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Intensity correlations in metal films with periodic-on-average random nanohole arrays

Randhir Kumar, Sushil Mujumdar *

Nano-optics and Mesoscopic Optics Laboratory, Tata Institute of Fundamental Research, Mumbai 400005, India

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ABSTRACT

We report detailed numerical studies based on three-dimensional finite-difference time domain computations of the intensity–intensity correlations in deliberately randomized, periodic-on-average systems. Correlation analyses are carried out in plasmonic thin films with nanohole arrays as a function of strength of disorder. We find that the intensity at certain uncharacteristic wavelengths remains strongly correlated with that in the periodic system, and these wavelengths do not match the global maxima of the periodic transmission spectrum. The study indicates that the strength of correlations is related to the pinning of the intensity to the holes. Since the intensity pinning is special characteristic of metals, the effect is only applicable in plasmonic systems.

1. Introduction

Wave propagation in random media has been a subject of immense importance, with a long-standing history of literature ranging from sub-fields such as atmospheric physics to the recent electromagnetic crystals [1,2]. Perhaps the most well-known exotic wave phenomenon originating from disorder is the Anderson localization of waves, manifested by the self-interference of wavelets scattered from centers of disorder. Originally proposed as a quantum phase transition of electrons in a crystal [3], the phenomenon has been observed in classical waves such as light [4,5] and sound [7]. Due to the ability to reveal new physics of disorder, a large research effort is focused on the study of light localization [6]. However, one particular area in which the experimental evidence of localization, although available [8], is rather scarce is the plasmonic domain. Interestingly, the physics of disordered systems depends critically on dimensionality, in that localization is easier to observe in lower dimensions. So plasmonic polaritonic systems, owing to their inherent two-dimensional nature, are excellent candidates for such studies. Experimental efforts have already given evidence of interference of plasmonic waves, which indicates their viability for further mesoscopic studies. However, the omnipresent dissipation offered by the real metals creates formidable challenges in studies aimed at localization.

An exciting variant in disorder configurations is a periodic-on-average random system (PARS). Essentially, PARS involves a periodic configuration that is deliberately randomized, with a view to studying the effects of randomness on Bloch modes existing in the erstwhile periodic system. Such a configuration was first discussed in the earliest works proposing photon localization [9], and has subsequently seen several theoretical and experimental developments [10–14]. The significant effects in such PARS systems arise due to the modification of bandedges and bandgaps in the underlying periodic structures. In the plasmonic domain, too, the presence of bands and bandgaps in periodic structures has been amply demonstrated [15,16]. Plasmonic quasicrystals have also been studied, wherein systematic departures from periodicity were introduced [17,18]. However, not many studies can be found in the plasmonic PARS system. To our belief, given the volume of research in periodic plasmonic systems, it is appropriate to extend it to periodic-on-average randomness. In this paper, we numerically address a plasmonic PARS system. In particular, the periodic structure that we choose for the study is the nanohole array. Such arrays have been shown to exhibit extra-ordinary transmission [19], and have been thoroughly studied in literature. As a precursor to our studies aimed at localization, we address the plasmonic intensity correlations in nanohole arrays as a function of increasing disorder. We find that the intensity distribution across configurations with varying disorder shows lasting correlations at uncharacteristic wavelengths. We quantify these correlations as a function of disorder strength and trace the behavior to intensity-pinning at the nanoholes in the metallic system. Since the concept of intensity-pinning is special to metals, we believe this study reveals significant departures in behavior of metallic systems vis-a-vis dielectric systems.

* Corresponding author.
E-mail address: mujumdar@tifr.res.in (S. Mujumdar).
URL: http://www.tifr.res.in/~mujumdar (S. Mujumdar).

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2. Computations

The virtual structure comprised a gold nanofilm (film thickness 100 nm and dimensions 10 μm × 10 μm) laid onto silica glass (n_{ref} = 1.46), with an overwritten nanohole array. Herein we discuss an array with a lattice constant of 700 nm and hole diameter of 300 nm, and mention that other combinations provided qualitatively similar results. We also studied thinner gold films (thickness 50 nm), which exhibited similar behavior, certifying the thickness-independence of the effect. The total hole matrix written was 12 by 12 over the entire film, with a clearance of 0.8 μm left at all edges. Fig. 1(a) exhibits a section of the periodic array. Randomness was introduced into the array by deviating the central positions of the holes by an amount δ, which was a Gaussian random variable with the width of the distribution given by σ, expressed hereafter as percentage disorder. Under this definition, the strongest case of 100% disorder still implies a configuration wherein an underlying periodicity can still be weakly present, and two holes can come just near each other but cannot touch each other. Any stronger randomness will conjoin two holes, leading to undesirable effects. Fig. 1(b) shows the section of the aperiodic array with a degree of disorder of η = 40%. The transmittance and intensity distributions were studied using finite difference time domain computations, using the commercial software Lumerical. The computational space was resolved into a grid with size 661 × 661 × 44 in X, Y and Z dimensions respectively. The simulation was run for a total time period of 100 fs. Adaptive gridding enabled us to set the spatial resolution to 15 nm in X and Y, and 5 nm in the metallic region along the Z-axis. These numbers are in tune with the smallest structures that can be lithographically created in real fabrication processes. The input intensity was polarized along the X -axis. Perfectly absorbing boundaries were laid at the edges of the computational box. Initially, a continuous plane wave was made normally incident onto the film from underneath, and the transmittance across the film was detected by a detector array at a distance 250 nm from the metal surface. The inbuilt DFT algorithm provided the spectral decomposition of the transmitted light. Fig. 1(c) depicts the transmittance spectra for various disorder strengths. The periodic film shows the characteristic peaks and valleys in the spectrum, indicating the collective modes over the entire film, showing that the nanohole array was acting as a plasmonic crystal. The addition of disorder essentially deteriorates this collective plasmonic mode by nullifying the peaks and valleys. By about η = 60%, the transmission is relatively flat across the spectrum. The persistent peak at $\lambda \sim 723$ nm indicates that it did not originate from a collective mode, and is not invoked in this analysis. The two vertical lines mark the wavelengths of maximum and minimum transmission, namely λ_{max}=588.2 nm, and λ_{min} =486.9 nm. The observed trends continue up to 100% disorder. In comparison, in a dielectric system comprising periodically arranged scatterers, the collective modes formed by the co-operative scattering of all scatterers realize the photonic modes of the crystal, whose frequency shifts upon introduction of the slightest disorder. This shift is particularly noticeable because of the high quality factor of the modes in the absence of absorptive loss. In contrast, the metallic system is lossy, increasing the bandwidth of the modes. Therefore, the shift in wavelength, although discernible, is very mild. The only noticeable effect it creates is the

