

# Coherent control of photocurrent in a disordered photovoltaic system

S. F. Liew<sup>1,†</sup>, S. M. Popoff<sup>1,2,†</sup>, S. W. Sheehan<sup>3</sup>, A. Goetschy<sup>1,4</sup>, C. A. Schmuttenmaer<sup>3</sup>, A. D. Stone<sup>1</sup> and H. Cao<sup>1\*</sup>

<sup>1</sup> Department of Applied Physics, Yale University, New Haven, CT 06520, USA

<sup>2</sup> CNRS LTCl, Telecom ParisTech, 46 rue Barrault, 75013 Paris, France

<sup>3</sup> Department of Chemistry, Yale University, New Haven, CT 06520, USA

<sup>4</sup> ESPCI ParisTech, PSL Research University, Institut Langevin, 1 rue Jussieu, 75005 Paris, France

<sup>†</sup> These two people contribute equally to this work. \*hui.cao@yale.edu

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**Optical absorption is commonly considered an intrinsic property of a medium, independent of the details of the illumination source. However, for a spatially coherent illumination of a disordered medium, interference effects can modify the spatial distribution of light inside the medium, allowing the global or local absorption to be tuned by adjusting an input beam's spatial characteristics. Here we demonstrate the coherent control of absorption by using the wavefront shaping technique to modulate photocurrent in a dye-sensitized solar cell. The incident wavefront of a laser beam is optimized to concentrate light near the front side of the porous photoanode where the collection efficiency of photo-generated electrons is highest. Destructive interference reduces light leakage through open boundaries, facilitating light absorption and conversion to current. This technique provides a powerful tool for probing and controlling photochemical processes inside nominally opaque media by manipulating the internal spatial distribution of light intensity.**

The absorption properties of a medium do not depend solely on the medium's bulk attributes but also on the coherence and spatial distribution of the input light. One recent example confirming this is the demonstration of nearly perfect absorption of light in a silicon wafer by tuning the relative phase of two counter-propagating incident beams [1]. This phenomenon, known as coherent perfect absorption (CPA), is the time-reversed analog of laser emission [2]. Similar to lasing, this effect is narrow-band and requires tuning of both the frequency of the input light and absorptivity of the material to achieve 100% absorption [3–5]. A related but distinct approach is to take advantage of the large number of degrees of freedom available for multiple scattering in a lossy environment to vary the absorption at any frequency. It has been shown theoretically [6, 7] that a suitable coherent superposition of input fields significantly enhances the coupling of light into the entire scattering medium or into local absorbing regions, thus increasing the absorption either globally or locally. This effect is termed coherently enhanced absorption (CEA) [6].

In the past decade, advances in wavefront shaping (WFS) techniques using spatial light modulators (SLMs) have enabled the generation of arbitrary wavefronts at the input of an optical system. It not only drives the development of optical imaging techniques through scattering media such as biological tissue and fog [8–11], but also makes a random medium

function as an optical element such as a lens [12–14], a phase plate [15, 16] or a spectral filter [17, 18]. A particularly dramatic form of control is to enhance the total transmission through a lossless opaque medium; this is more challenging than simple focusing or beam steering because it involves long range correlations of the input fields, but recently significant progress has been made toward the goal via WFS of the incident laser beam [7, 19–22].

WFS can enable CEA by delivering light deeper into a disordered medium, allowing weak bulk absorptivity to have a much larger effect than it would if most of the flux were reflected diffusively. However, the concept of coherent control of the interaction of light with a complex medium is more general and flexible than simply enhancing uniform absorption. We demonstrate here for the first time that WFS can be used to coherently control absorption in a space-dependent manner, both to enhance and suppress local absorption in a multiple scattering medium. For certain applications, targeted absorption of light in local regions of a random medium is preferred. For example, we study here a dye-sensitized solar cell (DSSC) that utilizes a highly scattering layer of dye-sensitized TiO<sub>2</sub> nanoparticles. Its photocurrent increases if light is preferentially absorbed near the front side of the TiO<sub>2</sub> layer, where the collection efficiency of photo-generated electrons is higher. Enhancing this local absorption close to an open boundary is potentially more challenging than increasing global absorption because light can easily leak out of the medium.

In this paper, we enhance or suppress the short-circuit current of a DSSC by tailoring the incident light's wavefront with a genetic optimization algorithm. When the incident beam is optimized to concentrate light near the front portion of the scattering TiO<sub>2</sub> layer, the collection efficiency of photo-generated electrons is improved, and the photocurrent is increased. We demonstrate via numerical modeling that the optimal wavefront excites the modes of the random medium in such a way that their destructive interference reduces light leakage through the open boundary. The buildup of light intensity inside the random medium, despite the proximity to the open boundary, facilitates light absorption and conversion to electrons. Alternatively, we show that WFS can suppress absorption by enhancing light reflection via constructive interference. This work demonstrates the great potential of WFS in depositing energy into targeted regions inside highly scattering media.

### Control of DSSC photocurrent

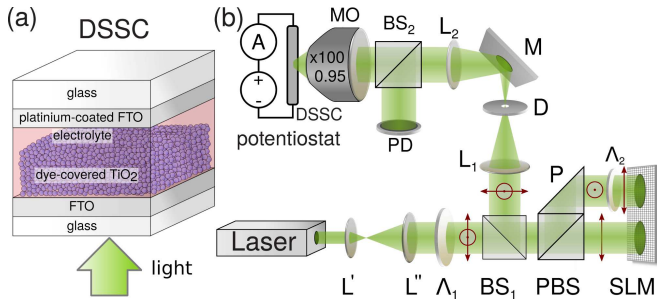


FIG. 1: **Experiment setup.** (a) Schematic of the dye-sensitized solar cell (DSSC) used in this study, which employs a thinner absorbing layer and a different electrolyte from the conventional DSSC to demonstrate coherent control of photovoltaic current. (b) Schematic of the experimental setup (see Methods). A laser beam at  $\lambda = 532\text{nm}$  is split to two orthogonal polarizations and projected onto the SLM. The phase front of both polarizations are modulated and then recombined at a beam splitter. The shaped wavefront is imaged onto the surface of the porous electrode in the DSSC using a microscope objective. The illuminated area is a disk of diameter  $\sim 8.3\ \mu\text{m}$ . The short-circuit current of the solar cell is measured using a potentiostat.

A DSSC comprises a porous layer of sintered  $\text{TiO}_2$  nanoparticles covered by a molecular dye, in this case the ruthenium-based N719 dye [23], deposited on a conductive glass substrate. It functions as the light-absorbing working electrode of the solar cell, while a platinum-loaded conductive glass slide acts as the counter electrode. An electrolyte fills the gap between the two electrodes and shuttles charges between them. The working electrode of a DSSC constructed in this manner can be considered as a scattering sample with homogeneous absorption [24]. Figure 1a presents a schematic of the solar cells created in this study. Light is incident onto the porous  $\text{TiO}_2$  layer through the glass substrate coated with transparent conducting oxide. After light absorption, the excited dye molecules rapidly inject electrons into the  $\text{TiO}_2$ . These electrons diffuse through the sintered particle network to be collected at the front side electrode, while the dye is regenerated via reduction by a redox mediator in the electrolyte, in this case the iodide/triiodide redox couple. The photo-induced current from a DSSC is typically measured without applying a bias voltage, and is known as the short-circuit current  $I_{sc}$ . Not all photo-generated electrons can diffuse through the  $\text{TiO}_2$  layer and reach the electrode, because electrons may recombine or get trapped on the way [25, 26]. The farther the electrons are generated from the electrode, the lower their probability of reaching it and contributing to the current. Hence,  $I_{sc}$  depends not only on the amount of light being absorbed but also on the position where the light is absorbed inside the  $\text{TiO}_2$  layer [27].

