Plasmonic Metasurface “Bullets” and other “Moving Objects”: Spatiotemporal Dispersion Cancellation for Linear Passive Subwavelength Slow Light
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A class of plasmonic meta-surfaces is introduced with the ability to tailor the dispersion surface of the associated plasmon-polariton into striking novel shapes. Examples include dispersion surfaces with hyperbolic curves, with multiple van Hove singularities of various types or with points of simultaneous spatiotemporal dispersion cancellation leading to unprecedented surface flatness. The latter effect, unseen before in linear passive systems, implies slow propagation of ultra-subwavelength wavepackets of any shape devoid of longitudinal or lateral broadening, limited only by absorption.

In the perennial effort to mold the flow of light at will, one fundamental issue has always stood as an obstacle: dispersion, the tendency of light wavepackets to spread out as they propagate. It appears in many types, according to the dimensionality of the propagation. In a one dimension x, such as a straight waveguide, curvature in the dependence kx(ω) of wavevector on frequency is called temporal dispersion or, equivalently seen as ω(kx), spatial-longitudinal dispersion (pulse broadening in the direction of propagation). Scientists have devised methods to compensate temporal dispersion, including via nonlinearities leading to temporal solitons [1], or waveguide designs to actually cancel (namely eliminate) it to high orders [2, 3].

For propagation along x in a two-dimensional (2d) system xy, a constant-frequency narrow beam will diverge/diffRACT, if the dependence ky(kx) of longitudinal on transverse wavevector has curvature, or, seen as curvature in ω(ky), a pulsed beam infinite in x will disperse spatially-transversely (in y). Diffraction has been countered either again via nonlinear compensation and spatial soliton beam formation [1, 4] or via cancellation in flat equi-frequency dispersion curves (EFDC), named super-collimation, of 2d photonic crystals [5, 6] and of hyperbolic plasmonic meta-surfaces [7, 8].

For pulse propagation in 2d, both temporal (spatial-longitudinal) and spatial-transverse dispersions occur, and only nonlinear spatiotemporal solitons [1, 4, 9–11] and linear wavepackets of pre-prepared specific shapes (Airy-Bessel) [12] have been shown to form ‘light bullets’ by balancing both types. They have never been simultaneously cancelled before. In this Letter, we present a linear passive plasmonic meta-surface platform, which accomplishes simultaneous SpatioTemporal Dispersion Cancellation (STDC) for subwavelength slow Surface Plasmon Polariton (SPP) modes [13], leading to propagation without broadening for ultra-short 2d pulses, namely plasmonic ‘bullets’, and generally for sub-wavelength 2d wavepackets of any shape, akin to ‘moving objects’. We emphasize that these effects relate to in-plane meta-surface propagation, in contrast to plasmonic meta-surfaces involving transmissive functionality [14, 15].

Tuning the Photonic Density of States (PDOS) of a structure is fundamental for controlling or enhancing light-matter interactions. Systems with hyperbolic dispersion [16–18] exhibit theoretically divergent PDOS, so are promising for boosting spontaneous or thermal emission and could have applications in imaging. In a 2d dispersion surface, Van-Hove extrema-type singularities [19] lead to steps in the PDOS and saddle-type points, previously encountered only in 2d photonic crystals [20, 21], to logarithmic PDOS divergences, both PDOS features extremely useful for high-frequency-sensitivity sensors and filters. Our currently proposed SPP platform can be tuned to also give a variety of exotic dispersion surfaces, including hyperbolic behavior, extrema- or saddle-type van-Hove singularities, or all of these simultaneously.

