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## High order standing-wave plasmon resonances in silver u-shaped nanowires

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Optical measurements of the transmission spectra through nanofabricated planar arrays of silver u-shaped nanowires on a silicon substrate resonating at infrared frequencies are performed. Good agreement with the numerically simulated surface plasmon standing wave resonances supported by the structures is found. Such resonances exhibit field enhancement and are able to provide magnetic and electric responses when used as the unit cell of a metamaterial. The magnetic excitation of the resonators using oblique incidence is shown to be drastically reduced by the existence of a high index substrate such as silicon. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4759444>]

### INTRODUCTION

The optical properties of metallic nanostructures with features smaller than the wavelength of incoming light have proven useful in numerous applications. The study of their behavior involves the extremely interesting interaction of light with the plasma of free electrons in the metal. The variety of phenomena that arise from such coupling gives rise to the field of plasmonics.<sup>1</sup> Surface plasmons are waves of collective excitations of the conduction electrons in metals coupled to light and propagating through the surface of metallic conductors. Such waves are of special interest since the associated field mode profile is confined to the surface and decays exponentially away from it, allowing surface plasmons to break the diffraction limit of conventional dielectric waveguides,<sup>2</sup> as well as achieving near field enhancement<sup>3</sup> and superlensing.<sup>4</sup> The possibility to tailor the nanostructure of metallic nanoparticles has enabled scientists to profoundly study and engineer such waves.

The interesting properties of such plasmons arise especially at and around the plasmonic resonances. Metallic nanoparticles show localized surface plasmon resonances (LSPR) in which strong collective electron oscillations are induced in the metallic nanostructures, with applications in spectroscopy and sensing<sup>5</sup> thanks to their field enhancement. These LSPRs are also responsible for strong electric and/or magnetic responses. The magnetic response in a collection of metallic particles arises when an incoming magnetic field excites plasmonic resonances such that they create an additional magnetic field, typically owing to the existence of closed loops of electric and/or displacement currents, called virtual current loops (VCLs).<sup>6</sup> The magnetic response of such an ensemble is interesting in itself, since it is able to mimic, at optical frequencies, the magnetic response (characterized by a relative magnetic permeability  $\mu_r \neq 1$ ) that exists in natural materials only at microwave and lower frequencies. A magnetic response is a necessary condition for many applications in the field of metamaterials, such as the

famed invisibility cloak<sup>7</sup> and the perfect lens.<sup>8</sup> It could also be useful as an optical security signature.<sup>9</sup>

In this paper, we numerically calculate and experimentally observe, through far field measurements, the plasmonic resonances existing in an array of u-shaped resonators. This work follows previous theoretical studies about the use of this kind of resonator as a metamaterial unit cell for negative electric and magnetic responses.<sup>10</sup>

### U-SHAPED RESONATOR RESONANCES

Arrays of u-shaped metallic resonators, and similarly arrays of split-ring resonators (SRRs), are known to exhibit multiple plasmonic resonances.<sup>11–16</sup> The first resonance appearing at lower frequencies is the well known LC-resonance [Fig. 1(a)], in which a virtual current loop is formed along the whole structure, with electrons moving collectively throughout the full length of the metallic wire forming the particle, and with a strong electric field closing the loop between the two tips of the U shape. The VCL created by such a resonance is known to produce a magnetic field perpendicular to the plane of the resonator, so it can be used as unit cell in magnetic metamaterials.<sup>6</sup> The second resonance appearing is characterized by electrons moving back and forth along both legs of the particle in the same direction [Fig. 1(b)]. Such a resonance creates strong opposite charges at both ends of the particle, generating an electric dipole, which can be associated with an electric response. This resonance is sometimes called the electrical resonance. Further high-order resonances are observed, the first of which [Fig. 1(c)] shows an electric quadrupole and two VCL's (one of them dominating over the other) which also implies a magnetic response. As we proposed in a previous theoretical paper, this magnetic response can be combined with the electrical resonance to yield an effective negative index metamaterial.<sup>10</sup> In this paper, we prefer the interpretation of all these resonances (LC, electric, and higher order resonances) as the plasmonic standing wave resonances of a nanowire.<sup>10,11,14</sup>