In the WFS experiment, the solar cell is excited by a narrow band laser beam at wavelength  $\lambda = 532\ \text{nm}$ . The input wavefront is modulated using the experimental setup schematically

shown in Fig. 1b. The two linear polarizations are modulated independently [22] (see Methods) and recombined into a single beam, which is projected onto the scattering medium in the solar cell using a microscope objective (NA = 0.95). We measure  $I_{sc}$  generated by the DSSC for each input phase mask. We use a genetic algorithm to find the wavefronts that maximize or minimize  $I_{sc}$ . Such algorithms are known to be robust against noise in WFS experiments [28, 29]. We define a cost function  $\eta_e$  that estimates the variation of the conversion efficiency of the solar cell:

$$\eta_e = \frac{(I_{sc} - I_{dark})/\mathcal{P}_0^{opt}}{(\langle I_{sc} \rangle - I_{dark})/\langle \mathcal{P}_0 \rangle}, \quad (1)$$

where  $\langle I_{sc} \rangle$  and  $\langle \mathcal{P}_0 \rangle$  are the average short-circuit current and input optical power, respectively, measured for a set of random phase masks.  $I_{dark}$  is the small dark current measured at  $\mathcal{P}_0 = 0\ \mu\text{W}$  [30].

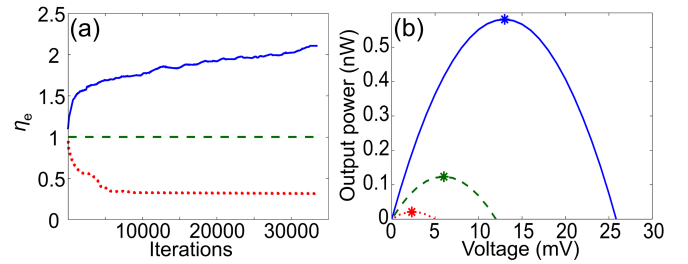


FIG. 2: **Control of the DSSC current and power by shaping the input wavefront of a laser beam.** (a) The measured cost function  $\eta_e$  for the short-circuit current, normalized to the value with random input wavefront (green dashed line), increases (blue solid line) or decreases (red dotted line) during the optimization of the input wavefront by a genetic algorithm. The photo-induced current is enhanced 2.1 times or suppressed 3.0 times by wavefront shaping. (b) Output electrical power of the DSSC illuminated by a random phase pattern (dashed green line), and the two optimized wavefronts for maximal  $\eta_e$  (solid blue line) and minimal  $\eta_e$  (dotted red line). The incident laser power is fixed at  $100\ \mu\text{W}$ . The asterisk indicates the maximal power, which is enhanced 4.7 times or reduced 5.7 times.

We constructed the DSSC using standard procedures (see Methods and Supplementary Information). The transport mean free path of light in the  $20 \pm 1\ \mu\text{m}$  thick  $\text{TiO}_2$  layer,  $\ell$ , is measured in a coherent backscattering experiment to be  $3.2\ \mu\text{m}$  [31]. To shorten the response time of the solar cell in order to perform WFS experiments on reasonable time scales, we use a concentrated non-volatile iodine-based electrolyte that also absorbs light at 532 nm. The ballistic absorption length  $\ell_a$  is about  $65\ \mu\text{m}$ , and the diffusive absorption length (the typical distance light will travel via multiple scattering prior to being absorbed),  $L_a = \sqrt{\ell\ell_a/3}$ , is  $8.4\ \mu\text{m}$  [31, 32]. Without absorption, the total transmission  $T$  of the  $\text{TiO}_2$  layer is estimated to be 26%, and the reflection  $R = 74\%$ . In the presence of absorption,  $T$  is reduced to 7%, and  $R$  to 47%, with the remaining 46% being absorbed (in the absence of wavefront optimization). These estimations agree well with the experimental measurement [30].

By optimizing the incident wavefront, the absorption cost function  $\eta_e$  is gradually increased or decreased as shown in Fig. 2a. With the optimal wavefronts, the photocurrent is enhanced by a factor of  $\sim 2.1$  or suppressed by a factor of  $\sim 3.0$ . We are thus able to coherently vary the energy conversion efficiency by a ratio of  $\sim 6.3$ . To quantify the variation of the output power due to the modification of the input optical pattern, we measure the current-voltage ( $I$ - $V$ ) curves for the two optimized wavefronts corresponding to maximum and minimum  $\eta_e$ , as well as for a random illumination pattern. Figure 2b displays the light-induced electrical output power, given by  $(I - I_{dark}) V$ , as a function of bias voltage for each scenario. The maximum output power is enhanced 4.7 times or reduced 5.7 times, leading to total variation by a factor of 27.

Since approximately 46% of the light is absorbed in the solar cell for a random illumination, the highest possible enhancement of the absorption would be a factor of 2.2. If  $I_{sc}$  were proportional to the total amount of light being absorbed [27], the experimental enhancement of 2.1 of the short circuit current would correspond to an almost perfect absorption of light in the absorbing medium. However, achieving the maximum absorption requires complete control over the amplitude and phase of the light in all input channels [6]. Experimentally we control only a limited fraction of the input channels due to the finite numerical aperture of the objective lens and the fact that the probe beam is incident only on one side of the scattering sample. Moreover, our SLM setup only modulates the phase and not the amplitude of the incident light, as would be necessary to achieve the global optimum. Consequently, the enhancement of total absorption that could be achieved with WFS is significantly reduced [7, 22].

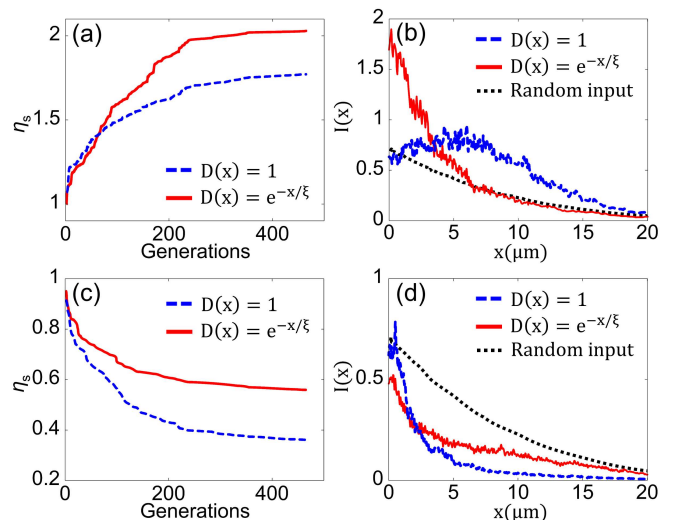
### Manipulation of local absorption

To investigate the cause for such a high enhancement factor measured experimentally, we perform numerical modeling. Without loss of generality, we simulate the scattering medium confined in a 2D waveguide with reflecting sidewalls to reduce the computation load; the system dimensions, as well as the amount of scattering and uniform absorption, are chosen to be near to the experimental values (see Methods). Light with transverse-magnetic polarization ( $E$  field perpendicular to the waveguide plane) is injected from one end of the waveguide, and its propagation in the random system is calculated with the recursive Green's function method [6, 33, 34]. The transmission and reflection of light for the systems with and without absorption, averaged over random input wavefronts, match the measured values of real samples. To model the effect of WFS in our experiment, we use the empty waveguide modes as the basis to shape the input field. During the optimization process, we modulate the phase of each empty waveguide modes while keeping their amplitude constant. The (linear) space-dependent absorption of light is proportional to the local intensity  $|E(x, y)|^2$ , and the total absorption is proportional to the spatially integrated intensity. To take

into account the spatial variation of the electron collection efficiency  $D(x) = \exp[-x/\xi]$ , where  $\xi$  is the electron diffusion length, we define a new cost function  $\eta_s$  which models the relative change of the short-circuit current similarly to  $\eta_e$ :

$$\eta_s = \frac{\int \int D(x) |E(x, y)|^2 dx dy}{\langle \int \int D(x) |E_0(x, y)|^2 dx dy \rangle}, \quad (2)$$

where  $E(x, y)$  is the electric field distribution in the scattering medium for the optimized input wavefront,  $E_0(x, y)$  is the field distribution for a random input wavefront, and  $\langle \dots \rangle$  represents the average over random input wavefronts. As in the solar cell, the absorption of light that occurs far from the front surface of the random medium contributes little to  $\eta_s$ . We used both a genetic algorithm and a sequential algorithm [12, 35] to maximize or minimize  $\eta_s$ , and found similar results, although the sequential algorithm converges faster than the genetic algorithm due to the absence of noise in simulation.



