Observation of linear plasmonic breathers and adiabatic elimination in a plasmonic multi-level coupled system

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Abstract: We provide experimental and numerical demonstrations of plasmonic propagation dynamics in a multi-level coupled system, and present the first observation of plasmonic breathers propagating in such systems. The effect is observed both for the simplest symmetric case of a thin metal layer surrounded by two identical dielectrics, and also for a more complex system that includes five and more layers. By a careful choice of the permittivities and thicknesses of the intermediate layers, we can adiabatically eliminate the plasmonic waves in all the intermediate interfaces, thus enabling efficient vertical delivery and extraction of plasmonic signals between the top layer and deeply buried layers. The observation relies on controlling the excited mode by breaking the symmetry of excitation, which is crucial for obtaining the results experimentally. We also observe this breathing effect for transversely shaped plasmonic beams, with Hermite-Gauss, Airy and Weber wavefronts, that despite the oscillatory nature of propagation in such systems, still preserve all their unique wavefront properties. Finally, we show that such approaches can be extended to plasmonic propagation in a general multi-layered system, opening a path for efficient three-dimensional integrated plasmonic circuitry.

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References and links
Surface-plasmon-polaritons are unique two-dimensional (2D) electromagnetic surface waves,
propagating at a metal-dielectric interface, coupled to charge oscillations in the metal [1]. Owing to their lower dimensionality and their unusual properties of confining and enhancing the optical field, they enable the observation of unique optical phenomena in a 2D evanescent wave system. Among these are plasmonic paraxial and non-paraxial self-accelerating beams [2–5], non-diffracting [6] and loss compensating beams [7], super-oscillations [8], and a plasmonic Bragg law [9–11]. Furthermore, the propagation of these high frequency surface waves on metals is very attractive for integration platforms, which can jointly support electrical and optical signals [12–14]. Thus, major scientific efforts have been done in order to realize plasmonic integrated elements, while mainly focusing on planar integration at a single plane. Recently, efforts have been made to achieve 3D routing of optical signal by using three-dimensional photonic crystals, while still trying to maintain confined waveguiding with a vertical footprint of several micrometers [15–17]. In order to allow similar robust and scalable plasmonic integration, 3D plasmonic circuitry should be also be enabled. The ideal platform would support different circuits residing at different layers, and allow the efficient delivery of plasmonic signals to/from these individual layers. Thus, a robust and compact way to enable this is essential.

In this work, we report the experimental observation of a new type of plasmonic solution – a plasmonic breather that periodically oscillates between different metal-dielectric interfaces, in a stack of metal-dielectric layers. We experimentally demonstrate the behavior of a plasmonic two-coupled-mode system (2CMS), and four-coupled-mode system (4CMS) in the adiabatic-elimination (AE) scheme, and show that it allows for efficient and controllable transfer of plasmonic signals from the most upper level to the lowest level, and vice-versa, with a vertical footprint of less than 0.5 µm. We show that this system can support arbitrary transverse plasmonic wavefronts, while periodically oscillating, or "breathing", between different layers. Finally, we show that this approach can be extended to a general plasmonic multi-layered system, providing a powerful platform for truly 3D integrated plasmonic circuitry, in sub-wavelength vertical spacing.

In order to achieve this, we utilize the fact that the underlying physical behavior that governs a vertical stack of metal/dielectric layers can be described by coupled multi-level systems, where each metal/dielectric interface acts as a single level. While eigenmodes modes of metal-dielectric layers have been previously analysed [42, 43], their non-eigenmodes analysis and excitation have not been studied until now. The simplest multi-layered coupled system is the 2CMS, which has many physical analogs, such as two coupled pendulum, atomic two-level systems, and two coupled identical optical waveguides. In the latter, once excited at a single waveguide (level), the system will exchange energy in an oscillatory manner between the coupled waveguides, and behave as a directional coupler [18]. Although plasmonic directional couplers have been extensively researched, these usually describe the interaction between two, Insulator-Metal-Insulator (IMI), Metal-Insulator-Metal (MIM) or hybrid waveguides, in close proximity on the same horizontal plane [19–26]. However, the fundamental physical understanding that these plasmonic systems are actually multi-level coupled systems implies that a single IMI waveguide is already a vertical plasmonic 2CMS with two levels (interfaces) (Fig. 1a). This approach enables to further extend this concept to a set of densely packed multiple plasmonic levels with nanometric spacing, while precisely controlling how energy is transferred between these levels. In order to achieve a robust and efficient transfer of the plasmonic waves from the most upper layer to the most lower layer, while minimizing the energy in all other modes, we have implemented the AE scheme in this system. This scheme is shown to be quite immune to additional loss and decoherence of the plasmonic system in the vertical pass. Such scheme allows a practical way of transferring plasmonic signals into/from deeply buried layers.