It is known that metallic straight nanowires support bound propagating plasmon waves along their length.<sup>17</sup> One

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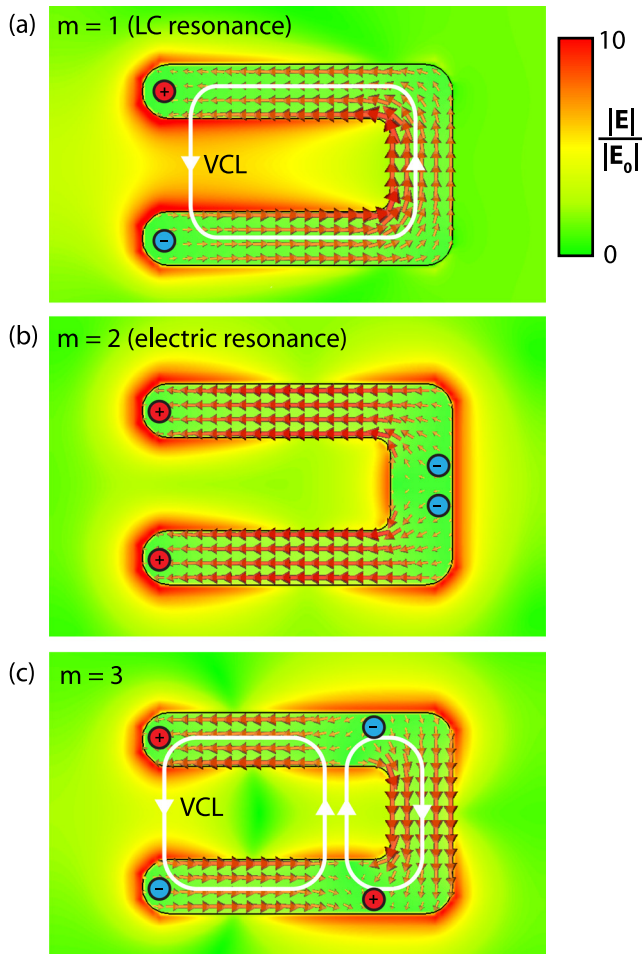


FIG. 1. Numerical simulation of the first three localized surface plasmon resonances in a unit cell of the u-shaped particle array. Both the electric current density (red arrows) and the amplitude of electric field (normalized to the amplitude of the electric field in the same plane in the absence of the metallic particle) are shown. An intuitive view of the virtual current loops (white loops) and the charge accumulations (+ and - charge symbols) has been included.

of these modes is the so-called slow, or short-range, surface plasmon, which is highly confined inside the metal. This mode is especially interesting in metallic nanowires because it is highly reflected at the ends of the nanowire (unlike other modes such as the long-range surface plasmon) and thus can form standing wave plasmon resonances along the length of the metallic nanowires.<sup>18,19</sup> A finite metallic nanowire of certain length then supports standing wave resonances with an integer number  $m$  of half-wavelengths of the short-range surface plasmon. Therefore, this resonance shows  $m$  sections of electric current in alternating directions along the length. One can intuitively picture the u-shaped resonator as a simple bending of a straight nanowire, so that the LC resonance corresponds to the  $m=1$  standing wave resonance and the electric resonance corresponds to the  $m=2$  standing wave resonance. Thus, the dispersion relation of slow plasmons in nanowires can be used to predict the position of the resonances in u-shaped nanowires,<sup>10</sup> although the predictions are not exact due to various effects.

