], and computed L_X for each of its s- and p-polarized eigenmodes (surface plasmons), as shown in Fig. 1. For any value of the eigenvalue X, the eigenmodes form a broad distribution about $L_X/R_c \simeq 0.5$. Consistent pictures are obtained for other individual clusters of this type. These results imply that, on average for a 2D-projected CCA, there is no strong localization expected. We have obtained similar results for other models CCA clusters transferred to a plane and also for 3D CCA clusters. In all these cases, for realistic values $|X| \lesssim R_0^{-3}$ relevant for [1], the distribution of eigenmode sizes is very wide, with no strong localization on average. The absorption into an eigenmode is $\propto L_X^3$[2], so that the delocalized eigenmodes are responsible for most of the local-field energy.

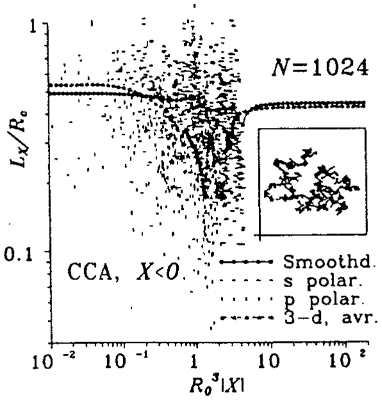

FIG. 1. Scaled L_X of eigenmodes (R_c is the cluster radius), vs their eigenvalues X (R_0 is the distance between monomers). The dependence $L_X(X)/R_x$ for the 2D-projected cluster (smoothed over $\Delta X = 0.1 R_0^{-3}$) is shown by solid circles and for 3D embedded CCA's ($N = 300$, averages over 10^3 clusters) by triangles. Inset, the 2D-projected cluster used.

Further, we note that the hot spots in Fig. 3 of Ref. [1] are strongly anisotropic, elongated in the direction of the wave vector **k** of light, with the maximum length of the spots in this direction $L_\parallel$ reaching almost 4000 nm ($\gg \lambda = 4\pi/k$) and several (up to 5) times smaller perpendicular dimension $L_\perp$. For such an elongated excited region, the spatial dependence of local fields differs dramatically from r^{-6} used in Ref. [1]. Actually, $r \simeq 400 - 600$ nm is considerably less than $L_\parallel$. Therefore, gradients of local intensity are predominantly perpendicular to **k** and, consequently, the longitudinal (large) dimension of a hot spot is seen by PSTM almost in its natural size. Independently from this argument, the strong elongation of the hot spots in the direction of **k** implies that the phase of the exciting light wave oscillates within the interaction region and, consequently, the size of this region is larger than λ. Generally, for any light-scattering effect, a strong anisotropy forward/across of forward/back (with respect to **k**) witnesses that the length of the interaction region is greater than λ. From these arguments, we conclude that the excitations of the clusters in [1] are delocalized over regions considerably larger than λ, in agreement with Fig. 1. The anisotropy of the hot spots is likely to be a polariton effect. Theses spots represent the fine structure of the eigenmodes, which themselves are fractal and have fluctuations on all scales within L_X.

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