We develop a compact semi-analytical theoretical approach that can be applied to a variety of time varying dispersive materials and is particularly well suited to describe the thin film ITO experiments discussed above. We find that the continuous wave reflection and transmission coefficients can be replaced with equivalent operator expressions that are simple to evaluate numerically and act on the spectrum of the incident wave. Although distinct from this work, aspects of our operator-based approach (where e.g. the wave–vector is treated as an operator) appear in the recent extension of Mie theory given by Ptitcyn et al. [23]. We find that the eigenfunctions of these operators represent spectra of incident waves that are e.g. unchanged on reflection from a TVM. Furthermore when the eigenvalue of e.g. the reflection operator is zero or infinite, we have respectively the superior performance of this method in terms of both accuracy and efficiency.

For a static material, the electric current $j$ is linked to the electric field $E$ through the conductivity $\sigma(t - t')$ that represents the movement of charge in response to the past behaviour of the electric field and depends only on the time difference $t - t'$. When the material is explicitly time-dependent, due to e.g. a pump pulse [17] (at optical frequencies) or electronic modulation (at radio frequencies), the conductivity can be replaced with a
two–time function such that,
\[ j(t) = \int_{-\infty}^{\infty} dt' \sigma(t, t-t') E(t'). \] (1)

As in the static case, causality requires \( \sigma(t, t-t') = 0 \) when \( t' > t \). We understand \( \sigma(t, t-t') \) as the response at time \( t \) to the electric field at the earlier time \( t' \), where this response is modulated as a function of the observation time \( t \). We could also develop the same formalism by taking the first argument of \( \sigma \) as \( t' \) instead of \( t \). As the two times are related by \( t' = t - (t-t') \), our results can be applied to either form of Eq. (1), with only minor modifications. We could also equally develop the formalism in terms of the permittivity and/or permeability instead of the conductivity.

Performing a Fourier transform of (1), \( \tilde{j}(\omega) = \int dt \tilde{j}(t) \exp(\omega t) \), the frequency dependent current can be written as
\[ \tilde{j}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \mathcal{E}(\omega') \int_{-\infty}^{\infty} dt \sigma(t, t') \exp(i(\omega - \omega')t) \] (2)
\[ = \int_{-\infty}^{\infty} d\omega \mathcal{E}(\omega') \hat{\sigma}(-i\partial_\omega, \omega') \delta(\omega - \omega') \] (3)
\[ = \hat{\sigma}(-i\partial_\omega, \omega) \mathcal{E}(\omega) \]

where \( \hat{\sigma} \) is the operator obtained by replacing the first argument \( t' \) with the operator \(-i\partial_\omega\). In the final line of (3), all derivatives \( \partial_\omega \) within \( \hat{\sigma} \) must be ordered such that they appear to the left of all the frequency dependence of \( \hat{\sigma} \). This prescription is reminiscent of the anti–normal operator ordering adopted in quantum mechanics. To use Eq. (3) we write the operator as e.g. \( \hat{\sigma} = \sum_n a_n (-i\partial_\omega) b_n(\omega) \). The derivative \( \partial_\omega \) is numerically constructed as an \( N \times N \) matrix acting on \( N \) frequency points, via the finite difference approximation or a Fourier transform. The operators \( a_n \) are then evaluated as matrix valued functions, and \( b_n(\omega) \) is a diagonal matrix. This idea of using an operator valued function is similar to the exponential function of the Hamiltonian operator used as the time evolution operator in quantum mechanics.

In this work we assume the magnetic permeability is unity, and use the Drude model with a time-varying plasma frequency \( \omega_p \) (see Supplementary Information)
\[ \hat{\sigma}(-i\partial_\omega, \omega) = \omega_p^2 (-i\partial_\omega) \frac{i\eta_0}{\omega + i\gamma}, \] (4)
where we have imposed the aforementioned anti–normal ordering, and \( 1/\gamma \) is the collision time. Note that throughout this work we use the symbol \( \omega_0 = \omega_p(-\infty) \), i.e. the plasma frequency before the time variation.

