Optimizing Light Storage in Scattering Media with the Dwell-Time Operator

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We prove that optimal control of light energy storage in disordered media can be reached by wave front shaping. For this purpose, we build an operator for dwell times from the scattering matrix and characterize its full eigenvalue distribution both numerically and analytically in the diffusive regime, where the thickness L of the medium is much larger than the mean free path ℓ . We show that the distribution has a finite support with a maximal dwell time larger than the most likely value by a factor $(L/\ell)^2 \gg 1$. This reveals that the highest dwell-time eigenstates deposit more energy than the open channels of the medium. Finally, we show that the dwell-time operator can be used to store energy in resonant targets buried in complex media, without any need for guide stars.

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Recent developments in wave front shaping protocols have allowed spectacular demonstrations of light manipulation in complex media [1,2], such as noninvasive imaging in biological tissues [3,4], focusing [5] or enhanced power delivery [6–8] behind opaque media, or focusing [9–11] and enhanced absorption [12] inside scattering materials. The large number of degrees of freedom supported by disordered systems has also been proposed as a resource for imaging with high resolution [13,14], controlling the strength of light-matter interaction [15–17], or performing optically complex and reconfigurable operations [18].

In this context, great attention has been given to the properties of the transmission matrix t [19–23]. This matrix admits a significant fraction of singular states, called open channels, responsible for complete destructive (constructive) interference in reflection (transmission), even if the medium is opaque on average [24,25]. The intensity map inside the medium resulting from the propagation of open channels has also been elucidated [26], revealing a bellshaped profile along the propagation direction very different from the linear decay obtained with plane wave illumination [27]. This property makes open channels good candidates for significantly enhancing energy deep inside disordered media [28-30]. However, these states are, by construction, those maximizing the output flux (they are eigenstates of the operator $t^{\dagger}t$ associated with the largest eigenvalues) and not necessarily the stored energy. The transverse localization of open channels, discovered recently [31], also supports the idea that they are not necessarily the ones optimizing energy storage.

In this Letter, we explicitly build an operator for dwell time (or stored energy) in complex media illuminated with monochromatic light. Its expression can be obtained directly from the scattering matrix of the medium and the dispersion properties of the surroundings. First, we

show that the dwell-time (DT) operator is not strictly identical to the Wigner-Smith matrix, introduced historically to characterize the duration of a scattering process [32], by identifying a contribution resulting from the interference between the incident and scattered fields, similar to that predicted in 1D for electrons [33]. Second, we study its eigenvalue distribution $p(\tau)$ for wave propagating through a disordered slab of thickness $L \gg \ell$, where ℓ is the light mean free path. We find that for nonresonant scattering $p(\tau)$ is parametrized by two timescales only: the scattering time $\tau_s \sim \ell/c$ (c being the speed of light in vacuum), as well as the mean time $\langle \tau \rangle \sim L/c$, which is known to be remarkably independent of the disorder strength [34,35]. It also exhibits a dominant peak at $\tau \sim \tau_s$ and has a finite support with a maximal DT eigenvalue $\tau^{\text{max}} \sim \langle \tau \rangle^2 / \tau_s$. This last result implies that the maximal energy that can be stored in a disordered medium by wave front shaping with fixed input power ϕ^{in} scales as $U^{\rm max} \sim \phi^{\rm in} \tau_{\rm Th}$, where $\tau_{\rm Th}$ is the Thouless time. Finally, we show that the DT operator is a powerful tool to selectively deposit energy on local resonant targets embedded in a given realization of a complex medium.

Let us start with the construction of the DT operator. For clarity, we restrict the present discussion to the propagation of scalar waves in nonresonant and nonabsorbing materials, described by the equation $[\nabla^2 + k^2 \epsilon(\mathbf{r})] \psi(\mathbf{r}) = 0$. Here, $\epsilon(\mathbf{r})$ is the (real) dielectric function, and ψ is the complex amplitude of the monochromatic wave with frequency $\omega = ck$. The quantity to maximize is the electromagnetic energy $U = \epsilon_0 \int_{\mathcal{V}} d\mathbf{r} \epsilon(\mathbf{r}) |\psi(\mathbf{r})|^2/2$, where \mathcal{V} is the volume occupied by the disordered slab. From the wave equation, we readily obtain the relation