**FIG. 3: Numerical simulation of WFS experiment.** (a) The cost function  $\eta_s$ , defined in Eq.(2), increases and then saturates when the input wavefront is optimized for  $D(x) = \exp[-x/\xi]$  (red solid line) and  $D(x) = 1$  (blue dashed line) using a sequential algorithm.  $\xi$  is taken to be  $3 \mu\text{m}$  (see Methods). (b) The cross-section integrated intensity in the random system illuminated by random input wavefronts (black dotted line) and the optimized wavefront obtained with the non-uniform cost function for  $D(x) = \exp[-x/\xi]$  (red solid line) and the uniform cost function for  $D(x) = 1$  (blue dashed line). The light is concentrated near the front side of the random system for  $D(x) = \exp[-x/\xi]$ , but penetrates deeper into the system for  $D(x) = 1$ . (c,d) Same as (a,b) for the minimization of  $\eta_s$ . For  $D(x) = \exp[-x/\xi]$ , the light intensity in the front side of the random system is reduced, but increased deeper into the system. For  $D(x) = 1$ , the light intensity is reduced in the interior of the system but remain almost the same near the front surface.

Figure 3a shows the enhancement of the cost function  $\eta_s$  in the random medium with a uniform absorption coefficient. For comparison we also simulate the case of  $D(x) = 1$ , which corresponds to a uniform cost function similar to the standard

CEA [6, 7]. Both the non-uniform ( $D(x) = \exp[-x/\xi]$ ) and uniform cost functions increase continuously in the optimization process and eventually saturate; however as expected, the final enhancement factor for  $D(x) = \exp[-x/\xi]$  significantly exceeds that for  $D(x) = 1$ . Figure 3b represents the integrated intensity  $I(x) = \int |E(x, y)|^2 dy$  inside the random medium. With random wavefront illumination,  $I(x)$  decays exponentially in the disordered system, as expected for light diffusion in the presence of absorption [32]. When the input wavefront is optimized for uniform cost function ( $D(x) = 1$ ),  $I(x)$  is peaked in the interior of the random system. The buildup of energy deep inside the scattering medium enhances the total absorption from 46% to 82%. Meanwhile, the reflection decreases from 47% to 9%, and the transmission slightly increases from 7% to 9%. This result confirms that the global absorption is enhanced by WFS through causing the light to penetrate deeper into the lossy disordered medium.

In contrast, the optimization for the non-uniform cost function ( $D(x) = \exp[-x/\xi]$ ) leads to a significant increase of light intensity close to the front surface of the random medium [Fig. 3b], since only this region contributes significantly to the cost function  $\eta_s$ . Naively, the proximity to the open boundary would be expected to facilitate light escape from the absorbing medium, thus reducing the absorption and increasing the reflection. Instead, for the optimal wavefront, the total absorption is increased from 46% to 67%, and the reflection decreased from 47% to 28%. This result suggests some partial trapping of the light near the surface, leading to a buildup of its intensity in the vicinity of the open boundary where the front side electrode is located. Hence, the numerical simulation illustrates that our experimental procedure, which does feedback optimization based on the short-circuit current, does not simply optimize total absorption, but optimizes absorption locally, near the working electrode, so as to preferentially enhance the collected current. The measured current increase by a factor 2.1 does not imply perfect absorption of all incident light and can be realized with the incomplete experimental control.

### Modal interference effects

To understand how the leakage rate of the light close to the open boundary is reduced, we investigate the contribution of different quasi-modes in the system to the reflected light. It has been shown previously that light propagation in random media can be well explained in terms of the interference effects of quasi-modes [36–39]. We take this approach by decomposing the electric field inside the random medium in terms of the quasi-modes of the system:

$$E(x, y) = \sum_m a_m u_m(x, y) + \sum_m b_m v_m(x, y), \quad (3)$$

where  $u_m$  ( $v_m$ ) is the  $m$ -th mode with purely outgoing (incoming) wave boundary condition, and  $u_m = v_m^*$  [40]. The reflected field at the front surface ( $x = 0$ ) of the disordered

medium can be expressed as  $E_r(y) = \sum_m a_m u_m(0, y)$ . The total intensity of reflected waves is  $R = \int |E_r(y)|^2 dy = \sum_m \alpha_m$ , where  $\alpha_m = a_m \int E_r^*(y) u_m(0, y) dy$  represents the contribution of the  $m$ -th mode to the reflection. While  $\alpha_m$  is a complex number,  $R$  is a real number, and the contributions of different modes to  $R$  interfere with each other. The interference effect can be quantified by  $C \equiv |\sum_m \alpha_m|^2 / \sum_m |\alpha_m|^2$  [30]. If  $C > 1$  ( $C < 1$ ), the modes interfere constructively (destructively) at the front surface to enhance (suppress) reflection.

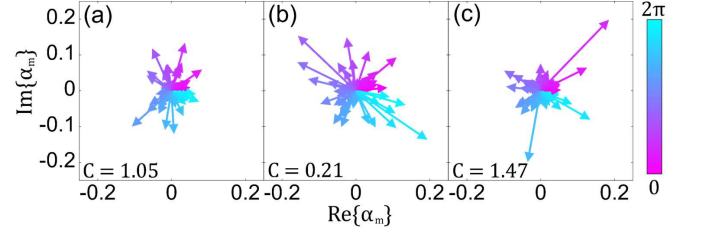


FIG. 4: **Modal analysis of the wavefront optimization.** Contributions  $\alpha_m$  of individual modes  $m$  supported by the random system to the total intensity of reflected waves. The modes are excited by a random input wavefront (a), the optimized wavefront for maximal  $\eta_s$  (b) and minimal  $\eta_s$  (c) with  $D(x) = \exp[-x/\xi]$ . In (a) the modes have random phases and their interference is averaged out,  $C = 1.05$ . In (b) the modes interfere destructively,  $C = 0.21 < 1$ , to reduce reflection and enhance absorption. In (c) the modes interfere constructively,  $C = 1.47 > 1$ , to enhance reflection and reduce absorption.

To reduce the computing time for the full modal analysis, the length and width of the system in the simulation is reduced by a factor of two. The scattering and absorption parameters are adjusted so that the total transmission, reflection and absorption for random input wavefronts are essentially unchanged. We use the finite-difference frequency-domain (FDFD) method to calculate the quasi-modes whose frequencies are close to the frequency of the input light. The calculated values of  $\alpha_m$  for the different scenarios with  $D(x) = \exp[-x/\xi]$  are plotted in the complex plane in Figs. 4a-c. The modes have uncorrelated phases for random wavefront illumination and their interference is averaged out (and therefore  $C$  is close to 1). We see that the optimized wavefront for enhanced local absorption causes a destructive interference among the modes, making  $C$  much smaller than 1, and more importantly, that reflection is minimized and absorption is maximized at the front surface. Despite the complexity of the medium, WFS exploits interferences to efficiently concentrate light in the targeted region of the system.

Similarly, we show that WFS can also reduce the energy in a chosen region by minimizing the cost function  $\eta_s$  [Figs. 3c and d]. For  $D(x) = 1$ , the reflection is greatly enhanced, decreasing the light intensity deep inside the system and thus the total amount of absorption. For  $D(x) = \exp[-x/\xi]$ , the minimization of the cost function is less efficient as it requires to *simultaneously* increase the reflection and decrease the absorption close to the front surface. By decomposing the fields

into the quasi-modes of the system, we confirm that the increase in reflection is caused by the constructive interference of excited modes [Fig. 4c], which facilitates light escape and suppresses absorption.