The resonances predicted with this model exist both in SRRs and in U-shaped resonators (which can be seen as a

simplified version of the SRRs in which the gap is made wider) showing analogous field distributions. However, the specific resonant frequencies are slightly different for a SRR and a U-shaped nanowire with the same metallic wire length. A strong and highly confined field exists in the gap of the SRRs, so the geometry of the gap has strong effects in the spectral position of the first resonance. The LC model, taking into account the appropriate capacitance of the gap, is therefore required to determine its frequency,<sup>20</sup> and careful refinements of the LC model can result in very accurate predictions for SRR resonant frequencies even in the optical region.<sup>21</sup> On the contrary, the field which closes the VCL in the first resonance of a u-shaped resonator is more evenly distributed across the length of the arms [Fig. 1(a)], and the resonant frequency does not depend strongly on small variations of the distance between them<sup>10</sup>—at least for the sizes under study here—so modeling the resonances as slow plasmon standing waves along a straight nanowire of the same length is a convenient way to approximately predict and understand the frequencies of all the resonances  $m=1, 2, 3, \dots$  using one same model, albeit lacking the high accuracy of specific LC models. In the case in which the gap is narrow, as happens on a SRR, the model becomes inaccurate to determine the spectral position of the resonances, but even in that case it is useful as a possible interpretation for the different resonance field profiles.<sup>14</sup>

## METHODS AND MATERIALS

Fabrication of silver u-shaped nanowires was performed on a silicon substrate by standard electron-beam-lithography. First, the clean silicon wafer was spin-coated with a 2 nm chromium layer to provide adhesion for the silver film. A layer of electron-sensitive resist was deposited upon the chromium layer. The sample pattern was then transferred to the electron resist via exposure to a focused electron beam. Subsequent chemical development removed the resist from all the regions exposed to the electron beam. A 20 nm silver metal film was then deposited on the sample by a sputtering process. Following deposition of the silver film, a chemical lift-off process removed the resist layer from the chromium surface. This left the periodic metallic u-shaped nanowires deposited directly on the chromium substrate through the openings in the resist mask. A schematic diagram of four unit cells of the resulting sample is depicted in Fig. 2. A scanning electron micrograph of the sample is shown in Fig. 3(a). The metallic u-shaped nanowires have the geometrical parameters indicated in Fig. 3(b). The infrared (IR) transmission spectra were obtained using a Bruker<sup>TM</sup> Fourier Transform IR spectrometer. The IR illumination was focused on the sample with a 0.4 numerical aperture 15 $\times$  objective, using square apertures in the microscope beam path to adjust the illumination region to the whole sample area. Our measurements are mostly at frequencies below Wood's anomaly, so all the far field transmission below 53 THz is associated directly to the zeroth order grating lobe exiting in a direction normal to the plane of the array into the substrate. The illumination was polarized using the polarizations indicated in Fig. 3(b).



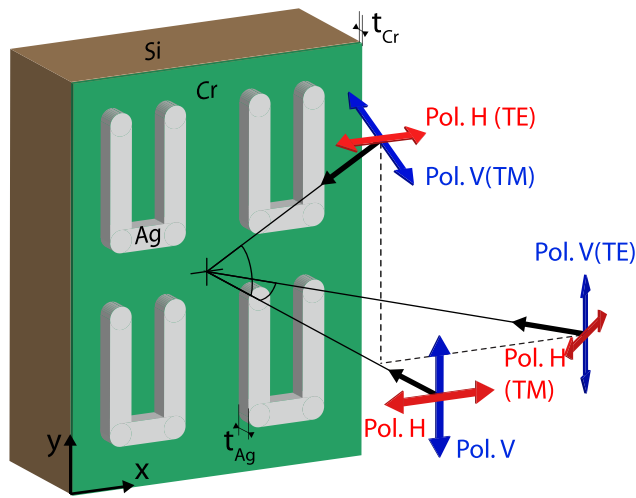


FIG. 2. Depiction of four unit cells of the sample showing the plane wave incidence for the normal and oblique cases.

The numerical simulations were performed using the commercial software CST MICROWAVE STUDIO. The transmission spectra through an infinite array of u-shaped nanowires were calculated by imposing periodic conditions at the boundaries of the unit cell. Both time domain and frequency domain simulations gave consistent results. The silicon substrate was modeled as a dielectric with refractive index  $n = 3.45$ , and the silver u-shaped nanowire was modeled using a Drude model  $\epsilon(\omega) = 1 - \omega_p^2 / (\omega^2 + i\omega\omega_c)$ , with plasma frequency  $\omega_p = 1.37 \times 10^{16}$  rad/s and collision frequency  $\omega_c = 8.5 \times 10^{13}$  rad/s.

