

Awaneesh Singh<sup>a</sup>, Olga Kuksenok<sup>b</sup>, and Anna C. Balazs<sup>a\*</sup>

<sup>a</sup>Chemical Engineering Department, University of Pittsburgh, Pittsburgh, PA, 15261, USA

<sup>b</sup>Department of Materials Science and Engineering, Clemson University, Clemson, SC 29634,  
USA

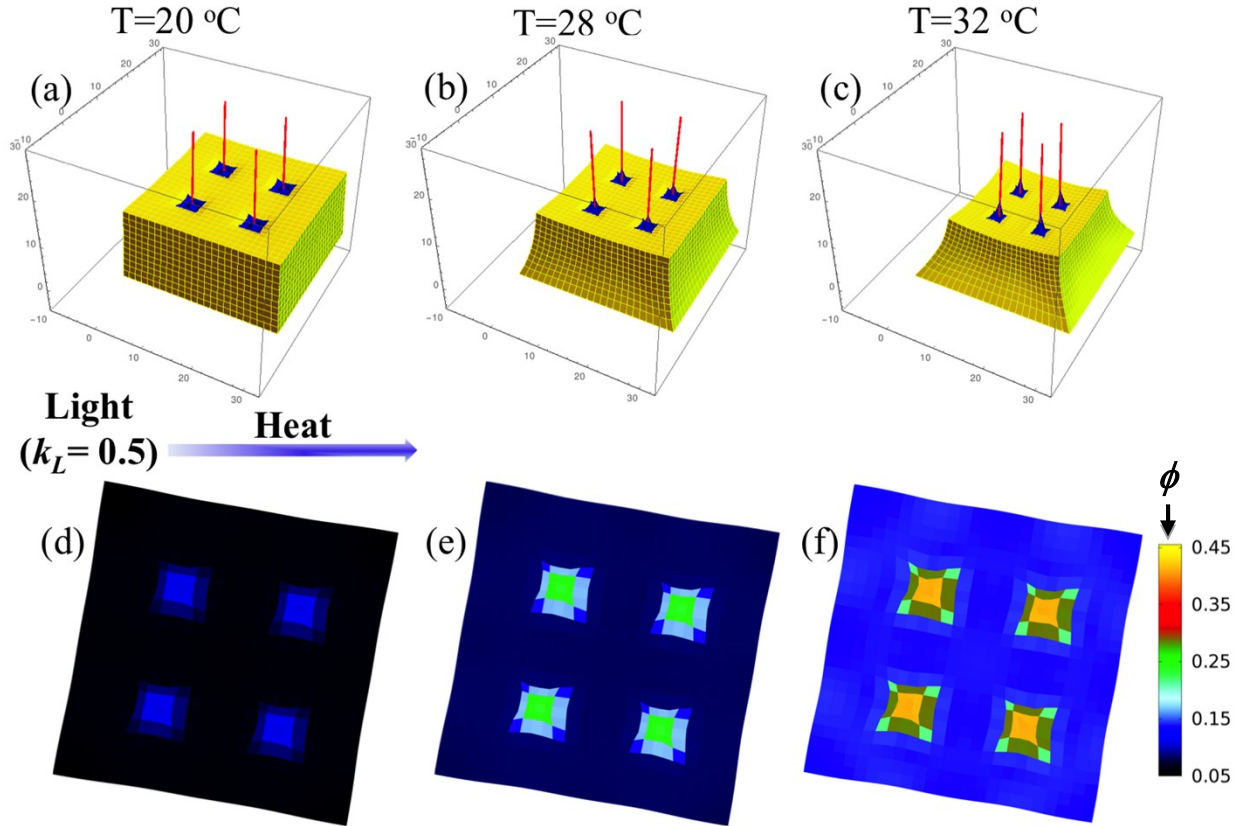
### Supplementary Information

The response of fiber-gel composite (gel embedded with four fibers arranged as in Figs. 2 and 3) under heat and light is depicted in Figs. S1 and S2, where the size of the SP-functionalized region around each fiber is set to  $R=3$  and  $R=4$ , respectively. Upon heating the sample under the light, the composite displays behavior similar to that in Fig. 3, where  $R=5$ . However, the composite response for  $R=5$  is more pronounced as shown by the temperature dependence of  $r_{ip}$  curves in Fig. S3.

