Localization versus Delocalization of Surface Plasmons in Nanosystems: Can One State Have Both Characteristics?

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From a partial-differential eigenproblem, without use of dipole approximation, we show that the eigenmodes (surface plasmons) of disordered nanosystems (modeled as random planar composites) are not universally Anderson localized, but can have properties of both localized and delocalized states simultaneously. Their topology is determined by separate small-scale "hot spots" that are distributed and coherent over a length that may be comparable to the total size of the system. Coherence lengths and oscillator strengths vary by orders of magnitude from mode to mode at nearby frequencies. The existence of dark vs luminous eigenmodes is established and attributed to the effect of charge- and parity-conservation laws. Possible applications are discussed.

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The nanoscale optical properties of nanosystems have recently attracted much interest; see, e.g., [1–3] and references cited therein. This interest may be due, in part, to the possibility of using near-field optical phenomena on those scales for ultradense and ultrafast optical computing and information storage. On the nanometer spatial scale and at femtosecond times, surface plasmons are the modes carrying electromagnetic energy. The possibility of Anderson localization of surface plasmons [4,5] is of crucial importance in this context. In particular, delocalized modes allow for transfer of energy over the entire extent of the system, while localized modes permit concentration of energy in a small part of it.

In this Letter, we study localization of surface plasmons in a random planar composite (RPC) as a model for a disordered planar film. Our model composite is made of two constituents, characterized by dielectric permittivities $\varepsilon_1(\omega)$ (inclusion) and $\varepsilon_2(\omega)$ (host). Because the nanosystem heterogeneity scale is much less than the wavelength of light, the quasistatic approximation is applicable. This does not imply that processes are slow, but rather the opposite: the small size prevents retardation effects even for ultrafast processes. The local system response is described by a space- and frequency-dependent dielectric function $\varepsilon(\mathbf{r},\omega)$. The electric potential $\varphi(\mathbf{r})$ satisfies the equation $\nabla\cdot[\varepsilon(\mathbf{r},\omega)\nabla\varphi(\mathbf{r})]=0$ in the rectangular-prism-shaped volume $0\le x\le L_x$, $0\le y\le L_y$, $0\le z\le L_z$. Conventional Dirichlet-Neumann boundary conditions are imposed,

$$\varphi(\mathbf{r}) = \varphi_0(\mathbf{r})|_{z=0,L_z}\,;$$
$$\partial_x\varphi(\mathbf{r})|_{x=0,L_x} = \partial_y\varphi(\mathbf{r})|_{y=0,L_y} = 0\,,$$

where $\varphi_0(\mathbf{r}) = -E_0 z$ is the potential of the uniform external or volume-average electric field. We also make use of the spectral parameter $s(\omega)\equiv[1-\varepsilon_1(\omega)/\varepsilon_2(\omega)]^{-1}$.

The material-independent eigenmodes of this problem are characterized by eigenvalues s_i and eigenfunctions $\varphi_i(\mathbf{r})$, which satisfy a generalized eigenproblem

$$s_i\nabla^2\varphi_i(\mathbf{r}) = \nabla\cdot[\theta(\mathbf{r})\nabla\varphi_i(\mathbf{r})]\,, \tag{1}$$

along with the homogeneous variant of the above-mentioned boundary conditions, i.e., for $\varphi_0(\mathbf{r})\to 0$. Here $\theta(\mathbf{r})$ is the characteristic function of the ε_1 constituent, equal to 1 inside that constituent and equal to 0 everywhere else. We set $\theta(\mathbf{r})=0$ near the system boundaries.

Using $\varphi_i(\mathbf{r})$ and s_i, spectral representations are exploited to compute the local potential $\varphi(\mathbf{r})$ and the macroscopic (bulk) effective dielectric permittivity ϵ [6]:

$$\varphi = \varphi_0 + \sum_i \frac{m_i^*\varphi_i}{s(\omega)-s_i}\,,$$
$$\frac{\epsilon(\omega)}{\varepsilon_2} = 1 - p\sum_i\frac{f_i}{s(\omega)-s_i}\,; \tag{2}$$
$$m_i = \frac{s_i\int dV\,\varphi_0^*\nabla^2\varphi_i}{\int dV\,\varphi_i^*\nabla^2\varphi_i}\,,$$
$$f_i = -\frac{s_i|\int dV\,\varphi_0^*\nabla^2\varphi_i|^2}{pVE_0^2\int dV\,\varphi_i^*\nabla^2\varphi_i}\,. \tag{3}$$

