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Ultra-thin condensers for optical subwavelength resolution microscopy

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We present optical subwavelength resolution images of periodic patterned nanostructures using ultra-thin condensers (UTCs) illuminated by evanescent waves. We demonstrate bright and dark field microscopy using UTCs based on two types of surface wave illumination: surface plasmon polaritons and evanescent waves related to total internal reflection. We provide a discussion about the potential of UTCs for deep subwavelength resolution microscopy, and we discuss the similarities and differences between proposed UTCs, traditional bulky optical condensers, and several demonstrated superlenses. © *2014 AIP Publishing LLC*.
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I. INTRODUCTION

Since the discovery of the optical condenser by Abbe in 1873, it is well known that illuminating a periodic structure with very inclined angles permits to reduce the minimum period (p_{min}) observable using an optical microscope from $p_{min} \sim \lambda_o/NA$ to $p_{min} \sim \lambda_o/(2\,NA)$,[1–3] where λ_o is the wavelength of the illumination source in vacuum and NA is the numerical aperture of the microscope objective lens. The limiting value, $\lambda_o/(2NA)$, is the well-known Rayleigh resolution limit of optical microscopes.[4] Typical condensers used in optical microscopes consist of a combination of bulky lenses (or mirrors) and diaphragms designed to illuminate the sample with a cone of light. Optical condensers are then the oldest and simplest known approach to obtain subwavelength imaging resolution. Nevertheless, the subwavelength resolution achievable using optical condensers,[5] $\lambda_o/(2NA) < p_{min} < \lambda_o/NA$, is comparable to reported subwavelength resolution values of several demonstrated superlenses,[6–13] which then are not ideal superlenses in the original sense of Pendry's proposal of achieving perfect image reconstruction.[14] For instance, in an experimental confirmation of the original Pendry's practical implementation of a near-field superlens by using a planar layer of silver illuminated near its plasma frequency,[14] super-resolution was demonstrated by imaging gratings immersed in a medium with refractive index $n \sim 1.5$ with periods down to 145 nm at a wavelength of 365 nm.[7] This corresponds to $p_{min} = 145\,\text{nm} > \lambda_o/(2n) = 121\,\text{nm}$, where we have considered that at the near-field the maximum angle of light collection is $\theta \sim 90°$, so that $NA = n\sin\theta \sim n$. Similarly, a far-field optical hyperlens was demonstrated by imaging a 150 nm spaced line-pair object illuminated with $\lambda = 364$ nm and using an objective lens with $NA = 1.4$.[8] This corresponds to $p_{min} = 150\,\text{nm} > \lambda_o/(2NA) = 130\,\text{nm}$. Optical condensers are routinely used in biomedical and material sciences research laboratories which employ optical microscopes as a basic imaging and characterization tool.[15] However, modern optical condensers continue to be as bulky as the original one designed by Abbe.

In this work we describe ultra-thin condensers (UTCs), which rely on the illumination of the object under observation with evanescent waves. We studied two types of UTCs. In plasmonic UTCs, surface plasmon polaritons (SPPs)[16] excited by fluorescence emitted near the metal-dielectric interface between two thin films[10–13,17–20] were the surface waves used as the illumination source. Surface waves related to total internal reflection[12,13] were used as the illumination source in the second type of UTCs. In both cases the light used for imaging is the light that couples to the evanescent waves and leaks to the microscope objective lens.[10–13,17–21] This is in contrast to image formation in microscopes using traditional bulky condensers, where the images are formed by light that traverses the sample and is diffracted without excitation of evanescent waves. In addition to the experiments demonstrating bright and dark field microscopy with optical subwavelength resolution implemented using both types of UTCs, we discuss the implications of the demonstration of dark field microscopy with optical subwavelength resolution using UTCs. Dark field microscopy using plasmonic condensers in a microscope with an objective lens with $NA < 1$ have been demonstrated previously.[22,23] In this work we present the first demonstration of dark field microscopy with subwavelength resolution using plasmonic condensers in a microscope with $NA > 1$. We point out that plasmonic UTCs may produce resolution values well below the Rayleigh resolution limit of optical microscopes, which may suggest there is a general and fundamental relationship between UTCs and the slab of silver superlens proposed by Pendry.[14]

This paper is organized as follows: In Secs. II and III we describe experiments with plasmonic and non-plasmonic UTCs, respectively. Section IV is dedicated to the discussion

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of the experimental results and their implications for subwavelength resolution microscopy. Finally, the conclusions of this work are given in Sec. V.

