Babinet to the Half: Coupling of Solid and Inverse Plasmonic Structures

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ABSTRACT: We study the coupling between the plasmonic resonances of solid and inverse metallic nanostructures. While the coupling between solid-solid and inverse-inverse plasmonic structures is well-understood, mixed solid-inverse systems have not yet been studied in detail. In particular, it remains unclear whether or not an efficient coupling is even possible and which prerequisites have to be met. We find that an efficient coupling between inverse and solid resonances is indeed possible, identify the necessary geometrical prerequisites, and demonstrate a novel solid-inverse plasmonic electromagnetically induced transparency (EIT) structure as well as a mixed chiral system. We furthermore show that for the coupling of asymmetric rod-shaped inverse and solid structures symmetry breaking is crucial. In contrast, highly symmetric structures such as nanodisks and nanoholes are straightforward to couple. Our results constitute a significant extension of the plasmonic coupling toolkit, and we thus envision the emergence of a large number of intriguing novel plasmonic coupling phenomena in mixed solid-inverse structures.

KEYWORDS: Plasmons, Babinet's principle, electromagnetically induced transparency, inverse nanostructures, chirality

In recent years complex and composite plasmonic systems of ever-increasing complexity and tunability have been demonstrated.1–5 All of these systems exhibit collective optical properties due to efficient coupling between the constituting metallic nanoparticles via the strongly enhanced local electric fields that are associated with their plasmonic resonances. As the overall design freedom in the creation of composite plasmonic systems is ultimately limited by the number of potential building blocks as well as by their optical and near-field properties, new building blocks are highly desirable. Apart from solid metallic nanostructures which are not only available from top-down nanostructuring but also from wet-chemical synthesis, so-called inverse structures have been studied. Here, the resonance is associated with a nanoaperture in a continuous metallic film. The overall behavior is governed by Babinet’s principle,6,7 which, broadly speaking, warrants the presence of a reflectance dip rather than a transmission dip in the optical spectrum of the structure. Apart from studying the nature of the exited modes,8–11 a number of interesting applications has been demonstrated, such as the manipulation of the polarization state of light,12,13 chiral structures,14–16 transmission lines,17 negative refraction,18 second harmonic generation from inverse split ring resonators,19 a polarization analyzer for the magnetic vector of light,20 a probe for the magnetic field of light in near-field measurements,21 as well as the concept of extraordinary optical transmission.22

Consequently, the coupling between the resonances in purely solid structures as well as the coupling between resonances in purely inverse systems is very well understood. It has been shown that the interaction in solid-solid systems can be intuitively understood in terms of the coupling of electric dipoles. The coupling in inverse-inverse systems on the other hand can be interpreted in terms of magnetic dipoles.5,7,23–25 Therefore, inverse structures are particularly interesting as they offer magnetic moments, modes, and resonances which are difficult to obtain from solid structures. Note, however, that this picture is simply a paraphrasing of electric field distributions. The coupling in a plasmonic system can be visualized either in terms of electric or magnetic near fields. These two pictures are fully equivalent. However, depending on the situation at hand, the coupling physics is often easier to understand by thinking about either the magnetic or the electric near fields. In particular, this does not imply that coupling only takes place via electric or magnetic fields; rather, it is simply a different way of looking at the same physics. For inverse structures it is convenient to look at magnetic rather than electric near fields, which offers a very intuitive picture in excellent analogy to the solid-solid case.

In contrast, the interaction in mixed solid-inverse composite systems is nearly unexplored despite the fact that this coupling scheme significantly expands the toolkit for the construction of complex plasmonic systems. By incorporating the solid and corresponding inverse building blocks into the toolkit, one greatly expands the degrees of freedom in structure design.

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However, a number of intriguing questions remain to be addressed: Is it possible to efficiently couple the modes in solid and inverse plasmonic structures? Which prerequisites have to be fulfilled to facilitate the interaction of the electric and magnetic modes? A limited number of pioneering studies has been devoted to this topic,

\[ \text{15,16,26−31} \] However, most of the proposed designs do not show an efficient coupling between the excited modes, mostly due to limitations in fabrication techniques. One potential technique involves the evaporation of gold through a resist mask which leads to identically shaped the excited modes, mostly due to limitations in fabrication techniques. One potential technique involves the evaporation of gold through a resist mask which leads to identically shaped and oriented solid and inverse structures.

\[ \text{13,15,32} \] Despite being a very elegant technique, it offers no design freedom for controlled and changeable coupling between the structures, which will prove to be of great importance.