$$\epsilon(\mathbf{r})|\psi(\mathbf{r})|^2 = \frac{c^2}{2\omega} \nabla \cdot (\partial_\omega \psi \nabla \psi^* - \psi^* \partial_\omega \nabla \psi), \quad (1)$$

which allows us to express the energy as the surface integral

$$U = \frac{\epsilon_0 c^2}{4\omega} \int_{\mathcal{S}} d\mathbf{r} \, \mathbf{n} \cdot (\partial_{\omega} \psi \nabla \psi^* - \psi^* \partial_{\omega} \nabla \psi), \qquad (2)$$

where S denotes the input and output surfaces of the slab and **n** is the outward normal on them. For a Schrödinger wave ψ , a similar relation holds [32], which involves the probability $\int_{\mathcal{V}} d\mathbf{r} |\psi(\mathbf{r})|^2$ instead of U. The former is independent of the explored potential, contrary to U that depends on $\epsilon(\mathbf{r})$. Equation (2) is the scalar version of the energy theorem known for electromagnetic fields [36]. Next, we express the field ψ on each surface in terms of the incident field ψ^{in} and the reflection and transmission matrices (including evanescent channels). Here, we consider a disordered slab embedded in a multimode waveguide supporting N propagating channels, so that the reflection and transmission matrices restricted to this channel basis, denoted r and t, respectively, are of size $N \times N$. Relegating the technical derivation to the Supplemental Material [37], we find that the stored energy can be expressed as

$$U = \phi^{\rm in} \langle \psi^{\rm in} | Q_d | \psi^{\rm in} \rangle, \tag{3}$$

where the DT operator Q_d is a sum of three contributions, with clear and distinct meanings discussed below,

$$Q_d = Q + Q_e + Q_i. (4)$$

Depending on the physical situation of interest (choice of ψ^{in} , size and scattering strength of the medium), each of these terms can produce the dominant contribution to U. We discuss their expressions below for an incident field without evanescent component and injected from one side of the slab only, which corresponds to the most common experimental situation. More general expressions are given in the Supplemental Material [37].

The first term on the right-hand side in Eq. (4) is the wellknown Wigner-Smith matrix $Q = -i(t^{\dagger}\partial_{\omega}t + r^{\dagger}\partial_{\omega}r)$, which characterizes the duration of the scattering process for quasimonochromatic signals [2,32,45]. For one-dimensional free space propagation, it would reduce to Q = L/c. The utility of this operator for controlling wave propagation in multimode fibers and disordered media has been demonstrated in recent years [10,46–50]. The second term captures scattering contributions into evanescent channels. It reads $Q_e = (t_e^{\dagger} D^e t_e + r_e^{\dagger} D^e r_e)/2$, where r_e and t_e are the reflection and transmission matrices into evanescent channels of the waveguide [51]. The matrix D^e is diagonal, with elements $D_{\alpha\beta}^e = (\partial_\omega \kappa_\alpha / \kappa_\alpha) \delta_{\alpha\beta}$, where $\kappa_\alpha = \sqrt{q_\alpha^2 - k^2}$ is the inverse decay length of the evanescent channel α $(q_{\alpha} = \alpha \pi/W \text{ in a 2D waveguide of width } W)$. The contribution of Q_e cannot be neglected close to the onset of a new propagating mode of the waveguide [37,52,53]. The important impact of evanescent channels on dwell times has also been revealed recently in the case of scattering by subwavelength particles [54]. However, the contribution of Q_e to the distribution $p(\tau)$ studied below turns out to be negligible for all frequencies except a discrete set (see Supplemental Material [37]) and will not be discussed further.

The third term in the decomposition (4) describes a qualitatively different contribution, due to the interference between the incident and reflected propagating fields. Since the total field is $\psi \sim \psi^{\text{in}} + r\psi^{\text{in}}$ on the front surface and $\psi \sim t\psi^{\text{in}}$ on the back surface, the field products in Eq. (2) involve cross terms in reflection only. The associated matrix reads $Q_i = -i(Dr - r^{\dagger}D)/2$, where D has elements $D_{\alpha\beta}=(\partial_{\omega}k_{\alpha}/k_{\alpha})\delta_{\alpha\beta}, \quad \text{and} \quad k_{\alpha}=\sqrt{k^2-q_{\alpha}^2}.$ In Supplemental Material [37], we provide an alternative proof of Eq. (4) based on the continuity equation that highlights the close connection between Q_i and the interference term between the incident and scattered field at the origin of the optical theorem [55]. The contribution of Q_i becomes appreciable for input states with large reflection in directions nearly parallel to the sample surface. For this reason, it cannot be neglected in strongly scattering media. In particular, it contributes substantially to the lower part of the DT eigenvalue distribution in the regime $L \gg \ell$ (see Supplemental Material [37]).