Consider a SPP propagating along x on a metallic surface defining the xy-plane with dielectrics above it (z > 0). For a common isotropic planar system with continuous xy-translational and z-rotational symmetries, the EFDC ω(kx, ky) are circles, with ω dependent only on \(\sqrt{k_x^2 + k_y^2}\), so \(D_{xy}^\omega = k_x \partial^2 \omega / \partial k_x^2|_{k_y=0}\), the leading dispersion coefficient causing diffraction, is \(D_{xy}^\omega = -1\). In order to instead achieve super-collimation \(D_{xy}^\omega = 0\), the rotational symmetry needs to be broken. The standard way is to use 2d periodicity in the xy-plane [5, 6], but here we begin by considering a uniform system with anisotropy. Let the space z > 0 be occupied by a uniform uniaxial dielectric with its optical axis along x [inset of Fig. 1(a)]. This system has been analyzed [7, 22] and shown to exhibit hyperbolic dispersion within some frequency range. The dispersion for \(\varepsilon_x = 6.45, \varepsilon_y = 1.845\) is shown in Fig. 1(a), where, at low frequencies, EFDC are ellipses, so \(D_{xy}^\omega < 0\), while, at higher frequencies, EFDC indeed become hyperbolas, so \(D_{xy}^\omega > 0\); therefore, there is a frequency where super-collimation \(D_{xy}^\omega = 0\) occurs (true for any 2d hyperbolic system [7, 8]). For definiteness, in all results presented in this Letter, the metal is silver (Ag) and was modeled by a Drude permittivity with \(\varepsilon_{\infty} = 4\), plasma frequency \(\omega_p = 9.3eV / h = 2e/\lambda_p\), and \(\gamma / \omega_p = 0.0025\) intrinsic bulk losses at room temperature [23–26], but nonlocal Landau surface-damping [27] was ignored to avoid complexity.
Refs. [3, 28] motivated using ultra-thin dielectric layers with alternating high/low index on top of the metal to create interesting $z$-rotationally-invariant SPP dispersion relations, including the cancellation of temporal dispersion to high orders. The underlying principle was that, at small $k$, the SPP field extends far from the metallic interface and thus ‘sees’ more of the outermost dielectric layer, while, as $k$ increases, the field is more tightly bound to the interface, so is affected more by the innermost layers. The same idea holds, when we include uniaxial layers. Figs. 1(b,c) show the dispersion for two such systems. Their qualitative difference is that, at large $k$, they asymptote to circular and hyperbolic dispersions respectively, because the closest layer to the metal is correspondingly air and a uniaxial dielectric. More importantly however, in both cases, very interesting and unusual features are present, specifically several van-Hove singularities [19]: two minima A, two sharp maxima D and four saddle points B,C. For Fig. 1(b), their associated impacts in the Total Density of States (TDOS) are, as expected, a large positive step at minimum A, log-peaks at saddles B,C and a small negative step at maximum D, as shown in Fig. S1 of the Supplementary Information section B (SupInfo-B) [29], where we describe also the TDOS calculation details [30–32]. Such singularities have so far only been associated with 2d-periodic systems [20, 21], but here we show that they are, perhaps counter-intuitively, possible also for $xy$-translationally-invariant systems, which combine the special properties of SPPs with anisotropy. The calculations for multi-layered structures including uniaxial dielectric media (in Figs. 1,2) were done using a scattering-matrix method [33, 34] based on the formulation in [35], as presented in detail in the SupInfo-A.