## EXPERIMENTAL RESULTS

Figure 3(c) shows the measured transmission spectrum through the fabricated sample, normalized to the transmission through the substrate, together with the numerically simulated normalized transmission. Three distinct resonances can be clearly observed in the measurements and in fairly good agreement with the numerical calculations, corresponding to the first three standing wave plasmonic resonances in the u-shaped nanowire,  $m = 1, 2, 3$ , whose electric current density and electric field strength are plotted in Fig. 1. We focus on these first three resonances. The position of these resonances as a function of geometrical parameters is discussed in detail in theoretical papers for u-shaped nanowires.<sup>10</sup> As can be seen, the periodicity of the u-shaped nanowire array is between 0.1 and 0.4 times the wavelength.

Resonance  $m = 2$  is the electric dipole resonance, and can be excited with the electric field parallel to the u-shaped arms. According to symmetry considerations, this resonance can only be excited by the incident polarization labeled as Pol. V. The resonance is broad in the frequency domain because it couples easily to radiated plane waves, thereby showing high radiative losses and a low quality factor.

Resonances  $m = 1$  and  $m = 3$  can be excited at normal incidence using an electric field oriented along the gap formed between the two ends of the U shape arms, and according to symmetry considerations, both resonances are only excited for the incident polarization labeled as Pol. H.

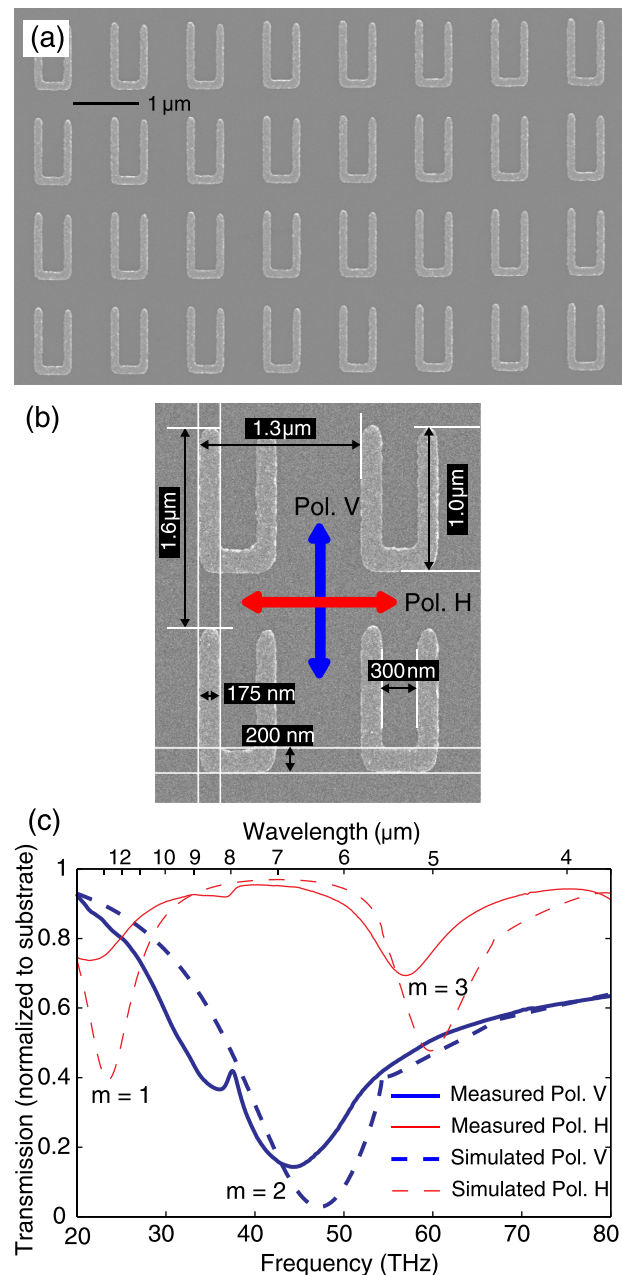


FIG. 3. (a) Scanning electron micrograph of a region of the fabricated sample. (b) Geometric sizes of the fabricated and simulated u-shaped resonators. (c) Optical measured and simulated transmission spectra for both horizontal and vertical electric field polarizations.

