scale using two photon excitation microscopy combined with spectral and polarization information. The main result of this work is the use of size-resonant local field "hot spots" in order to enhance the optical response of non-linear chromophores in close contact with such structures. The molecules have been deposited using the Langmuir-Blodgett technique (figure 1) on gold fractal supporting layers fabricated from metal evaporation under high vacuum, with a controlled mass thickness ranging from 20Å to 150Å. Samples of mass thickness from 45Å to 90Å present a fractal morphology as evidenced by Transmission Electron Microscopy measurements.

A 120 fs-pulsed source excitation was used at a fundamental wavelength of ~1020 nm, and two-photon detection was coupled to an inverted microscope with a spatial resolution of 500 nm. The surface SHG response from the gold substrates themselves showed highly localized enhancements with values up to two orders of magnitude higher than observed previously using the NSOM technique.[4] We show a strong effect of the localization induced exaltation effect depending on the granularity of the samples (figure 2), with SHG enhancement factors ranging from 10 to 300. Simultaneous spectral information allowed us to evidence the presence of continuum emission from the structures at the onset and above damage threshold. We report for the first time on such structures a strong polarization dependence of the SHG "hot spots" localized signal, which reflects the sensibility of SHG to the local surface morphology.

The deposition of a molecular monolayer on such substrates resulted in a strong effect on their non-linear response. Molecules in close contact to the metal show SHG signals up to 10^3 times their value on metal-free glass substrates. A fast decay of this enhancement is however observed, which might originate from molecule to metal energy transfer. This effect is analyzed in conjunction with two-photon fluorescence response.

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Local-Field Correction for a Nanosource in a Crystal

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The influence of the local environment on the electromagnetic properties of a nanosource (atom or molecule at a surface, quantum dot in a microcavity, . . .) is of crucial importance in modern optics and optoelectronics. We present a self-consistent calculation of the local-field correction experienced by an interstitial nanosource in a crystal with cubic symmetry. The local-field correction is obtained from a rigorous description of spontaneous emission[1] which includes the Purcell effect and the dipole-dipole coupling between the source and the host. This rigorous description is used as a reference theory to devise simpler approaches to the local-field problem that can be applied to a wider range of crystal symmetries.

We consider a two-level atom that is located at r_0 in a microcavity and has an electric dipole transition moment along direction $\sigma = x$, y or z. The normalized decay rate is given by[2]

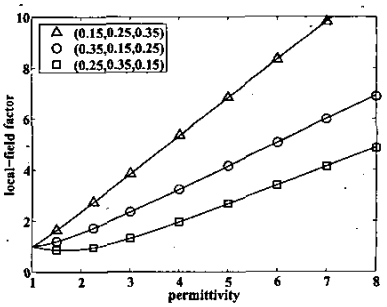
$$\frac{\Gamma_\sigma}{\Gamma_0} = 1 + \frac{3}{2k_0^3}\,\mathrm{Im}[F_{\sigma\sigma}(\vec{r}_0, \vec{r}_0; \omega)], \qquad (1)$$

where Γ_0 and $k_0 = \omega/c$ are the decay rate and wave vector in free space, and F is the electric field-susceptibility which represents the electromagnetic response of the cavity. Finding the field susceptibility entering the expressions for the decay rate amounts to finding the electric field reflected back at the source by the cavity. To solve this problem we use the coupled dipole method (CDM) which was developed to study the scattering of light by particles with arbitrary shapes.[3,4,5,6] In the CDM, the scatterer is discretized over a cubic lattice. The details of the theory are given elsewhere.[1,7] The local field correction L is defined as[7]

$$\Gamma = L^2\Gamma_{cav} + \delta\Gamma, \qquad (2)$$

where Γ_{cav} represent the effect of the cavity geometry (reflection at the boundaries of the cavity). The term $\delta\Gamma$ represents the dissipative part of the dipole-dipole interaction between the source and the polarizable elements of the host medium (for a lossless medium $\delta\Gamma = 0$). When we consider a spherical cavity, we can use the theory of Chew[8] to compute Γ_{cav}. By comparing to the CDM result, Eq. (2) allows us to find $|L|$.[7] Therefore the CDM allows us to compute rigorously the retarded, self-consistent local-field correction, and separate the contributions from the geometry of the microcavity and the local-field effect.

We plot in Fig. 1 the local-field factor (absolute value) found with the CDM (the direction of the transition moment of the source defines direction z). In some special cases, like the standard virtual cavity factor $(\varepsilon + 2)/3$, the local-field factor depends linearly on the permittivity ε. In the general case the linearity of the local-field factor with respect to the permittivity is only asymptotic (for large ε). The CDM shows that we can separate the influence on the dynamics of a nanosource in an arbitrary microcavity of the geometry of the cavity from the local-field effect. Thus, for a given geometry of the cavity, the local-field factors corresponding to different lattice symmetries of the host crystal lattice can be computed as lattice sums of an infinite crystal. This is illustrated in Fig. 1 where we compute the local-field factor L directly as a rapidly converging sum[9,10] for an infinite crystal, and we compare our lattice-sum result to our rigorous, self-consistent, CDM calculation for sources at arbitrary locations in the lattice. The agreement is excellent, opening the way to a simple description of local-field effects for interstitial nanosources in finite crystals with complex symmetries.



QMI5 Fig. 1. Absolute value of the local-field factor versus the permittivity ε for an interstitial source. Symbols: self-consistent CDM calculation. Solid lines: lattice-sum results. The numbers between parenthesis give the (x, y, z) normalized coordinates of the source in the lattice. The transition moment of the source defines the z direction.

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