Electronic Supplementary Material (ESI) for Nanoscale Horizons.

Supplementary Materials

Photonic artificial synapse with reversible multifaceted photochromic compound

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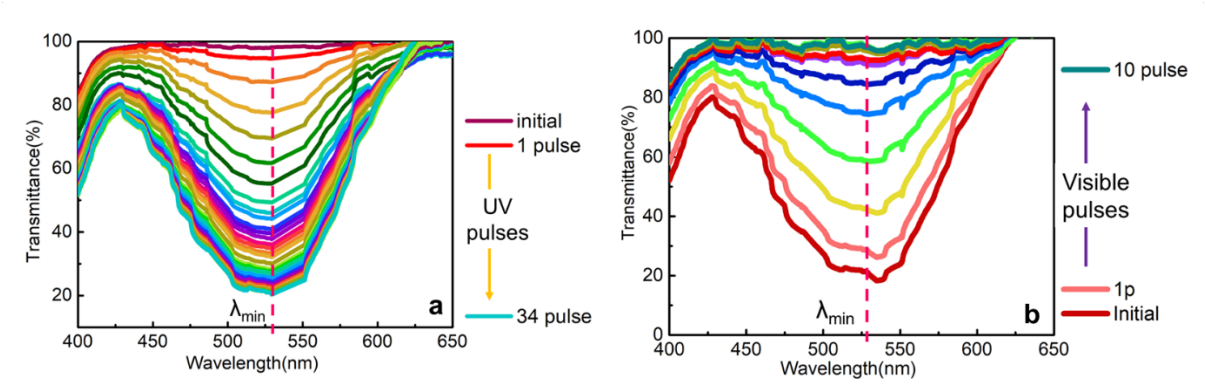
1. Impact of UV and Visible Light Pulses on the transmission spectra-

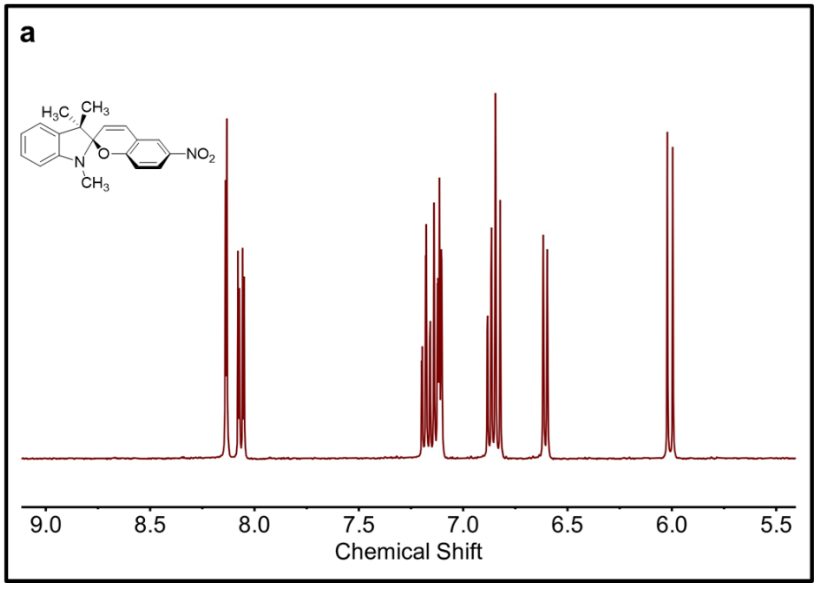
The full impact of UV and visible light pulses on the transmission spectra of spiropyran is presented. The 30 seconds pulse in each case results in the transmission intensity changes. Considering the colorless state as the initial state, the transmission reduces due to the UV pulses to 20%, leading to more merocyanine isomers in the solution with a peak minimum at 527.5 nm due to more merocyanine isomers in the solution (Figure S1(a)). While with the dark pink state as the initial state, visible light pulses turn the solution colorless, signifying high spiropyran isomer concentration (Figure S1(b)). The arrows show the direction of transmission spectra change.