These two resonances show VCL's and therefore exhibit a magnetic field in the direction perpendicular to the plane of the u-shape. In this case, we are performing an electrical excitation of the magnetic resonances.<sup>6</sup>

A noticeable kink is observed in the measurements around  $8 \mu\text{m}$  (38 THz). We do not know the origin of this sharp kink, and we did not manage to replicate it in the numerical simulations. Any effect of the surrounding environment and the measurement apparatus is taken care of when normalizing the measurement to the transmission through the substrate alone. A slight asymmetry in the arm length also cannot explain this kink. Alternatively, it could come from some material property of the silver or the chromium adhesion layer not taken into account in the simulation.

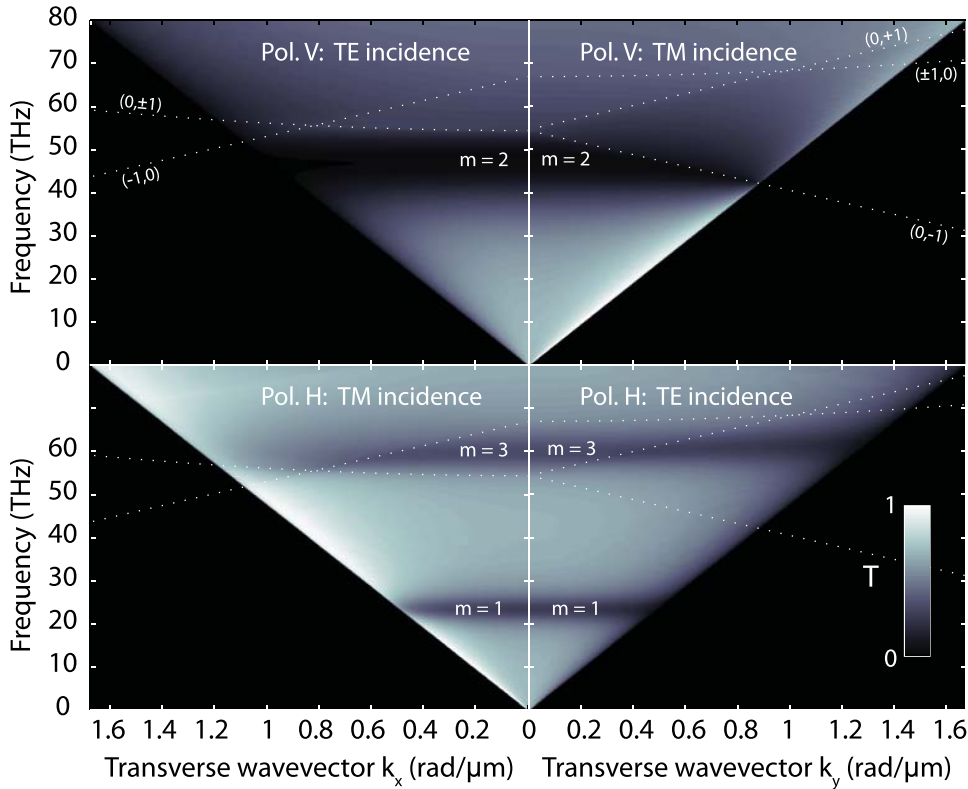


FIG. 4. Simulated power transmission spectra through the sample for oblique incidence (no normalization to the substrate was made). The power transmission for the two different TE and TM angles for each of the two different polarizations is shown. The angle of incidence was converted into the corresponding transverse wavevector. The plotted transmitted power corresponds only to the (0,0) grating order. The frequencies at which higher grating orders arise at the output substrate are shown as white dotted lines.

