Laser Physics, Vol. 11, No. 1, 2001, pp. 26–30.

Giant Optical Responses in Microcavity–Fractal Composites

V. A. Podolskiy and V. M. Shalaev

Department of Physics, New Mexico State University, Las Cruces, NM, 88003 USA

e-mail: vshalaev@nmsu.edu

Received July 16, 2000

Abstract—Optical properties of fractal nanostructured composite materials are considered. The fractal geometry results in localization of plasmon excitations in the "hot" spots, where the local field can exceed the applied field by several orders of magnitude. The high local fields of the localized fractal modes result in dramatic enhancement of optical responses, making feasible the surface-enhanced nonlinear spectroscopy of single molecules and nanocrystals. The field enhancement becomes especially large when fractals are placed inside a microcavity. A theory for the enhanced Raman and hyper-Raman surface-enhanced scattering in microcavity–fractal composites is developed.

INTRODUCTION TO FRACTALS

A great deal of the progress in the physics of random media arose because of breakthroughs in modeling the irregular structures of complex disordered systems. Among the many descriptions proposed, one of the most powerful relies on concepts of fractals, encompassing self-similar or self-affine structures [1]. These models are capable of satisfactorily simulating a large class of the random media.

A fractal cluster with radius of gyration R_c consisting of N identical particles would obey a scaling law of the form $N = (R_c/R_0)^D$, where D, called the fractal Hausdorff dimension is, in general, fractional and, for a true fractal, less than the dimension of the embedding space d. R_0 is a constant which expresses the scale in terms of which the description of the cluster is framed. Its minimum value, for a cluster composed of particles, is of the order of the minimum separation between particles.

Fractals are self-similar or self-affine. A self-similar cluster is characterized by a single value of D and scales equivalently in all three dimensions. In contrast, self-affine structures have different scaling properties in the (x, y) plane from those in the direction, z, normal to that plane. Deposited films, the roughness of etched surfaces, and self-similar fractals projected onto a plane are often self-affine structures.

The most intriguing discovery involving fractals is that dynamical excitations of fractals are, in general, localized, while for compact systems, they are, in general, delocalized over the body of the excited object. This is because all physical excitations of a translationally invariant crystal: phonons, polaritons, magnons, etc., can be represented by running plane waves, i.e., by Fourier harmonics, which are eigenfunctions of the shift operator, ∇, representing the translational invariance symmetry. However, running waves are not eigenfunctions of the dilation symmetry operator, and fractals are not translationally invariant. Hence, in many cases, they cannot transmit propagating running waves.

The strongest scattering occurs from inhomogeneities whose dimensions are of the same scale as the wavelength of the dynamical excitations. Anderson localization, for example, may result from coherent multiple scattering in random systems with uncorrelated disorder. A fractal, although in some sense disordered, is, nevertheless, correlated on all length scales lying between the size of its constituent particles to one corresponding to the dimensions of the fractal. Fractals are unique in that, because of their scale-invariance, there is no characteristic length for the inhomogeneities. Instead, inhomogeneities of all sizes (raging between the aforementioned lower and upper limits) are present in the fractal. Hence, whatever the wavelength of excitation there will always be fluctuations of the requisite size, resulting in strong scattering that can lead to localization.

Localization of excitations in fractals was first predicted for acoustic excitations [2] and demonstrated in an ingenious experiment by Sapoval *et al.* on a fractal drum-head [2], and for optical excitations by Stockman and Shalaev [3]. Recently, Stockman *et al.* [4], showed that localization in fractals is inhomogeneous in the sense that eigenmodes with very different coherence radii can coexist at the same wavelength; at frequencies close to the resonance of individual particles even chaotic behavior of the eigenmodes can occur.

What all of this means is that for a fractal object the optically excited distribution of the resonant electrical polarization will not be homogeneous on the fractal but concentrated in "hot spots" much smaller in size than the size of the fractal and often much smaller than the wavelength [5, 6]. When sufficiently concentrated, the large electromagnetic fields in the hot spots can result in very large enhancements of optical nonlinearities and in other effects, such as photochemistry that is usually associated with high fields.