In this Letter we demonstrate that efficient coupling in mixed solid-inverse systems is possible. We show that the coupling behavior and the resulting modes can no longer be interpreted using the intuitive picture of pure electric or magnetic dipoles. To unravel the coupling and mode formation, electric near-fields have to be considered. For anisotropic structures, such as nanorods, we find novel and intriguing coupling scenarios, such as an unusual plasmonic electromagnetically induced transparency (EIT) structure. In-plane isotropic disks and holes on the other hand do readily couple and thus allow for composite inverse-solid systems, a finding which is experimentally verified in a mixed solid-inverse chiral structure.

Figure 1a depicts a metallic nanorod. When illuminating the rod with light polarized along its long axis, a plasmonic resonance can be excited. The signature of this resonance will immediately infer the nature of the mode and the optical response.\[ \text{6,7} \] According to this principle, and given not too high dissipative losses, in the inverse system several quantities exchange their respective roles: transmission and reflection, electric and magnetic field, as well as the two polarization directions. Therefore, the rod-shaped aperture will exhibit a pronounced dip in the reflection spectrum for incident light polarized perpendicular to the long axis of the aperture. Moreover, the near-field distribution will exhibit a dipolar pattern in the \( z \)-component of the magnetic near-field distribution. The resonance is therefore associated with a magnetic dipole lying within the sample plane.

It is important to note, however, that the picture of a magnetic dipole is only a reformulation of electric fields which ultimately mediate the coupling in the system. When examining the electric near-field distribution of such a rod-shaped aperture, we find strongly enhanced electric fields in the equatorial plane around the aperture. This behavior is intuitively straightforward to understand: The magnetic moment is created by a ring current flowing around it. Even though this picture is a very simple one, it offers intuitive comprehension.

If one now intends to couple the plasmonic modes of a solid rod and rod-shaped aperture, one has to spatially overlap the electric near-fields of both structures. Coupling will be most efficient for best overlap of the corresponding plasmonic mode profiles. As the electric near-field is largest at the end of the rod and nearly zero at the end of the rod-shaped aperture, a side-by-side configuration will not lead to any interaction due to vanishing mode overlap. Breaking the systems symmetry by displacing the two structures and thus overlapping the electric near-fields will, however, lead to plasmonic interaction.

Figure 1c and d depicts the same case for a solid metallic disk and a hole-shaped aperture, respectively. In principle the explanations given above for the case of rods hold as well. However, there is one very important difference: The disk is in-plane isotropic; therefore the fundamental plasmonic resonance can be excited with any given incident polarization under normal incidence illumination. Accordingly, the hole-shaped aperture exhibits a plasmonic resonance for the same incident polarization as the disk does. Moreover, the electric near-fields do overlap when placing the two structures side-by-side which readily allows for efficient coupling.

It has been demonstrated that in purely solid or purely inverse systems the concept of electric or magnetic dipoles, respectively, is a straightforward way to interpret the observed spectra.\[ \text{6,7,23−25} \] However, these concepts are no longer readily applicable to intuitively understand solid-inverse interactions. Even though one can still regard the structures as being composed of magnetic and electric dipoles, the interaction on the basis of these dipoles is no longer intuitive. If one intends to couple inverse and solid structures we have found that it is thus much easier to interpret the coupling in terms of electric fields. Therefore, one has to examine the local resonant electric fields associated with electric and magnetic dipoles and make sure that those fields spatially and spectrally overlap; that is, one has to ensure modal overlap.

