Apertureless Near-field Optical Microscopy Investigations of Long-range Indirect Interactions in Unordered Metamaterials

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Abstract

Statistical near-field optical microscopy analysis of unordered metamaterials reveals long-range interactions between distant "metaatoms", not observed with far-field optical spectroscopy. The system is characterized by a dense interaction matrix, in contrast to the sparse hopping-type interactions encountered in solid state materials

1. Introduction

In metamaterials, metallic particles are often used in a periodic, crystalline arrangement to gain critical control of the light-matter interaction in a predictable manner. Their localized plasmonic response to *direct* electromagnetic radiation is amenable to numerical prediction as a function of size, shape and placement, which may be used to define spectral properties[1,2]. A second path to control is through hybrization of plasmonic eigenmodes of closely situated metaatoms, which interact via their near-fields[3]. Here we show that in addition a *long-range, indirect* mode of excitation is present in extended nanoplasmonic systems, which can be critical to understanding near-field spectral and spatial behaviour. In particular, the local response of individual particles in unordered arrays of plasmonic nanodiscs depends sensitively on the collective and coherent influence of all active scatterers in its extended neighbourhood.

2. Methods and Results

We present recent studies of deterministically aperiodic and amorphous metamaterials using apertureless scanning near-field optical microscopy (aSNOM) and finite-difference-time-domain simulations (FDTD). aSNOM achieves lateral resolution of better than 10nm - well beyond the far-field diffraction limit - by utilizing the field enhancement at the apex of sharp tips. An interferometric measurement scheme allows detecting both near-field intensity and optical phase. The proper choice of tip position, together with optimal polarizer and analyzer angles of our cross-polarization scheme ensures a background free plasmonic eigenmode mapping. Comparison with simulation data not including the tip shows that the measurement has little to no influence on the bare sample fields[4,5].

Using this approach, we study the near-field optical interactions in unordered metamaterial systems. A large number of dipole-excitable metaatoms (gold nanodisks) have been investigated. In the far-field

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extinction spectra, samples of different densities yield the same fundamental spectral features. In the near-field optical data, however, the statistical behaviour analysed in terms of the local dipole orientation observed at each disk results in a clear correlation with nearest-neighbour inter-particle separation.

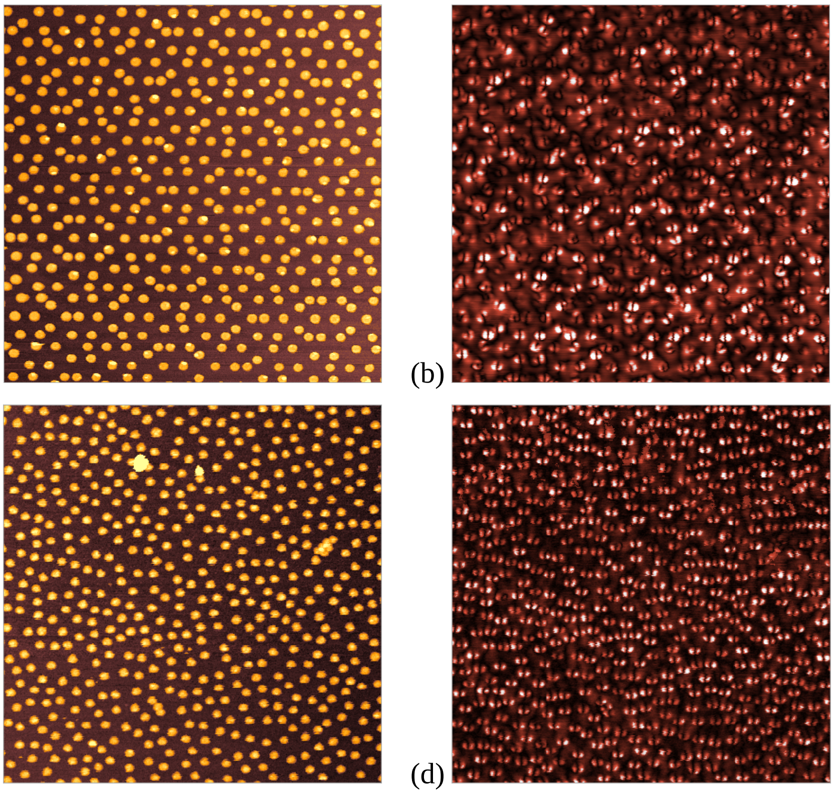

Fig. 1: Apertureless Scanning Near-field Optical Microscopy (aSNOM) study of unordered metamaterials. Typical 10µm x 10µm area scans of metamaterials composed of dense arrays of dipole-resonant gold nanodisks. Topography (a,c) and near-field optical field strength (b,d) of a deterministic aperiodic structure (a,b) and a truly random structure (c,d) showing only weak short range order.

When the identical large area topographies were simulated with FDTD, the same analysis yields a quantitatively different behaviour, at first. We demonstrate how these differences can be traced back not to varying sample properties, but are a result of different excitation geometries. The coherent interaction is found to extend far beyond that reported for simple plasmonic dimer systems, suggesting a collective, long-range interaction.

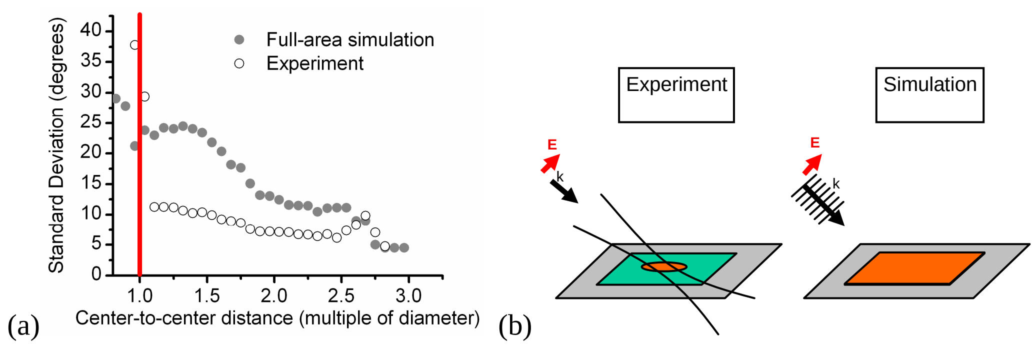

Fig. 2: (a) Statistical analysis of the distributions of the in-plane dipole orientation as a function of nearest-neighbour distance. A clear discrepancy reveals itself between experimentally and numerically obtained results. (b) Different kinds of excitations in experiment and simulation. Systematic analysis of their influence uncovers the strong dependence of local variations in the optical response on distant scatterers.

3. Summary and Outlook

The large local fluctuations observed in the near-field optical response are not necessarily reflected in corresponding far-field optical properties. Two immediate conclusions of these findings can be drawn: Firstly, as the situation is reminiscent of the seemingly unpredictable behaviour of plasmonic "hot spots" in roughened metallic surfaces, related stochastic phenomena such as surface enhanced Raman scattering might be governed by similar long-distance effects. Secondly, the dependence of local field responses on excitation conditions at distant areas suggests novel ways to engineer specific electro-magnetic fields through spatially controlled illumination.

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