II. PLASMONIC ULTRA-THIN CONDENSERS

Figure 1(a) shows a schematic of the transversal cross-section of plasmonic samples studied in this work. The plasmonic UTC comprises a ~150 μm glass substrate covered with a 50 nm thick gold layer and coated with a ~110 nm thick layer of poly methyl methacrylate doped with Rhodamine-6G (PMMA-R6G). The object under observation is a periodic array consisting of cylindrical Cr pillars 20 nm tall with varying periodicity p, and diameters $d = p/2$ arranged in a lattice with square symmetry. Using a combination of electron-beam lithography and lift-off techniques, the arrays of Cr pillars were fabricated on top of the gold layer before covering it with the PMMA-R6G layer. The sample was illuminated from the top (patterned side) with a continuous-wave laser emitting at 532 nm wavelength. The fluorescent emission centered at ~568 nm wavelength excites SPPs in the sample, and their propagation directions are modified, from that in the un-patterned layer, by the periodic structure.[24,25] Light coupled to the excited SPPs leaks, forming a hollow cone[17–19] similar to the hollow cone of light produced by traditional bulky optical condensers producing circular illumination.[5] Leaked light is collected by an immersion oil objective lens with $NA = 1.3$, band-pass filtered at $\lambda_o = 568$ nm wavelength, and then imaged at both real and Fourier planes using a commercial inverted microscope.[10,11,19] It is worth noting that when a condenser is used, bright field microscopy results if the non-diffracted hollow cone of light produced by the condenser is captured by the microscope objective lens; however, dark field microscopy results if the non-diffracted hollow cone of light cannot be captured by the microscope objective lens because the light has a large inclination. A schematic illustration of the experimental setup is depicted in Fig. 2. Real plane (RP) and Fourier plane (FP) images corresponding to a Cr array with a period of 400 nm are shown in Fig. 3. The RP and FP images shown in Figs. 3(a) and 3(b), respectively, were

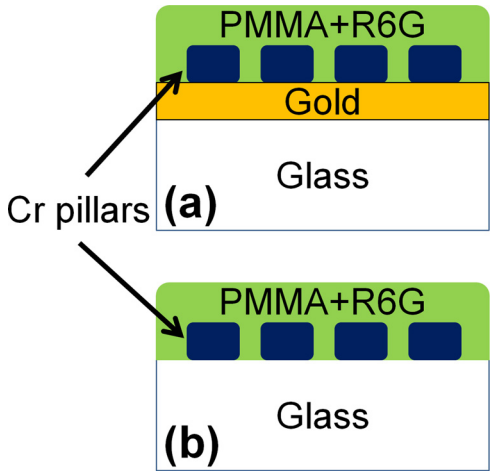

FIG. 1. Schematic of the cross-section of fabricated (a) plasmonic UTCs and (b) non-plasmonic UTCs, with an array of Cr pillars.

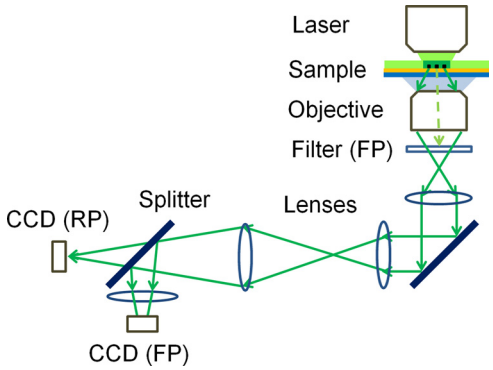

FIG. 2. Schematic illustration of the experimental set up. The sample is the ultra-thin condenser with an array of Cr pillars.

obtained by substituting the laser used to produce fluorescence in the sample (shown in Fig. 2) by the built-in white--light illumination source of the inverted microscope. In this configuration, no SPPs were excited in the sample, which implies that a plasmonic UTC does not contribute to image formation in this case. However, the RP and FP images shown in Figs. 3(c) and 3(d), respectively, were obtained using the experimental set-up sketched in Fig. 2. The appearance of bright rings in the FP image shown in Fig. 3(d) indicates that SPPs were excited in the sample,[10,11,17–20,24,25] which implies that a plasmonic UTC contributed to the formation of the images shown in Figs. 3(c) and 3(d). Also, the appearance of the centered bright thick ring pointed by the arrow in the FP image shown in Fig. 3(d) indicates that the RP and FP images shown in Figs. 3(c) and 3(d) were obtained using a bright field microscope arrangement. The *NA* of the plasmonic UTC can be directly found from the radius of the rings (in refractive index units) formed in the FP image, which can directly be determined from the FP image.[26] Dark field microscopy requires that the *NA* of the condenser (NA_c) should be larger than the *NA* of the microscope objective lens (NA_o). In order to increase the *NA* of the plasmonic UTC, we added a drop of water on top of the sample. Corresponding RP and FP images are shown in Figs. 4(a) and 4(b), respectively. As expected, the centered bright thick ring, pointed by the arrow in the FP image shown in