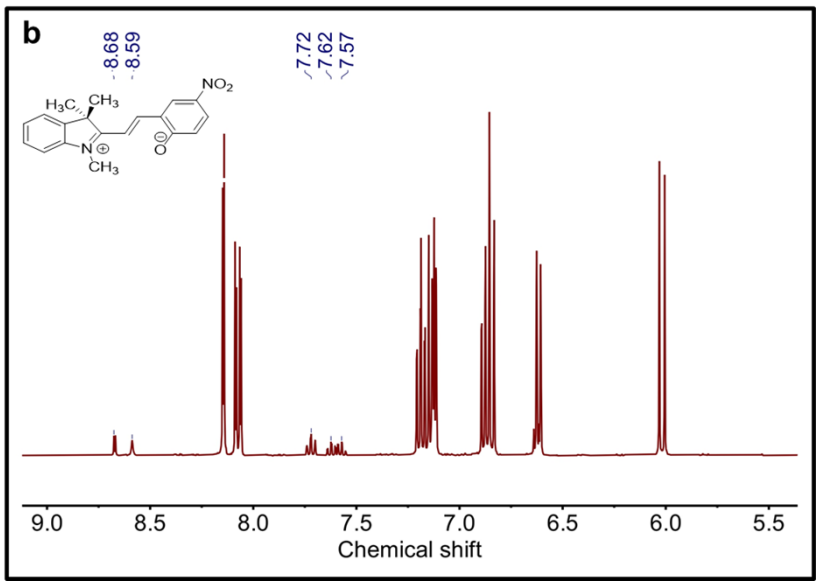
FigureS1. (a) The colorless-to-dark pink transition of the transmission spectra with UV light pulses of 30 seconds. (b) The dark pink-to-colorless change of transmission spectra with visible light pulses of 30 seconds.

2. ^{1}H NMR spectra of Spiropyran and Merocyanine-

A deuterated version of methanol (CD_3OD) was used for the ^{1}H NMR studies of both spiropyran and merocyanine isomers. Spiropyran solution was irradiated with visible light to convert it into a colorless form. When illuminated with ultraviolet, the same solution shifts to a dark pink solution. The NMR spectra of merocyanine (figure. S2(b)) show extra peaks, proving the existence of open chain merocyanine isomers[1].



FigureS2(a). ^{1}H NMR spectra of Spiropyran showing the intensity peaks of the molecule.



FigureS2(b). ^{1}H NMR spectra of merocyanine showing extra intensity peaks signifying the presence of merocyanine isomers.

3. Spiropyran-to-merocyanine conversion in a polar and non-polar solvent

The charge distribution in the spiropyran isomer differs from that in merocyanine, which is easily affected by the polarity. The polar nature of the solvent is responsible for forming merocyanine without UV radiation when placed in the dark. The strong solute-solvent interactions will change the energy levels of the ground and the excited state of merocyanine form, where the molecule experiences electronic transition without any external excitation affecting the colour of the molecule. Due to such interactions, the C-O bond breaks in spiropyran, forming merocyanine, giving rise to the light pink intermediate state. This effect is not observed in the case of non-polar solvents, where an external UV is required to achieve merocyanine to form as the reaction is non-spontaneous (figure S3). Considering polar solvents such as IPA, ethanol, and methanol, with increasing polarity, the interaction of merocyanine changes with the solvent, which increases the band gap and hence the color[2].

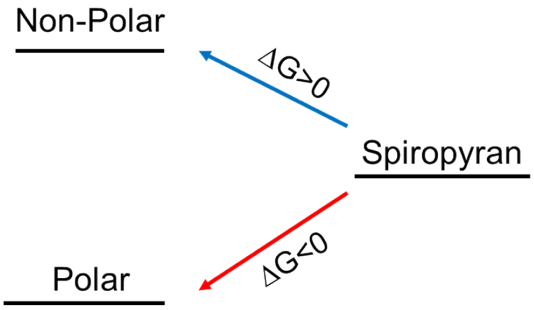


Figure S3. Spiropyran-to-merocyanine conversion in a polar and non-polar solvent. The reaction is spontaneous in a polar solvent, whereas in a non-polar solvent, the process is non-spontaneous.