Here p is the fill factor (volume fraction) of the inclusion constituent $\varepsilon_1(\omega)$, $V=L_xL_yL_z$ is the system volume, and f_i is the oscillator strength (weight) of the eigenmode i. The localization radius L_i of an eigenmode is defined as the gyration radius of its electric field intensity $|\mathbf{E}_i(\mathbf{r})|^2$,

$$L_i^2 \equiv \frac{\int dV\,\mathbf{r}^2|\mathbf{E}_i|^2}{\int dV\,|\mathbf{E}_i|^2} - \left(\frac{\int dV\,\mathbf{r}|\mathbf{E}_i|^2}{\int dV\,|\mathbf{E}_i|^2}\right)^2, \quad \mathbf{E}_i \equiv -\nabla\varphi_i\,. \tag{4}$$

All numerical calculations have been performed on a discrete version of Eqs. (1)–(4). We have generated an RPC on a cubic lattice by randomly positioning cubes of

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size $2 \times 2 \times 2$ (all sizes are in units of the grid step) with some probability p_0. In the examples discussed below, we limit ourselves to a thin monolayer of width 2 around the central yz plane, with $p_0 = 0.5$ as the filling probability *in that layer*. The system obtained is a thin film composite in three-dimensional space. Such systems have recently been studied extensively (see, e.g., Ref. [7] and references therein). The eigenmodes of the finite matrix eigenvalue problem, obtained by the above-described spatial discretization, were found using LAPACK [8]. The error is expected to be $\propto 1/L_i^4$. Because the step function $\theta(\mathbf{r})$ has a singular derivative, we decided to smooth its discontinuities using Gaussian smoothing with an RMS width equal to the grid step size. Such smoothing does not affect the properties of the system at intermediate or large scales, and it actually makes the small scale structure more realistic by eliminating sharp edges.

We tested these techniques by computing $\epsilon(\omega)$ for a uniform planar dielectric layer, reproducing the exact result with a $\lesssim 10\%$ error. We also tested our solutions with the exact sum rules of the problem [6]:

$$\frac{1}{\pi}\int_{-\infty}^{\infty} ds \,\mathrm{Im}\,\frac{\epsilon(s+i\delta)}{\varepsilon_2} = p \Longleftrightarrow \sum_i f_i = 1\,; \qquad (5)$$

$$\Phi_i(z) \equiv \int D_{iz}(x,y,z)\,dx\,dy = \mathrm{const}\,, \qquad (6)$$

where $\mathbf{D}_i = (1 - \theta s_i^{-1})\mathbf{E}_i$ is the electric displacement field of the eigenmode i, and $\Phi_i(z)$ is its normal flux through the xy planes, which is *independent of z*. Equations (5) express the Thomas-Reiche-Kuhn (TRK) dipole sum rule, while Eq. (6) follows from Gauss' theorem. Numerically, Eqs. (5) and (6) are satisfied with a relative error $\lesssim 10^{-5}$.

In Fig. 1 we show the smoothed, discretized nanostructure of one particular composite sample, as well as all of its eigenmodes (surface plasmons) in a plot of oscillator strength f_i vs localization length L_i. These eigenmodes are strikingly unusual. First, there is a large number of eigenmodes with negligible weights $f_i \lesssim 10^{-5}$. Such eigenmodes do not couple to propagating waves, and they can be neither observed nor excited from the far (wave) zone. We call them *dark modes*. They can, however, be excited and observed by near-field scanning optical microscope-type probes in the near-field region. There are also many eigenmodes with relatively large weights, $f_i \gtrsim 1/V \sim 10^{-4}$, which we call *luminous modes,* that couple efficiently to the far-zone fields. Second, both the luminous and dark modes have localization radii L_i with all possible values, from zero to one-half of the diagonal system size, and with very little correlation between f_i and L_i, except for the superlocalized (zero-size) eigenmodes which are all dark. This wide range of L_i shows that *Anderson localization does not occur for most of the modes, including the luminous modes,* in contradiction to what was found in Ref. [7]. Deviations from simple Anderson localization were also seen in some previous studies of the spatial structure of vibrational modes [9,10] and dephasing rates [11] in disordered solids. Similarly to our findings, those deviations were also caused by long-range (dipole-type) interactions.