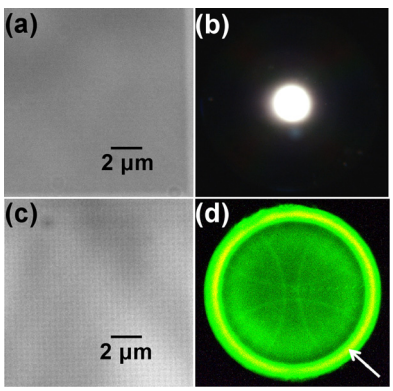

FIG. 3. RP (a), (c) and FP images (b), (d) of a Cr array sample with 400 nm period obtained with the microscope's built-in white-light illumination source (a), (b), and a plasmonic UTC (c), (d). The arrow in (d) points to the centered bright thick ring appearing within the *NA* of objective lens, suggesting subwavelength resolution in bright field microscope arrangement.

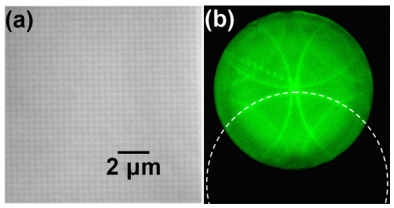

FIG. 4. (a) RP and (b) FP images obtained with a plasmonic UTC of a Cr array with period of 400 nm with a drop of water over the UTC. One of the shifted rings is highlighted in (b), the radius of the shifted rings is larger than the *NA* of objective lens, suggesting subwavelength resolution in dark field microscope arrangement.

Fig. 3(d), is not observed in Fig. 4(b) because with the drop of water $NA_c > NA_o$; therefore, the RP and FP images shown in Figs. 4(a) and 4(b) were obtained using a dark field microscope arrangement. Nevertheless, shifted bright rings can be observed in the FP image shown in Fig. 4(b), where a dashed-arc was overlaid on one of the shifted rings to help visualizing the size of the shifted rings in comparison with the numerical aperture of the objective lens. The presence of these rings in the FP image shown in Fig. 4(b) indicates that SPPs were excited in the sample, which means that a plasmonic UTC contributed to the formation of the images shown in Fig. 4. The array of Cr pillars is not visible (since it cannot be resolved) in the RP obtained using microscope's built-in white-light illumination source (Fig. 3(a)); therefore, a comparison of the RP images shown in Figs. 3(a), 3(c), and 4(a) demonstrates the subwavelength resolution provided by a plasmonic UTC. In addition, a comparison of the RP images shown in Figs. 3(c) and 4(a) demonstrates that it is not necessary that the centered bright ring formed in the FP of the microscope has to be collected by the objective lens for the formation of a correct RP image in the microscope; in other words, it demonstrates that dark field microscopy with plasmonic UTCs produces RP images with subwavelength resolution.

III. NON-PLASMONIC ULTRA-THIN CONDENSERS

Figure 1(b) shows a schematic of the cross-section of fabricated non-plasmonic samples. We fabricated periodic structures consisting of cylindrical Cr pillars 15 nm tall with varying periodicity p, and diameters $d = p/2$ arranged in a lattice with square symmetry, over a $\sim$150 μm thick glass cover slip. The glass cover slip with the Cr array, which is the object to be imaged, was then covered with $\sim$110 nm thick layer of PMMA-R6G. All the images shown in Fig. 5, corresponding to an array of Cr pillars with square symmetry with 300 nm period, were obtained using the experimental set up sketched in Fig. 2. The observation of thick bright rings in the FP image shown in Figs. 5(b) and 5(d) indicates that evanescent waves related to total internal reflection were excited in the sample,[12,13] which means a non-plasmonic UTC contributed in the formation of the images shown in Fig. 5. The appearance of the centered bright thick ring in the FP image shown in Fig. 5(b) indicates that the RP and FP images shown in Figs. 5(a) and 5(b), respectively, were obtained using a bright field microscope arrangement. The RP and FP images shown in Figs. 5(c) and 5(d), respectively, were obtained with a drop of water placed on top of the