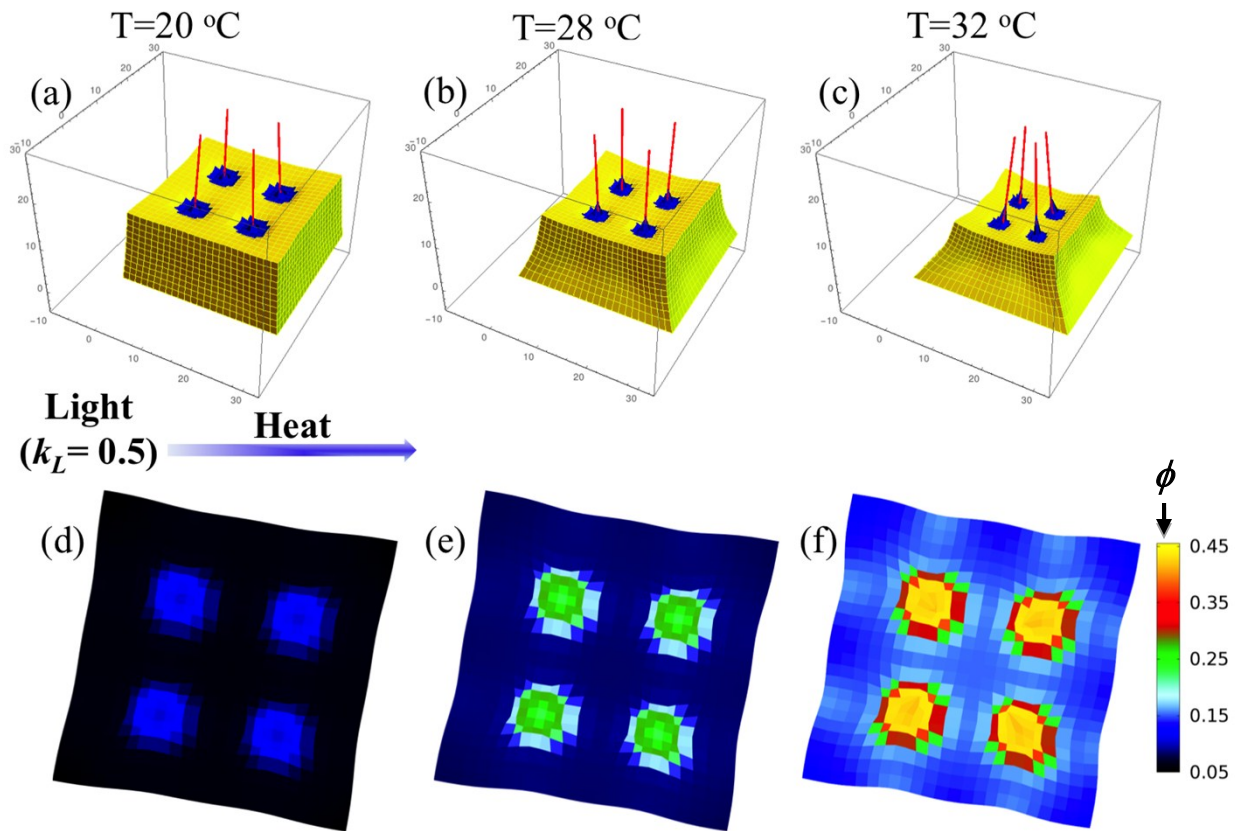
Further, we consider a sample with four fibers arranged symmetrically in a square pattern where each fiber lays ten elements (along the diagonal direction) from the center of the gel (see Fig. S4). The structural variation of the nonilluminated and illuminated samples under heat are displayed in Figs. S4 and S5, respectively. The size of the SP-functionalized region around each fiber is set to  $R=5$ . The dynamic response of heating the sample in the dark and the light are exhibited in Fig. S6. The structural variation and the dynamics of the sample are similar to the case displayed in Figs. 9-11, where the number of fibers was eight.