## Conclusion

In summary, we have experimentally demonstrated control of photocurrent in a dye-sensitized solar cell (DSSC) by tailoring the input wavefront of a laser beam. To maximize the short-circuit current, the input light is concentrated on the front side of the porous electrode where the photo-generated electrons are most efficiently collected. Despite the proximity to the open boundary, the destructive interference of the excited quasi-modes at the front surface minimizes light leakage and facilitates absorption. Although in the current WFS experiment a narrow-band laser is used for coherent control of absorption, recent theoretical study suggests that this approach is applicable to a relatively broadband light [41]. More generally, this approach opens new pathways to probe and characterize photo-induced processes inside opaque media such as solar cell materials. Our study shows that WFS and optimization allows coherent control of absorption by causing light to be concentrated into arbitrary local regions within an open random medium, including regions very close to the open boundaries. Using this method one may investigate photochemical processes in targeted regions inside a dense scattering medium. Such studies will provide physical insight into the behavior of photovoltaic and photoelectrochemical systems, which may lead to the development of methods to improve their performance.

## Methods

*Sample preparation.* To facilitate the WFS experiment, we made the DSSC with a thicker  $\text{TiO}_2$  scattering layer, reduced loading of N719 dye, and a thin (175  $\mu\text{m}$  thick) ITO coated glass substrate for the working electrode [30]. The porous  $\text{TiO}_2$  working electrode is made from  $\sim 20\text{nm}$   $\text{TiO}_2$  nanoparticle paste (P25, Degussa), which is doctor-bladed onto a tin-doped indium oxide (ITO) coated glass slide (SPI, Product No. 06477B-AB). The slide with doctor-bladed paste is then sintered at  $450^\circ\text{C}$ . The thickness of the  $\text{TiO}_2$  layer is  $20 \pm 1 \mu\text{m}$ . The working electrode is sintered at  $450^\circ\text{C}$  for 1 hour, then immersed in a solution of N719 (ditetrabutylammonium cis-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4'-dicarboxylato) ruthenium(II) for 30 minutes [23]. The counter electrode is made by heating hexachloroplatinic acid in ethanol (2 mg/mL) to  $400^\circ\text{C}$  for 15 minutes on a fluorine-doped tin oxide (FTO) coated glass slide. The solar cells are assembled with a 25  $\mu\text{m}$  Surlyn spacer hot-pressed between the electrodes and filled with a non-volatile iodine-based electrolyte (Mosalyte TDE-250, Solaronix SA, Switzerland). The absorption of light by the electrolyte is proportional to that by the dye at any location, as both scale linearly with local field intensity. Thus the short-circuit current, which results from light absorption by the dye, also reflects the absorption of the electrolyte, which does not contribute directly to the current.

*Optical setup.* A shown schematically in Fig. 1, the output beam from a continuous-wave Nd:YAG laser is expanded (lenses  $L'$  and  $L''$ ) and projected onto a phase-only SLM (Hamamatsu, X10468-01). A half-wave-plate ( $\Lambda_1$ ) is used to balance the light intensities of two orthogonal linear polarizations. To control independently the two polarizations, a polarizing beam splitter (PBS)

attached to a right-angle prism (P) separates the two polarizations of the laser beam, which are modulated by different areas of the SLM. The cube and the prism are mounted in the same holder to eliminate fluctuations from independent reflective elements in the two paths. A second half-wave-plate ( $\Lambda_2$ ) rotates the polarization to match that of the SLM. The two illuminated areas on the SLM corresponding to the two polarizations of the input light are divided into 1740 macropixels. The modulated wavefronts are then recombined and projected onto the pupil of a microscope objective (MO) with numerical aperture 0.95. A beam splitter ( $\text{BS}_2$ ) along with a photo-detector (PD) are used to measure the optical power  $\mathcal{P}_0$  impinging on the solar cell. The photo-induced current of the DSSC is measured by a potentiostat (Digi-Ivy DY2116b).

*Numerical simulation.* In the 2D model, dielectric cylinders with radius 50 nm and refractive index 2.5 are randomly positioned inside a waveguide of width  $W = 20 \mu\text{m}$  and refractive index 1.5. The length of the random structure is  $L = 20 \mu\text{m}$ , and the effective index of refraction is 1.9. The wavelength of input light is  $\lambda = 532 \text{ nm}$ , and the number of transverse guided modes in the waveguide is 114. The transport mean free path  $\ell$  is 3.7  $\mu\text{m}$ . Without absorption and wavefront optimization, the transmission is  $T = 26\%$  and the reflection is  $R = 74\%$ . A uniform absorption coefficient is introduced via the imaginary part of the refractive index to the random system. The diffusive absorption length is 8.5  $\mu\text{m}$ .  $T$  is reduced to 7%, and reflection to 47%, thus the absorption  $A = 46\%$ . These numbers match the measured values of our DSSC. The electron diffusion length  $\xi$  is chosen to be 3  $\mu\text{m}$ , close to the value of DSSCs similar to ours [25, 26].

In the modal analysis to decompose the field distribution in the random medium with the quasi-modes, we keep the wavelength  $\lambda$  the same, but reduce the number of waveguide modes to 37.  $\ell$  is reduced to 2  $\mu\text{m}$ , and the diffusive absorption length to 4.2  $\mu\text{m}$ . Without absorption,  $T = 24\%$  and  $R = 76\%$ ; with absorption,  $T = 7\%$ ,  $R = 46\%$ , and  $A = 47\%$ . The electron diffusion length  $\xi$  is also shortened to 1.5  $\mu\text{m}$ .

## References

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- [1] Wan, W. *et al.* Time-reversed lasing and interferometric control of absorption. *Science* **331**, 889–892 (2011).
  - [2] Chong, Y. D., Ge, L., Cao, H. & Stone, A. D. Coherent perfect absorbers: time-reversed lasers. *Phys. Rev. Lett.* **105**, 053901 (2010).
  - [3] Noh, H., Chong, Y. D., Stone, A. D. & Cao, H. Perfect coupling of light to surface plasmons by coherent absorption. *Phys. Rev. Lett.* **108**, 186805 (2012).
  - [4] Noh, H., Popoff, S. M. & Cao, H. Broadband subwavelength focusing of light using a passive sink. *Opt. Express* **21**, 17435–17446 (2013).
  - [5] Li, S. *et al.* An equivalent realization of coherent perfect absorption under single beam illumination. *Sci. Rep.* **4**, 7369 (2014).
  - [6] Chong, Y. D. & Stone, A. D. Hidden black: coherent enhancement of absorption in strongly scattering media. *Phys. Rev. Lett.* **107**, 163901 (2011).
  - [7] Goetschy, A. & Stone, A. D. Filtering random matrices: the effect of incomplete channel control in multiple scattering. *Phys. Rev. Lett.* **111**, 063901 (2013).
  - [8] Yaqoob, Z., Psaltis, D., Feld, M. S., & Yang, C. Optical phase conjugation for turbidity suppression in biological samples. *Nat. Photon.* **2**, 110–115 (2008).
  - [9] Popoff, S., Lerosey, G., Fink, M., Boccaro, A. C. & Gigan, S. Image transmission through an opaque material. *Nat. Commun.* **1**, 81 (2010).
  - [10] Mosk, A. P., Lagendijk, A., Lerosey, G. & Fink, M. Controlling waves in space and time for imaging and focusing in complex media. *Nat. Photon.* **6**, 283–292 (2012).