GIANT OPTICAL RESPONSES IN FRACTALS

An electron microscope image of a typical fractal aggregate of colloidal Ag particles is shown in Fig. 1. Its fractal dimension, $D \approx 1.78$, suggests that it was formed through cluster-cluster aggregation [1], a process that can be readily simulated numerically. Cluster-cluster aggregates are formed [1] when initially isolated nanoparticles diffuse randomly in solution adhering to other nanoparticles they encounter. The resulting clusters, of all size, themselves diffuse and aggregate. The process never ends but growth slows dramatically when all of the particles are subsumed in very large, slowly-diffusing and infrequently colliding clusters.

When the constituent particles of a fractal cluster are irradiated by light, oscillating dipole moments are induced in them which interact strongly through dipolar forces leading to the formation of collective optical modes. Under such circumstances, the local field distribution can be calculated to an acceptable level of approximation by solving the coupled-dipole equation (CDE) [5, 6] $\mathbf{d}_i = \alpha_0(\mathbf{E}^{(0)} + \sum_{j \neq i} \hat{W}_{ij}\, \mathbf{d}_j)$, where $\mathbf{d}_i$ is the local dipole induced on the ith particle (related to the local field $\mathbf{E}_i$ by $\mathbf{d}_i = \alpha_0 \mathbf{E}_i$), α_0 is the polarizability of the individual particles composing the cluster, $\mathbf{E}^{(0)}$ is the incident electromagnetic wave, and $\hat{W}_{ij}$ is the operator describing the coupling between the induced dipoles on particles i and j. Hence, the second term in the brackets represents the contribution to the local field $\mathbf{E}_i$ due to the dipole moments induced on all of the particles in the cluster. The polarizability for spheres is given by $\alpha_0 = R^3(\epsilon - 1)/(\epsilon + 2)$, where R is the radius of spheres and ϵ is the dielectric constant of the material.

The local field distribution excited by light of wavelength $\lambda = 1$ μm computed in this manner at the surface of a simulated self-affine silver CCA is shown in Fig. 2. The largest fields are extremely localized; and the local field intensity in the "hot" spots can exceed the applied field by up to 10^6, while the average enhancement is only $\sim 10^2$ to 10^3. The localized optical excitations have been first experimentally observed by Tsai *et al.* [3].

In fractals, the resonance frequency of a localized surface plasmon mode depends on the local configuration of particles at the mode location. A random fractal is composed of a large variety of local geometries, each possessing a different plasmon resonance frequency; as a result, the range of frequencies spanned by the plasmon modes in a fractal cluster is unusually broad covering the whole of the visible and infrared portions of the spectrum. Additionally, for most metals, the electromagnetic energy and the enhancement of nonlinear optical effects and Raman scattering increase towards longer wavelengths [6]. This is because for most metals, the real part of the dielectric function is negative (that is why metals are such good reflectors) and its magnitude increases strongly towards longer wavelengths, resulting in a parallel increase of the quality-factor of the plasmon resonances of fractal clusters composed of metal particles.

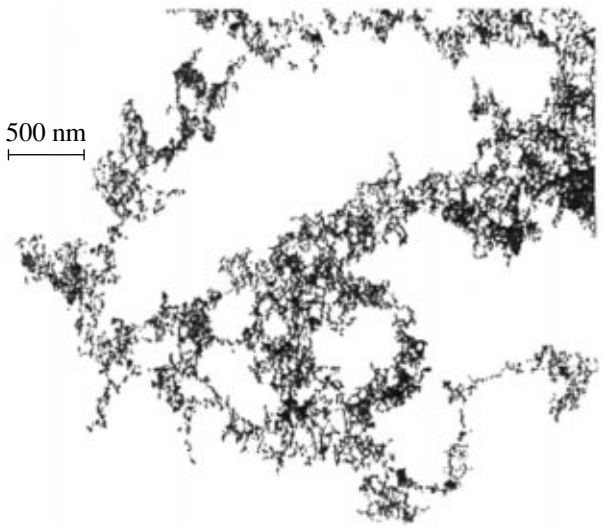

Fig. 1. Electron micrograph of a fractal colloid aggregate. Voids corresponding to all length scales are present, the minimum being the size of a single particle, the maximum one the size of the entire cluster. This is a graphic illustration of the statistical self-similarity and hence the fractal nature of the cluster. The radii of the individual particles is ~10 nm, while the size of the cluster is ~1 μm.