When the four fibers are placed at the edges of the sample (fourteen elements away from the center of the sample along the diagonal direction) with  $R=5$ , fibers bend towards center under the influence of heat in the dark (see Fig. S7); this response is more significant at  $T=32^{\circ}C$ . In the presence of light at  $T=20^{\circ}C$  fibers bend away from the center as displayed in Fig. S8a. Further, on heating the sample under the light, fibers again bend towards center due to global heating effect (see Fig. S8b-c). The dynamic behavior of the composites shown in Figs. S7 and S8, are displayed in Fig. S9.

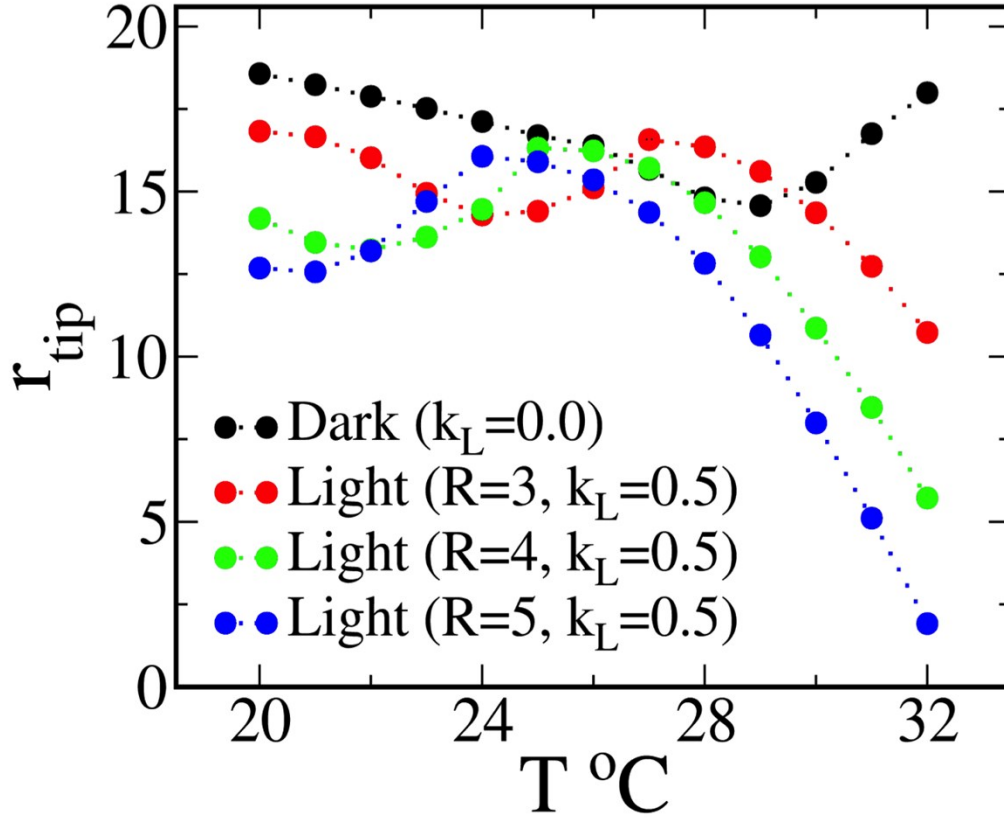
Finally, in Fig. S10 we demonstrate the structural variation of the sample and the dynamics (i.e., temporal variation of  $r_{ip}$ ) at  $T=32^{\circ}C$  when few selective fibers (“fingers”) are illuminated at a time. Figures S10a and S10b show the time evolution of  $r_{ip}$  for the case of four fibers (displayed in Figs. S7 and S8 respectively) when few selective fibers (indicated with symbols in the legend) are illuminated at a time. These plots are also compared with nonilluminated sample (solid blue line), and all illuminated fibers (dashed blue line). The inset shows the fiber numbering on the sample and the corresponding structural variation are presented in (I), (II), and (III).