Furthermore, since a planar distribution of ultra-thin isotropic layers can lead to temporal (or spatial-longitudinal) dispersion cancellation [3] and uniaxial layers can result in super-collimation (spatial-transverse dispersion cancellation), our main goal is to examine if the presently proposed combination of isotropic and uniaxial dielectric layers can lead to a SPP with simultaneous (longitudinal and transverse) STDC. Considering all optical axes of uniaxial layers to lie in the $x$ direction, then $\omega(k_x, -k_y) = \omega(k_x, k_y)$. Therefore, if $D_{n,m} \equiv \omega_0^{-n-1} \frac{\partial^{n+m} \omega}{\partial k_x^n \partial k_y^m} \big|_{k_x=0}$ are the normalized dimensionless dispersion constants of $(n,m)^{th}$ order at the point $(k_{x0}, 0, \omega_0)$, then $D_{n,m} = 0$ for $m$ odd and $v_{g0} = cD_{1,0}$ is the group velocity along $x$ ($D_{0,1} = 0$). STDC would mean that we can design a system with $D_{2,0} = D_{0,2} = 0$ (note $D_{1,1} = 0$ anyway). Even better then, in Figs. 2 we present results for a structure which supports a SPP mode with $D_{2,0} = D_{0,2} = D_{3,0} = 0$, namely the first non-zero dispersion terms are $D_{1,2}, D_{4,0}, D_{2,2}$ and $D_{0,4}$, which are high-order terms with small impact to pulse propagation, as we will see. Although several configurations within our proposed system can achieve this cancellation, the simplest structure presented here (shown in inset of Fig. 2(a)) entails placing on the metal interface only a very thin dielectric ($\varepsilon = 11.9$) layer and an even thinner uniaxial layer ($\varepsilon = 6.45, \varepsilon = 1.845$). The dispersion surface shown in Fig. 2(a) around the cancellation point is the flattest plane ever demonstrated, to our knowledge, for a linear passive photonic structure. Note that, since $D_{y2} = k_x \frac{\partial^2 \omega}{\partial k_y^2} = k_x \frac{\partial}{\partial k_y} \left( -\frac{\partial \omega}{\partial k_x} / \frac{\partial \omega}{\partial k_y} \right) = k_x \left( -\frac{\partial^2 \omega}{\partial k_x^2} + \frac{\partial \omega}{\partial k_x} \frac{\partial^2 \omega}{\partial k_y^2} \right) v_{g0}^2 = -k_x D_{0,2} / \omega_0 v_{g0} = 0$, super-collimation is achieved also in the traditional sense, as indicated with the EFDC shown in Fig. 2(a).
dielectrics (GaP-air) with same thickness (right). (b) The structure of Fig. 2 (left) emulated via (b-right) with $d = 0.0483\lambda_p$: note the extremely flat regime around $(k_{x0}/k_p = 1.540, k_{y0}/k_p = 0, \omega_0/\omega_p = 0.237 - 0.001034i)$ with $v_{g0}/c = 0.0053$; 3 EFDC demonstrate super-collimation at the cancellation point. (b) (all lines except green) $D_{n,m}$ dispersion coefficients (namely derivatives of (a)) along $k_x$: $D_{2,0} = D_{3,0} = 0 = \text{cancelled at } (k_{x0}, 0)$. (green line) $\Gamma = -\text{Im}(\omega)$ loss rate; note that, as $k_x \to \infty$, $\Gamma \to \gamma/2$ intrinsic limit [3, 36]. (c) $D_{0,m}$ coefficients ($k_y$-derivatives of (a)) along $k_y$: again $D_{0,1} = D_{0,2} = D_{0,3} = 0$.

Figure 3: (a) A uniaxial dielectric with optical axis along $x$ (left) created effectively via $x$-periodic layering of two isotropic dielectrics (GaP-air) with same thickness (right). (b) The structure of Fig. 2 (left) emulated via z-slicing the effective uniaxial of (a) into a GaP grating meta-surface (right). (c) Periodic part of Bloch mode $F(k_{x0}, k_{y0}; x, z)$ for the SPP meta-surface in (b-right) with $d_{\text{film}} = 0.13\lambda_p = 17.3\text{nm}$, $d_{\text{grating}} = 0.059\lambda_p = 7.9\text{nm}$, $a = 0.15\lambda_p = 20\text{nm}$, at $(k_{x0}/k_p = 1.576, k_{y0} = 0, \omega_0/\omega_p = 0.238 - 0.00104i)$ and with $v_{g0}/c = 0.004$. (d) $\omega(k_x, k_y)$ dispersion surface: exactly same qualitative features as Fig. 2.