In the following we are thus studying two different cases: The first example demonstrates the emergence of the plasmonic analogue of EIT in a structure which consists of two anisotropic rod-shaped apertures and a single anisotropic solid rod. The second one demonstrates a chiral optical response in a handed arrangement of individually isotropic holes and disks.
Generally speaking, the plasmonic analogue of EIT can be observed when coupling a “bright” to a “dark” plasmonic mode.\textsuperscript{33} In most studied systems the fundamental dipolar resonance of a single nanorod serves as the bright resonance. The dark mode is formed by a wire pair which supports an out-of-phase oscillation of the respective electric dipoles, thus forming a quadrupolar resonance.\textsuperscript{34–36} In case of efficient coupling between the two modes, the resulting spectrum will exhibit a peak of nearly perfect transmission within the former transmission dip of the dipolar resonance. In our mixed solid-inverse EIT structure two rod-shaped apertures serve as the dark mode, as depicted in Figure 2a. Such an arrangement supports an out-of-phase oscillation of the two magnetic dipole moments of the individual apertures, as sketched. The resulting mode can therefore be considered to be a magnetic dark mode which cannot be excited directly by an incident light field. A single solid rod arranged in parallel to the apertures and displaced from the symmetric position. In this case, the modes can interact via their respective electric near-fields, as sketched, and will lead to the formation of plasmonic EIT. (c) Geometrical parameters of the studied structures (identical in the simulations and experiments): $L_{Q} = 300 \text{ nm}$, $L_{D} = 280 \text{ nm}$, $B_{Q} = B_{D} = 80 \text{ nm}$, $D = 60 \text{ nm}$, the thickness of the gold film and of the solid dipole is $d_{Au} = 40 \text{ nm}$, and the vertical spacing between the gold film and the solid rod is 30 nm; the structure is repeated periodically along $x$ and $y$ directions with periodicity 800 nm.

As mentioned earlier, one has to visualize the electric near-field distribution to understand the resulting coupling phenomenon. For the aperture pair one observes strong electric near-fields in the equatorial plane around the apertures, leading to a charge distribution as depicted in the snapshot in Figure 2a, which is very similar to the case of a single rod-shaped aperture (cf. Figure 1b). It becomes immediately apparent how to couple the bright dipolar resonance of a single solid rod to this mode: In order for most efficient coupling the electric near-field overlap has to be maximized. This maximum overlap is achieved when the rod is arranged parallel to the apertures and displaced by half the length of the rod-shaped apertures, cf. Figure 2b. The field strength for the rod is largest at its ends; for the apertures it is largest right in the center. Therefore, varying the lateral shift $S$ of the solid rod versus the aperture pair allows tuning of the coupling strength between the bright and the dark mode and therefore the strength of the EIT feature. For zero offset, that is, for a symmetric placement of the rod between the apertures, the mode overlap and therefore the coupling vanish, and only the spectral signature of the bare solid rod can be observed.

Simulations are based on the Fourier modal method (also known as rigorous coupled-wave analysis) improved by adaptive coordinates and have been cross-checked by a commercial finite integration time domain software (CST).\textsuperscript{37,38} Figure 3 depicts simulated reflectance spectra as well as near-field distributions of the solid-inverse EIT structure for 0 and 150 nm offset. In the fully symmetric case one observes a single pronounced reflectance dip. To understand the nature of the excited modes we examine the normal component (that is the $z$-component) of the electric near-field distributions. In general, one has to study the modes in all three spatial dimensions. However, it has proven sufficient to only investigate the normal components of the near fields which nearly fully reveals the mode character. As expected from the explanations given above on resonance one clearly recognizes the dipolar character of the fundamental resonance of the solid rod at spectral position A. It is interesting to note that this dipolar resonance in fact hybridizes with a mirror plasmon excited in the metallic film which modifies the expected field distribution slightly, which becomes more pronounced when removing the aperture. However, the overall bright character of this plasmonic mode is not affected.

In breaking the system symmetry by laterally shifting the solid rod by 150 nm, one opens a pronounced reflectance window at the spectral position of the former reflectance minimum. If this phenomenon is indeed a plasmonic EIT effect, the near-field distributions at spectral position B right at

![Figure 2. (a) A pair of rod-shaped apertures in a metallic film supports an out-of-phase oscillation of two magnetic dipoles. This mode is a dark mode as it cannot be excited by an external light field. (b) The plasmonic dipolar resonance of a solid rod can couple to this magnetic dark mode if the rod is arranged parallel to the apertures and displaced from the symmetric position. In this case, the modes can interact via their respective electric near-fields, as sketched, and will lead to the formation of plasmonic EIT. (c) Geometrical parameters of the studied structures (identical in the simulations and experiments): $L_{Q} = 300 \text{ nm}$, $L_{D} = 280 \text{ nm}$, $B_{Q} = B_{D} = 80 \text{ nm}$, $D = 60 \text{ nm}$, the thickness of the gold film and of the solid dipole is $d_{Au} = 40 \text{ nm}$, and the vertical spacing between the gold film and the solid rod is 30 nm; the structure is repeated periodically along $x$ and $y$ directions with periodicity 800 nm.](image1)