The simplest application of Eq. (3) is where the medium is homogeneous, propagation is along \( x \), and the field is polarized such that \( \mathcal{H} = He_z \). In this case Maxwell’s equations become \( \partial_x^2 \mathcal{H} + \hat{K}^2 \mathcal{H} = 0 \), the solutions to which are
\[ \mathcal{H}(x, \omega) = \exp(\pm i\hat{K}x) H_0(0, \omega) \] (5)
where \( k_0 = \omega/c \) and \( \hat{K}_s \) is the matrix square root of \( \hat{K}^2 = i\eta_0 k_0 \sigma_0 + k_0^2 [27] \) and \( H(0, \omega) \) is the Fourier amplitude of the wave at \( x = 0 \), which we are free to choose. The quantity \( \eta_0 = \sqrt{\mu_0 / \epsilon_0} \) is the impedance of free space. Although the solutions (5) have the appearance of plane waves, the operator \( \exp(\pm i\hat{K}_p x) \) modifies the spectral content of the wave as the observation point \( x \) is changed, describing the reshaping of the pulse during propagation. Eq. (5) shows that those spectra \( H(0, \omega) \) that are eigenfunctions of \( \hat{K}_p \) with eigenvalue \( \lambda \) have a plane wave spatial dependence \( \exp(\pm i\lambda x) \) and retain the same spectrum during propagation, despite the time variation of the material parameters. Note also that Eq. (5) is similar to the aforementioned time evolution operator in quantum mechanics, where a state \( \ket{\psi} \) evolves in time as \( \ket{\psi(t)} = \exp(-iHt/\hbar) \ket{\psi(0)} \), where \( H \) is the Hamiltonian operator.

Fresnel coefficients for a dispersive, time–varying interface: Consider a pulse incident from vacuum onto a dispersive half–space. For further details).

Assuming incidence in the \( x-y \) plane with in–plane wave–vector \( k_\parallel \), TE polarized waves have an electric field \( \mathcal{E} = E e_z \) obeying the operator Helmholtz equation \( \partial_x^2 E + \hat{K}^2 E = 0 \) where \( \hat{K}_s^2 = i\eta_0 k_0 \sigma_0 + k_0^2 - k_\parallel^2 \). Inside the TVM \((x > 0)\) the solution is given by Eq. (3). \( E(x > 0, \omega) = \exp(i\hat{K}_p x)C_s(\omega) \). Meanwhile, on the entrance side the field is a sum of plane waves for each frequency \( E(x < 0, \omega) = A_s(\omega) \exp(ik_\parallel x) + B_s(\omega) \exp(-ik_\parallel x) \) where \( k_\parallel = [k_0^2 - k_\parallel^2]^{1/2} \). The spatial boundary conditions are the same as for static media, with both electric \( E \) and magnetic \( \eta_0 H_y = ik_\parallel E \) fields continuous across \( x = 0 \). Substituting the forms of the fields in the respective regions leads to the following reflection and transmission operators
\[ \hat{r}_s = (1 - \hat{Z}_s) (1 + \hat{Z}_s)^{-1} \]
\[ \hat{t}_s = 2 (1 + \hat{Z}_s)^{-1} \] (6)
where \( \hat{Z}_s = k_\parallel^{-1} \hat{K}_s \), \( B_s = \hat{r}_s A_s \) and \( C_s = \hat{t}_s A_s \). Eqns. (6) are the TE Fresnel coefficients [28], with an operator replacing the usual expression for the wave–vector in the material.
two impedance operators are then simply related by

\[ \hat{Z}_p = \hat{K}_s^{-1}k_0 = \hat{Z}_s^{-1}, \]

making the reflection operators (6–7) differ by a minus sign \( \hat{r}_p = -\hat{r}_p \) as expected for the two polarizations at normal incidence [29].

As discussed above, it is again interesting to examine the eigenvalues and eigenvectors of the reflection and transmission operators. These reveal that there are pulse spectra (‘eigenpulses’) that retain an identical spectrum after interaction with the TVM (bar an overall multiplicative factor). Alternatively, taking a singular value decomposition of the reflection and transmission operators, we can find pulse spectra that are scaled by a set level (the singular value), but have a different spectral content after interaction with the material. We can see from expressions (6–7) that, in the case of a single interface, the eigenvalues are the eigenfunctions of the impedance operators \( \hat{Z}_{s,p} \) and thus both transmitted and reflected spectra are unchanged. Fig. 2 shows a comparison between the reflection of a Gaussian pulse and an eigenpulse from a TVM (plots show the incident field just before the interface). While the Gaussian pulse is significantly broadened and reshaped by the interaction with the TVM, the eigenpulse reduces in frequency in tandem with the plasma frequency, retaining an identical spectrum upon reflection. In this case (modulus of eigenvalue \( |\tau| = 1 \)), the eigenpulse is also entirely reflected by the medium, as if it were a mirror.