- [11] Bertolotti, J. *et al.* Non-invasive imaging through opaque scattering layers. *Nature* **491**, 232–234 (2012).
- [12] Vellekoop, I. M. & Mosk, A. P. Focusing coherent light through opaque strongly scattering media. *Opt. Lett.* **32**, 2309–2311 (2007).
- [13] Vellekoop, I. M., van Putten, E. G., Lagendijk, A. & Mosk, A. P. Demixing light paths inside disordered metamaterials. *Opt. Express* **16**, 67–80 (2008).
- [14] Vellekoop, I. M., Lagendijk, A. & Mosk, A. P. Exploiting disorder for perfect focusing. *Nat. Photon.* **4**, 320–322 (2010).
- [15] Guan, Y., Katz, O., Small, E., Zhou, J. & Silberberg, Y. Polarization control of multiply scattered light through random media by wavefront shaping. *Opt. Lett.* **37**, 4663–4665 (2012).
- [16] Park, J. H., Park, C. H., Yu, H., Cho, Y. H. & Park, Y. K. Dynamic active wave plate using random nanoparticles. *Opt. Express* **20**, 17010–17016 (2012).
- [17] Park, J. H., Park, C. H., Yu, H., Cho, Y. H. & Park, Y. K. Active spectral filtering through turbid media. *Opt. Lett.* **37**, 3261–3263 (2012).
- [18] Small, E., Katz, O., Guan, Y. & Silberberg, Y. Spectral control of broadband light through random media by wavefront shaping. *Opt. Express* **37**, 3429–3431 (2012).
- [19] Vellekoop, I. M. & Mosk, A. P. Universal optimal transmission of light through disordered materials. *Phys. Rev. Lett.* **101**, 120601 (2008).
- [20] Kim, M. *et al.* Maximal energy transport through disordered media with the implementation of transmission eigenchannels. *Nat. Photon.* **6**, 581–585 (2012).
- [21] Yu, H. *et al.* Measuring large optical transmission matrices of disordered media. *Phys. Rev. Lett.* **111**, 153902 (2013).
- [22] Popoff, S. M., Goetschy, A., Liew, S. F., Stone, A. D. & Cao, H. Coherent control of total transmission of light through disordered media. *Phys. Rev. Lett.* **112**, 133903 (2014).
- [23] O’Regan, B. & Grätzel, M. A low-cost, high-efficiency solar cell based on dye-sensitized colloidal TiO<sub>2</sub> films. *Nature* **353**, 737–740 (1991).
- [24] Galvez, F. E., Barnes, P. R. F., Halme, J. & Miguez, H. Dye sensitized solar cells as optically random photovoltaic media. *Energy Environ. Sci.* **7**, 689–697 (2014).
- [25] Nakade, S. *et al.* Dependence of TiO<sub>2</sub> nanoparticle preparation methods and annealing temperature on the efficiency of dye-sensitized solar cells. *J. Phys. Chem. B* **106**, 10004–10010 (2002).
- [26] Oekermann, T., Zhang, D., Yoshida, T. & Minoura, H. Electron transport and back reaction in nanocrystalline TiO<sub>2</sub> films prepared by hydrothermal crystallization. *J. Phys. Chem. B* **108**, 2227–2235 (2004).
- [27] Sheehan, S. W., Noh, H., Brudvig, G. W., Cao, H. & Schmuttenmaer, C. A. Plasmonic enhancement of dye-sensitized solar cells using core-shell-shell nanostructures. *J. Phys. Chem. C* **117**, 927–934 (2013).
- [28] Katz, O., Small, E., Bromberg, Y. & Silberberg, Y. Focusing and compression of ultrashort pulses through scattering media. *Nat. Photon.* **5**, 372–377 (2011).
- [29] Conkey, D. B., Brown, A. N., Caravaca-Aguirre, A. M. & Piestun, R. Genetic algorithm optimization for focusing through turbid media in noisy environments. *Opt. Express* **20**, 4840–4849 (2012).
- [30] Supplementary information
- [31] Akkermans, E. & Montambaux, G. *Mesoscopic Physics of Electrons and Photons* (Cambridge University Press, 2007).
- [32] van Rossum, M. C. W. & Nieuwenhuizen, Th. M. Multiple scattering of classical waves: microscopy, mesoscopy, and diffusion. *Rev. Mod. Phys.* **71**, 313–371 (1999).
- [33] Lee, P. A. & Fisher, D. S. Anderson localization in two dimensions. *Phys. Rev. Lett.* **47**, 882–885 (1981).
- [34] Beenakker, C. W. J., Paasschens, J. C. J. & Brouwer, P. W. Probability of reflection by a random laser. *Phys. Rev. Lett.* **76**, 1368–1371 (1996).
- [35] Vellekoop, I. M. & Mosk, A. P. Phase control algorithms for focusing light through turbid media. *Opt. Commun.* **281**, 3071–3080 (2008).
- [36] Wang, J. & Genack, A. Z. Transport through modes in random media. *Nature* **471**, 345–348 (2011).
- [37] Wang, J., Shi, Z., Davy, M. & Genack, A. Z. Wave propagation and localization via quasi-normal modes and transmission eigenchannels. *Int. J. Mod. Phys. Conf. Ser.* **11**, 1–11 (2012).
- [38] Liew, S. F., Popoff, S. M., Mosk, A. P., Vos, W. L. & Cao, H. Transmission channels for light in absorbing random media: From diffusive to ballistic-like transport. *Phys. Rev. B* **89**, 224202 (2014).
- [39] Shi, Z. & Genack, A. Z. Modal makeup of transmission eigenchannels. arXiv:1406.3673 (2014).
- [40] Cerjan, A. & Stone, A. D. Steady-state *ab initio* theory of lasers with injected signals. *Phys. Rev. A* **90**, 013840 (2014).
- [41] Hsu, C. W. *et al.*, unpublished (2015)

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### Author contributions

H.C. and A.D.S. initiated the study. S.F.L. and S.M.P. conducted the experiments and numerical modeling. S.W.S. and C.A.S. fabricated the samples and did structural characterization. A.G. and A.D.S. did an theoretical analysis. S.F.L. and S.M.P. analyzed the data and drafted the paper, the rest of the authors contributed to data interpretation and manuscript preparation.

## Supplementary Material

### Coherent control of photocurrent in a disordered photovoltaic system

S. F. Liew<sup>1,†</sup>, S. M. Popoff<sup>1,2,†</sup>, S. W. Sheehan<sup>3</sup>, A. Goetschy<sup>1,4</sup>, C. A. Schmuttenmaer<sup>3</sup>, A. D. Stone<sup>1</sup> and H. Cao<sup>1\*</sup>

<sup>1</sup> Department of Applied Physics, Yale University, New Haven, CT 06520, USA

<sup>2</sup> CNRS LTCI, Telecom ParisTech, 46 rue Barrault, 75013 Paris, France

<sup>3</sup> Department of Chemistry, Yale University, New Haven, Connecticut 06520, USA

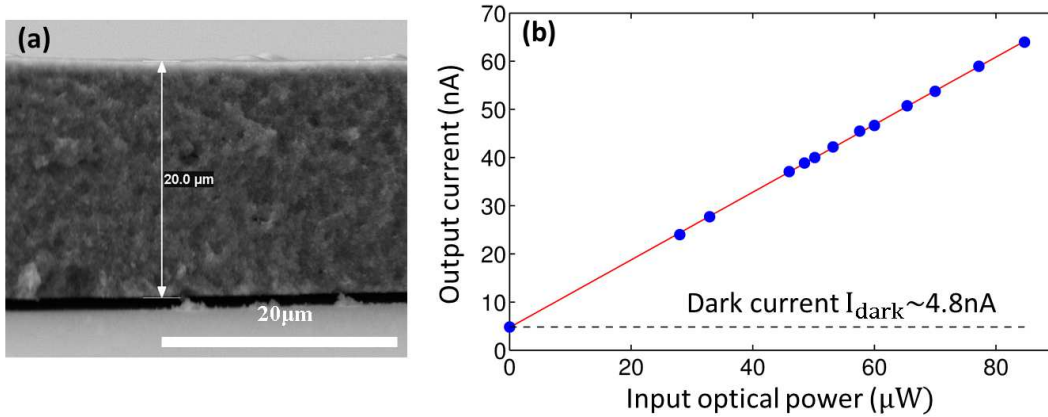
<sup>4</sup> ESPCI ParisTech, PSL Research University, Institut Langevin, 1 rue Jussieu, 75005 Paris, France

<sup>†</sup> These two people contribute equally to this work. \*hui.cao@yale.edu

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#### SYNTHESIS AND CHARACTERIZATION OF THE DSSC

The DSSC in this study was modified to demonstrate coherent control of photovoltaic current, we made the TiO<sub>2</sub> layer thicker than in the common DSSC to ensure multiple scattering and used a concentrated non-volatile iodine-based electrolyte to shorten the response time of the DSSC. Degussa P25 TiO<sub>2</sub> nanoparticles were made into a terpeneol-based screen printing paste containing ethyl cellulose, using the procedure outlined by Ito *et al.* [1]. The paste was doctor bladed on to a 175 μm thick ITO-coated glass coverslip 5 times, which was heated to 200°C for 10 min then cooled to room temperature between each application of a layer of paste. The thin substrate was used so that the incident light could be coupled by a high-NA objective lens with short working distance into the DSSC. After application of the screen-printing paste, the working electrodes were heated in a tube furnace to 450°C for one hour, with a 4°C per minute heating and cooling rate. Figure S1(a) is a representative SEM image of the resulting 20 μm thick TiO<sub>2</sub> layer. The thickness of the working electrodes was confirmed using a profilometer (KLA Tencor Alphastep 200).