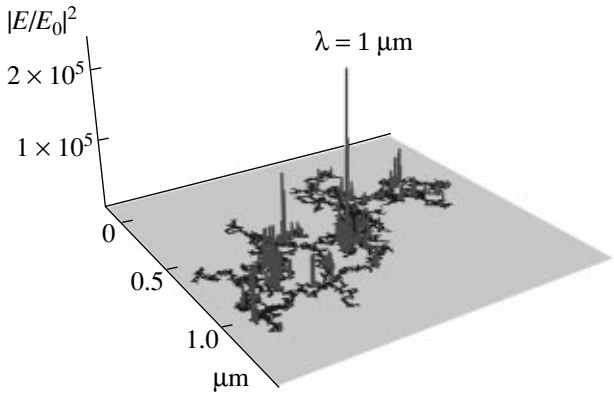

Fig. 2. Calculated field distributions on the surface of a silver fractal aggregate deposited on the plane.

The various nanoscale areas, where the resonant fractal excitations are localized, can be thought of as a set of different optical nanoresonators, each having different resonance frequencies, and altogether, resonating in the visible and IR spectral ranges. These fractal nanoresonators have large resonance quality factors (representing the local-field enhancement), which increase from the visible to the IR and reach 10^6 in the near-IR.

Locally intense fields of this magnitude suggest a large number of unusual local optical and photochemical effects, among them, single-molecule spectroscopy.

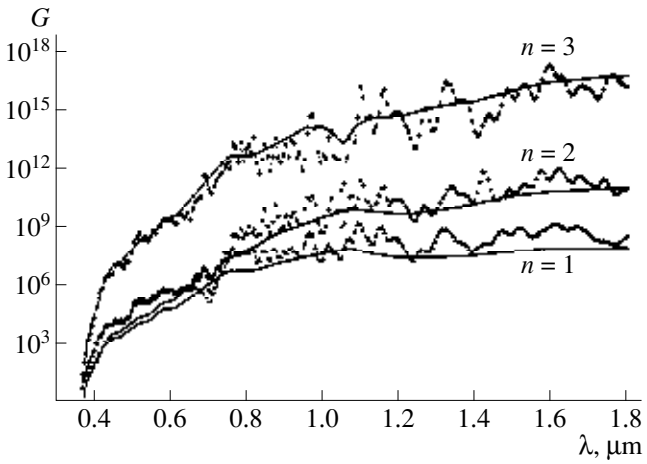

Fig. 3. Average enhancement of Raman and hyper-Raman (two- and three-photon pumped) scattering in silver fractal aggregates.

Specifically, because for surface-enhanced Raman scattering (SERS) the local enhancement factor $\propto |E|^4$ [7], it can reach magnitudes of 10^{12} or greater, making Raman spectroscopy of single molecules possible [8].

The local-field enhancement for a nonlinear optical process can, in general, be written as

$$G_n \sim \langle \left|E_i/E^{(0)}\right|^k [E_i/E^{(0)}]^m \rangle, \qquad (1)$$

where $n = k + m$. In particular, $k = 4$, $m = 0$ describes the field enhancement for SERS, and $k = 2$, $m = 2$ for the nonlinear refraction (optical Kerr effect). Signals associated with coherent nonlinear light scattering, such as four-wave mixing (FWM), are proportional to the average square of the nonlinear polarization; hence the FWM enhancement, for example, is [6] $G_{FWM} = |G_4|^2$.