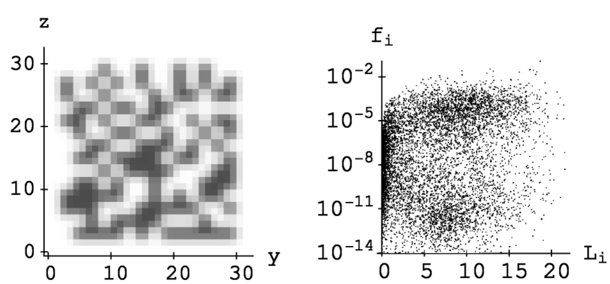

FIG. 1. For a planar random continuum composite (in the yz plane), the density of the inclusion component (left panel) and all eigenmodes plotted as oscillator strength f_i vs localization radius L_i (right panel). The size of the embedding space is $8 \times 32 \times 32$ in grid units.

One can rigorously prove that all Anderson-localized modes are dark. To quantify the eigenmodes, we normalize them as $\int dV\,|\mathbf{E}_i|^2 = 1$. Then the weight can be expressed as $f_i = s_i L_z^2 |\Phi_i(z)|^2/(pV)$. If an eigenmode is Anderson localized [12], then the electric field near at least one of the xy boundary planes must be exponentially small. Therefore the flux $\Phi_i(z)$ will be exponentially small at all xy planes, and consequently also f_i will be exponentially small. Thus these localized modes cannot significantly couple to fields in the far zone and must be dark. A corollary of this theorem is that if a composite existed in which *all* eigenmodes were localized, then it would be transparent at all frequencies. That is impossible due to the TRK dipole sum rule of Eq. (5).

To gain more insight, we show in Fig. 2 the local electric field intensities $|\mathbf{E}_i(\mathbf{r})|^2$ for particular eigenmodes of four extreme types, all with eigenvalues very close to $s_i = 0.2$. The data of Fig. 2 confirm the above-discussed absence of correlation between localization length and oscillator strength and also show that there is no correlation between the topology of $|\mathbf{E}_i(\mathbf{r})|^2$ and that strength—compare the pairs of eigenmodes: $s_i = 0.1996$ with $s_i = 0.2015$, and $s_i = 0.2$ with $s_i = 0.2011$.

Clearly, the hot spots seen in Fig. 2 cannot yield dominant contributions to the weight f_i or the amplitude $m_i = s_i L_z \Phi_i(z)$, since we could have calculated $\Phi_i(z)$ at a value of z where those hot spots are absent. This implies that a hot spot actually consists of regions where $D_{iz}(\mathbf{r})$ has large values but with *opposite signs,* which nearly cancel out in the evaluation of $\Phi_i(z)$. The occurrence of "very dark" modes (of the type shown in the lower panels of Fig. 2) is due to the symmetry of the microstructure under reflection in the midplane of the composite. The odd-parity eigenmodes exhibit a strictly vanishing total flux $\Phi_i(z) = 0$.

To illustrate these conclusions, we show the local displacement field $D_{iz}(\mathbf{r})$ in two adjacent yz planes ($x = 5$ and $x = 6$) that are symmetric with respect to the

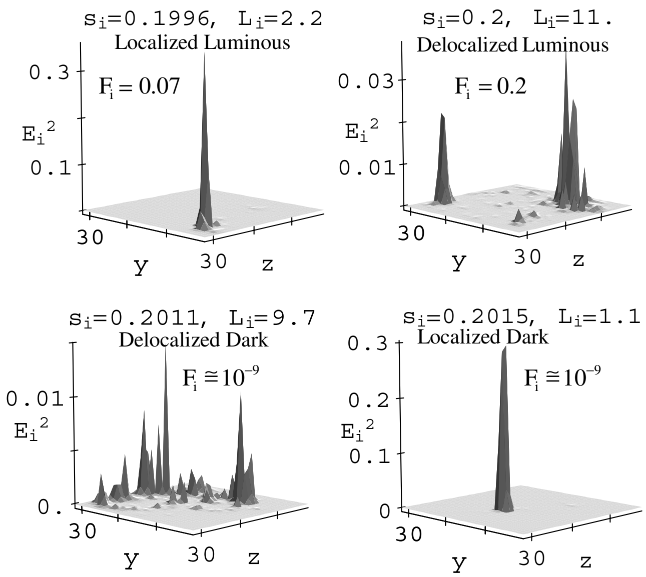

FIG. 2. Local field intensities $|\mathbf{E}_i(x, y, z)|^2$ of eigenmodes at the midplane of the sample shown in Fig. 1, vs spatial coordinates y, z in that plane. The oscillator strength per monomer shown is defined as $F_i \equiv pVf_i$. Localization radius L_i is also indicated for each eigenmode.