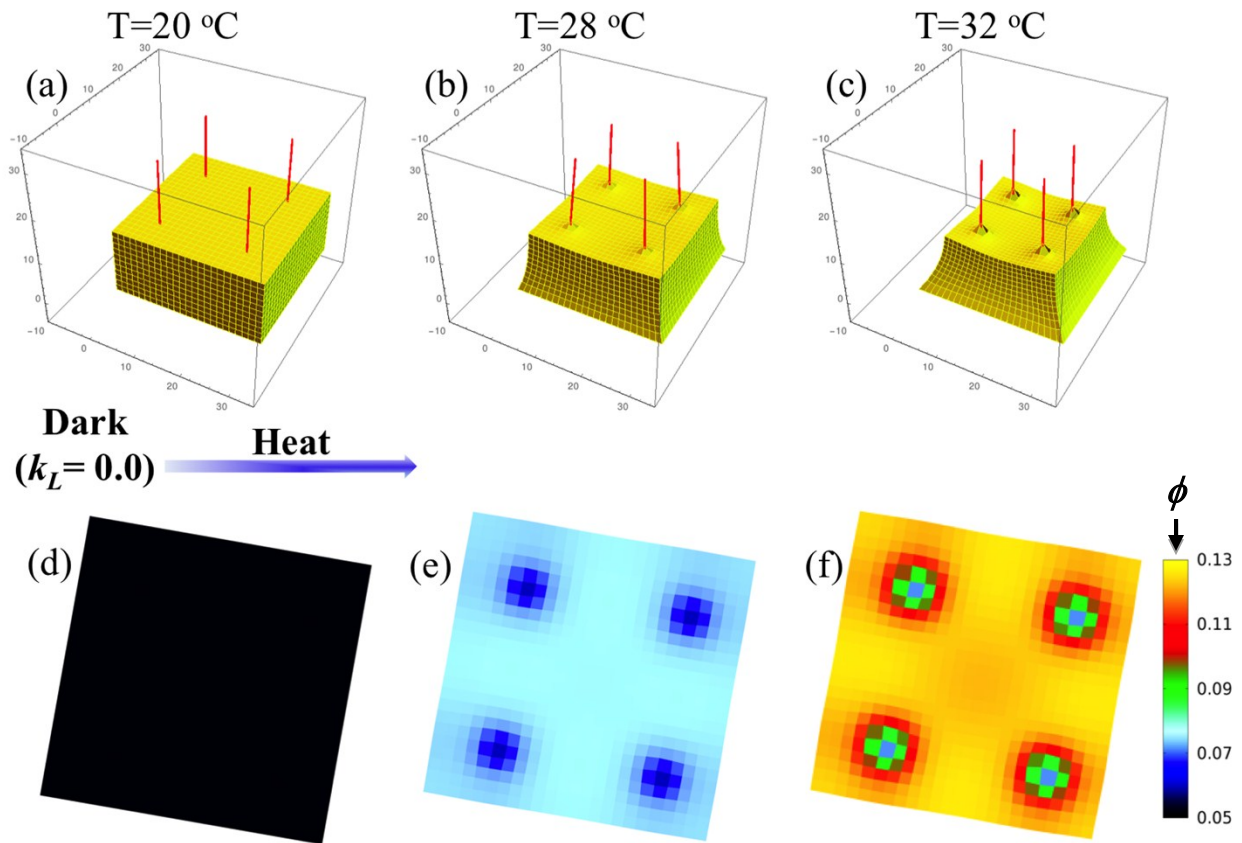
**Figure S1:** The late time response of the composite to light ( $k_L = 0.5$ ) for the size of SP-functionalized region  $R = 3$ . Initially, the fibers are arranged in the diagonals with separation between the fibers being equal to  $r_{tip} = 12$  elements. The gel sample size is  $25 \times 25 \times 12$ , the length of the fiber extending out of the gel is 14 dimensionless unit. (a) Late time response to the light at  $T = 20\text{ }^{\circ}\text{C}$ . In (b) and (c), the sample is heated to  $T = 28\text{ }^{\circ}\text{C}$  and  $T = 32\text{ }^{\circ}\text{C}$ , respectively, under light ( $k_L = 0.5$ ). (d-f) The polymer volume fraction,  $\phi$ , within the top layer of the gel, for the cases shown in (a-c).