Equation (4) generalizes to arbitrary scattering media and arbitrary dimension (arbitrary N) the relation established for electrons [33] or electromagnetic waves [56] interacting with a simple barrier (for which Q_e is zero), in the case N=1. For electrons, the trace of the matrix Q_i is known as a correction to the Friedel sum rule [45], which relates the density of states (\sim Tr Q_d) to the Wigner time delay (\sim TrQ). In its operator form, the difference between Q_d and Q has also been pointed out in Refs. [2,57,58], but not expressed in the explicit and computationally useful expansion given by Eq. (4).

To characterize the properties of the matrix Q_d , we performed extensive numerical simulations of scalar wave propagation through two-dimensional disordered slabs placed in a multimode waveguide using the recursive Green's function method [37] and computed Q_d as defined in Eq. (4). The eigenvalue distribution $p(\tau)$ of Q_d is represented in Fig. 1 for three values of the disorder strength $1/k\ell$, in the regime of diffusive transport $kL \gg$ $k\ell \gg 1$ and large conductance $g = N\ell/L \gg 1$. We find that $p(\tau)$ has a pronounced peak that shifts toward small time as $k\ell$ decreases. This illustrates the fact that most of the light experiences a few scattering events before being reflected after a time $\sim \ell/c$. On the other hand, a close look at the largest eigenvalues (see inset) reveals that $p(\tau)$ is bounded, with an upper edge τ^{max} that increases with the disorder strength. This effect is triggered by light crossing the sample by diffusion, a process that takes more time when the mean free path is reduced since the number of scattering events is increased.

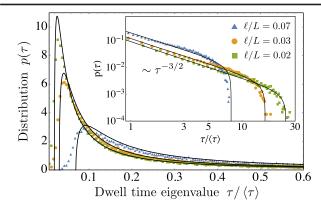


FIG. 1. Eigenvalue distribution of the dwell-time operator Q_d , evaluated for a disordered slab (length kL=300) embedded in a multimode waveguide (N=287). Analytical predictions (solid lines) are compared with numerical results (dots) obtained from the solution of the wave equation for 128 realizations of the slab, with dielectric function $\epsilon(\mathbf{r})=n_1^2+\delta\epsilon(\mathbf{r}); n_1=1.5$ and $\delta\epsilon(\mathbf{r})$ is uniformly distributed in [-a,a]. Results for three values of a are represented, corresponding to $k\ell=21.4, 9.3, 5.8$.

To support the previous observations, we developed an analytical model for $p(\tau)$. Since Q_d and Q have similar spectra, with differences observed at small times only (see Supplemental Material [37]), we work with Q in the theoretical development. First, following Ref. [59], we use the simple relation that links the scattering matrix $S = \binom{r}{t}$ in absence of absorption, to the scattering matrix $S^{a}(\omega) = S(\omega + i/2\tau_{a})$ with uniform absorption, where τ_{a} is the absorption time. A first-order expansion gives $Q(\omega) \simeq \tau_a [1 - S^a(\omega)^{\dagger} S^a(\omega)]$ for $\omega \tau_a \gg 1$. The advantage of this relation lies in the fact that the joint probability distribution (JPD) of the eigenvalues of the operator $S^{a}(\omega)^{\dagger}S^{a}(\omega)$ is known, for disordered media excited from one side, in the limit $L \to \infty$ [60,61]. Denoting by τ_n the eigenvalues of Q, the JPD of the decay rates $\gamma_n = 1/\tau_n$ takes the form of the Gibbs distribution $p(\{\gamma_n\}) \sim e^{-\mathcal{H}}$, with

