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Author(s)	Jayachandra, Bangi; Hemalatha, M.; Anita, R. W.; Vijayan, C.; Murukeshan, Vadakke Matham
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Asymmetric transmission and optical low-pass filtering in a stack of random media with graded transport mean free path.

Jayachandra B [1,2], Hemalatha M[2], Anita R W[2], Vijayan C[2], Murukeshan V M[1]*

1. Center for Optical and Laser Engineering (COLE), School of Mechanical and Aerospace Engineering, Nanyang Technological University, SINGAPORE.
2. Department of Physics, Indian Institute of Technology Madras, Chennai-INDIA.

*Correspondence E-mail: mmurukeshan@ntu.edu.sg

Abstract: Light transport and the physical phenomena related to light propagation in random media are very intriguing, they also provide scope for new paradigms of device functionality, most of which remain unexplored. Here we demonstrate, experimentally and by simulation, a novel kind of asymmetric light transmission (diffusion) in a stack of random media (SRM) with graded transport mean free path. The structure is studied in terms transmission of photons propagated through and photons generated within the SRM. It is observed that the SRM exhibits asymmetric transmission property with a transmission contrast of 0.25. In addition, it is shown that the SRM works as a perfect optical low-pass filter with a well-defined cutoff wavelength at 580 nm . Further, the photons generated within the SRM found to exhibit functionality similar to an optical diode with a transmission contrast of 0.62. The basis of this functionality is explained in terms of wavelength dependent photon randomization and the graded transport mean free path of SRM.

Key words: Asymmetric transmission, random media, optical low pass filter, optical diode.

Introduction

A random medium can host very interesting physical processes that form the basis for development of photonic devices. Many interesting concepts and properties of these disordered media were reported in the recent past [1-7]. They include reciprocity in multiple scattering [1], strong localization (Anderson localization) [2], interference effects (Speckle pattern) [3], random lansing (intense light-matter interaction) [4] and near and far field optical imaging [5]. However, the fabrication of photonic devices based on random medium is a relatively unexplored area, except for a limited number of attempts reported in literature, such as a compact spectrometer [6] by Brandon Redding et al, and a random super prism wavelength meter [7] by Michael Mazilu et al. Both these reports were based on the wavelength dependent speckle pattern in random media.

Scattering in disordered media depends on the particle shape, size, refractive index and the refractive index of the surrounding medium. Controlling these parameters enable tuning of the scattering wavelength windows. Wavelengths that undergo high scattering have a lower value of the transport mean free path, l_t, that determines the scattering strength of disordered medium. Short transports mean free paths imply a larger extent of randomization of photons with longer path lengths inside the medium. Transmission through the disordered medium in diffusive regime is proportional to l_t/L, where L is thickness of the medium. Photons with low light, stay in the medium for longer time and can be harvested efficiently [6, 8]. Those wavelengths within this scattering window cannot travel through the random medium, which could function as a stop band filter for this window. This virtue of photon filtering can be easily tunable as per the choice. Hence, a stack of such filters with different stop bands can result an asymmetric structure with graded transport mean free path. The phenomena and optical properties of asymmetric random environments are explored only in theoretical stands [9] not much explored experimentally. In this context, this work focusses on the light propagation in a stack of random media with graded transport mean free path and demonstrates the asymmetric transmission phenomena with broadband operation and reasonable transmission contrast. Here we considered two cases, first, how the photons from outside to transmit through the SRM. Second, how the photons generated within the SRM transmit through it. This work finds the applications in display technologies [10] and random media based photonic devices such as low-pass filters and optical diodes.