**NUMERICAL RESULTS**

To further clarify the nature of these resonances, we performed additional numerical simulations. We computed the transmission spectra for oblique angles of incidence, as depicted in Fig. 2. The resulting transmission maps are shown in Fig. 4, where the numerically calculated power transmission through the sample is plotted as a function of frequency and transverse wavevector. The spectral position of the three resonances does not vary with the angle of incidence. This is a further indication that we are dealing with localized surface plasmon resonances in the individual nanoparticles, rather than propagating modes along the plane of the array. It is also an indication that the coupling between the nanoparticles of the array is relatively weak. The only instance of dispersive behavior in our results is seen at those frequencies at which higher order grating modes change from evanescent to propagating. These frequencies are usually associated with sudden changes in the transmission levels (Wood’s anomaly) of the zeroth order mode and they depend on the angle of incidence according to the relation  $2\pi f_{(n_x, n_y)} = (c/n_{subs}) \sqrt{(k_x^{inc} + n_x 2\pi/a_x)^2 + (k_y^{inc} + n_y 2\pi/a_y)^2}$ , where  $(n_x, n_y)$  is the grating order,  $n_{subs}$  is the refractive index of the substrate,  $a_x$  and  $a_y$  are the periodicities in the  $x$  and  $y$  directions, and  $k_x^{inc}$  and  $k_y^{inc}$  are the transverse incident wavevectors. The frequencies for the first higher order modes are plotted as white dotted lines in Fig. 3 and their effect on the transmission spectra can be noticed.

In addition, we now turn our attention to the magnetic excitation of the  $m = 1$  and  $m = 3$  resonances, expected at high incident TE angles for V. polarization. Under those conditions, it is known that the incoming magnetic field has a component normal to the plane of the array and therefore

can excite the LC ( $m = 1$ ) resonance (the phase difference in the electric field arriving at both arms can be another source of coupling to this resonance, but can be neglected for highly subwavelength particles). This magnetic excitation is of special importance in many applications of SRRs and U-shaped nanowires. Figure 5 shows—in blue—the simulated transmission spectra for the TE incidence for V. polarization at two different incidence angles (corresponding to cross sections of Fig. 4). At normal incidence, the V. polarization can excite only the  $m = 2$  resonance, however, under oblique incidence, the  $m = 1$  resonance can be very slightly excited by the incident magnetic field. Indeed, a very smooth shallow dip exists around 25 THz for angles

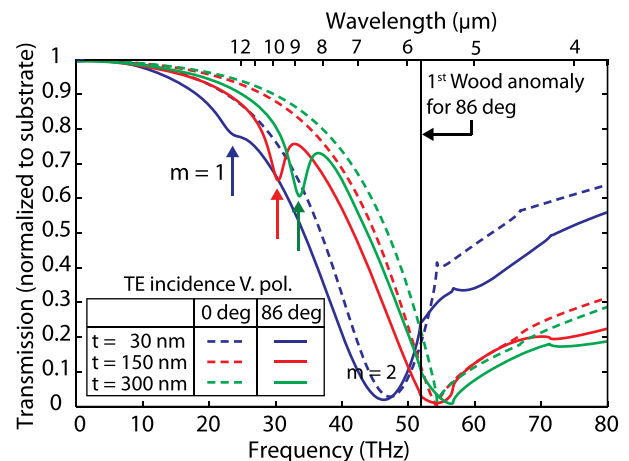


FIG. 5. Normalized transmitted power for V. polarization at normal incidence and at oblique 86° TE incidence, showing the magnetic excitation of the  $m = 1$  resonance. Three different thicknesses of the silver u-shaped nanowire are used: 30 nm (as in the fabricated samples), and higher thicknesses 150 nm and 300 nm, to improve the magnetic excitation.