For fractals, an estimate for G_n (at $k \neq 0$) can be expressed in terms of the polarizability $\alpha_0 \equiv -[X(\omega) + i\delta(\omega)]^{-1} = R^3(\epsilon - 1)/(\epsilon + 2)$, of the individual particles composing the fractal as [6]

$$G_n \sim c_n |X|^n \delta^{1-n} \mathrm{Im}[\alpha(X)], \qquad (2)$$

where c_n is a frequency-independent constant. The variable X indicates the proximity of ω to the resonance of an individual particle, occurring for a spherical particle at $\epsilon' = -2$ (that corresponds to $X \approx 0$), and it plays the role of a frequency parameter; δ characterizes dielectric losses. The resonance quality factor is proportional to δ^{-1}. In the formula above, $(|X|/\delta)^n$ gives the resonant local field (normalized to the applied field) raised to the n-th power and $\delta \mathrm{Im}[\alpha(X)]$ is the approximate fraction of resonant particles in the fractal, for a given light frequency. The factor $\mathrm{Im}[\alpha(X)]$ represents the average light extinction, which differs significantly for fractals ($D < d$) and non-fractals ($D = d$).

For the average local-field intensity ($n = 2$), it can be shown that the *exact* result is given by [3, 6]

$$G = G_2 = \frac{(X^2 + \delta^2)}{\delta} \mathrm{Im}[\alpha(X)], \qquad (3)$$

in agreement with the estimate (2).

It is worth noting here that the zero-point field fluctuations, which are responsible for spontaneous emission, are expected to be enhanced in the same manner as their "classical" counterpart so that the formula (3) also characterizes enhancement of spontaneous emission in fractals.

For nonfractal random systems, the extinction $\mathrm{Im}[\alpha(X)]$ typically peaks near the resonance frequency of the individual particles (where $X(\omega) \approx 0$) becoming negligible for $|X| \gg \delta$, so that G_n, according to (2) is relatively small [6]. Contrariwise, for fractals the factor $\mathrm{Im}[\alpha(X)]$ remains significant even in the long wavelength part of the spectrum, where $|X(\omega)| \gg \delta$, leading to very large values of G_n [6] ($\mathrm{Im}\alpha(X)$ has roughly a box-like distribution in the interval $-4\pi/3 \leq X \leq 4\pi/3$, which in terms of λ includes the whole of the visible and infrared parts of the spectrum [6]). Thus the large enhancement in a broad spectral range, as mentioned, is a direct result of the localization of the optical excitations and of the broad variety of resonating local structures.

We also note here that by exciting all (or most of) the fractal modes and matching their phases one can produce attosecond light pulses; this is because the extremely large spectral range of fractal modes (from the near-UV to the far-IR) in the spectral domain corresponds to the attosecond time intervals in the time domains.

As an example of enhanced optical phenomena, we consider now in more detail Raman Scattering (RS) and hyper-Raman scattering (HRS) for molecules adsorbed on the surface of metal particles in a fractal aggregate. The HRS occurs typically because of vibrations of molecules at, say, frequency Ω. The frequency of the scattered wave ($\omega_s = 2\pi(c/\lambda_s)$) in n-photon pumped hyper-Raman scattering is given then by $\omega_s = n\omega - \Omega$. The case of $n = 1$ corresponds to conventional RS. For simplicity, we assume below that the shift Ω is small (less than the plasmon width Γ).

The enhancement $G_{n-\mathrm{HRS}}$ is defined through the ratio of the local (enhanced) and the external (or probe) fields ($E^{(0)}$):

$$G_{n-\mathrm{HRS}} = \langle \left|E_\lambda\right|^{2n} \left|E_{\lambda_s}\right|^2 \rangle / \left|E_\lambda^{(0)}\right|^{2n} \left|E_{\lambda_s}^{(0)}\right|^2. \qquad (4)$$

For conventional Raman ($n = 1$), with a relatively small Stokes shift (so that the fundamental and Stokes fields are correlated in space):

$$G_{\mathrm{RS}} = G_{1-\mathrm{HRS}} = G_4 \approx \left|\frac{X}{\delta}\right|^4 \delta \mathrm{Im}[\alpha(X)]. \qquad (5)$$

For hyper-Raman scattering, where the hot spots for the fundamental and Stokes fields almost do not correlate ($n \neq 1$):

$$G_{n-\mathrm{HRS}} \approx \left(\left|\frac{X}{\delta}\right|^{2n} \delta \,\mathrm{Im}[\alpha(X)]\right) \times \left(\left|\frac{X_s}{\delta_s}\right|^{2} \delta_s \,\mathrm{Im}[\alpha(X_s)]\right), \quad (6)$$

where $X_s = X(\lambda_s)$.