![Figure 3. Simulated reflectance spectra and near-field distributions of the mixed solid-inverse EIT structure for $S = 0 \text{ nm}$ (upper row) and $S = 150 \text{ nm}$ offset (lower row) for incident excitation with its $E$-field along the solid rod (for geometrical parameters see caption of Figure 2). For the fully symmetric structure one observes a single dipolar resonance which originates form the excitation of the single solid rod. The near-field distribution ($z$-component, normal to the surface, of the electric field) exhibits a dipolar pattern, confirming this interpretation. For $S = 150 \text{ nm}$ one observes the formation of a reflectance window due to the interaction of the modes of the solid and inverse structures. As expected, the magnetic near-field distribution ($z$-component) at spectral position B clearly shows the signature of two out-of-phase oscillating magnetic dipoles, thus being a magnetic dark mode. When comparing the electric field strength associated with the solid bar at spectral positions B and C inside and outside the reflectance window, respectively, one observes a clear reduction in excitation of the solid bar, in accordance with the expectations for a plasmonic EIT-like phenomenon. The near-field plots represent cuts 10 nm below the plane of the dipole and are given in units of the incident field.](image2)
the maximum of the reflectance window should exhibit the characteristics of the dark plasmonic mode. In our case, as argued above, this dark mode is a magnetic one. Therefore one has to study the magnetic near-field rather than the electric one. Indeed, the z-component of the magnetic field clearly shows an out-of-phase oscillation of two magnetic dipoles, thus forming a dark quadrupole-type resonance. At this spectral position the z-component of the electric field associated with the solid rod exhibits a significantly reduced magnitude when compared to spectral position C. This finding further underlines our interpretation as an EIT-like phenomenon: Due to the energy transfer involved in the coupling of the bright and dark modes the scattering of the solid rod is reduced, causing the opening of a pronounced reflectance window.

The structure exhibits another intriguing asymmetry: The modulation of the reflectance window increases with increasing offset until S equals half of the length of the apertures (which is S = 150 nm in this case) as the mode overlap increases. For larger offsets the electric mode overlap diminishes again. Ultimately, the coupling will vanish, and a single dipolar resonance is observed in the spectrum. It is interesting to note that this resonance is not at the same spectral position as for the fully symmetric case with 0 nm offset (see Figure S1 in the Supporting Information for simulated reflectance spectra in dependence of the offset S). The reason is as follows: In the case of the symmetric structure the solid rod is surrounded by the two rod-shaped apertures. On the one hand the dielectric surrounding is strongly influenced by the presence of the holes, which results in a resonance shift due to the coupling of the plasmon with the nonresonant background. On the other hand, the air-holes diminish the strength of the induced mirror plasmon. As soon as the solid rod is shifted by an S larger than the length of the apertures the plasmonic mode will hybridize with a strong mirror plasmon in a now basically unperforated metallic film.

To experimentally verify our findings we have fabricated a set of samples via two-step electron beam exposure. The inverse layer is fabricated by argon milling through a resist mask and planarized with a spin-coatable spacer layer. The second layer is afterward fabricated via a standard PMMA lift-off procedure (for details please refer to the Methods section in the Supporting Information). Figure 4a depicts tilted and normal view scanning electron microscope (SEM) images of the fabricated structures. The pairs of rod-shaped apertures appear as dark structures in the otherwise comparably bright gold surface. The single solid rods appear very bright, in particular as they are closer to the sample surface as compared to the gold film. The normal view SEM images underline the excellent alignment of the two layers with respect to one another. Overall, the images demonstrate the feasibility of fabricating mixed solid-inverse systems.