4. Comparison between all-optical artificial synapse with three terminal transistors.

We can compare the all-optical artificial synapse with a three-terminal transistor-like device. The changes in current in the transistor due to an electrical spike in gate voltage are replaced with the optical transmission, either using a UV pulse or a visible light pulse. When a gate pulse voltage is applied, it changes the source-drain current, which is nothing but a modification of synaptic weight. The UV and visible optical pulse trigger a photochromic spiropyran-merocyanine isomerization that changes optical transmission through the sample. This change in the transmission is utilized in designing the all-optical synapse. The comparison of an all-optical artificial synapse with a three-terminal device-like structure is shown in figure.

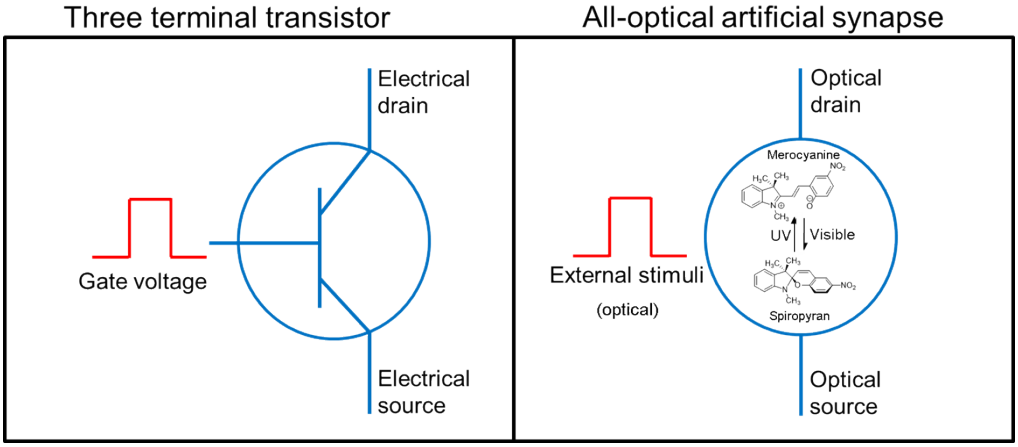


Figure S4. Comparison between three-terminal transistor and all-optical artificial synapse.

5. Spiropyran polymer composite

Poly (methyl methacrylate) PMMA polymer is used in the experiment to prepare spiropyran polymer film. In 5mL of CH_2Cl_2 (dichloromethane or DCM), a 0.3g polymer was added and stirred well for about 20 minutes. After the polymer solution is ready, spiropyran (2 wt% of polymer) is added and mixed until a homogenous solution is observed. The prepared liquid polymer was spin-coated on the quartz substrate[3].

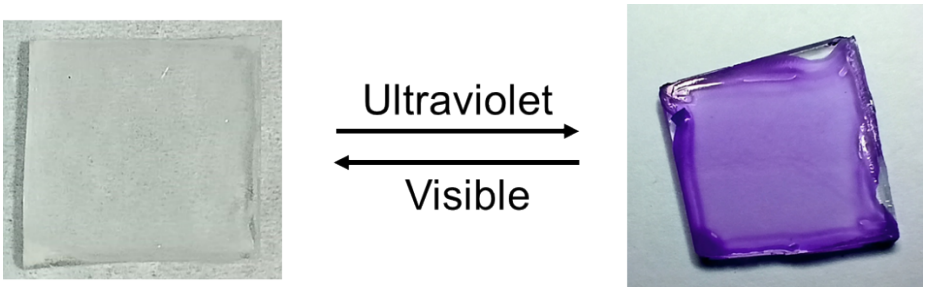


Figure S5. Spiropyran and merocyanine isomers are switched when the molecule is dispersed in PMMA and spin-coated on a glass substrate.

The polymer deposited on the quartz substrate was irradiated with visible-UV light. The transmission spectra show a minimum of 571 nm in the case of PMMA polymer film with UV irradiation. This might be due to the changing environment and different solute-solvent interactions. Spiropyran thin films coated on quartz appear uniform and smooth in optical images. The optical image and its optical transmission are given in Figures S6(a & b). In the case of the spiropyran polymer film, there is no intermediate state observed as in the case of the liquid form (when the solution was kept in the dark, spiropyran spontaneously converted to merocyanine, giving light pink colour), so our measurement is only restricted to mimic the inhibitory nature of the synapse. Thus, the colorless (spiropyran) polymer changes to purple (merocyanine) on a UV pulse and returns to its original state in the dark, indicating the memory.

