

Magnetic response of chlorophyll self-assembly within hydrogel: a mechanistic approach towards enhanced photo harvesting

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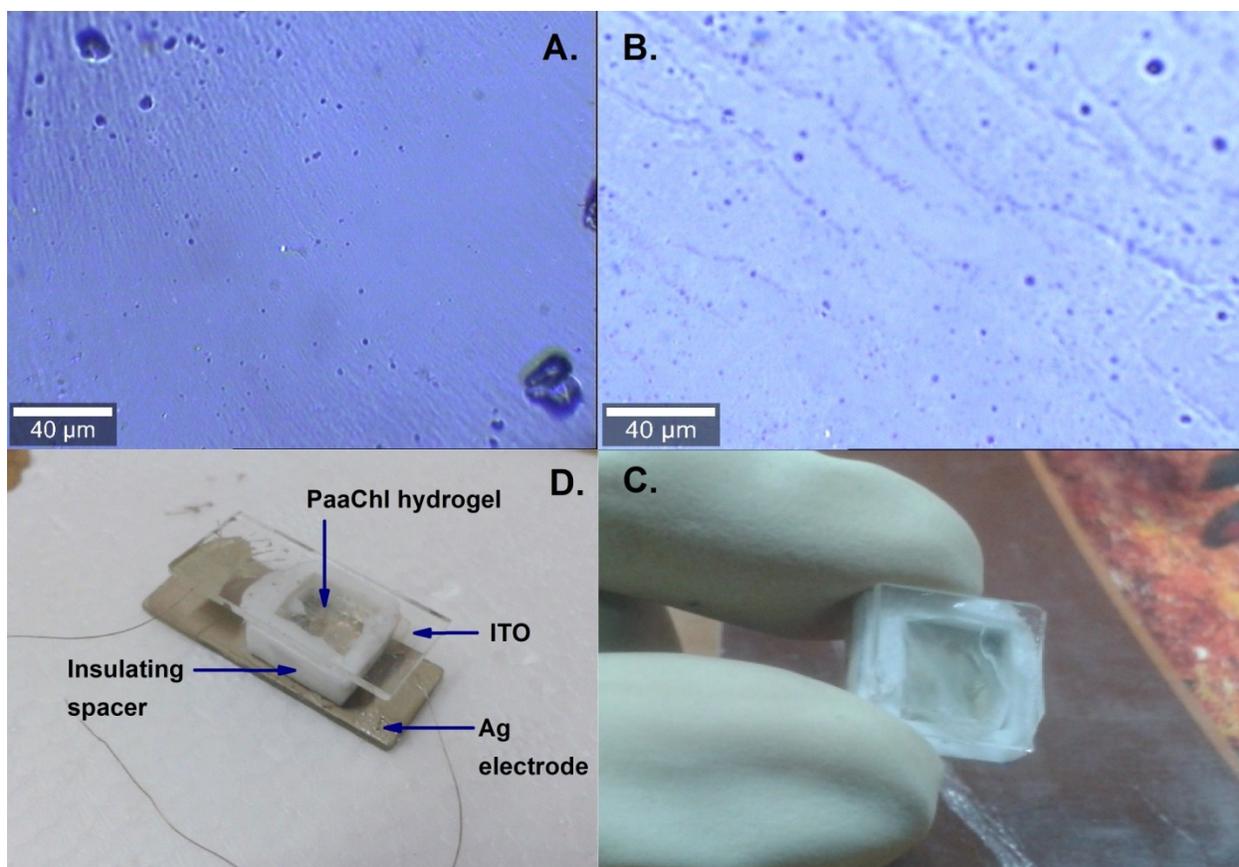


Fig. S1. Material and device images. A. & B. are the optical microscopic view of PaaChl entrapped Chl-a regions, C. & D. are the device image which was fabricated for the electronic characterizations.