In addition, the scattering operators can also exhibit poles. For example in Eqns. (6–7) these poles occur where \( \text{det}(1 + \hat{Z}_{s,p}) = 0 \). The vectors in the null-space of \( (1 + \hat{Z}_{s,p}) \) then represent non–time harmonic modes that are—in this case—confined to the interface of the material. In the Supplementary Information we find the surface plasmon–like eigenpulses that are confined to the interface of a TVM.

**Time varying layer:** We can straightforwardly extend this approach to any multilayer and any simple geometry (e.g., a spherical, cylindrical, or ellipsoidal object) that admits an analytic solution to Maxwell’s equations in the static case. Broadly speaking, the results for the scattering operators will have an identical form but with an operator replacing the material parameters. To illustrate this in a non–trivial case we calculate the reflection and transmission operators for a slab of thickness \( d \), which is relevant to the experiments reported in [9, 10, 18, 19, 21].

Assuming TM polarization, the magnetic field within the layer is taken to be of the form

\[ H(0 < x < d, \omega) = e^{i\hat{K}_s x}C_p(\omega) + e^{-i\hat{K}_s x}D_p(\omega) \]

with the field in the external regions equal to \( H(x < 0) = \exp(ik_0 x)A_p(\omega) + \exp(-ik_0 x)B_p(\omega) \) and \( H(x > d) = \exp(ik_0(x - d))F_p(\omega) \). Imposing the same boundary conditions described above we obtain the reflection and
FIG. 2. Reflection of an eigenpulse from a time varying dispersive half–space. (a) We compare the reflection of two different incident pulses; a Gaussian pulse (upper curve), and an eigenpulse (lower curve) computed from the reflection operator (7), with eigenvalue $|r| = 1.00$ (the zero level is displaced to aid visualization). (b) Time variation of $N(t) = \omega_p^2(t)/\omega_0^2$ in Eq. (4). (c–d) Magnitude of incident and reflected Fourier spectra computed via a numerical integration of Maxwell’s equations (See Supplementary Information), for an incident (c) eigenpulse, and (d) Gaussian pulse.

transmission coefficients for the slab

$$\hat{r}_{\text{slab}} = \left[ \hat{A}_+ e^{i K_p d} \hat{A}_- - \hat{A}_- e^{-i K_p d} \hat{A}_+ \right] \times \left[ \hat{A}_- e^{-i K_p d} \hat{A}_- - \hat{A}_+ e^{i K_p d} \hat{A}_+ \right]^{-1}$$

(9)

and

$$\hat{t}_{\text{slab}} = 4\hat{Z}_p \left[ \hat{A}_+ e^{-i K_p d} \hat{A}_- - \hat{A}_- e^{i K_p d} \hat{A}_+ \right]^{-1}$$

(10)

where $\hat{A}_\pm = 1 \pm \hat{Z}_p$. Expressions (9) and (10) reduce to the familiar reflection and transmission coefficients of a dielectric slab [28] when the operators are replaced with their scalar counterparts. When $d = 0$ the reflection operator (9) is identically zero, and the transmission operator (10) becomes the identity, as they should.

In Fig. 3 we give a comparison between results obtained using COMSOL Multiphysics (see SI), and calculations made using the reflection and transmission operators (9,10). We plot the normalized transmitted spectra as a function of pulse delay time $\Delta t$. As shown in the lower panel of this figure, there is excellent agreement between the finite element calculation and our operator approach. Additional comparisons to an adiabatic multiple-timescale approach used to model past experiments [18,21] are also available in the SI. Importantly, these tests demonstrate the advantage of this method for the efficient modelling of structures that feature extremely subwavelength layers, circumventing the need for expensive numerical calculations.

Summary and Conclusions: We have developed a compact theoretical approach for treating the problem of scattering from dispersive TVM. We have shown that our analytic expressions match full wave numerical simulations well. Although the theory is formally similar to the case of static materials, the TVM parameters are given in terms of operators that depend on both the frequency and frequency derivatives, which must be carefully ordered. The advantage of our theory is that it is semi–analytical, allowing us to give explicit operator expressions for scattering coefficients from the TVM, and thus determine conditions for e.g. incoming modes that are bound, not reflected, or completely reflected by the material. We have numerically constructed these operators and found the ‘eigenpulses’ of a time–modulated Drude half–space, numerically verifying that there are input pulse spectra that e.g. reflect as if the TVM was a static mirror. This approach may be readily extended to other areas of wave physics such as pressure acoustics [30] and elasticity and may be of interest to those working on TVM as well as multiple scattering, where our reflectionless eigenpulses are analogous to the con-
cept of open scattering channels in disordered media (see e.g. [31]).

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