Reflectance measurements for the different structures were performed using a Fourier transform infrared spectrometer with a 15× Cassegrain objective (NA = 0.4) and a liquid nitrogen cooled MCT detector. Figure 4b depicts the measured reflectance spectra of our fabricated samples for increasing lateral offset S. For the fully symmetric structure, that is, for S = 0 nm we observe a single dipolar reflectance dip. Already for S as small as 30 nm the onset of a reflectance window can be clearly observed. Further increasing the lateral offset increases the modulation of the reflectance window. For S = 150 nm we observe a well-modulated reflectance peak at
Figure 5. Normal and tilted view scanning electron micrographs and measured circular dichroism (CD) spectra of the fabricated mixed solid-inverse chiral structures. The first layer consists of three holes arranged in an L-shape. A solid disk placed in a second layer determines the handedness of the resulting structure. The cluster exhibits a clear CD signal underlying the straightforward coupling of disks and holes. The holes and disks have a diameter of 200 nm and an interparticle spacing of 20 nm. The thickness of the gold layers is 40 nm. The vertical stacking distance is 30 nm. The scale bars are 500 nm for the overview image and 200 nm for the close-ups.

As an additional example of a mixed solid-inverse system we study a chiral plasmonic structure. It has been shown theoretically and experimentally that the handed arrangement of equally sized particles will result in a chiral optical response of the cluster, that is, in a different response of the structure to left- and right-handed circularly polarized light. The original design of the structure adapted here consists of three equally sized disks, three arranged in an L-shape in the first layer, one placed in a second layer, and thus determining the handedness of the resulting cluster. As discussed earlier, solid disks and holes do readily couple to one another as they are isotropic structures. Hence, in contrast to the mixed solid-inverse EIT system there is no need for a redesign of this chiral structure. We expect that every possible combination of disks and holes will facilitate efficient plasmonic coupling and hence the formation of collective behavior and a chiral optical response from the cluster.

Figure 5 shows normal and tilted view SEM micrographs as well as measured spectra of the fabricated structures. The fabrication procedure is identical to the mixed solid-inverse EIT structure. The first layer consists of three holes arranged in an L-shape in a continuous gold film. A solid disk is placed on top of a spacer in a second layer and determines the handedness of the resulting cluster. Again, the difference of the solid and inverse structures is clearly visible. The clusters are arranged in a C4 symmetric lattice to suppress contribution of polarization conversion. To characterize the chiral optical response of the cluster, transmission and reflection spectra for circularly polarized light have been measured. Subsequently, the absorbance spectra can be calculated via $A_{XCP} = 1 - T_{XCP} - R_{XCP}$. From the absorbance the circular dichroism (CD) spectra can be calculated as $CD = A_{LCP} - A_{RCP}$. The obtained spectra are displayed in the right panel of Figure 5. We observe a clear CD response from the clusters. Moreover, the spectra change sign upon change of the handedness of the cluster, as expected. The overall strength of the signal is weaker as compared to previously reported results. We believe that this is related to the challenging fabrication and the difficulty of obtaining well-pronounced resonances in inverse structures rather than to an inherent design flaw. Therefore, we have experimentally demonstrated that in in-plane isotropic structures efficient plasmonic coupling is indeed possible.

In conclusion we have shown that it is possible to efficiently couple the plasmonic resonances in solid and inverse plasmonic structures. In adapting a dolmen-type plasmonic EIT structure we have demonstrated the efficient exchange of energy between modes in inverse and solid structures. Importantly, for structures comprising of anisotropic building blocks the overlap of the electric near-fields, that is, modal overlap, has to be ensured. In case of, for example, our mixed solid-inverse EIT structure this coupling prerequisite leads to a somewhat counterintuitive parallel arrangement of the two apertures and the single solid rod. Moreover we have shown that, for in-plane isotropic structures, such as disks and holes, efficient coupling is automatically ensured as both structures exhibit plasmonic resonances for identical illumination conditions. We have experimentally verified this finding in a mixed solid-inverse chiral structure which consist of three holes and one disk and found a clear plasmonic CD response. The incorporation of inverse structures into solid ones offers a new tool for the construction of complex plasmonic structures. In general inverse structures are particularly interesting due to the magnetic nature of their modes and can thus be expected to be a suitable antenna for magnetic emitters. By utilizing the greatly enhanced toolbox presented in this paper, inverse magnetic slot antennas could couple via adjacent solid electric dipole antennas strongly to the propagating far field. This would enhance the efficiency of magnetic emission dramatically and would have extremely beneficial advantages for single quantum emitters.

ASSOCIATED CONTENT

Supporting Information
A description of methods and Figure S1. This material is available free of charge via the Internet at http://pubs.acs.org.

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