midplane of the composite, for the two "most counterintuitive" eigenmodes (delocalized dark and localized luminous)—see Fig. 3. Evidently, the dark mode (upper panels) has odd parity: the local fields in the symmetric sections are identical in magnitude and opposite in sign; thus their displacements completely cancel each other out in the total flux $\Phi_i(z)$. This explains the entire band of dark modes seen in Fig. 1 for $f_i \lesssim 10^{-8}$. However, a significant cancellation occurs also for luminous, even-parity eigenmodes—see, e.g., the eigenmode exhibited in the

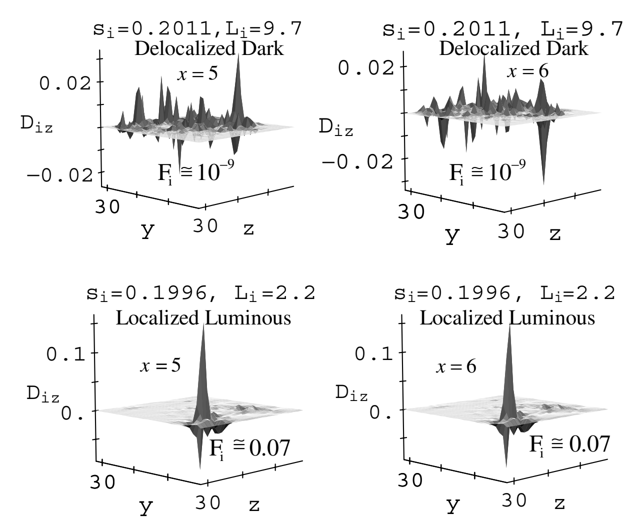

FIG. 3. Electric displacement field $D_{iz}(x, y, z)$ of two eigenmodes ($s_i = 0.2011$ and $s_i = 0.1996$) as function of the coordinates y, z. These displacements are calculated for two adjacent planes, $x = 5$ and $x = 6$ (indicated on the graphs), symmetric with respect to the mirror-symmetry plane of the composite.

lower panels of Fig. 3. This occurs due to the flux conservation constraint of Eq. (6). Although this mode ($s_i = 0.1996$) has even parity, the displacement field D_z still changes its sign at adjacent points *in the midplane within the hot spot*. As a result, the big peak in $\mathbf{E}_i^2$ almost completely cancels out, and almost the entire contribution to the flux $\Phi_i(z)$ comes from the small "ripple" seen outside of this peak. This near cancellation in the even-parity modes explains the very wide range of oscillator strengths seen in Fig. 1. Such a near cancellation will likely occur even in the absence of reflection symmetry.

The numerical results presented above deal with individual surface plasmons. In order to gain a statistical perspective, we show spectral distributions of localization lengths $P(L, s) = \langle \sum_i \delta(L - L_i)\delta(s - s_i)\rangle$, averaged over a representative ensemble of systems for all eigenmodes—see Fig. 4. We have verified that the corresponding distributions for luminous modes only are similar. Evidently, most eigenmodes are concentrated at comparatively small eigenvalues, $s_i \leq 0.3$, where the optical absorption is large. In this spectral region, the eigenmodes at any s are distributed over the maximum range of localization lengths, from one-half of the diagonal system size down to zero. This confirms that there is no uniform Anderson localization for all eigenmodes. The coexistence of eigenmodes with very different localization radii at the same frequency (eigenvalue) supports the concept of inhomogeneous localization introduced earlier for dipolar eigenmodes [13,14]. Note that the present eigenmodes are obtained without invoking the dipole approximation. Therefore, the hot spots of the local fields on the small scale and modes with small L_i are obtained more reliably.

The maximum size of the eigenmodes and their density decreases with increasing s. Comparing panels on the left and right of Fig. 4, we conclude that these distributions scale with the total size of the composite film, which limits the maximum extension of an eigenmode. This also implies that the size of the system in our computations is large enough and that no new information would be obtained if this size were increased. Comparing the panels in different rows of that figure, one sees that as the transverse system size decreases, the number of delocalized eigenmodes also decreases, and the overall distribution shifts to larger values of s. In the 2D case only [Figs. 4(e) and 4(f)], there is evidence for predominantly strongly localized eigenmodes at s close to 1.