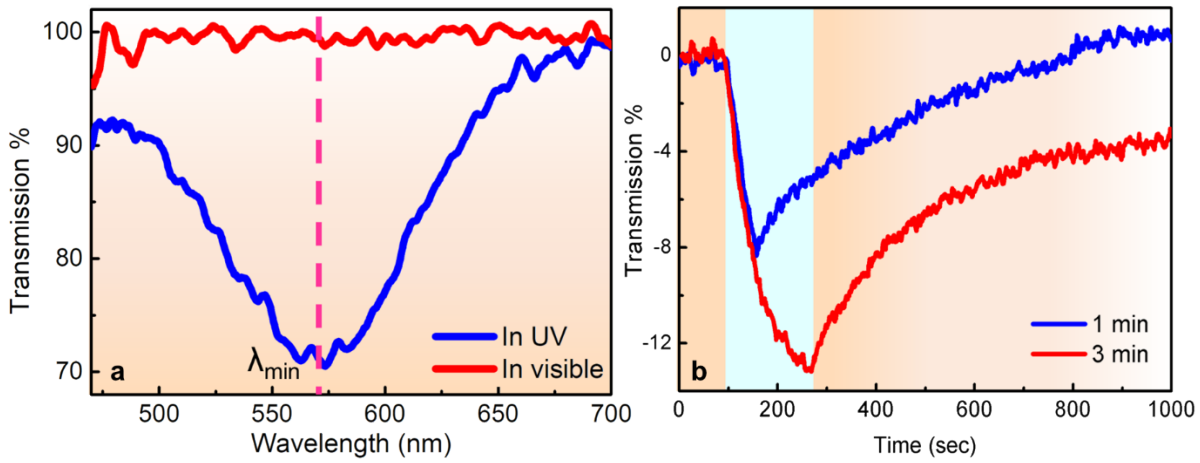


Figure S6(a). Transmission spectra show the colorless and purple transmission state intensities with λ_{min} at 571 nm. (b) Inhibitory synapse by changing time of the pulses in the spiropyran polymer film.

In the future, an artificial neural network from an all-optical artificial synapse can be constructed with a thin film or a polymer film of spiropyran showing all the functionalities as in the liquid state. An

intermediate state is required to observe both excitatory and inhibitory synapses. The films can be kept in the matrix format where a source light (I_0) is incident with a very low intensity similar to the resting potential in the biological synapses. This can be used to determine the changes in the optical transmission caused by the external stimuli in these films. For changing the synaptic weight of each film by optical pulses, each film can be supported with a separate excitation visible or UV light (can be LED). As per the requirement, the stimulating optical pulse (excitatory or inhibitory) can be given to change the transmission state of the film, which will modify the synaptic weights. As in the figure shown below, the transmission intensity of each stimulated synapse can vary from I_0 to I_2 depending on the nature of the stimuli (figure S7).

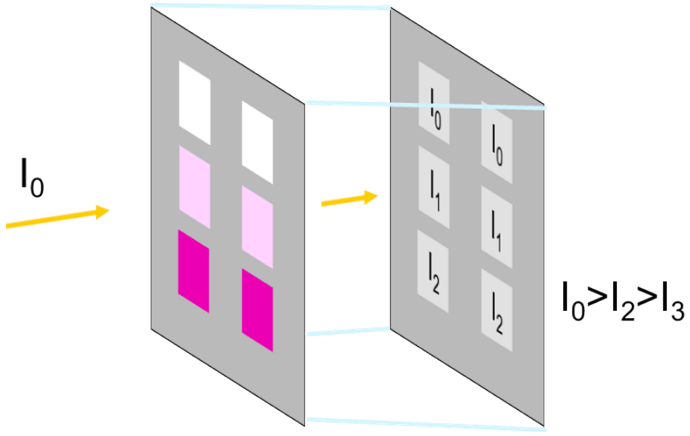


Figure S7. Proposed neural network for an all-optical artificial synapse.

References

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