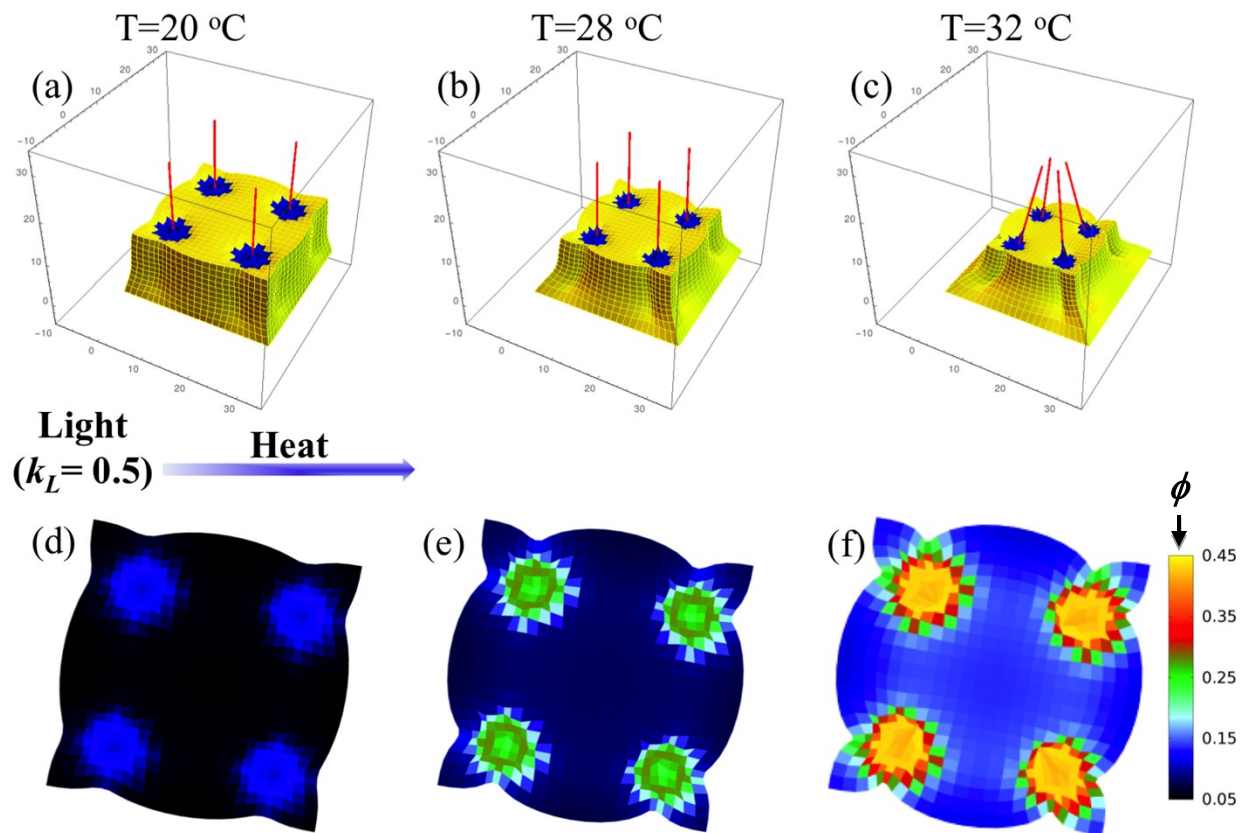
**Figure S2:** The gel-fiber composite response to light ( $k_L = 0.5$ ) for the size of SP-functionalized region  $R = 4$ . (a) Response to the light at  $T = 20^{\circ}\text{C}$ . In (b) and (c), the sample is heated to  $T = 28^{\circ}\text{C}$  and  $T = 32^{\circ}\text{C}$ , respectively, under light ( $k_L = 0.5$ ). (d-f) The polymer volume fraction,  $\phi$ , within the top layer of the gel, for the cases shown in (a-c)