In Fig. 3 we compare results of our numerical simulations based on the exact formula (4) with theoretical formulas (5) and (6), for enhancement of RS and HRS in fractal cluster-cluster aggregates of silver nanoparticle ($D \approx 1.78$). According to the figure, theoretical estimates are in accord with numerical simulations and predict giant enhancement: $G_{\mathrm{RS}} \sim 10^7$, $G_{2-\mathrm{HRS}} \sim 10^{10}$, and $G_{3-\mathrm{HRS}} \sim 10^{17}$. It is important to emphasize that the local field enhancement in the hot spots can significantly, by several orders of magnitude, exceed the average enhancement above. Thus, the local enhancement for RS in the hot spots can be between 10^{10} to 10^{12}, whereas the average is on the order of 10^7 only. The local enhancements in the hot spots for nonlinear optical processes, such as *n*-photon HRS, FWM, and others, can be extremely large, twenty and even more orders of magnitude. This level of local enhancement makes feasible nonlinear surface-enhanced spectroscopy of single molecules and nanocrystals.

A large variety of optical processes can be enhanced and otherwise modified by incorporating fractal clusters in the media or by ensuring that aggregation results in fractals. For example, fractals can be utilized to improve the performance of random lasers, such as powder lasers and laser paints [9], where lasing emissions can take place as a result of coherent multiple light-scattering in a disordered dielectric (or semiconductor) with appropriate structural elements. The notion of creating a "ring" laser cavity in a random medium through a sequence of multiple coherent scattering events along a closed path is itself, a fascinating, and almost counter-intuitive prospect. Scattering is normally considered detrimental to lasing; since, in a conventional laser cavity, it tends to remove photons from the lasing mode. In a properly constructed random medium, however, strong multiple scattering could return photons to the amplification region resulting in mode amplification [9]. By doping the laser powder or paint medium with fractal aggregates, or by imparting a fractal character to the medium as a whole, one could significantly decrease the pump power needed to effect lasing, in other words, one could decrease the lasing threshold.

FRACTALS IN MICROCAVITIES

Combining the energy-concentrating effects due to localization of optical excitations in fractals with other means for producing strong resonances can result in truly gargantuan local fields. For example, morphology-dependent resonances (MDRs) in dielectric microcavities are well known phenomena which can produce large intensity enhancements in the resonances that can lead to lasing [10]. These resonances, which may have extremely high quality factors ($Q = 10^5$ to 10^9), result from confinement of the radiation within the microcavity by total internal reflection. (We note that in contrast to the Q_f introduced above, characterizing the resonance enhancement for the field amplitude, the cavity's quality-factor $Q \equiv Q_I$ gives the resonance enhancement for the intensity; accordingly, in terms of the enhancement efficiency, Q in cavities should be compared with Q_f^2 in particles and fractals.) Light emitted or scattered from a source within the microcavity may couple to the high-Q MDRs in its spectral bandwidth, leading to the enhancement of the spontaneous and stimulated optical emissions. Hence, seeding fractal aggregates into appropriate microcavities should further increase the local fields.

Huge multiplicative field enhancement factors of this sort were obtained in [11], where it was found that lasing emission can be observed from Rhodamine 6G (R6G) dye molecules adsorbed on silver colloidal aggregates residing inside a cylindrical, quartz microcavity, even for dye molarity and threshold pump intensity both 3 orders of magnitude lower than for the corresponding dye laser in the absence of the colloidal aggregates. The fractal aggregates provide the resonant enhancement which is sufficient to lower the lasing threshold to the level of a He–Ne pump laser, at which point the overall observed enhancement of the light emission due to the combined effects of the microcavity and the fractal resonances was between 10^{10} to 10^{12}.