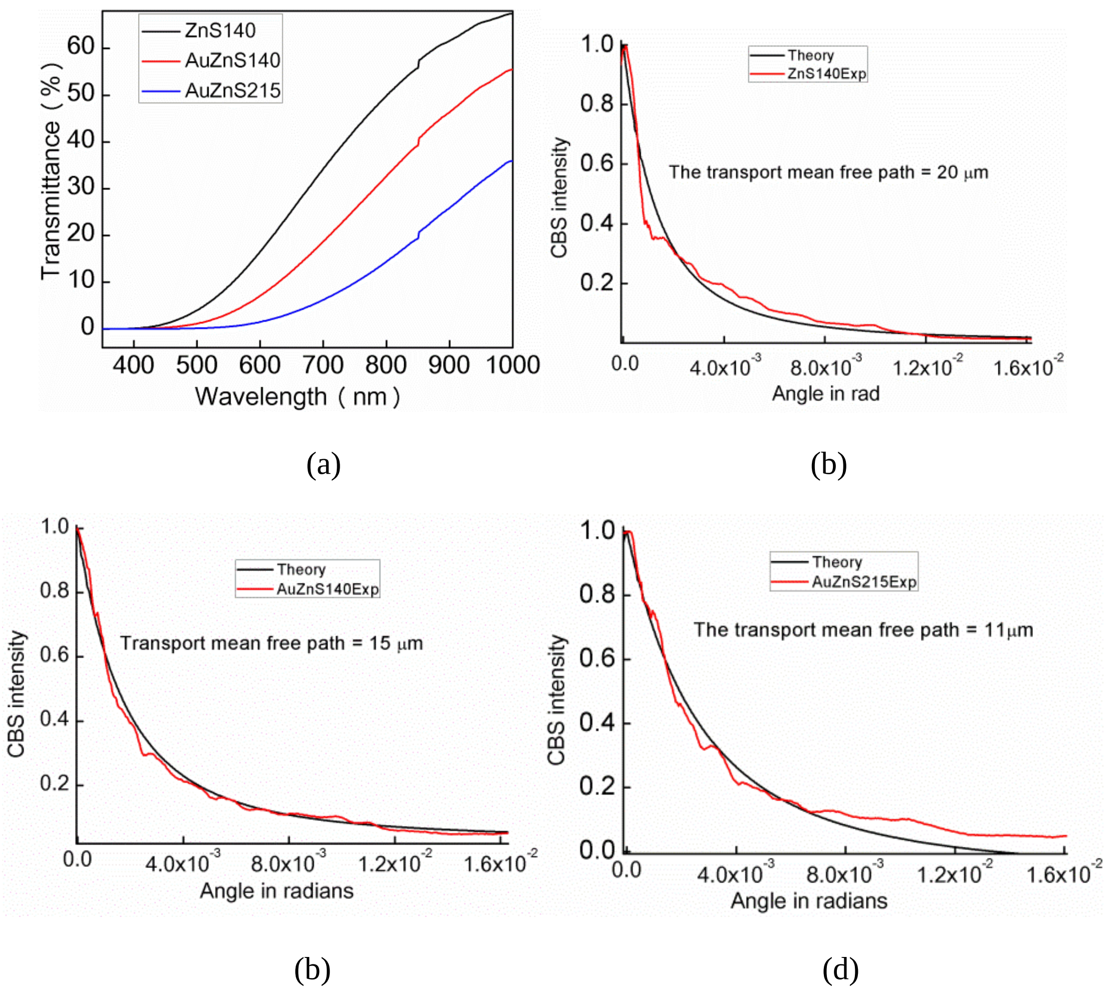

Fig.1. (*a*) Transmission Spectra of different stop band filters showing the filtering effect of random media with differing particle sizes. The coherent backscattering profiles of the samples

(*b*) ZnS140, (*c*) AuZnS140, and (*d*) AuZnS215 with calculated transport mean free path 20 µm, 15 µm and 11 µm respectively.

The basic structure proposed here is a random medium based stop band filter which is a stack of three different random media (See supplementary information for ZnS, Au particle synthesis). Three colloidal random media coded as ZnS140, AuZnS140, and AuZnS215, are prepared by using ZnS nanospheres of mean diameter 140nm, 215 nm respectively and Au nanoparticles of size 10-15 nm. The colloid is prepared by mixing 5mg/ml of ZnS140, ZnS215 spheres in Ethylene glycol - gold colloid. The random medium is characterized by transmission and coherent backscattering experiments. ZnS140, AuZnS140, and AuZnS215 work as a stop band filters up to 400 nm, 500 nm and 600 nm respectively as shown in Fig.1a. Here the gold nano particles role is to strengthen the scattering.

