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Spatial coherence of light measured by nanoscattering: supplementary material

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This document provides supplementary information to "Spatial coherence of light measured by nanoscattering," https://doi.org/10.1364/optica.5.000067. Here we describe in detail the estimation of the beam focus at the sample, as well as the fabrication of nanoparticles, and light interaction between the particles and substrate surface. Also, we describe the data analysis methods used to interpret the measured data.

BEAM FOCUS

The beam diameter at the focus was estimated in terms of two observations. First, by moving a CCD camera (pixel size 5.2 μm) longitudinally we measured the minimum focus diameter at the sample to be approximately 70 μm when the beam width is defined as the distance over which the intensity drops to 10 percent of its maximum. The same definition for the beam width is used in connection with Fig. 1c (of the main text) for unfocused beam, implying the scaling factor of 20 for the degree of coherence. Second, by moving the focus spot over a vertical 70 μm wide Au slab located in the vicinity of the particles, we noticed very little edge diffraction at the center of the slab, suggesting that the minimum spot size indeed is at the plane of the nanoparticles.

PARTICLE FABRICATION

The gold nanocube particles were fabricated on a standard <100> Si wafer. First the wafer was cleaned using O_2 plasma. The wafer was next coated with a 200 nm layer of ZEP7000 electron beam resist (Zeon Chemicals) and soft baked at hot plate at 180°C for 180 seconds. The nanoparticles were defined with electron beam exposure with previously optimized parameters using Vistec EBPG 5000+ ES HR system. The exposed resist layer was developed in ethyl 3-ethoxy propionate (EEP) and rinsed in isopropanol (IPA) and deionized water. Next, a 3 nm Cr layer was sputtered on the resist film. This layer acts as an adhesion layer for gold that generally has poor adhesion to silicon. This was followed by a thermal evaporation of gold with desired film thickness. Finally, a lift-off was performed in 40°C anisol and the sample was rinsed in IPA.

PARTICLE-SURFACE INTERACTION

The particles used in the measurement are Au cubes with dimensions much smaller than the wavelength of light. We may approximate them as electric dipoles, whose scattered electric fields are proportional to the incident field at the particle site. The proportionality coefficient is the polarizability which is specified by the size, shape, and material of the particle. For a cubical particle located in free space, the polarizability has been derived earlier [1]. However, since the particles are placed on a Si substrate, we need to take the reflections from the surface into account. The reflected field consists of two contributions, the direct reflection of the incident light from the surface and the field that is scattered by the dipole and reflected back from the surface. These are included in the analysis to form an effective polarizability [2]. Furthermore, we assume that the particle separation is large enough to ignore the interaction between the particles [3].

Taking the reflections into account the total far field at point $\mathbf{r}$ generated by a particle of effective (3×3 matrix) polarizability $\boldsymbol{\alpha}(\omega)$ and located at $\mathbf{r}_i$, $i \in (1,2)$, is given as [2]

$$\mathbf{E}_i^{(\mathrm{f})}(\mathbf{r},\omega) = \frac{\mu_0 \omega^2 \mathrm{e}^{ikr_i}}{4\pi r_i} \times \left[A\hat{\mathbf{u}}_{i,\varphi}\hat{\mathbf{u}}_{i,\varphi}^{\mathrm{T}} + \hat{\mathbf{u}}_{i,\theta}\hat{\mathbf{u}}_{i,\theta}^{\mathrm{T}} + B\hat{\mathbf{u}}_{i,\theta}\hat{\mathbf{u}}_{i,-\theta}^{\mathrm{T}} \right] \boldsymbol{\alpha}(\omega)\mathbf{E}_{\mathrm{in}}(\mathbf{r}_i,\omega). \tag{S1}$$

In this expression, μ_0 is the vacuum permeability, ω is the angular frequency, k is the wavenumber, $r_i = |\mathbf{r} - \mathbf{r}_i|$, $A = 1 + R_s \exp(ik\Delta r)$, and $B = -R_p \exp(ik\Delta r)$. Further, R_s and R_p are the Fresnel reflection coefficients for s-polarized and p-polarized light on the air-substrate interface, respectively, and Δr is the additional distance that the reflected field propagates. The unit vectors $\hat{\mathbf{u}}_{i,j}$, $i \in (1,2)$ and $j \in (\varphi, \theta)$, refer to the spherical polar coordinates when the origin is at the particle site $\mathbf{r}_i$, while $\hat{\mathbf{u}}_{n,-\theta}$ is $\hat{\mathbf{u}}_{i,\theta}$ but in the $-\theta$ direction, and T denotes the transpose. Finally, $\mathbf{E}_{\mathrm{in}}(\mathbf{r}_i,\omega) = E_{\mathrm{in}}(\mathbf{r}_i,\omega)\hat{\mathbf{y}}$ is the incident field, taken to be linearly polarized in the y direction represented by the unit vector $\hat{\mathbf{y}}$. The effective polarizability matrix of a material nanocube on a substrate is given as [2]

$$\boldsymbol{\alpha}(\omega) = \left(1 - r_p e^{i2kh}\right) \frac{32\pi h^3}{c^2 \mu_0 r_p} \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1/2 \end{bmatrix}, \tag{S2}$$

where r_p is the quasi-static limit of the Fresnel reflection coefficient at the particle-substrate interface, h is the distance from the particle center to the surface, and c the speed of light in vacuum.