We start by demonstrating the evolution of the plasmonic 2CMS. A general 2CMS can be described by two eigenmodes - symmetric and anti-symmetric, which when excited will propagate through the system unchanged. Such symmetric and anti-symmetric eigenmodes also
exist in the plasmonic IMI system, and are known as long- and short-range surface plasmons. While these eigenmodes excitations were extensively researched [27, 28], the excitation of a single level (interface), which is a superposition of the symmetric and anti-symmetric modes, is challenging and thus usually not considered. The coupled equations for the 2CMS can be written in the basis of the modes that propagate in the upper and lower levels as:

$$\frac{d}{dy} \begin{bmatrix} A_1 \\ A_2 \end{bmatrix} = i \begin{bmatrix} \beta_1 & \kappa_{12} \\ \kappa_{21} & \beta_2 \end{bmatrix} \begin{bmatrix} A_1 \\ A_2 \end{bmatrix}$$

(1)

where $A_i$ represent the fields of mode $i = 1, 2$, $\beta_i$ are the modes’ propagation constant, $\kappa_{12}, \kappa_{21}$ are the coupling strengths between modes 1, 2, where $\kappa_{12} = \kappa_{21}^{*}$, and $y$ is the propagation direction. The intensity distribution in one of the waveguides including losses can be described by $\exp(-2L_P \sin^2 \frac{\pi}{2L_{C}})$, where $L_C \equiv \frac{\pi}{|\beta_S - \beta_A|}$ is the coupling length, $L_P$ is the mean plasmonic propagation length, and $\beta_S, \beta_A$ are the wave-vectors of the symmetric and anti-symmetric modes [29, 30]. Hence, the intensity will oscillate with a period of $\Lambda = 2L_C$. To avoid any possible confusion, we note that $\beta_1, \beta_2$ are the propagation constants of a single interface plasmon, and the detuning of the system, $\Delta$, is defined by $\Delta \equiv \beta_1 - \beta_2$. Thus, for the symmetric structure where $\beta_1 = \beta_2$, the detuning is zero and we also have $L_C \equiv \frac{\pi}{|\beta_S - \beta_A|} \equiv \pi/\kappa_{12}$. To validate this prediction, in Fig. 1(b) we show COMSOL simulation of a 2CMS with 30nm Ag layer surrounded by BK7, excited on a single (top) level, and the predicted oscillations, having a period of $\Lambda = 14.5\mu m$.
are in complete agreement with this calculation. It can also be seen that since the structure is symmetric, the energy is transferred from one level to the other, in full agreement with the 2CMS theory.

To observe this phenomenon experimentally, it is required to excite the correct mode at only one, out of the two, levels. Such excitation, which requires the symmetry of excitation to be broken, is challenging and has not been explored nor observed until now. Commonly, a grating is etched as slits into the metal layer, which with the appropriate period, matches the momentum between the free-space illuminating beam and the excited plasmon. However, this results in the excitation of both the top and bottom levels in a symmetric environment, thus eliminating the possibility to observe the above phenomenon, as seen in the full-wave simulations presented in Fig 2(a). The small perturbations in plasmon intensity is related to some reflections from the PML boundary. To break this symmetry, we fabricate the sample in Fig. 1(a), by depositing a 30nm thick Ag grating on top of the metal layer, whose wave-vector \(2\pi\Lambda_G\), where \(\Lambda_G\) is the grating period, matches that of the single level plasmon mode, e.g. \(\Lambda_G = 2\pi\beta_1\). By placing the grating on the top layer we are breaking the symmetry of excitation, thus exciting the plasmon only at one interface, which results in the predicted oscillations, as seen in Fig. 2(b).