Photophysical properties analysis:

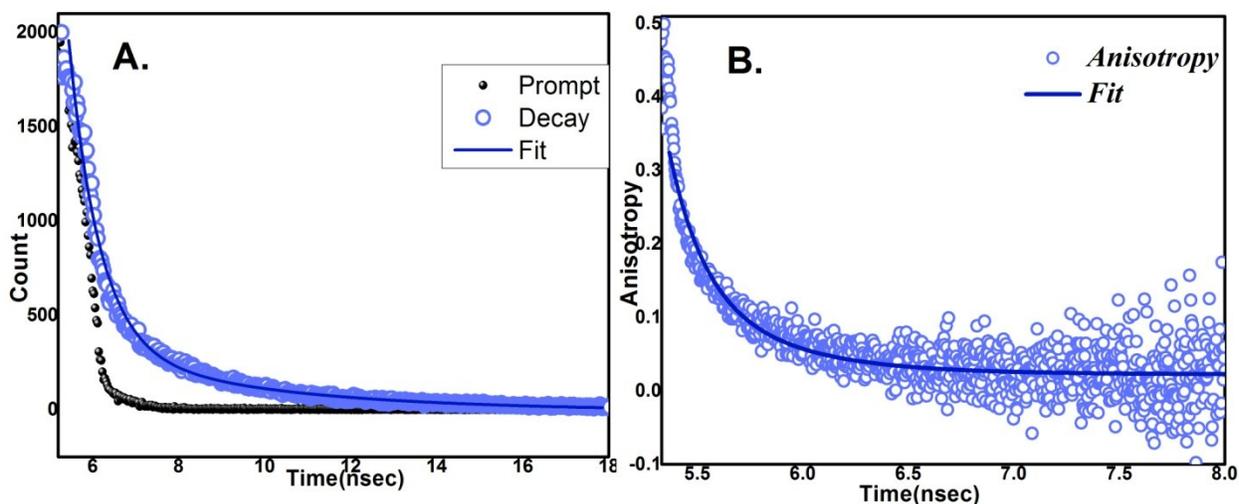


Fig. S2. A. Time correlated fluorescence spectroscopy & B. Fluorescence anisotropy of PaaChl revealing the presence of delocalized exciton and restricted spatial dynamics respectively in two different Chl-*a* assembly.

Exciton generation over chiral Chl-*a* assembly is evident through the life time calculation which suggests a coordinative homogeneous long range molecular pattern having different excitonic probability. Bi-exponentially fitted decay curve [Fig. S2.A] related to two Chl-*a* conformer [corroborating the presence of enantiomer in NMR spectra] having lifetime of 1.915 ns with 0.92 distribution ratio and excitonic-coherence length count of 4 Chl-*a* molecules [Table S1] indicates the presence of disorders as a result of site specific inhomogeneity [supported by steady state UV-absorption spectra; Fig. 1.A], which can induce dephasing to assist the excitonic quantum transport towards photo current generation. Decrease in average excited state lifetime from monomeric Chl-*a* can be correlated to intermolecular coordinative strength in 3D space. The homogeneous biphasic stress induced arrangement of chiral Chl-*a* assembly is also evident from bi exponentially fitted optical anisotropy curve [Fig. S2.B] showing relatively slower and steady decay at long time intervals, arising due to slow molecular motion as the assemblies are spatially

restricted in hydrogel matrix. Initial high optical anisotropy 0.5 suggests the strong dipolar interaction among excitonically coupled self-assembled Chl-a pointing towards restricted dynamics and lesser degree of spatial freedom of Chl-a assembly and supporting the presence of topological restriction.

Table S1. Parameters calculated from time correlated fluorescence decay data, Excitation wavelength: 405 nm; emission wavelength: 676 nm

Avg. excited state lifetime (ns)	Quantum yield	Radiative decay rate (ns ⁻¹)	Non-radiative decay rate(ns ⁻¹)	Dipole strength (D)	No. of molecules in coherence
1.915	0.34	0.178	0.34	1.03E-2	4

Table S2. Parameters calculated from bi-exponential fitting of fluorescence anisotropy decay curve of PaaChl

R	Exciton migration distance	τ_{ET}	τ_{Rot}
61 A°	58 A°	55 ps	0.44ns

R: Effective Radius of interaction; τ_{ET} : Faster decay component of anisotropy associated with energy transport; τ_{Rot} : Slower decay component of anisotropy associated with rotational relaxation.