The transport mean free path (at 405 nm wavelength) which is the measure of scattering strength and randomness, is calculated from coherent backscattering profiles [11] from the respective random media ZnS140, AuZnS140, AuZnS215 shown in figure.2. The transport mean free path values are 20±1 µm, 15±1µm and 11±1 µm for ZnS140, AuZnS140, and AuZnS215 respectively. Hence, using these random media in series results the graded transport mean free path structure and at the same time structure is asymmetric. This indicates that the wave travelling through the three stage device experience the asymmetry with respect to transport mean free path from forward to reverse directions.

Figure 2 shows the schematic diagram of the experimental configuration used for transmission properties of the structure and the inset shows a representative schematic diagram of the SRM. The stacked media consisting of three zones is fabricated using ordinary cover glass slides (refractive index – 1.5) and each zone is about 2 mm, 5 mm, 10 mm thickness, width and height respectively. Each zone is separated with other zone by a glass slide of thickness 1mm. It is confirmed the same integrated intensity output without sample in both directions (Forward and reverse). Colloids ZnS140, AuZnS140 and AuZnS215 are filled in zone1, 2 and 3 respectively. This stack with three zones of random media as detailed earlier was fabricated using ordinary cover glass slides and each zone is separated by a glass plate as shown in the inset of Figure. 2. The three random media (ZnS140, AuZnS140, and AuZnS215) were stacked such that a certain wavelength band is filtered in each stage. Zone1, 2 and 3 represent the ~400 nm, 500 nm, and 600 nm filters respectively. Here the light propagation is in two directions, one is the forward propagation (Zone 1→2→3) and the reverse propagation (Zone 3→2→1). The integrated light signal is collected at the exit window of Zone 3, or Zone 1 for forward and reverse propagation directions respectively by a convex lens of 40 mm diameter.

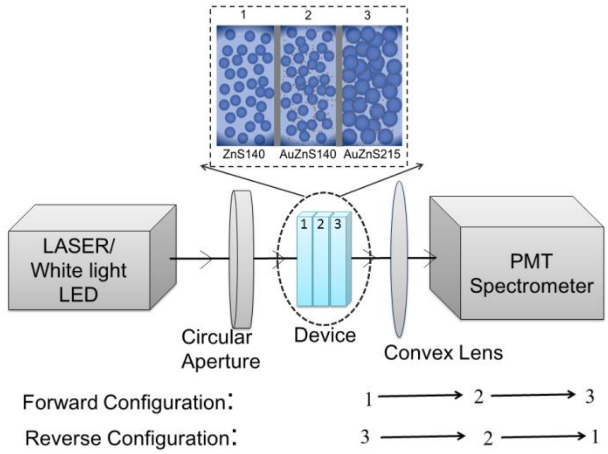

Fig. 2. The optical configuration used to study the performance of photonic diode. Inset shows the random media based stop band filter stack, which functions as the photon diode device.

The first case is to apply the photons from outside the structure and study the transmission characteristics of SRM. In this case, a white light LED is fixed at one end of the SRM and recorded the transmission of broadband spectra at the other end both in forward and reverse directions of SRM. Figure 3 shows the normalized transmitted intensity as a function of wavelength in both forward (Black) and reverse (Red) directions of the SRM. The blue curve is representing the spectrum of incident broad band light from LED. It can be inferred from Fig. 3 that the intensity propagated in the reverse direction is lower than that observed for the forward propagation indicating the asymmetric transmission property. In addition to the asymmetric transmission, it is clear that in both directions the propagation of wavelengths up to 580 nm is completely suppressed by SRM. Hence, from figure 3the SRM is working as an efficient optical low pass filter also, with a cutoff wavelength around 580 nm.