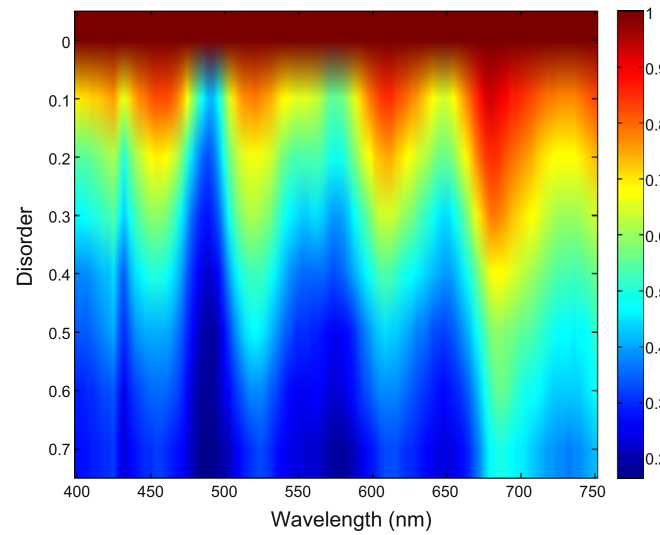

Fig. 2. Intensity–intensity correlation coefficient ξ as a function of wavelength across disorder strengths (Y -axis). Intensity at certain wavelengths exhibits strong correlations with that in the periodic system.

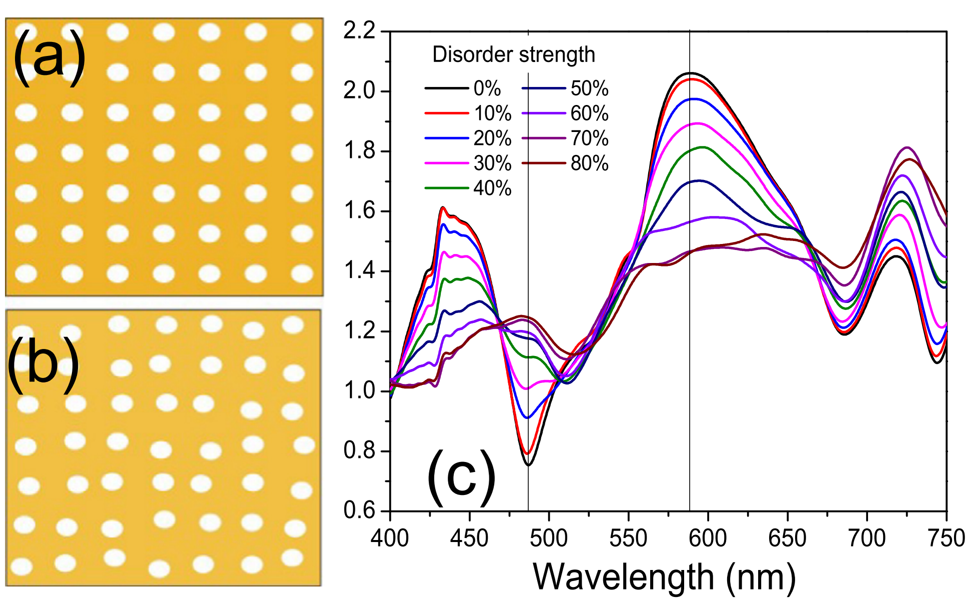

Fig. 1. (a) Section of gold film with a periodic nanohole array, with dimensions as mentioned in the text. (b) Section of the film with 60% disorder introduced in the periodic nanohole array. (c) Transmission spectra through various arrays with changing disorder, showing the modification of the photonic mode of the film. Vertical dotted lines mark the wavelengths of maximum and minimum transmission, namely λ_{max} = 588.2 nm, and λ_{min} = 486.9 nm.