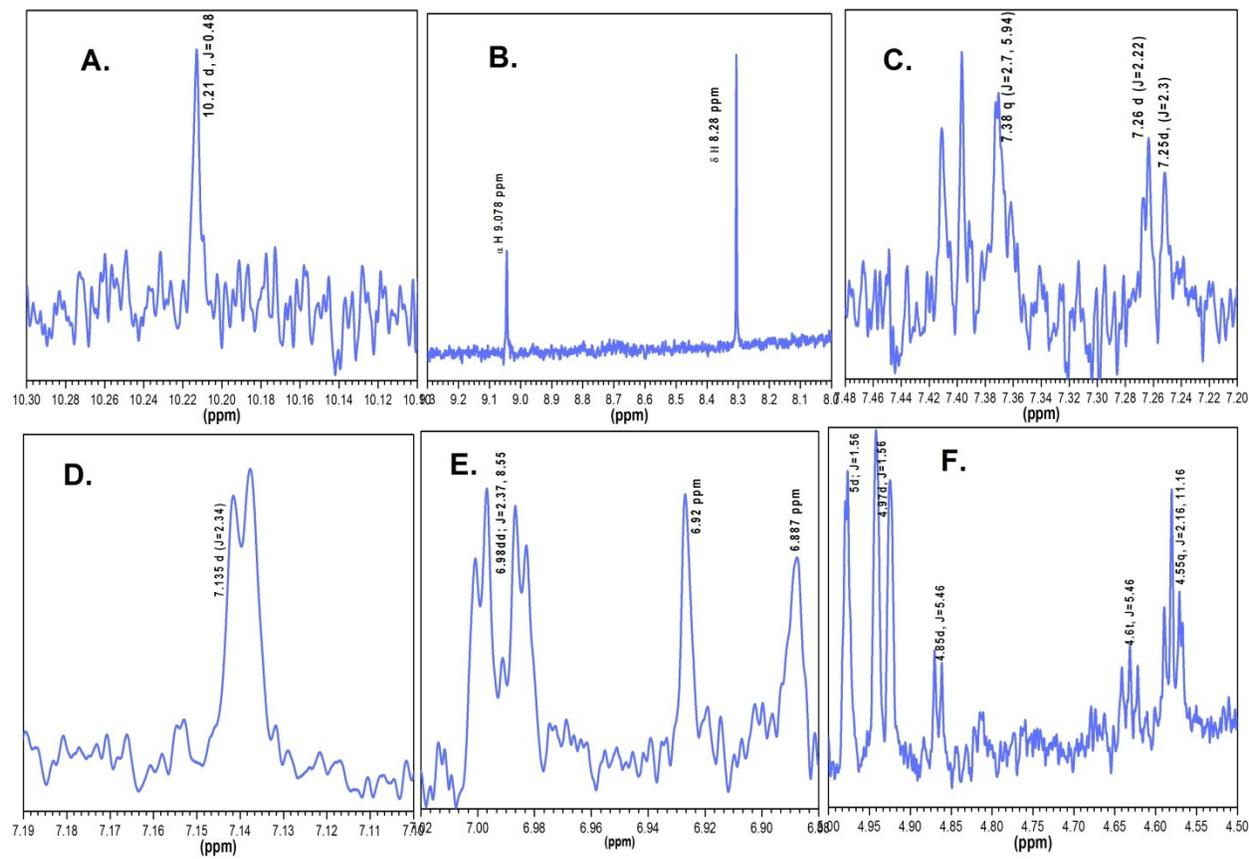


Fig. S3. ^1H NMR spectrum of PaaChl explicitly showing zones of dd splitting via Chl-a and Chl-a/matrix interaction.

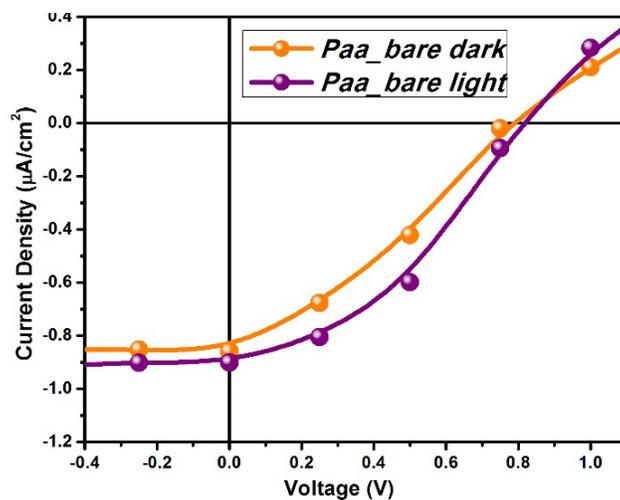


Fig. S4. Photo induced I-V curve of bare polyacrylamide hydrogel (Paa) showing no significant photocurrent generation.

Table S3. Photo I-V response of Paa

Jsc (μA/cm ²)	Voc (Volt)	FF	PCE (%)
0.91	0.8	0.55	0.025