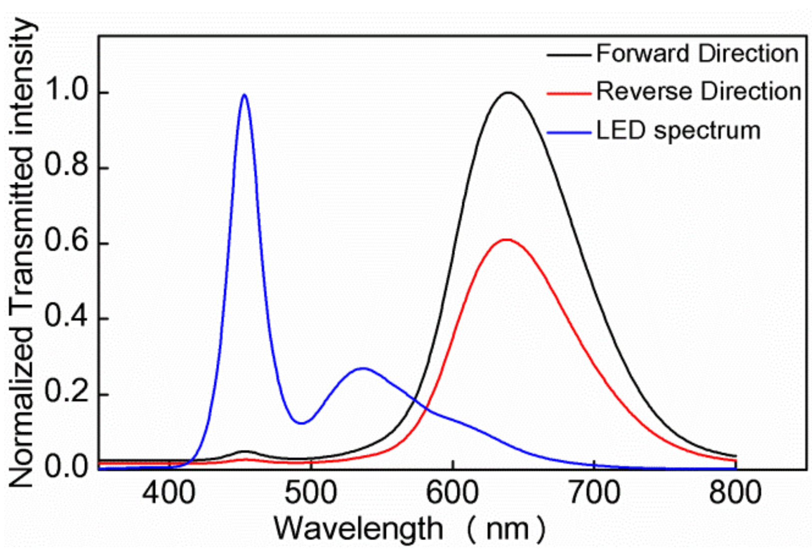

Fig.3. Experimental Normalized transmission in forward (Black line) and reverse (Red line) directions, the blue curve is the broadband light spectra from LED.

The asymmetric transmission is characterized by a parameter, transmission contrast (efficiency)

$$T_{Cont} = \frac{T_{For} - T_{Rev}}{T_{For} + T_{Rev}}$$

However, it is to be mentioned that the contrast can be altered by varying the random medium parameters such as particle size, refractive index, concentration (packing fraction) and the thickness of the random media zone. In the present case, the transmission contrast of broadband light through the SRM is calculated as 0.25.

In the second case, the propagation of photons generated within the SRM is studied. For this, a dye pyridine 2 of 1 μM concentration is introduced into each random medium and made this to distribute uniformly by stirring. So, now each stage became an active random medium with absorption and emission bands of 400 nm- 600 nm and 650-750 nm, respectively. The choice of emitter (dye) is to be optimized in order for it to absorb in the visible region as well as to induce emission at greater wavelengths beyond 600 nm. Explicitly, as the SRM working as a low pass filter, wavelengths < 600 nm are halted or attenuated inside the SRM due to strong scattering, at the same time these are absorbed by dye molecules. Now, the emitted photons $>$ 600 nm by dye molecules within SRM can transmit from excited end to the other end. Here the excitation can be made by source wavelengths between 400 nm to 600 nm. The signal at the other end (forward or reverse) can be between 650 nm to 750 nm.

The SRM is excited by 405 nm in both forward and reverse directions, and respective emission wavelengths were detected on the other end face using the same experimental setup shown in fig. 2. Figure 4a shows the emission signal peak at 690 nm, where it is evident that the forward signal is high compared to reverse signal with a transmission contrast of 0.625. It is to be mentioned here that the increased transmission is due to the dye as the emission source. Figure 4b shows the normalized output signal intensity as a function of input excitation power at an applied wavelength of 405 nm, in forward and reverse propagation directions. It is evident from the Fig. 4b that by increasing the input excitation power, the forward propagation intensity increases rapidly compared to that of reverse propagation. This behavior is similar to the electronic diode where electron flux (current) increases with increase in applied voltage. Figure 4c,d show the top view of the SRM in forward and reverse direction respectively. From the figure it is evident that the photons generated within the system after the excitation with 405 nm, have longer propagation distance in forward direction than reverse direction.