We have found that, without smoothing, the distributions corresponding to Fig. 4 are considerably different. The singular hot spots, sensitivity to the system structure at the minimum scale, eigenmodes delocalized over the entire system—all suggest that the theory may not be renormalizable and that a universal description (e.g., based on a percolation model) may be impossible.

In summary, we have established that eigenmodes (surface plasmons) of a disordered planar nanostructured system (modeled as an RPC film) are not generally Anderson

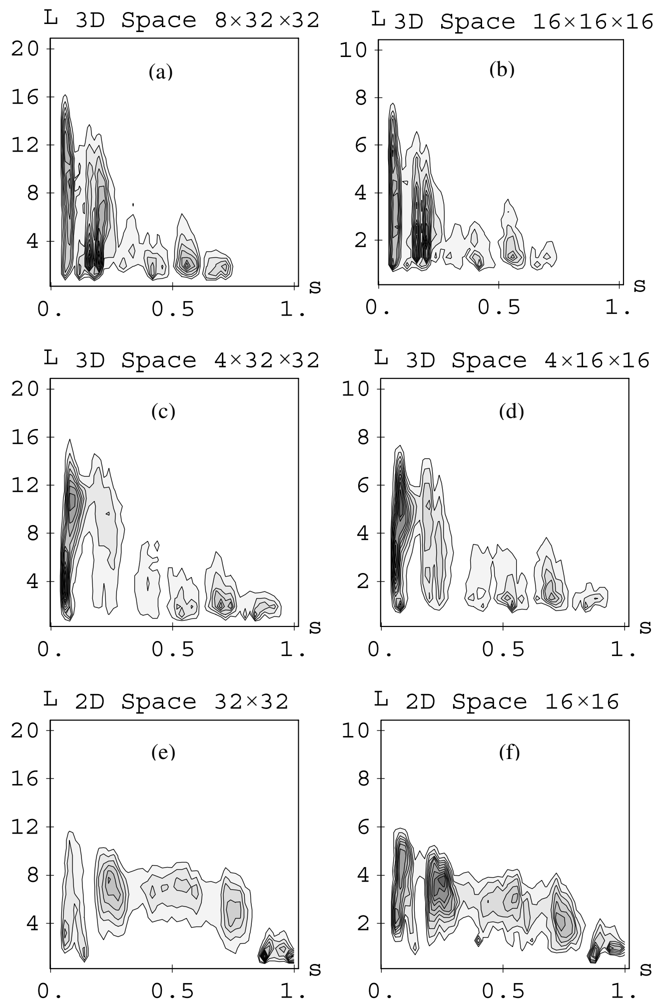

FIG. 4. Contour plots of the spectral distribution $P(L, s)$ of eigenmodes over their localization length L and spectral variable s. The system size $L_x \times L_y \times L_z$ is indicated above each plot (the composite film lies in the central yz plane). The lower panel is for 2D systems of size $L_y \times L_z$. The data are averaged over an ensemble of randomly generated systems large enough to make the statistical error negligible on the scale of the plots.

localized. This we attribute to the long-range dipolar interactions. We have proved that Anderson localization of surface plasmons in the entire spectral range is impossible, because that would conflict with the TRK sum rule. The localization radii L_i are distributed over the entire range from the minimum scale to the total size of the system, which is a signature of inhomogeneous localization. A typical eigenmode has properties of both localized and delocalized states: it includes hot spots on the small scale, but those are distributed and coherent over the large scale. The surface plasmons are extremely inhomogeneous in their oscillator strengths f_i that range over more than 10 orders of magnitude. A significant fraction of all the eigenmodes are dark and can be excited or observed only from the near-field zone with nanometer scanning probes, but not from the far zone. At the same time, the dipole-allowed (luminous) eigenmodes also exhibit a very wide range of f_i values. This full or partial suppression of oscillator strengths is due to the independent effects of two conservation laws: parity and flux (gauge invariance). Our conclusions *vis-à-vis* nonoccurrence of Anderson localization of the surface plasmon modes in RPC films followed from a careful analysis of the extensive numerical calculations with special attention to the luminosity of those modes. Such an analysis was not performed in previous studies of metal-dielectric films, like the one described in Ref. [7].

Finally, these unique properties of surface plasmons make them interesting candidates for retaining and transporting electromagnetic energy and information in ultrafast and ultradense computing applications: The hot spots of such modes could allow for concentration of electromagnetic energy in small parts of the system, while the long coherence range would permit transfer of energy and information across the entire extent of the system. The dark eigenmodes can be excited from within the nanosystem but will not radiate and cannot be observed or interfered with from the outside.

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