The experimental measurements are obtained by covering the sample with an index matching fluid having the same index as that of the lower BK7 layer. The sample was illuminated with a 1.064\(\mu\)m laser, and a Nanonics MultiView 2000™ NSOM system is used to collect the near-field of the excited plasmons at the top level, by measuring with the NSOM tip inside the fluid. All the experiments and simulations were designed to exhibit the desired phenomena within 80\(\mu\)mX80\(\mu\)m area, corresponding to the scanning range limit of the NSOM system. Fig. 3(a), shows the measured propagating plasmon intensity on the top interface of the 2CMS, and the plasmonic field oscillations can be clearly seen. The oscillation period is inversely proportional to the coupling coefficient, which depends on the thickness of the metal, refractive indices of the dielectrics, wavelength and so on. Accordingly, our theory and simulations predicts a change in the period from \(\Lambda = 14.5\mu\)m, for the 30nm layer, to \(\Lambda = 25.6\mu\)m period for a 40nm layer. Fig. 3(b) shows the results for a 40nm Ag layer thickness, and from Fourier analysis we find the measured periods to be \(\Lambda \approx 14.2\mu\)m and \(\Lambda \approx 25\mu\)m, respectively, which are in good agreement with our predictions. The expected exponential decay of the plasmons in the propagation direction can be also observed as the envelope of the oscillations.

So far we have shown that a single metal layer sandwiched between two dielectric layers can be accurately modeled as a coupled two-level system. Let us now extend the analysis to a system that includes multiple layers of metal and dielectric, such as a 4CMS. This system can

![Fig. 2. COMSOL full-wave simulations of the intensity distribution for the case of (a) a grating milled down in to the metal layer as slits, which results in a symmetric excitation and lack of oscillations, and (b) a metal grating deposited on top of the metal layer, which breaks the symmetry of excitation and results in the clearly observed oscillations.](image-url)
be modeled as a multi-level system assuming that the plasmonic mode in each metal-dielectric interface couples only to the nearest interfaces, which is a good approximation owing to their evanescent nature. The coupled mode equations for the 4CMS case, similarly to Eq. (1), can therefore be written as:

$$\frac{d}{dy} \begin{bmatrix} A_1 \\ A_2 \\ A_3 \\ A_4 \end{bmatrix} = i \begin{pmatrix} \beta_1 & \kappa_{12} & 0 & 0 \\ \kappa_{21} & \beta_2 & \kappa_{23} & 0 \\ 0 & \kappa_{32} & \beta_3 & \kappa_{34} \\ 0 & 0 & \kappa_{43} & \beta_4 \end{pmatrix} \begin{bmatrix} A_1 \\ A_2 \\ A_3 \\ A_4 \end{bmatrix}$$

where $\kappa_{12} = \kappa_{21}^*, \kappa_{23} = \kappa_{32}^*, \kappa_{34} = \kappa_{43}^*$. Analytically, this multi-level system is described by a set of four linearly coupled equations, exhibiting an $SUN$ symmetry with $N^2 - 1$ degree of freedom. Thus, we can now implement the AE procedure on the system, which basically reduces the symmetry of the system to $SU2 + U1 \ast N - 1$, which has only 3 degrees of freedom [31]. This means that any N-level system can be reduced to a system which, to a good approximation, behaves as a two-level system between the top and bottom levels. The procedure in fact put the constraints and requirements that the amplitude derivatives of the intermediate plasmons will be asymptotically zero along the entire propagation,

$$\frac{dA_i}{dz} \to 0.$$

In general, this can happen only when the interior plasmon modes have different (quite far) mode indices than the outer ones. Only in this way very low excitation of the interior layers can occur. In contrary, if all the plasmonic waves were identical one cannot obtain that the derivatives of the interior modes will be asymptotically zero along the entire propagation, thus resulting in their significant excitation, and the system cannot be considered as an effective two mode system (between the outer levels). For this reason, we change the material of the inner dielectrics to be different than the outer ones. This introduces a mismatch in the coupling between the inner and outer modes thus reducing or "eliminating" the amount of energy residing within them, as compared to the outer waveguide. In the last several years, this approach has been successfully applied in controlling the dynamics of a variety of physical phenomena, such as nonlinear optics [32], and nano-photonics [33, 34], and is quite different from previously non-adiabatic focusing methods [35], or decoupling methods [36].