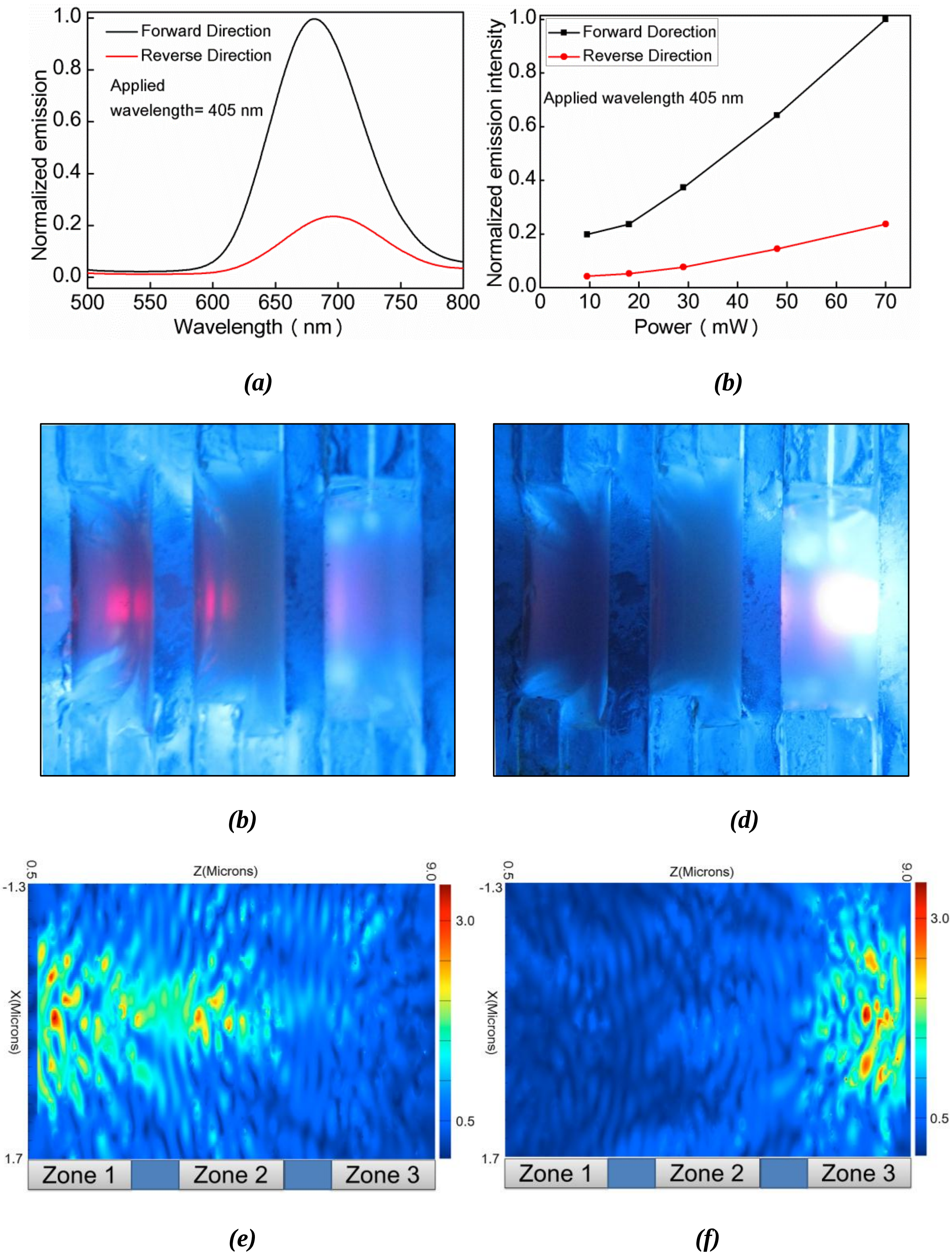

Fig. 4. (*a*, *b*) Emission field: forward and reverse field propagation (Gaussian beam of 405 nm wavelength). (*c*, *d*) The respective emission propagation captured on the device (Images taken from the top). (*e*) The plot of output emission signals in forward (Black curve) and reverse (Red

curve) propagations with excitation wavelength at 405 nm. (*f*) The plot showing the output emission signal variation with applied wavelength power.

