THE RESPONSE OF NANOSTRUCTURED SURFACES IN THE NEAR FIELD

ARISING FROM: P. LALANNE & J. P. HUGONIN, *NATURE PHYS.* **2,** 551–558 (2006)

To the Editors — The authors of ref. 1 state that their numerical calculation of the transmission intensity dependence of very simple subwavelength planar structures in a silver film agrees well with an earlier-developed model[2], but both model and numerical simulation show significant disagreement with the experimental results of ref. 3. The authors[1] speculate that the silver surfaces of the subwavelength structures used in ref. 3 were contaminated by an 11 nm overlayer of silver sulphide, because such a layer would bring the reported experimental results and their calculations into better agreement.

We have analysed the physical–chemical surface properties of the single-slit, single-groove subwavelength-structured silver films used in the experiments with high-resolution transmission electron microscopy (HRTEM), and we have calculated fully vectorial numerical solutions to Maxwell's equations for the relevant structures using the finite-difference-time-domain (FDTD) technique[4]. The HRTEM analysis shows that the silver films are free of detectable contaminants with a detection limit orders of magnitude below the 11 nm layer suggested by ref. 1. Furthermore, the FDTD calculations are in excellent agreement with experiment (Fig. 1a), showing a rapid fringe amplitude decrease in the near-zone (slit–groove distance out to 3–4 wavelengths). Extended FDTD calculations to slit–groove distances beyond the near-zone (Fig. 1b) show that the surface wave evolves to the expected bound surface plasmon polariton (SPP). The key finding is that a surface wave in the near-zone consists of a distribution of transient surface modes adjacent and in addition to the bound SPP. Beyond the near-zone the results confirm that the transients dissipate with only the bound SPP mode surviving. This result has important implications for the interpretation of light transmission through arrays of subwavelength structures with subwavelength pitch. The common assumption that only the SPP mode is populated over the entire surface is not justified in the near-zone.

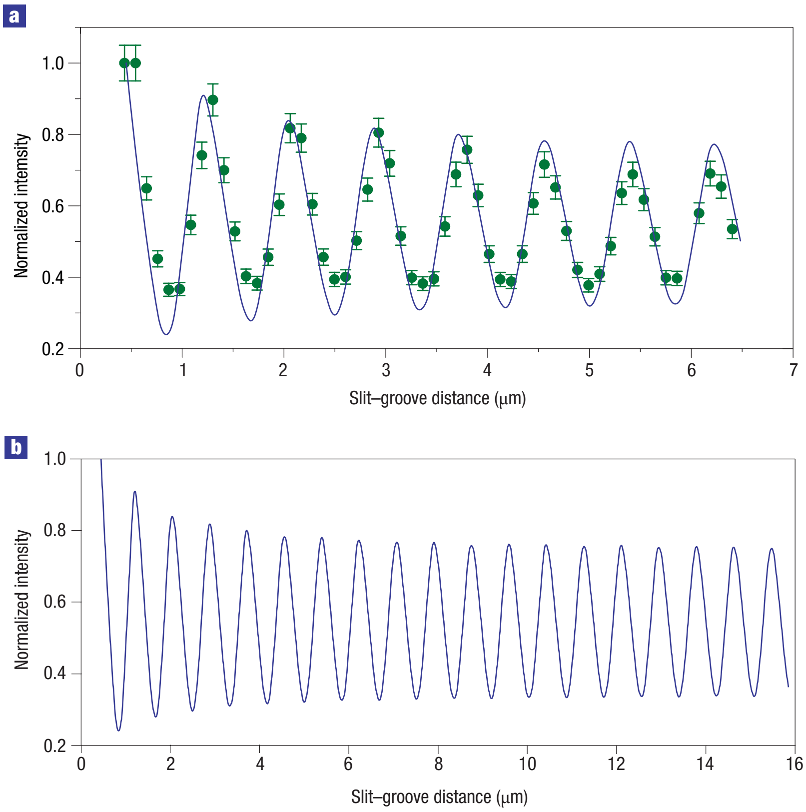

Figure 1 Comparison of FDTD simulation and experiment. **a**, The green points are experimental data taken from ref. 3. The blue curve shows our FDTD result. **b**, The blue curve plots the same FDTD calculation as in **a** but extended to 16 μm slit–groove distance.