The field at the detector is the sum of the fields scattered by the two particles, i.e., $\mathbf{E}_{\mathrm{tot}}(\mathbf{r},\omega) = \mathbf{E}_1^{(\mathrm{f})}(\mathbf{r},\omega) + \mathbf{E}_2^{(\mathrm{f})}(\mathbf{r},\omega)$. Furthermore, the intensity captured by the detector is $I(\mathbf{r},\omega) = \mathrm{tr}\left[\langle \mathbf{E}^*_{\mathrm{tot}}(\mathbf{r},\omega)\mathbf{E}^{\mathrm{T}}_{\mathrm{tot}}(\mathbf{r},\omega)\rangle\right]$, where tr and the angle brackets denote the trace and the ensemble average, respectively. After straightforward developments, we find that

$$\begin{aligned} I(\mathbf{r},\omega) &= I_1(\mathbf{r},\omega) + I_2(\mathbf{r},\omega) + 2\sqrt{I(\mathbf{r}_1,\omega)I(\mathbf{r}_2,\omega)} \\ &\times \mathrm{Re}\left\{\mathrm{tr}\left[\mathbf{M}^*(\mathbf{r},\mathbf{r}_1,\omega)\hat{\mathbf{y}}\hat{\mathbf{y}}^{\mathrm{T}}\mathbf{M}^{\mathrm{T}}(\mathbf{r},\mathbf{r}_2,\omega)\right]\right\} |\mu(\mathbf{r}_1,\mathbf{r}_2,\omega)| \\ &\times \cos\left\{\arg[\mu(\mathbf{r}_1,\mathbf{r}_2,\omega)] + k(r_2 - r_1)\right\}, \end{aligned} \tag{S3}$$

where $I_i(\mathbf{r},\omega)$ is the far-field intensity due to the particle located at $\mathbf{r}_i$ and $I(\mathbf{r}_i,\omega)$ is the intensity incident on it, $i \in (1,2)$, $\mu(\mathbf{r}_1,\mathbf{r}_2,\omega)$ is the (complex) degree of coherence of the field at the particles, whereas Re and arg denote the real part and the phase of a complex number, respectively. Furthermore, the matrix

$$\begin{aligned} \mathbf{M}(\mathbf{r},\mathbf{r}_i) &= \frac{8h^3k^2(1 - r_p e^{i2kh})}{r_i r_p} \\ &\times \left[A\hat{\mathbf{u}}_{i,\varphi}\hat{\mathbf{u}}^{\mathrm{T}}_{i,\varphi} + \hat{\mathbf{u}}_{i,\theta}\hat{\mathbf{u}}^{\mathrm{T}}_{i,\theta} + B\hat{\mathbf{u}}_{i,\theta}\hat{\mathbf{u}}^{\mathrm{T}}_{i,-\theta}\right], \end{aligned} \tag{S4}$$

with $i \in (1,2)$ is, to a good approximation, constant over the detector scanning range (± 6 mm at 55 mm distance). It further satisfies $\mathbf{M}(\mathbf{r},\mathbf{r}_1) \approx \mathbf{M}(\mathbf{r},\mathbf{r}_2)$ (since $r_1 \approx r_2$), implying that equation (S3) assumes the form

$$\begin{aligned} I(\mathbf{r},\omega) &= I_1(\mathbf{r},\omega) + I_2(\mathbf{r},\omega) \\ &+ 2\sqrt{I_1(\mathbf{r},\omega)I_2(\mathbf{r},\omega)}|\mu(\mathbf{r}_1,\mathbf{r}_2,\omega)| \\ &\times \cos\left\{\arg[\mu(\mathbf{r}_1,\mathbf{r}_2,\omega)] + k(r_2 - r_1)\right\}. \end{aligned} \tag{S5}$$

According to the above equation, the far-field intensity varies sinusoidally with position and the visibility of the modulation is obtained from equation (1) of the main text.

DATA ANALYSIS

The analysis of the recorded far-field intensity-fringe patterns was carried out in three steps. First, the average of numerous intensity distributions was taken to even out random fluctuations. Second, the background, comprising of reflections from the silicon surface as well as stray light, was subtracted from the averaged data. The background was measured by scanning the intensity the same way as in the particle scattering measurement, but with the beam focused onto the substrate approximately 50 μm away from the nanocubes. This ensures that the measured background is close to the actual background noise of the particle site. Finally, the noise-reduced intensity patterns were subjected to data fitting. In this process the fitting function was

$$f(s) = f_1 \cos^2\left[\frac{\pi a}{\lambda}\sin\left(\arctan\frac{s}{D}\right) + f_2\right] + f_3, \tag{S6}$$

where D is the distance between the particles and the detector and f_n, $n \in (1,2,3)$, are fitting coefficients. The nonlinear least-squares method was employed to find the fitting parameters.

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