$$\mathcal{H} = 2(N+1)\tau_s \sum_{n=1}^{N} \gamma_n - \sum_{n < m} \ln |\gamma_n - \gamma_m|, \qquad (5)$$

where τ_s is the scattering time. In 2D, for nonresonant scattering, it has to be defined as $\tau_s = (\pi/2) \ell/v$, where v = c/n is the phase velocity (that coincides with the energy velocity), n being the effective refractive index (see Supplemental Material [37]). The Laguerre distribution defined by Eq. (5) implies that Q^{-1} behaves as a Wishart matrix in a disordered medium, a property which is also true in a chaotic cavity [62,63]. In the first case, Q^{-1} has the same JPD as the matrix HH^{\dagger}/τ_s , where H is an $N\times(N+1)$ Gaussian random matrix, while in the second case, Q^{-1} has the JPD of $HH^{\dagger}/\langle \tau \rangle$, where H is of size $N\times(2N+1)$.

The result in Eq. (5) was obtained for infinite-size systems. In this limit, the marginal distribution $p(\tau)$ depends on τ_s only, with infinite $\langle \tau \rangle = \langle \sum_{n=1}^N \tau_n \rangle / N$. In nonresonant systems of finite size L, it is known that $\langle \tau \rangle$ scales as $\sim L/c$, both in the quasiballistic and diffusive regimes [34,35]; in 2D, $\langle \tau \rangle = (\pi/2)L/v$. To restore a finite mean time, we make the ansatz that $\mathcal H$ is still well approximated by expression (5) in the regime $g \gg 1$ and search for $p(\tau)$ that minimizes $\mathcal H$ under the constraint $\int d\tau p(\tau) = \langle \tau \rangle$. We find (see Supplemental Material [37])

$$p(\tau) \simeq \frac{2\tau_s}{\pi \tau^2} \sqrt{\left(\alpha \frac{\tau}{\tau_s} - 1\right) \left(1 - \beta \frac{\tau}{\tau_s}\right)} \left(1 + \gamma \frac{\tau}{\tau_s}\right), \quad (6)$$

where α , β , and γ obey coupled equations depending on the single parameter $b \equiv \langle \tau \rangle / \tau_s = L/\ell$. At large optical thickness $(b \gg 1)$, they reduce to $\alpha \simeq 1$, $\beta \simeq 9/[4b(b-4)]$, and $\gamma \simeq 2\beta$. The distribution (6) is maximum for $\tau \simeq 4\tau_s/3$ and has a finite support $[\tau^{\min}, \tau^{\max}]$, with $\tau^{\min} \simeq \tau_s$ and

$$\tau^{\text{max}} \simeq \frac{4}{9} \langle \tau \rangle \left(\frac{\langle \tau \rangle}{\tau_s} - 4 \right),$$
(7)

which scales as the Thouless time $\tau_{\rm Th}$. This indicates that the existence of states with dwell time parametrically larger than $\tau_{\rm Th}$ is unlikely in the limit of large conductance considered here. Note that at moderate g, rare disorder realizations may support prelocalized states, responsible for a log-normal profile of $p(\tau)$ for $\tau \gtrsim \tau_{\rm Th}$ [64,65]. Our theoretical results are in excellent agreement with the simulations, without adjustable parameter, as shown in Fig. 1. The distribution $p(\tau)$ is notably different from the distribution of the spectral derivative of a speckle phase pattern [66], exhibiting in particular a power law $p(\tau) \sim \tau^{-3/2}$ for $\langle \tau \rangle \lesssim \tau \lesssim \tau^{\rm max}$. This is a hallmark of diffusion, also observed in numerical simulation of the 2D kicked rotor dynamics [65].

Our analysis reveals the existence of a finite maximal eigenvalue τ^{max} of Q^d . According to Eq. (3), the corresponding eigenstate $|\psi^{\max}\rangle$ should yield the largest amount U^{max} of stored energy. To check this prediction, we compared the intensity pattern produced inside the slab by $|\psi^{\rm max}\rangle$ with that resulting from the propagation of other remarkable wave fronts, such as the most open channel $|\psi^{\text{oc}}\rangle$ (the eigenstate of $t^{\dagger}t$ associated with the largest transmission eigenvalue $T \simeq 1$). Representative results are shown in Fig. 2(a). Contrary to the intensity profile created by the first mode of the waveguide (which behaves as a plane wave), both $|\psi^{oc}\rangle$ and $|\psi^{max}\rangle$ give rise to a concentration of energy deep inside the medium. In addition, the intensity pattern due to $|\psi^{\max}\rangle$ is significantly larger than that due to $|\psi^{oc}\rangle$, when integrated over the transverse dimension y. This clearly shows that $U^{\text{max}} > U^{\text{oc}}$.