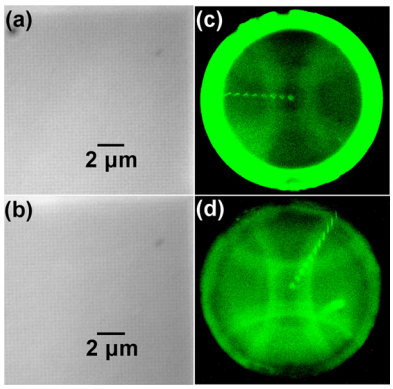

FIG. 5. RP (a), (c) and FP images (b), (d) obtained with a non-plasmonic UTC comprising a Cr array with 300 nm period without (a), (b) and with (c), (d) a drop of water on the top of the UTC.

sample. No centered bright thick ring is observed in Fig. 5(d) because with the drop of water $NA_c > NA_o$; therefore, the RP and FP images shown in Figs. 5(c) and 5(d) were obtained using a dark field microscope arrangement. A comparison of the RP images shown in Figs. 3(a), 5(a), and 5(c) demonstrates subwavelength resolution provided by a non-plasmonic UTC. In addition, a comparison of the RP images shown in Figs. 5(a) and 5(c) demonstrates that that it is not necessary that the centered bright thick ring formed in the FP of the microscope has to be collected by the objective lens for the formation of a correct RP image in the microscope; in other words, it demonstrates that dark field microscopy with non-plasmonic UTCs produces RP images with subwavelength resolution.

IV. DISCUSSION OF THE EXPERIMENTAL RESULTS

The minimum observable period (p_{min}) in a microscope using a traditional bulky condenser is given by the following equation:[1]

$$p_{\min} = \frac{\lambda_o}{NA_o + NA_c}. \quad (1)$$

When $NA_o = NA_c$, Eq. (1) gives the Rayleigh resolution limit. From the FP image shown in Fig. 3(b) follows that $NA_c \sim 0$ when the built-in white-light illumination source was used; therefore, using Eq. (1) we estimated a value of $p_{min} \sim 434$ nm. This explains why the RP image shown in Fig. 3(a) did not reveal the periodic structure with a period of 400 nm present in the sample. Equation (1) also is valid for UTCs. For plasmonic UTCs, $NA_c = n_{eff}$, where n_{eff} is the effective refractive index experienced by the excited SPPs.[10,11,13] A FP image can be considered a map of the effective refractive indexes corresponding to the SPPs or, in general, surface waves excited in the sample.[26] From the FP image shown in Fig. 3(d), the extracted values of n_{eff} are $\sim$1.12 and $\sim$1.31 corresponding to the centered bright thick ring pointed by the arrow, and the four narrow, less intense, shifted rings, respectively.[19,26] Therefore, using Eq. (1) with $n_{eff} \sim 1.31$, we estimate a value of $p_{min} \sim 217$ nm, which is in correspondence with the observation of a periodic structure with a 400 nm period in the RP image shown in Fig. 3(c).

For periodic and non-periodic samples, deeper subwavelength resolution using plasmonic UTCs in a bright field microscope arrangement has already been demonstrated.[10,11] In Refs. 10 and 11, the plasmonic UTCs were called superlenses. We believe that the term *condensers* provides a better description of the physical principles involved in the observed subwavelength resolution. Traditional bulky optical condensers contain lenses, whose function is to illuminate the sample with inclined light. UTCs illuminate the object under observation with surface waves, i.e., extremely inclined light. In addition, as for traditional bulky condensers,[5] the subwavelength resolution provided by UTC is determined by the size of the bright rings observed in the FP images. The observed bright rings are due to the intersection of hollow cones of light leaking from the sample with the microscope's FP.[17–19] If a fraction of the shifted rings can be captured by the microscope objective lens, then the periodic structure of the object will be visible in the RP image.[10,13] The value of n_{eff}, and hence NA_c increased when a drop of water was placed on top of the sample. This is in good correspondence with the values of $n_{eff} \sim 1.38$ obtained from the bright shifted rings observed in Fig. 4(b). The drop of water increases the effective refractive index of the SPP related to the centered bright thick ring pointed by the arrow in the FP image shown in Fig. 3(d). As a result, the centered bright thick ring is not present ($n_{eff} > NA_o$) in the FP image shown in Fig. 4(b), as it was not captured by the collecting objective lens. Consequently, the observation of a periodic structure in the RP image shown in Fig. 4(a) constitutes the first demonstration of subwavelength resolution in a dark field microscope arrangement using plasmonic UTCs or superlenses.