Specifically for the plasmonic system, it implies that by carefully selecting the thicknesses and permittivities of the metal and dielectric layers, we can implement the AE method, so that the excitation of the intermediate interfaces will be minimal along the entire propagation distance,
and the plasmonic beam will oscillate between the top and bottom interfaces. This enables an important application for 3D integration of on-chip plasmonics, as it allows the delivery and extraction of plasmonic signal from the top layer into deeply buried layers. The plasmonic structure for such an AE scheme is achieved by choosing an identical refractive index for the most top and bottom dielectrics (hence $\beta_1 = \beta_4$), but different from the intermediate dielectric layers (thus $\beta_1 \neq \beta_2$), and properly setting the coupling strengths and detuning of the intermediate interfaces. Specifically, in our configuration the two intermediate interfaces are obtained for a dielectric layer sandwiched between two silver layers, hence $\beta_2 = \beta_3$. We therefore have only two different propagation constants -- $\beta_1 = \beta_4$ for the top and bottom interface, and $\beta_2 = \beta_3$ for the two intermediate interfaces. This setting enables to couple the first and fourth interfaces, while adiabatically eliminating the coupling to the second and third intermediate interfaces. Specifically, we obtain the coupling coefficients of the 2CMS for different thickness and fit it exponentially to obtain the metal-thickness-dependent real and imaginary part of the coupling coefficient ("Rabi"-coupling), and then use it in determining the coupling between two adjacent plasmons in the multi-layers analysis. Once these are determined, full-wave simulations can be performed to obtain the exact behavior of the system.

Such a plasmonic 4CMS is shown in Fig. 1(b), and is composed of a BK7 substrate, 30nm Ag layer, 300nm Al$_2$O$_3$ layer, 30nm Ag layer and BK7 as the top dielectric. The contrast between the BK7 and Al$_2$O$_3$ refractive indices provides the detuning required for the AE scheme, whereas the thicknesses of the different layers enables to control the coupling strengths. Fig. 1(d) shows the simulation of the structure, and the oscillations between the 1st and 4th levels are clearly observed. The cross-section of the intensities residing in each level is presented in the inset of Fig. 1(d), and the AE behavior of the system is clearly seen, e.g. most of the energy is oscillating between the 1st and 4th levels, with a small portion residing in the 2nd and 3rd levels. Thus,
Fig. 5. Near-field measurements of shaped plasmonic breathers. (a), (b) HG first order mode propagating on the 2CMS and 4CMS, respectively. (c), (d) Self-accelerating Airy and Weber plasmon breathers, respectively, propagating in the 2CMS. Pink curves represent the designed trajectories of acceleration. All beams are seen to maintain their wavefront properties despite propagating on different spatial layers.

we have successfully reduced the 4CMS to a practical 2CMS. The fabrication yielded 22 nm top Ag layer, 190 nm Al$_2$O$_3$ layer and 30 nm bottom Ag layer. From simulations we deduce the period of oscillation to be $\Lambda \approx 38 \mu m$. Figure 4(a) shows the experimental measurement of the plasmonic 4CMS together with SEM images of the samples’ cross-section, and the oscillations are clearly seen with a period of $\Lambda \approx 40 \mu m$, which is in good agreement with the simulation, validating the AE characteristic of the system. We note that the increased noise level observed in the 4CMS, compared to the 2CMS, is due to the accumulated surface roughness of the larger 4CMS structure, also affecting the intensity distribution among levels.