Unfortunately, in practice, there are no materials with such large anisotropy. Nevertheless, we can create an effective uniaxial meta-material with $\varepsilon_o = (\varepsilon_1 + \varepsilon_2)/2$ and $\varepsilon_e^{-1} = (\varepsilon_1^{-1} + \varepsilon_2^{-1})/2$, by layering two dielectrics with permittivities $\varepsilon_1, \varepsilon_2$ and the same thickness along the desired optical axis with a period smaller than the propagation wavelength $\lambda_{x0} = 2\pi/k_{x0}$ (Fig. 3(a)) [7, 37]. To get $\varepsilon_o = 6.45, \varepsilon_e = 1.845$, we have to use $\varepsilon_1 = 1$ (air) and $\varepsilon_2 = 11.9$, which is the permittivity of GaP at the frequency $\hbar\omega_0 \approx 0.237 \cdot 9.3\text{eV} = 2.2\text{eV}$, where GaP is also transparent (see SupInfo-C). As shown in Fig. 3(b), slicing a thin $z$-layer of this meta-material creates simply a grating on top of the uniform GaP layer. Using an analytical model for the material dispersion of GaP (see SupInfo Fig. S2(a)) [38], the Bloch modes of this grating meta-surface were calculated with COMSOL [39]. Our desired STDC ($D_{2,0} = D_{0,2} = D_{3,0} = 0$) is achieved also for this periodic structure, for period $a = 0.236\lambda_{x0}$ and optimized parameters close to those of the $xy$-uniform system with non-dispersive dielectric materials. This justifies the effective meta-material design approach and shows that dielectric material dispersion, although included here for accuracy, is a much weaker effect than the geometric dispersion control imposed by our mechanism in plasmonics. The periodic part of the modal magnetic field $\mathbf{H}$ at $(k_{x0}, 0, \omega_0)$ is shown in Fig. 3(c), and the $\omega(k_x, k_y)$ dispersion in Fig. 3(d) is super-flat and almost identical to Fig. 2(a). With $\lambda_0 = c\Gamma_0 = 2\pi c/\omega_0$ the free-space wavelength, this unique SPP STDC is accomplished for a very subwavelength mode, truly-plasmonic longitudinally ($\lambda_{x0}\mu_{GaP}/\lambda_0 = 0.52$) and vertically ($d_{\text{film}}\mu_{GaP}/\lambda_0 = 0.28$, where $d_{\text{film}}^2 a = \iint \mu_0 |\mathbf{H}|^2 \,dx\,dz/\max(\mu_0 |\mathbf{H}|^2))$, for very slow light at $v_{g0} = 0.004c$, and with quality factor due to the metal absorption $Q = \omega_0/2\Gamma_0 = 115$. 
To understand the benefits of STDC in wavepacket propagation, we compare the evolution of a pulse centered at \((k_{x0}, 0, \omega_0)\) for our meta-surface of Fig. 3 to a classic SPP on the interface between silver and an isotropic dielectric of \(\varepsilon = 10.405\), such that this SPP crosses \((k_{x0}, 0, \omega_0)\), with similar \(Q = 118\), but larger \(v_{g0} = 0.0293c\) and nonzero \(D_{2,0} = -0.0115\), \(D_{0,2} = 0.00443\) (note \(D_{y2} = -k_{x0}D_{0,2}/\omega_0 v_{g0} = -1\) is confirmed). Specifically, we consider an initial Gaussian pulse envelope \(|H(x,y,t)|^2 = \exp (-x^2/2\sigma_x^2 - y^2/2\sigma_y^2)\) with ultra-short widths \(\sigma_x = \sigma_y = \lambda_0/20 = 28nm\), and this green-light \((\lambda_0 = 560nm)\) pulse is shown in Fig. 4(a). Then we calculate numerically (details in SupInfo-E), at any time \(t\), the field, whose intensity is reduced due to both absorption and dispersion. In Figs. 4(b,c), it is shown for the classic SPP and our meta-surface at \(t = 36T_0\), chosen to correspond to a reduction of peak intensity to 10% for the latter. Impressively, this pulse maintains its shape for much longer, basically until extinguished by absorption (as shown in a SupInfo video for \(t \leq 100T_0\)), and can be thought of as a ‘plasmonic bullet’. In contrast, the classic-SPP system has led to significant broadening, both longitudinally and transversely, and much worse intensity reduction. The pulse-widths \(\sigma_x, \sigma_y\) and peak-intensity \(I_{pt}\) are calculated numerically (shown in SupInfo Fig. S2(b) for \(t \leq 100T_0\)) and fit respectively to an approximate time-dependence \(\sqrt{\sigma_x^2, \sigma_y^2}/\lambda_0 \approx 0.024, 0.013 \tau, I_{pt} \exp (2\pi \tau/Q) \approx 1/(1 + 0.12\tau + 0.014r^2 + 0.00013r^3)\), \(\tau \equiv t/T_0\) for the classic SPP and \(\approx [0.0064, 0.0040]\tau, \approx 1/(1 + 0.0085\tau + 3.7 \cdot 10^{-5}\tau^2)\) for the meta-surface. The latter broadening is 3.7 times smaller and would be substantially smaller for broader initial pulses (in the SupInfo-E, we calculate \(\sigma_x, \sigma_y\) also analytically, using a procedure similar to [40]). For the simple SPP, absorption decay \(\exp (-2\pi \tau/Q)\) is weaker than dispersion decay for \(t < 110T_0\) (e.g. at \(t = 36T_0\) in Fig. 4(b), 0.147 vs 0.005/0.147 = 0.034 respectively). Therefore, despite the notorious losses in plasmonics, dispersion can surprisingly be a much more detrimental effect than absorption, for short enough pulses. STDC can thus maintain large localized field intensities for longer time, which could be useful to prolong interactions of plasmonic bullets with comoving nanoparticles and for dynamic nanosensors.