Supplementary Figure 1: **Characterization of the DSSC.** (a) Scanning electron microscope (SEM) image of photoanode TiO<sub>2</sub> film, with thickness 20 μm ± 1 μm. (b) Measured short-circuit current  $I_{sc}$  as a function of the input optical power  $\mathcal{P}_0$  for a fixed random input waveform. The Experimental data (blue dots) are well fitted by a linear function (solid red line) with a dark current  $I_{dark} \approx 4.8$  nA.

Since strong absorption would suppress the interference of multiply scattered light, we lowered the loading of dye by reducing the time that the working electrodes were immersed in a low molarity N719 (Dyesol) solution (0.05 mM in dry ethanol). The dye-coated TiO<sub>2</sub> film was then assembled into a device, with a Pt-coated FTO cathode hot-pressed to the anode with a 25 μm Surlyn spacer (Meltonix Solaronix SA, Switzerland). The DSSCs were filled, through holes drilled into the cathode, with Mosalyte TDE-250 (Solaronix SA, Switzerland), which is a highly viscous, non-volatile, and stable ionic liquid electrolyte, before being sealed shut via hot-press with Surlyn. The conductive copper tape was attached to the working and counter electrodes separately to improve electrical contact with the potentiostat.

Due to the slow diffusion of charges through the liquid electrolyte and the relatively long charge carrier lifetime, the response of the solar cell is slow (typically on the order of 10 seconds). With a large number of degrees of freedom to optimize (quantified by the number of SLM segments), the search algorithm needs at least tenth of thousands of iterations to reach an optimal value. To shorten the response time, we replace the volatile electrolyte by the

non-volatile one. However, the conversion efficiency of the DSSC is reduced due to the lack of efficiency-enhancing additives such as 4-*tert*-butylpyridine.

The transmission and reflection of the TiO<sub>2</sub> layer in the DSSC can be estimated with the following formula [2, 3].

$$T = \frac{\sinh(z_{\text{inj}}/L_a) + (z_{e1}/L_a) \cosh(z_{\text{inj}}/L_a)}{(1 + z_{e1}z_{e2}/L_a^2) \sinh(L/L_a) + ((z_{e1} + z_{e2})/L_a) \cosh(L/L_a)}, \quad (1)$$

and

$$R = \frac{\sinh((L - z_{\text{inj}})/L_a) + (z_{e2}/L_a) \cosh((L - z_{\text{inj}})/L_a)}{(1 + z_{e1}z_{e2}/L_a^2) \sinh(L/L_a) + ((z_{e1} + z_{e2})/L_a) \cosh(L/L_a)}, \quad (2)$$

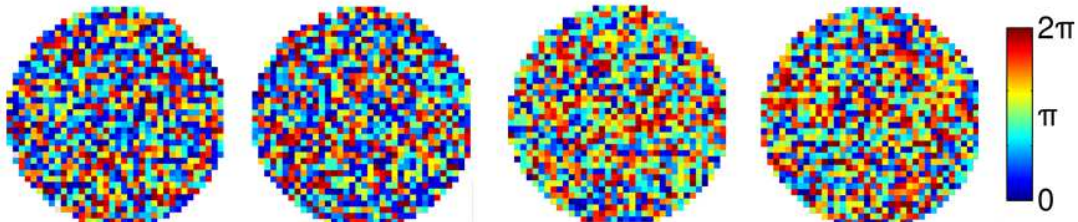
where  $L_a = \sqrt{\ell\ell_a/3}$  is the diffusive absorption length,  $z_{e1}$  and  $z_{e2}$  are the extrapolation lengths on the front and rear surfaces of the sample [4, 5]. In addition,  $z_{\text{inj}}$  is the injection length estimated using the formula in Ref. [6]. Using the measured values of  $\ell = 3.2 \mu\text{m}$ , we obtain  $T = 26\%$ ,  $R = 74\%$  if there is no absorption, and  $T = 7\%$ ,  $R = 47\%$  for the DSSC with absorption. After taking into account the reflections from the surfaces of the glasses, the measured values for the fabricated DSSC are  $T = 7\%$ ,  $R = 42\%$ , in good agreement to the above estimation.

The short-circuit current  $I_{sc}$  of the DSSC can be written as:

$$I_{sc} = I_{dark} + \beta \mathcal{P}_0, \quad (3)$$

where  $I_{dark}$  is the dark current,  $\mathcal{P}_0$  is the incident optical power, and  $\beta$  is a constant. The  $I_{dark}$  is independent of the incident wavefront, and its value is obtained experimentally. As shown in Fig. S1(b), the measured  $I_{sc}$  scales linearly with  $\mathcal{P}_0$  for a fixed input wavefront. The dark current is about 4.8 nA.

Even though the short-circuit current is lower than the that of the standard DSSC, it still exceeds the measurement noise and ensures the convergence of the optimization algorithm. Figure S2 shows the phase masks produced by the SLM to maximize or minimize the short-circuit currents. As expected, the patterns look completely random.



Supplementary Figure 2: **Phase masks to produce the optimal wavefronts.** The optimal phase masks for the two orthogonal linear polarizations when the cost function  $\eta_c$  is maximized (left two patterns) or minimized (right two patterns) in the DSSC, as shown in Fig. 2 of the main text.

## NUMERICAL SIMULATION OF WFS EXPERIMENT

We use the recursive Green's function method to calculate light transport in a two-dimensional (2D) random system with a uniform absorption coefficient. The scattering and absorption parameters are close to the experimental values. Tables 1a and 1b gives the values of the transmission, reflection, and absorption for random incident wavefront (averaged over 500 random wavefronts), and the numerically optimized wavefront to maximize or minimize the cost function  $\eta_s$  for  $D(x) = \exp[-x/\xi]$  and  $D(x) = 1$ , where  $\xi = 3\mu\text{m}$ .

Supplementary Table 1a: **Values of the transmission, reflection, and absorption for random incident wavefront, and the optimal wavefront to maximize the cost function  $\eta_s$  for  $D(x) = \exp[-x/\xi]$  and  $D(x) = 1$ , where  $\xi = 3\mu\text{m}$ .**

	Random wavefront (averaged)	Optimized wavefront $D(x) = \exp[-x/\xi]$	Optimized wavefront $D(x) = 1$
Transmission	0.07	0.05	0.09
Reflection	0.47	0.28	0.09
Absorption	0.46	0.67	0.82

Supplementary Table 1b: **Values of the transmission, reflection, and absorption for random incident wavefront, and the optimal wavefront to minimize the cost function  $\eta_s$  for  $D(x) = \exp[-x/\xi]$  and  $D(x) = 1$ , where  $\xi = 3\mu\text{m}$ .**

	Random wavefront (averaged)	Optimized wavefront $D(x) = \exp[-x/\xi]$	Optimized wavefront $D(x) = 1$
Transmission	0.07	0.05	0.01
Reflection	0.47	0.70	0.82
Absorption	0.46	0.25	0.17

## MODAL ANALYSIS

To decompose the electric field distribution inside the random system on the quasi-modes basis, we used the finite-difference frequency-domain (FDFD) method to calculate the quasi-modes of the random system. There are two types of such modes: (i)  $u_m$  - the eigenmodes of the Maxwell equations which satisfy the boundary conditions that there are only outgoing waves to the infinity; (ii)  $v_m$  - the eigenmodes for the boundary conditions of only incoming waves from infinity. In the absence of absorption,  $u_m = v_m^*$ . Tables 2a and 2b gives the values of the transmission, reflection, and absorption for random incident wavefront, and the optimal wavefront to maximize or minimize the cost function  $\eta_s$  for  $D(x) = \exp[-x/\xi]$  and  $D(x) = 1$ , where  $\xi = 1.5\mu\text{m}$ . The values are close to the ones in Table 1, despite of the reduction in the system parameters.