Huge enhancement of RS (from the sodium citrate present in minute concentration) in the fractal-microcavity composites has also been observed in [11]. The measurements show that the combined *average* RS-enhancement in fractal-microcavity composites is, at least, 10^{12} (with the factor 10^7 "coming from" the fractal colloids and the "extra" 10^5 being due to the microcavity). Because of concentration of the enhancement in the fractal hot spots, the *local* RS-enhancement in fractal-microcavities composite can be as large as 10^{18}, which is probably a record large SERS. In microcavities with larger Q (which are available), the enhancement can be even larger.

Surface-enhancement factor for hyper-Raman scattering from fractals is given by $G_{n-\mathrm{HRS}}$ in (4) and (6). In fractal-microcavity composites HRS can be further enhanced by the microcavity's MDRs. In the simplest approximation when the fractal and cavity's modes are "decoupled," the resultant enhancement for HRS is

given by the product $G_{n-\mathrm{HRS}} g_{\mathrm{cav}}^{(n)}$, where $g_{\mathrm{cav}}^{(n)}$ describes the cavity enhancement. This factor depends on whether the fundamental and Stokes waves couple to the MDRs of the cavity. If only the Stokes wave at ω_s is coupled to MDRs than $g_{\mathrm{cav}}^{(n)} \sim Q(\lambda_s)$, where $Q(\lambda_s)$ is the cavity quality-factor at the Stokes frequency (note that, if needed, Q can also take into account the decrease of the resonance quality due to the presence of fractals inside the cavity). If only the fundamental wave is coupled to the MDRs, then $g_{\mathrm{cav}}^{(n)} \sim Q^n(\lambda)$, where $Q(\lambda)$ is the cavity quality-factor at the fundamental frequency. When both waves are coupled to MDRs, $g_{\mathrm{cav}}^{(n)} \sim Q^n(\lambda)Q(\lambda_s)$; and if none of them is coupled to MDRs, than $g_{\mathrm{cav}}^{(n)} \sim 1$.

By combining the field enhancement in fractals with the microcavity's MDR enhancement, a truly gigantic enhancement can be obtained, especially, when both the fundamental and Stokes waves are coupled to the cavity modes [11]. Therefore, despite the fact that Raman hyper-polarizabilities are extremely small (so that under the normal, no-enhancement conditions, they can hardly be observed), in the fractal–microcavity composites, hyper-Raman scattering can be detected even at very low laser pumps. In recent experiments [11] the dramatically enhanced two- and three-photon pumped hyper-Raman scattering in fractal–microcavity composites was observed at extremely low pump powers provided by a He–Ne laser. Thus, placing fractal nanostructures in a microcavity opens new avenues for spectroscopy, including the nonlinear one, of single molecules.

Another cavity effect arises from the possibility of enhancing the spontaneous emission (SE) rate [12] of an emitter placed in a cavity (Purcell effect). Originally formulated for a localized dipole in resonance with a single cavity mode with quality-factor Q, the SE rate in the cavity mode referenced to the SE rate in a homogeneous medium, is given by the Purcell figure of merit $F \sim Q\lambda^3/V$, where V is the effective cavity volume. By placing a fractal aggregate in a microcavity further enhancement of the zero-point fields (and thus of the SE rate) can be achieved because of the high-quality factors of the aggregate's optical excitations and their localization in sub-wavelength regions of space. Under optimum conditions, the SE rate can be enhanced by the factor given by a product of G defined in (3) and the Purcell figure of merit F.

In summary, the optics of fractal media display a rich variety of effects some of which are hardly intuitive. Field localization of various sorts occur and recur in a wide gamut of disordered systems, most strikingly in those possessing dilational symmetry, leading to the enhancement of many optical phenomena, especially non-linear processes. Making judicious use of these enhancement effects and of other aspects of the many complex resonances that distinguish these systems can lead to new and unexpected physics, and to such applications as very low threshold lasers whose cavities are self-organizing loops of coherently scattered events in highly unconventional media. When developed, in the fullness of time, these disordered materials may attain a level of practical importance and versatility that might rival or surpass their geometrically ordered counterparts.