Furthermore, this system is more sensitive to the thickness of the Ag layers than those of the dielectrics. For example, when changing the bottom Ag thickness from 30 nm to 50 nm, and the Al$_2$O$_3$ thickness from 190 nm to 260 nm the energy transfer changes drastically, as shown in Fig. 4(b), and the plasmon seems as a regular decaying Gaussian plasmon. Indeed, simulations of the structure reveals that the period of oscillation has changed dramatically, from $\sim 38 \mu m$ to over $80 \mu m$, which is larger than our measurement capabilities, thus it can be seen in Figure 4(b) as a lack of oscillations. It also reveals that the AE does not hold anymore, owing to the different structure. We note that this sensitivity to the variation in the permittivity may be quite useful for sensing application and the probing of thin layers.

Unlike other atomic/quantum multi-level systems, the plasmonic system possesses an additional and unique degree of freedom, which resides in its transverse wave-function. We can describe the propagation of plasmons in this multi-layered system as a single beam with a
well-defined transverse shape, propagating periodically and discretely on different levels, while maintaining this transverse wavefront. We refer to this unique behavior as plasmonic breather beams, as contrived by [37] (and not to be confused with nonlinear breathers). To launch such a plasmon, we replace the square grating with plasmonic binary hologram, which is capable of arbitrarily shaping the transverse plasmon’s wavefront [3, 38–40]. The advantages of this approach are twofold: first, the hologram couples the shaped wavefront to a specific level, thus, if we observe propagating plasmons oscillating with the same wavefront, we can safely deduce that it went through all the different layers. Second, a shaped and transversely confined wavefront plasmon may be useful for other applications, like on-chip communication and routing. Figure 5(a) and 5(b) shows the experimental measurements of such plasmonic breathers, on both 2CMS and 4CMS, having the transverse wavefront of a Hermite-Gauss (HG) first order mode. It can be clearly seen that this HG plasmon keeps its transverse shape although propagating discretely and periodically through the system, with the period of oscillation according to the 2CMS, and 4CMS observed above. We note that the intensity measured at the bottom left side of Fig. 5(b), is the illumination picked-up by the NSOM tip. No oscillations are observed from that signal, these are observed only where the binary hologram exists and couples the illumination into the system.

Another striking example is that of self-accelerating plasmon breathers. Airy plasmons are non-diffracting beams that keep their transverse shape while accelerating, e.g. propagating along curved trajectories [2, 4, 5]. Although coupled Airy breathers were theoretically predicted in a general coupled equation theory [37], this is their first experimental demonstration. Moreover, while Airy plasmons obey the paraxial approximation, even non-paraxial accelerating plasmons, such as the Weber beam [3, 41], are supported in this system. Fig. 5(c) and 5(d) shows both Airy and Weber plasmon breathers propagation in the 2CMS, which are accelerating on the two different levels along a curved trajectory. It is seen that their unique features of non-diffraction and self-acceleration are preserved in their breathers form as well.

Finally, showing that this approach can be generalized to a plasmonic N-level system, we implement AE to a plasmonic 6-couple-modes system. The 7-layer structure is thus, BK7 substrate, 20nm Ag layer, 150nm Al2O3, 20nm Ag layer, 150nm Al2O3 layer, 20nm Ag layer and BK7 as the top dielectric. The simulation results are presented in Fig. 6, and the AE behavior of the system is clearly observed. We note that the increased loss in this case, compared to the 4CM, is due to the thinner Ag layers, and can be tailored with the structure.

In conclusion, we provided analytical and experimental demonstrations of plasmonic propagation dynamics in multi-level coupled plasmonic system. We demonstrated a 2CMS, and AE in a
4CMS and 6CMS, which are capable of efficiently delivering signals between the top and bottom levels, with few hundred of manometers footprint. In addition, we demonstrated the existence of plasmonic breathers propagating on these multi-layered systems, while remarkably maintaining their wavefront properties. With the vast variety of configurations of vertical and planar IMI and MIM structures, having different geometries and materials, together with the sensitivity of the structures and additional schemes available from coupled-mode-theory and atomic physics, we believe this approach is of significant value for plasmonic on-chip circuitry and sensing, and will pave the way to new possibilities for 3D integration technologies. Moreover, the system can support multiple wavelengths, making it attractive to multiplexing applications, and can be further extended to other well-known concepts for efficient and robust coupling, such as adiabatic-following and STIRAP [31].

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