Supplementary Table 2a: **Values of the transmission, reflection, and absorption for random incident wavefront, and the optimal wavefront to maximize the cost function  $\eta_s$  for  $D(x) = \exp[-x/\xi]$  and  $D(x) = 1$ , where  $\xi = 1.5\mu\text{m}$ .**

	Random wavefront (averaged)	Optimized wavefront $D(x) = \exp[-x/\xi]$	Optimized wavefront $D(x) = 1$
Transmission	0.07	0.05	0.08
Reflection	0.46	0.31	0.13
Absorption	0.47	0.64	0.79

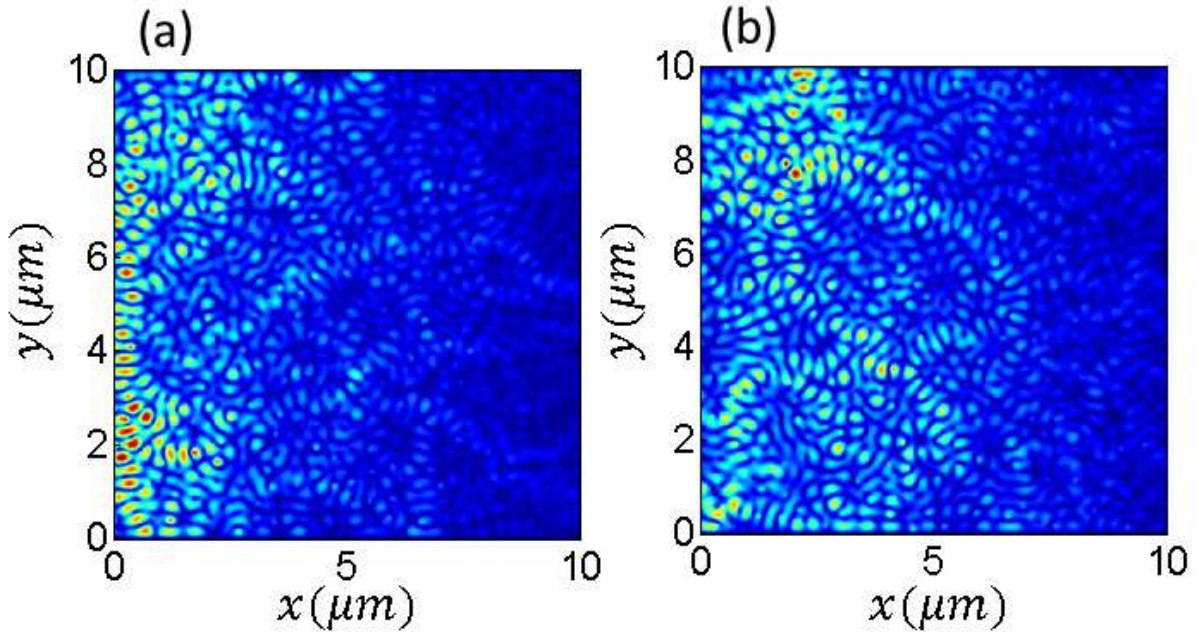
Supplementary Table 2b: **Values of the transmission, reflection, and absorption for random incident wavefront, and the optimal wavefront to minimize the cost function  $\eta_s$  for  $D(x) = \exp[-x/\xi]$  and  $D(x) = 1$ , where  $\xi = 1.5\mu\text{m}$ .**

	Random wavefront (averaged)	Optimized wavefront $D(x) = \exp[-x/\xi]$	Optimized wavefront $D(x) = 1$
Transmission	0.07	0.05	0.01
Reflection	0.46	0.69	0.80
Absorption	0.47	0.26	0.19

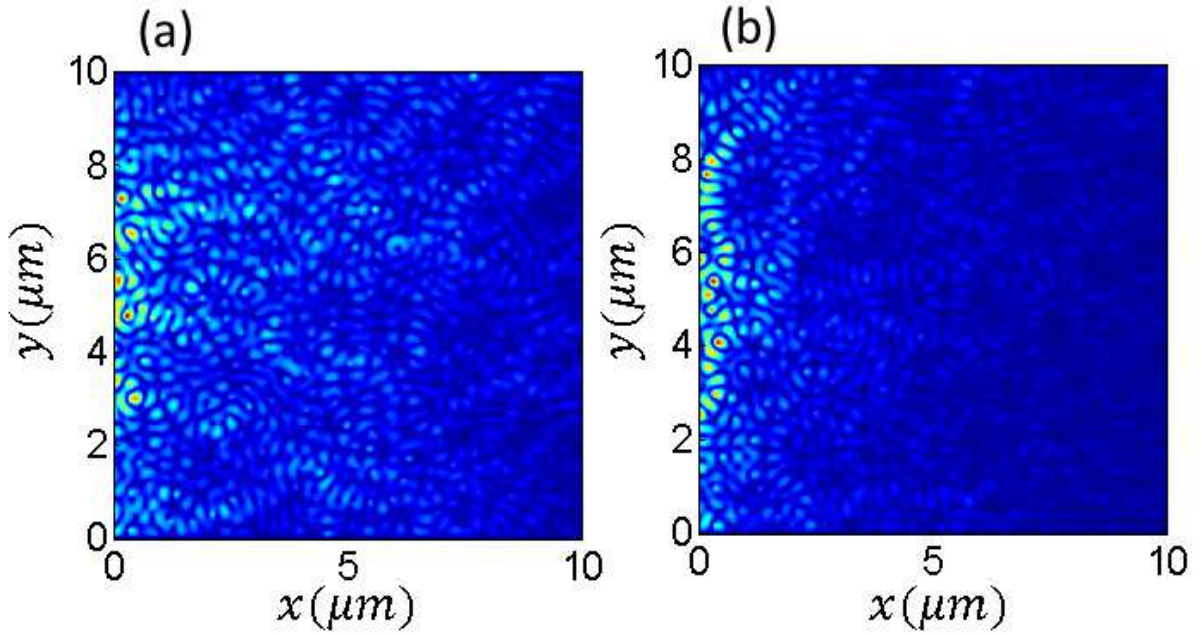
Figure S3 shows the spatial distribution of the electric field amplitude  $|E(x, y)|$  inside the random system with the optimized wavefront for maximal cost function  $\eta_s$  with  $D(x) = \exp[-x/1.5\mu\text{m}]$  (a) and  $D(x) = 1$  (b). In Fig. S3(a), the field intensity near the front surface of the random system is increased to enhance the absorption at the front side, while in Fig. S3(b) light penetrates deeper into the system to maximize the global absorption.

Figure S4 compares the spatial distribution of the electric field amplitude  $|E(x, y)|$  inside the random system with the optimized wavefront for minimal cost function  $\eta_s$  with  $D(x) = \exp[-x/1.5\mu\text{m}]$  (a) and  $D(x) = 1$  (b). While in Fig. S4(a) the field intensity near the front surface of the random system is reduced to suppress the absorption at the front side, in Fig. S4(b) the intensity deep inside the system is decreased to minimize the global absorption.