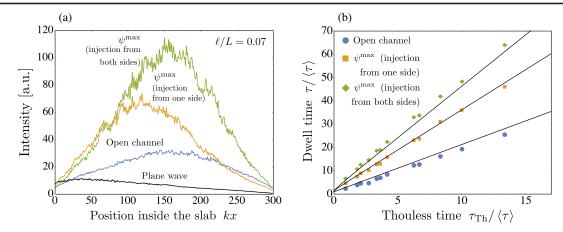


FIG. 2. (a) Intensity profiles inside a disordered slab (integrated over the transverse dimension) resulting from the propagation of different input states $\psi^{\rm in}$: the first mode of the waveguide (similar to a plane wave), the most open channel, and the eigenstate $\psi^{\rm max}$ of Q_d associated with the largest eigenvalue $\tau^{\rm max}$. (b) Dwell times $\tau = \langle \psi^{\rm in} | Q_d | \psi^{\rm in} \rangle$, averaged over 128 configurations (dots), corresponding to the different states $\psi^{\rm in}$ shown in (a) versus the Thouless time $\tau_{\rm Th} = L^2/\pi^2 D_B = 2L^2/\pi^2 \ell v$. All times are normalized by $\langle \tau \rangle = (\pi/2)L/v$. Solid lines correspond to analytical predictions (see text).

Using the analytical result (6), the ratio U^{\max}/U^{oc} can be evaluated precisely. In Ref. [26], it was shown that the intensity profile created by $|\psi^{\text{oc}}\rangle$ in 2D is $I^{\text{oc}}(x) = \int dy |\psi^{\text{oc}}(x,y)|^2 = (\pi/2)[1+(\pi/2)(L/\ell)x(L-x)/L^2]/k$, where x is the direction perpendicular to the slab. After integration over x, we obtain $U^{\text{oc}}=\phi^{\text{in}}\tau^{\text{oc}}$, with

$$\tau^{\rm oc} \simeq \frac{\pi}{12} \langle \tau \rangle \left(\frac{\langle \tau \rangle}{\tau_s} + \frac{12}{\pi} \right).$$
(8)

Hence, U^{oc} grows quadratically with the sample thickness L. It is much larger than the energy U^{pw} deposited by a plane wave that grows linearly with $L[I^{pw}(x) \sim (L-x)/$ (kL) gives $U^{\text{pw}} \sim \langle \tau \rangle \phi^{\text{in}}$, but it is smaller than U^{max} . Indeed, Eqs. (7) and (8) give $U^{\text{max}}/U^{\text{oc}} \simeq 16/3\pi > 1$. These predictions are confirmed by the results of numerical simulations presented in Fig. 2(b), where $\tau^{\rm max} = U^{\rm max}/\phi^{\rm in}$ and $\tau^{\rm oc} = U^{\rm oc}/\phi^{\rm in}$ are shown to be both larger than the Thouless time $\tau_{\rm Th} = L^2/(\pi^2 D_B)$, which is the longest mode lifetime of the diffusion equation. In 2D, the light diffusion constant is $D_B = \ell v/2$, so that $\tau^{\rm max} \simeq (\pi^3/9) \tau^{\rm Th}$ and $\tau^{\rm oc} \simeq (\pi^4/48) \tau^{\rm Th}$ for $L \gg \ell$. In Fig. 2(b), we also show that the ratio $\tau^{\text{max}}/\tau^{\text{Th}}$ can be increased by injecting light from both sides of the sample. In this case, the average intensity profile $I^{\max}(x)$ presents a mirror symmetry with respect to the middle of the slab x = L/2, as imposed by statistical invariance [see Fig. 2(a)]. Inspired by the microscopic approach developed in Ref. [67], we could establish (see Supplemental Material [37]) an expression for τ^{max} in this situation, which reads

$$\tau^{\text{max}} \simeq \tau_s \left[\zeta \left(\frac{\langle \tau \rangle^2}{\tau_s^2} + 4 \frac{\langle \tau \rangle}{\tau_s} - 4 \right) - 1 \right]$$
(9)

at large optical thickness, where $\zeta \simeq 0.57$ is the solution of a transcendental equation. This prediction also agrees with numerical simulations [see Fig. 2(b)].