The main advantage of linear STDC is that dispersionless propagation is pulse-shape independent. In Fig. 4(d-f), an initial green-light packet arbitrarily-shaped to spell “MiT” with subwavelength features is let propagate through the two compared structures. It is utterly unrecognizable at \(t = 36T_0\) on the isotropic SPP, but an almost-intact-shape ‘moving object’ on our STDC Ag/GaP meta-surface SPP, just attenuated from the silver absorption. The regime around super-collimation in hyperbolic materials has been termed ‘hyperlens’ and its relation to subwavelength imaging over short ranges discussed [7, 41, 42]. With our added temporal dispersion cancellation, one could now extend this discussion to realtime sub-diffraction short-range video imaging.

Figure 4: Propagating-wavepacket field intensities \(|H(x,y,t)|^2\) centered around \(x = v_{g0}t\) for initial ultra-short wavepackets: (a-c) a Gaussian pulse and (d-f) a packet spelling “MiT”; the colorbar is scaled on each plot to show the peak intensity. (a,d) Wavepacket at \(t = 0\) (initial), (b,e) at \(t = 36T_0\) for classic isotropic SPP, whose dispersion crosses the same \((k_{x0}, 0, \omega_0)\) point, and (c,f) for meta-surface of Fig. 3. In (e), peak intensity is relatively large due to superposition in one location of different parts of the heavily-dispersed wavepacket.
The unavoidable metal absorption loss is the major practical drawback of plasmonics. At least, since our proposed platform uses only one flat unpatterned metallic surface, it maintains the advantage of the lower expected losses associated with Dielectric-Loaded-SPPs [28] by avoiding boundary electronic scattering [26], present in common plasmonic meta-surfaces involving patterning of the metal. Furthermore, in the SupInfo-D we show the spectacular result that, if we used a semiconductor (Ga\textsubscript{i}In\textsubscript{i−x}P) [43] with direct bandgap slightly lower than the STDC frequency and if we could achieve the necessary population inversion [44, 45] (difficult via current injection for such subwavelength systems [46], but possible with optical pumping), the net modal loss-gain rate \( \Gamma (k_x, k_y) = -\text{Im} \{ \omega (k_x, k_y) \} \) could be designed to still always be positive, thus preventing ‘spasing’ at any frequency and ensuring stable propagation, but minimum and zero at the STDC point to achieve full loss-compensation \((\Gamma_0 = 0)\) with simultaneous loss-dispersion cancellation \((\partial \Gamma_0 / \partial k_x = 0)\) (and hence slightly reducing too the huge spontaneous emission \[47\]). This would signify true unprecedented ‘moving light-objects’.

Different metal/semiconductor combinations can also be used for operation at other visible frequencies, for example Ag/AlN for violet (\(\sim 400\text{nm}\)), Ag/ZnS for blue (\(\sim 440\text{nm}\)), Au/GaP for red (\(\sim 660\text{nm}\)), and lower frequencies could be reached by making the metal a thin film [13]. Just as in [3], by including additional layers, higher-order cancellations are possible, but there are limits to \( k_{x0}, \nu_{g0} \), for which they can be attained. Moreover, we confirmed our proposed effective transformation works not only for dispersion cancellation but also for the exotic dispersions in Fig. 1.

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