In Fig. S5, the electric field inside the scattering system for a random input wavefront is decomposed by the quasi-modes. After the decomposition, the reconstructed field distribution in Fig. S5(b) matches well the original one in Fig. S5(a). The good agreement is further shown in the cross-section integrated intensity  $I(x) = \int |E(x, y)|^2 dy$  as well as

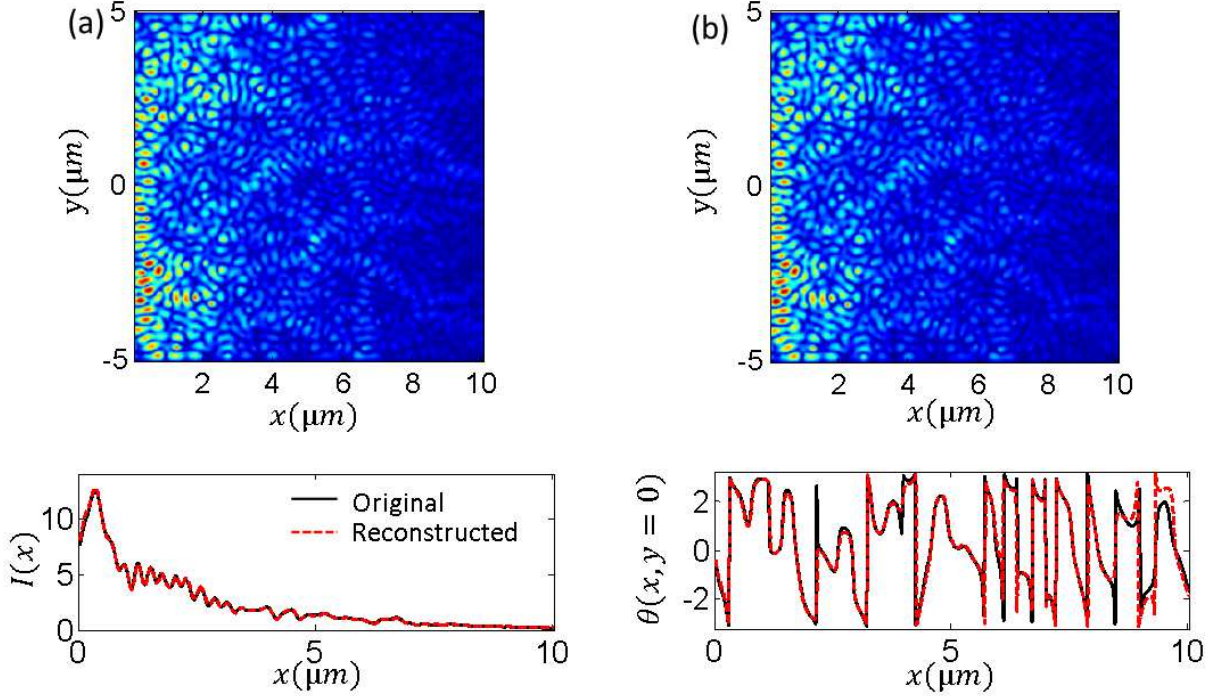


Supplementary Figure 3: **Spatial field distribution for enhanced local or global absorption in the random medium.** FDFD simulation of spatial field distribution for the optimized wavefront for maximal cost function with  $D(x) = \exp[-x/\xi]$ ,  $\xi = 1.5\mu\text{m}$  (a), and  $D(x) = 1$  (b).



Supplementary Figure 4: **Spatial field distribution for suppressed local or global absorption in random medium.** FDFD simulation of spatial field distribution for the optimized wavefront for minimal cost function with  $D(x) = \exp[-x/\xi]$ ,  $\xi = 1.5\mu\text{m}$  (a), and  $D(x) = 1$  (b).

the phase of the electric field  $E(x, 0)$  in Figs. S5(c) and S5(d). The error between the original field amplitude  $|E_S(x, y)|$  and the reconstructed one  $|E_C(x, y)|$  is computed by  $\int \int |E_C(x, y) - E_S(x, y)|^2 dx dy / \int \int |E_S(x, y)|^2 dx dy = 0.005$ .



Supplementary Figure 5: **Decomposition of the spatial distribution of electric field by the quasi-modes of the random system.** The spatial electric field profile inside the random medium for a random input wavefront  $|E_S(x, y)|$  (a) and after reconstruction  $|E_C(x, y)|$  (b). The cross-section integrated intensity  $I(x) = \int |E(x, y)|^2 dy$  and phase of the electric field  $E(x, y = 0)$  between the original and reconstructed field profiles are shown in (c) and (d) respectively. The error, given by the relative difference between the two field profiles, is  $\iint |E_C(x, y) - E_S(x, y)|^2 dx dy / \iint |E_S(x, y)|^2 dx dy = 0.005$ .

Supplementary Table 3: **Coherent and incoherent sums for the optimized wavefronts to maximize or minimize the cost function for  $D(x) = \exp[-x/\xi]$  with  $\xi = 1.5 \mu\text{m}$ .**

	Random input	Enhancement	Suppression
$S_c$	0.24	0.095	0.47
$S_i$	0.23	0.45	0.32
$C \equiv S_c/S_i$	1.05	0.21	1.47

As given in the main text, the total intensity of the reflected waves is  $R = \sum_m \alpha_m$ , where  $\alpha_m$  is the contribution of the  $m$ -th mode to the reflection.  $\alpha_m = |\alpha_m| e^{i\theta_m}$  is a complex number, where  $\theta_m$  represents its phase.

$$S_c \equiv \left| \sum_m \alpha_m \right|^2 = \sum_m |\alpha_m|^2 + \sum_{i,j,j \neq i} |\alpha_i| |\alpha_j| \cos(\theta_i - \theta_j) = S_i + S_f, \quad (4)$$

where  $S_c \equiv |\sum_m \alpha_m|^2$  is called the coherent sum,  $S_i \equiv \sum_m |\alpha_m|^2$  is the incoherent sum, and  $S_f \equiv \sum_{i,j,j \neq i} |\alpha_i| |\alpha_j| \cos(\theta_i - \theta_j)$  is the interference term. Thus the ratio of coherent sum to the incoherent sum,  $C \equiv S_c/S_i = 1 + S_f/S_i$ , describes the effect of modal interference on the reflection.

Table 3 gives the values of the coherent sum and incoherent sum, as well as their ratio for the random input wavefront and the optimized wavefronts to maximize or minimize the cost function  $\eta_s$  for  $D(x) = \exp[-x/\xi]$  with  $\xi = 1.5 \mu\text{m}$ . With random wavefront illumination, the coherent sum is nearly equal to the incoherent sum, indicating the interference term is almost zero. With the optimized wavefront for maximal  $\eta_s$ , the coherent sum is significantly smaller than the incoherent sum, and the interference term is negative, thus the modes interfere destructively to suppress reflection. With the optimized wavefront for minimal  $\eta_s$ , the coherent sum becomes larger than the incoherent sum, and the interference term is positive, thus the modes interfere constructively to enhance reflection.

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- [1] Ito, S. *et al.* Fabrication of screen-printing pastes from TiO<sub>2</sub> powders for dye-sensitised solar cells. *Prog. Photovoltaics* **15**, 603–612 (2007).
  - [2] Garcia, N., Genack, A. Z. & Lisyansky, A. A. Measurement of the transport mean free path of diffusing photons. *Phys. Rev. B* **46**, 14475–14479 (1992).
  - [3] Rivas, J. G., Sprik, R., Soukoulis, C. M., Busch, K. & Lagendijk, A. Optical transmission through strong scattering and highly polydisperse media. *Europhys. Lett.* **48**, 22–28 (1999).
  - [4] Lagendijk, A., Vreeker, R. & de Vries, P. Influence of internal reflection on diffusive transport in strongly scattering media. *Phys. Lett. A* **136**, 81–88 (1989).
  - [5] Zhu, J. X., Pine, D. J. & Weitz, D. A. Internal reflection of diffusive light in random media. *Phys. Rev. A* **44**, 3948–3959 (1991).
  - [6] van Rossum, M. C. W. & Nieuwenhuizen, Th. M. Multiple scattering of classical waves: microscopy, mesoscopy, and diffusion. *Rev. Mod. Phys.* **71**, 313–371 (1999).