We have discussed the properties of Q_d in statistically homogeneous nonresonant disordered materials and demonstrated quantitatively superior (yet qualitatively similar) performances of the largest DT eigenstates for energy storage, compared to open channels. However, Q_d is specifically constructed to optimize the quality factor of an arbitrary complex structure. This concept is radically different from the monochromatic scattering properties captured by $t^{\dagger}t$ or $r^{\dagger}r$. To illustrate this last point, let us consider a set of small absorbers buried in an otherwise nonabsorbing disordered medium. One of them, resonant at frequency ω_0 with quality factor $\omega_0/\Gamma \gg 1$, is described by the Lorentzian dielectric function $\epsilon = 1 + \omega_0^2 \mathcal{F}/(\omega_0^2 - \omega^2 - i\omega\Gamma)$. Our goal is to compare the performance of the absorption matrix A = $1 - t^{\dagger}t - r^{\dagger}r$ and the DT matrix in terms of focusing. In the case of an arbitrary complex dielectric function, the matrix Q_d defined in Eq. (4) becomes non-Hermitian. By generalizing relation (2) to this situation (see Supplemental Material [37]), we established that the energy inside the medium can be tuned by considering the eigenstates of the Hermitian part of Q_d defined as $Q_d^H = (Q_d + Q_d^{\dagger})/2$. The effect of the absorbing resonators on the spectrum of Q_d^H is discussed in the Supplemental Material [37]. We show in Fig. 3 the intensity patterns calculated inside the medium at the resonance frequency ω_0 and resulting from the propagation of the states $|\psi^a\rangle$, associated with the largest absorption eigenvalue, and $|\psi^H\rangle$, associated with the extremal eigenvalue of Q_d^H . We clearly see that $|\psi^a\rangle$ deposits energy indistinctly on all absorbers, whereas

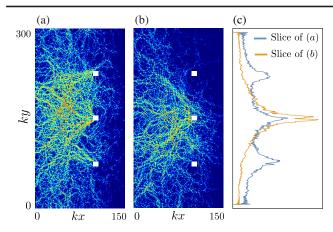


FIG. 3. (a), (b) Typical intensity patterns I(x,y) resulting from the propagation of the states associated with the maximal eigenvalue of the absorption operator (a) and the extremal eigenvalue of Q_d^H (b), in a disordered medium (kL=150, $k\ell=21.4$) containing three absorbers placed at depth kx=100 (white squares). Only the second absorber is resonant ($\mathcal{F}=10^{-4}$, $\omega_0/\Gamma=10^4$). (c) Slices $I(y)=\int_{80/k}^{95/k} dx I(x,y)$ of the patterns (a) and (b), averaged over 32 configurations.

 $|\psi^H\rangle$ focuses specifically on the resonant scatterer that will induce the largest dwell time.

In summary, we have introduced a general setting for tuning light storage in a complex medium based on the dwell-time operator Q_d . We showed that the distribution of its eigenvalues takes the universal form (6) in the diffusive regime. We demonstrated that the energy stored inside the medium can be increased by more than 100% by using eigenstates of Q_d , instead of open channels. Finally, we established that Q_d can be used to address hidden resonant targets without any need for guide stars, thus providing a powerful tool for wave front shaping experiments.

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Note added.—It was recently brought to our attention that a decomposition of Q_d for electrons similar to Eq. (4) is contained in Ref. [68]. We thank S. Rotter for pointing this out.

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- [37] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.123.243901 for (i) the proof of formulas (3) and (4) and their generalization to arbitrary dielectric function, (ii) details about numerical simulations, (iii) a study of the contributions of Q_e and Q_i to Q_d , (iv) the proof of the analytical predictions given in Eqs. (6) and (7), and (v) a study of $p(\tau)$ for light injected from both sides of the sample and the proof of Eq. (9). Supplementary Material includes Refs. [38–44].
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