For non-plasmonic UTCs based on the excitation of surface waves related to total internal reflection $NA_o = n_{sub}$, $NA_c = n_{sup}$, where n_{sub} and n_{sup} are the refractive indexes of the medium below and above the sample, respectively.[12,13] Therefore, using $n_{sub} \sim 1.5$ in Eq. (1) with, $n_{sup} \sim 1$ (air) and ~ 1.33 (water), we estimate $p_{min} \sim 227$ nm and ~ 201 nm, respectively, which are in correspondence with the observation of a periodic structure with $p = 300$ nm in the RP images shown in Figs. 5(a) and 5(c). Again, deeper subwavelength resolution using non-plasmonic UTCs in a bright field microscope arrangement has already been demonstrated.[12,13] However, the RP and FP images shown in Figs. 5(c) and 5(d) constitute the first demonstration of subwavelength resolution in a dark field arrangement using non-plasmonic UTCs. In dark field microscopy, $NA_c > NA_o$; therefore, from Eq. (1) $p_{min} < \lambda_o/(2NA_o)$, i.e., the microscope resolution is better than the Rayleigh resolution limit. Subwavelength resolution in a dark field microscope arrangement using UTCs is particularly interesting because it offers the possibility for obtaining extremely deep subwavelength resolution. Using non-plasmonic UTCs the resolution may be further increased by immersing the object under observation in a medium with $n_{sup} = NA_c \gg n_{sub} = NA_o$. In this case, as in the FP image shown in Fig. 5(d)), a centered bright thick ring, similar to the one observed in the FP images shown in Fig. 5(b), would not be observed in the FP image; however, as long as the shifted rings are collected by the objective lens as they were in the FP image shown in Fig. 5(d)), a correct image of the object would be formed directly in the RP of the microscope without the need of scanning, numerical calculation, or post-processing. This approach to deep subwavelength resolution is practically limited by the availability of transparent materials with high refractive index. It should be noticed that the Abbe's resolution limit given by Eq. (1) corresponds to the minimum period discernible with a condenser-microscope arrangement. This corresponds to the capture by the microscope objective lens of at least a fraction of the first order diffraction rings.[10,13] However, for practical microscopy applications, high quality contrast images is of great importance. This can be only accomplished if the second order diffraction features are also collected by the microscope objective lens.[2] Second order diffraction rings can be observed in the FP image shown in Fig. 4(b) but they are not visible in Figs. 5(a) and 5(b). This results in a better definition of the periodic structure present in the RP image shown in Fig. 4(b) when compared to the low quality RP images shown in Figs. 5(c) and 5(d). Nevertheless, these images are sufficient to verify the use of Eq. (1) to UTCs.

Equation (1) with $NA_c \gg NA_o$ shows that using plasmonic or non-plasmonic UTCs, it may be possible to obtain resolution values significantly smaller than the Rayleigh resolution limit. Therefore plasmonic UTCs can be used to obtain images with resolution well below diffraction limit of light. Due to the non-linear dispersion of SPPs,[16] it is in principle possible to conceive a plasmonic UTC where dark SPPs are excited with an extremely large value of n_{eff}, i.e., $n_{eff} \gg n_{sub}$, $n_{eff} \gg n_{sup}$. Such dark SPPs would not be able to leak to the objective lens of the microscope; therefore, a centered bright ring would not be observed in the FP images obtained using such an extreme plasmonic UTC, which means that the UTC-microscope combination would be in this case a dark-field microscope arrangement. However, using plasmonic UTCs, the light diffracted by the presence of an object formed by spatial frequencies much larger than $2NA_o/\lambda_o$ may be captured by the microscope's objective lens, resulting in bright shifted rings similar to the ones observed in the FP image shown in Fig. 4(b). This corresponds to resolution much smaller than the Rayleigh limit, suggesting that plasmonic UTCs are somehow similar to the slab of silver superlens originally proposed by Pendry,[14] which is not an ideal superlens permitting the perfect reconstruction of the image.[14]

V. CONCLUSIONS

In summary, we experimentally demonstrated that UTCs, which illuminate the object under observation with evanescent waves, produce optical subwavelength resolution. We demonstrated bright and dark field microscopy using UTCs based on two types of surface waves: surface plasmon polaritons and evanescent waves related to total internal reflection. We presented a theoretical discussion about the potential of UTCs, used in a dark-field microscope arrangement, for deep subwavelength resolution microscopy, and we discussed the similarities and differences between presented UTCs, traditional bulky optical condensers, and some previously demonstrated superlenses.

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