REFERENCES

1. Mandelbrot, B.B., 1982, *The Fractal Geometry of Nature* (San Francisco: Freeman); Barbasi, A.L. and Stanley, H.E., 1995, *Fractal Concepts in Surface Growth*, (Cambridge: Cambridge Univ. Press); Jullien, R. and Botet, R., 1987, *Aggregation and Fractal Aggregates*, (Singapore: World Scientific).
2. Alexander, S. and Orbach, R., 1982, *J. Phys. Lett.*, **43**, 625; 1991, *Fractals and Disordered Systems*, Bunde, A. and Havlin, S., Eds. (Heidelberg: Springer Verlag); 1986, *Fractals in Physics*, Pietroniero, L. and Tosatti, E., Eds., (Amsterdam: North-Holland); Sapoval, B., Gorbon, Th., and Margolina, A., 1991, *Phys. Rev. Lett.*, **67**, 2974.
3. Shalaev, V.M. and Stockman, M.I., 1987, *Sov. Phys. JETP*, **65**, 287; Butenko, A.V. *et al.*, 1988, *Z. Phys. D*, **10**, 81; Markel, V.A. *et al.*, 1991, *Phys. Rev. B*, **43**, 8183; Stockman, M.I. *et al.*, 1994, *Phys. Rev. Lett.*, **72**, 2486; Tsai, D.P. *et al.*, 1994, *Phys. Rev. Lett.*, **72**, 4149.
4. Stockman, M.I., Pandey, L.N., and George, T.F., 1996, *Phys. Rev. B*, **53**, 2183; Stockman, M.I., 1997, *Phys. Rev. E*, **56**, 6494; 1997, *Phys. Rev. Lett.*, **79**, 4562.
5. Safonov, V.P. *et al.*, 1998, *Phys. Rev. Lett.*, **80**, 1102; Shalaev, V.M. *et al.*, 1996, *Phys. Rev. B*, **53**, 2437; Markel, V.A. *et al.*, 1996, *Phys. Rev. B*, **53**, 2425; Shalaev, V.M., 1996, *Phys. Rep.*, **272**, 61.
6. Shalaev, V.M., 2000, *Nonlinear Optics of Random Media: Fractal Composites and Metal–Dielectric Films* (Berlin: Springer-Verlag).
7. Moskovits, M., 1985, *Rev. Mod. Phys.*, **57**, 783; 1982, *Surface Enhance Raman Scattering*, Chang, R.K. and Furtak, T.E., Eds. (New York: Plenum).
8. Kneipp, K. *et al.*, 1997, *Phys. Rev. Lett.*, **78**, 1667; Nie, S. and Emory, S.R., 1997, *Science*, **275**, 1102.
9. Letokhov, V.S., 1968, *Sov. Phys. JETP*, **26**, 835; Lawandy, N.M. *et al.*, 1994, *Nature*, **368**, 436; Wiersma, D.S. and Lagendijk, A., 1996, *Phys. Rev. E*, **54**, 4256; John, S. and Pang, G., 1996, *Phys. Rev. A*, **54**, 3642; Berger, G.A., Kempe, M., and Genack, A.Z., 1997, *Phys. Rev. E*, **56**, 6118; Cao, H. *et al.*, 1999, *Phys. Rev. Lett.*, **82**, 41; 2000, *ibid.*, **84**, 4276.
10. 1996, *Optical Processes in Microcavities*, Chang, R.K. and Campillo, A.J., Eds. (Singapore: World Scientific).
11. Kim, W., Safonov, V.P., Shalaev, V.M., and Armstrong R.L., 1999, *Phys. Rev. Lett.*, **82**, 4811; Kim, W. *et al.*, in preparation for publication.
12. Purcell, E.M., 1946, *Phys. Rev.*, **69**, 681; Haroche, S. and Klepper, D., 1989, *Phys. Today*, **42**, no. 1, 24; 1996, *Microcavities and Photonic Bandgaps: Physics and Applications*, Weisbuch, C. and Rarity, J., Eds., NATO ASI, ser. E, vol. 324 (Dordrecht: Kluwer); Yablonovitch, E., 1995, Light Emission in Photonic Crystal Micro-Cavities, *Confined Electrons and Photons*, Burstein, E. and Weisbuch, C., Eds. (New York: Plenum), p. 635.