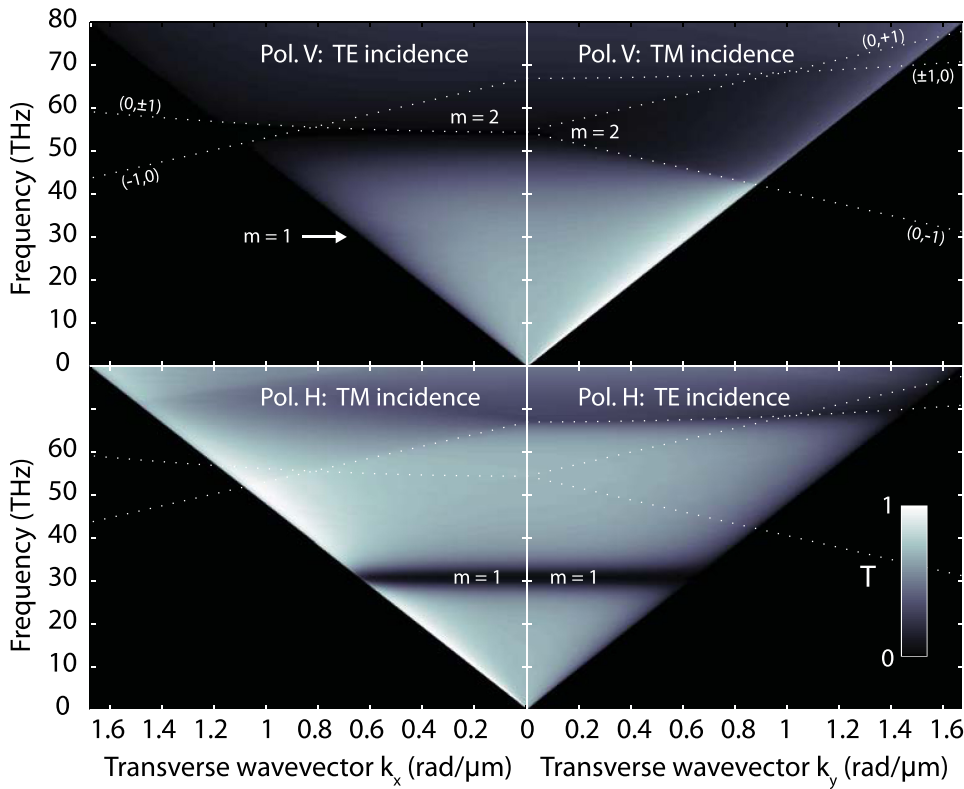


FIG. 6. Simulated power transmission spectra through the sample for oblique incidence, similarly to Fig. 4, for an increased thickness of silver of 150 nm. The  $m = 1$  resonance is stronger, and it can be slightly observed at high oblique angles for Pol. V at TE incidence.

close to  $90^\circ$  corresponding to such excitation, but it is very weak and is barely visible in Fig. 4. This is actually consistent with previous experimental studies, where the magnetic excitation of resonances has been experimentally measured to be very small at oblique incidence in arrays of SRRs.<sup>6</sup> This is in contrast to the theoretical predictions of free standing U-shaped nanowires or SRRs where the resonan-

ces can be strongly excited by the magnetic field,<sup>10</sup> so it is important to understand the reasons behind this reduced excitation.

A first attempt to increase this excitation is to make the  $m = 1$  resonance stronger by increasing the thickness of the silver layer. Fig. 6 shows the transmission maps for a geometry with a silver thickness of 150 nm. All resonances are