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Authors' reponse — Because Maxwell's equations for linear dielectric materials are exact, computation plays a crucial role in the analysis of light scattering by subwavelength structures. In ref. 1, two of the present authors use a fully vectorial method to analyse the normalized coupled power $|S|^2/|S_0|^2$ into the fundamental propagating slit mode of a slit–groove geometry in a silver film previously studied in ref. 2, see Fig. 1a. In agreement with the experimental results[2], $|S|^2/|S_0|^2$ exhibits an oscillatory behaviour as a function of the slit–groove distance d (Fig. 1b). However, the theoretical oscillation frequency, which perfectly matches the propagation constant $k_{SP} = k_0(\varepsilon_{Ag}/(\varepsilon_{Ag}+1))^{1/2}$ of the Ag–air surface plasmon polariton (SPP), significantly differs from the experimental one. Thus, an SPP-mediated interaction mechanism is promoted in ref 1, leading to an interpretation very different from that in ref. 2.

In the above comment, the authors provide FDTD computational results that nicely agree with the experimental results in ref. 2, and state that high-resolution TEM images show that their silver samples are basically free of detectable contaminants. This seems to question (1) the accuracy of the numerical results in ref. 1, and (2) the validity of the SPP-driven mechanism proposed in ref. 1.

In Fig. 1b, we show computational results for the slit–groove geometry obtained with three different fully vectorial frequency-domain methods. The black curve has been obtained with a Fourier modal method that relies on a supercell approach incorporating a complex nonlinear coordinate transform along the x coordinate[3]. It is the same as that in Fig. 1 in ref. 1. We have used 601 Fourier harmonics for the computation. A finite-element method in a commercial software package[4] has been used to obtain the red curve by using a space discretization with second-order nodal Whitney elements, and a refined mesh at the groove and slit edges and at the Ag–air interface. The blue curve was obtained with a local-eigenmode modal method CAMFR[5]. The method is similar to the Fourier modal method, except that the field expansion uses the local eigenmodes, whose propagation constants are obtained by solving for the roots of a transcendental equation. The numerical results have been obtained with 300 modes and with perfectly matched layers implemented as a complex stretching[6]. The predicted oscillation amplitudes differ from one curve to another, but remarkably in the present context, all results exactly show the same oscillation frequency k_{SP}. To summarize, computational results obtained with three totally different methods do not corroborate the FDTD results in the above comment, although the same permittivity and the same geometrical parameters have been used in all calculations. These consistent data therefore question the accuracy of the FDTD results. Additionally, they convincingly support our initial statement that the oscillations observed in ref. 2 are essentially due to the Ag–air SPP, even for short distances. Concerning the possibility of a contamination of silver, experimental data with a gold metal are currently under progress in our laboratory.

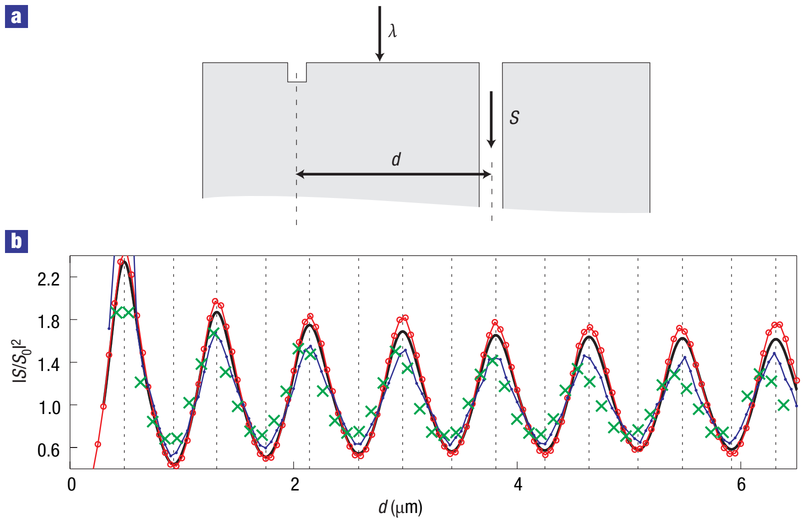

Figure 1 Three totally different methods provide the same oscillation frequency for the slit–groove scattering problem and for all distances. **a**, Geometry under consideration. **b**, The black curve (same as that in Fig. 1 in ref. 1), and the red and blue curves are computational data obtained with the methods of refs 3, 4 and 5, respectively. The green crosses are experimental data from ref. 2. All computations have been obtained for $\varepsilon_{Ag} = -33.22 + 1.1700i$ as in ref. 2, for $\lambda = 852$ nm, for a groove depth and width of 100 nm. For the sake of frequency comparison, the dotted vertical lines indicate the black curve extrema.

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