To prove the concept qualitatively, the emission field propagation in the forward and reverse directions is simulated. An FDTD (finite difference time domain method, FDTD Lumerical solutions software) analysis was carried out by simulating the proposed SRM by applying PML boundary conditions and using suitable power monitors. Particle sizes were taken as 140 nm, 215 nm (for ZnS nanospheres) and 10-15 nm (for gold nanoparticles) respectively. The refractive indices of background medium and ZnS are taken as 1.45 and 2.35 respectively. The actual dimensions of SRM are (thickness-2 mm, width- 5 mm, height- 10 mm and zone separation- 1mm). For simulation purpose, SRM dimensions the sizes were taken as Thickness- 2 µm, width- 5µm, height- µm and zone separation = 1 µm) to validate the proof of concept. The field (650 nm-750 nm) propagation is quite similar to the wave propagation shown in fig.4 c, d, which proves that there is an asymmetric wave transmission for the wavelengths beyond the 600 nm.

The basic principle can be explained based on the randomization effect and the extended path lengths. As shown in Fig. 5, the excitation beam can propagate to a certain distance during forward propagation depending on its wavelength. Along the path the intensity of the beam gets reduced and some of the absorbed light gets re-emitted by fluorescence. The emission wavelength in this case observed to be above 600 nm. So if randomization is comparatively less in Zone 1 and 2, it can enter the zone 3. This results in a certain quantity of photon flux still reaching the output end face of the device after zone 3. At the same time in the reverse configuration both the excited beam and the emitted wavelengths get the strong randomization in zone 3 due to high scattering strength, after which the emission field has to travel across zone 1,2 where the signal get attenuated further due to extended path lengths. In other words the penetration depth is more for excitation beam in the forward direction than that for reverse configuration due to randomized photon paths with increased path lengths

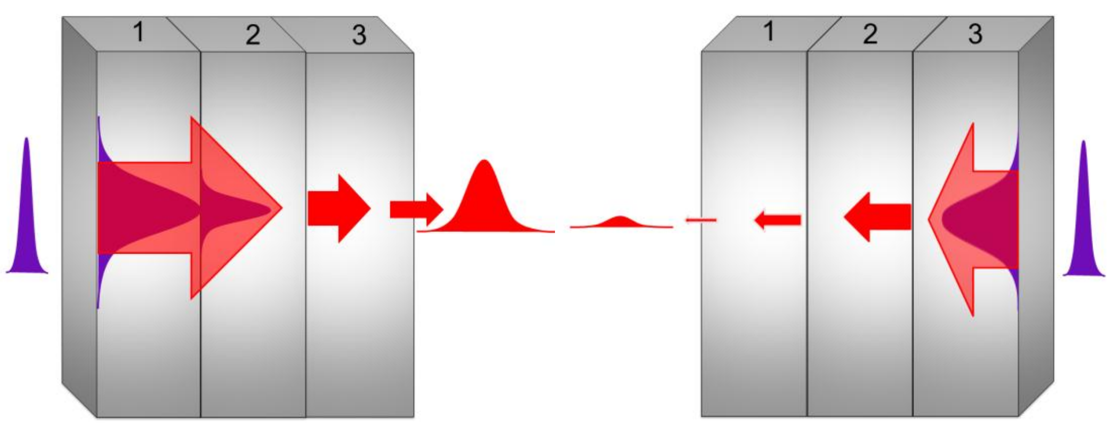

Fig.5. Photonic diode- schematic diagram representing the principle of field propagation in the (a) forward and (b) reverse propagation directions.

When the applied power is increased, the penetration depth increases in the forward configuration which increases the output signal significantly. However, in reverse direction, penetration depth will not change much, this results in the slowly increasing output signal even though input power is increased (See supplementary information for laser power dependent penetration depth explanation).

In conclusion, the asymmetric random medium is fabricated by stacking colloidal random media with graded transport mean free path. The broadband light propagation through the SRM revealed that the system is exhibiting an asymmetric transmission. Further, it is illustrated that the SRM also shows a good optical low pass filter property with a cutoff at 580 nm wavelength. The propagation study of photons which are generated within the SRM has shown an asymmetric behavior which indicate that this functionality can be extended to the optical diode behavior. With transmission contrast 0.62. The physical concept is explained with simple picture based on the photon randomization in SRM with graded transport mean free path. It is envisaged that this demonstrated gradient transport mean free path structures and new concepts can enable the realization of using random media with micro and macro size for different device applications.

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