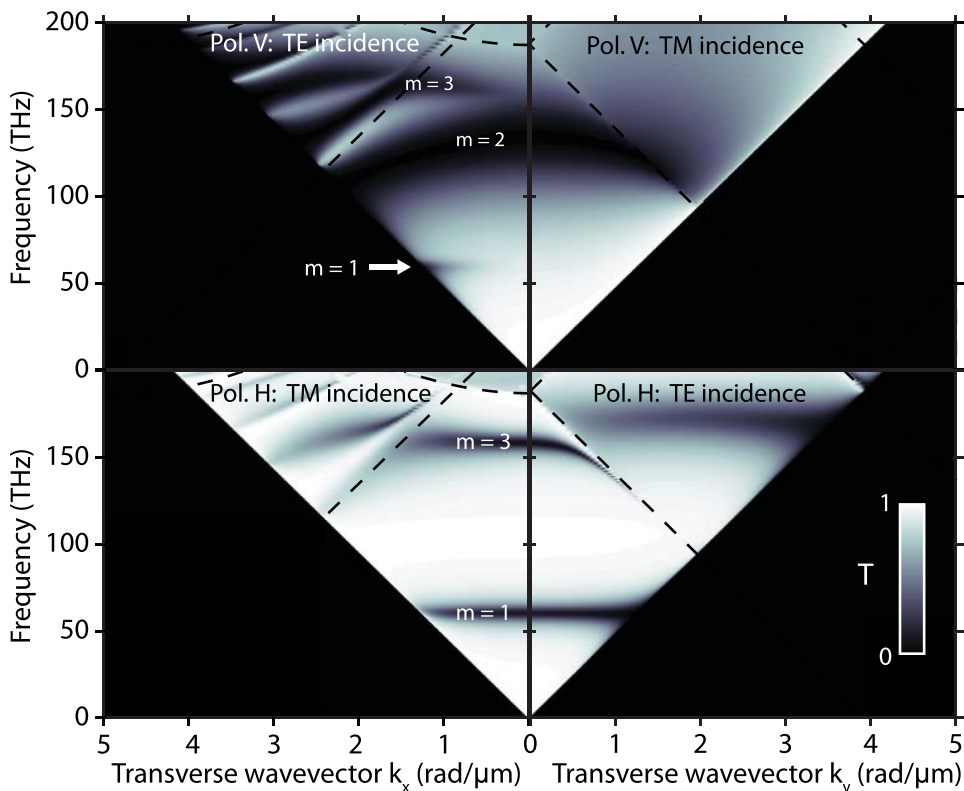


FIG. 7. Simulated power transmission spectra through the sample geometry for oblique incidence, similarly to Fig. 4, but removing the substrate. The  $m = 1$  resonance is clearly excited at high oblique angles for V polarization at TE incidence. The black dashed lines are the frequencies of Wood's anomalies.



slightly blueshifted, and clearly the  $m = 1$  resonance is stronger. The excitation of the  $m = 1$  resonance at TE incidence for V. polarization, however, still cannot be seen clearly in the figure, although it can be seen in the cross section shown in Fig. 5. In addition, Fig. 5 includes results for bigger thickness. We see that by increasing the metal thickness, we can increase the magnetic coupling to the  $m = 1$  resonance at high angles of TE incidence in V. polarization, but we always obtain a weak excitation, even for metal thickness as big as 300 nm, which is impractical.

A second attempt at increasing the magnetic excitation of resonances under oblique incidence is to use a substrate with a lower refractive index. We simulated the fabricated geometry (considering again a thin 30 nm layer of silver) but getting rid of the substrate (free standing particles), and we obtained the transmission map shown in Fig. 7. In this case, there is a blueshift in all resonances due to the big decrease in refractive index (notice the scale in the figure). This time, in the absence of substrate, the magnetic excitation of the modes under oblique TE incidence for V. polarization at high angles is strong: the modes  $m = 1, 3$  (and even higher order odd modes) that are not seen at normal incidence, are clearly excited at oblique incidences. This is true even when using a very thin metal. This suggests that the presence of a high index substrate greatly decreases the efficiency of the magnetic excitation of resonances at oblique angles of incidence, and this should be taken into account when designing practical applications for such structures.

## CONCLUSIONS

We have experimentally observed plasmon resonances on arrays of silver u-shaped nanowires. Our complete numerical modeling agrees well with the optical measurements, and shows that we are dealing with localized surface plasmon resonances associated with the slow-plasmon standing waves in the geometry, the first of which corresponds to the well known LC resonance. Owing to their plasmonic nature, the observed resonances present high field enhancement, with potential applications for spectroscopy and sensing. They also present electric and magnetic responses, with potential application for use as unit cell in metamaterials. The magnetic excitation of resonances using oblique incidence is not strong in our geometry due the high index substrate, and to a lesser extent due to the small thickness of the silver layer. These results should apply also for SRRs in addition to the u-shaped nanowires under study here, since both structures show similar field profiles and electric currents in their LC magnetic resonance.

## ACKNOWLEDGMENTS

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