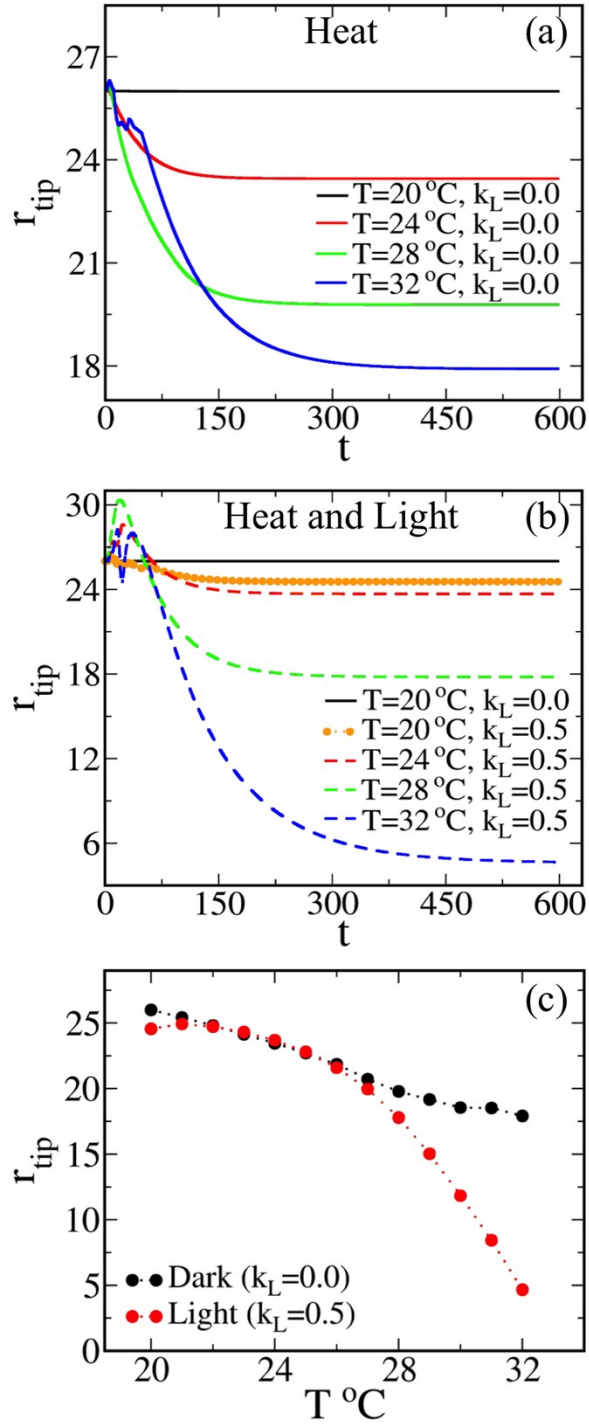
**Figure S3:** Comparison of the temperature dependence of  $r_{tip}$  after the sample has reached the equilibrium under the light ( $k_L = 0.5$ ) for  $R = 3$  (red curve),  $R = 4$  (green curve), and  $R = 5$  (blue curve). The black curve represents the sample in the dark ( $k_L = 0.0$ ). The temperature dependence of  $r_{tip}$  for this fibers arrangement emphasizes the non-monotonic behavior for each  $R$  value.



**Figure S4:** (a) Initial setup of the gel-fiber composite with four fibers arranged in the diagonals with separation between the tips being equal to  $r_{tip} = 20$  elements at  $T = 20^{\circ}\text{C}$ . The gel sample size is  $25 \times 25 \times 12$ , the length of the fiber extending out of the gel is 14 dimensionless unit. (b-c) The late time response of the composite to heat (at  $T = 28^{\circ}\text{C}$  and  $T = 32^{\circ}\text{C}$ ) in the dark ( $k_L = 0.0$ ). (d-f) The polymer volume fraction,  $\phi$ , within the top layer of the gel, for the cases shown in (a-c).

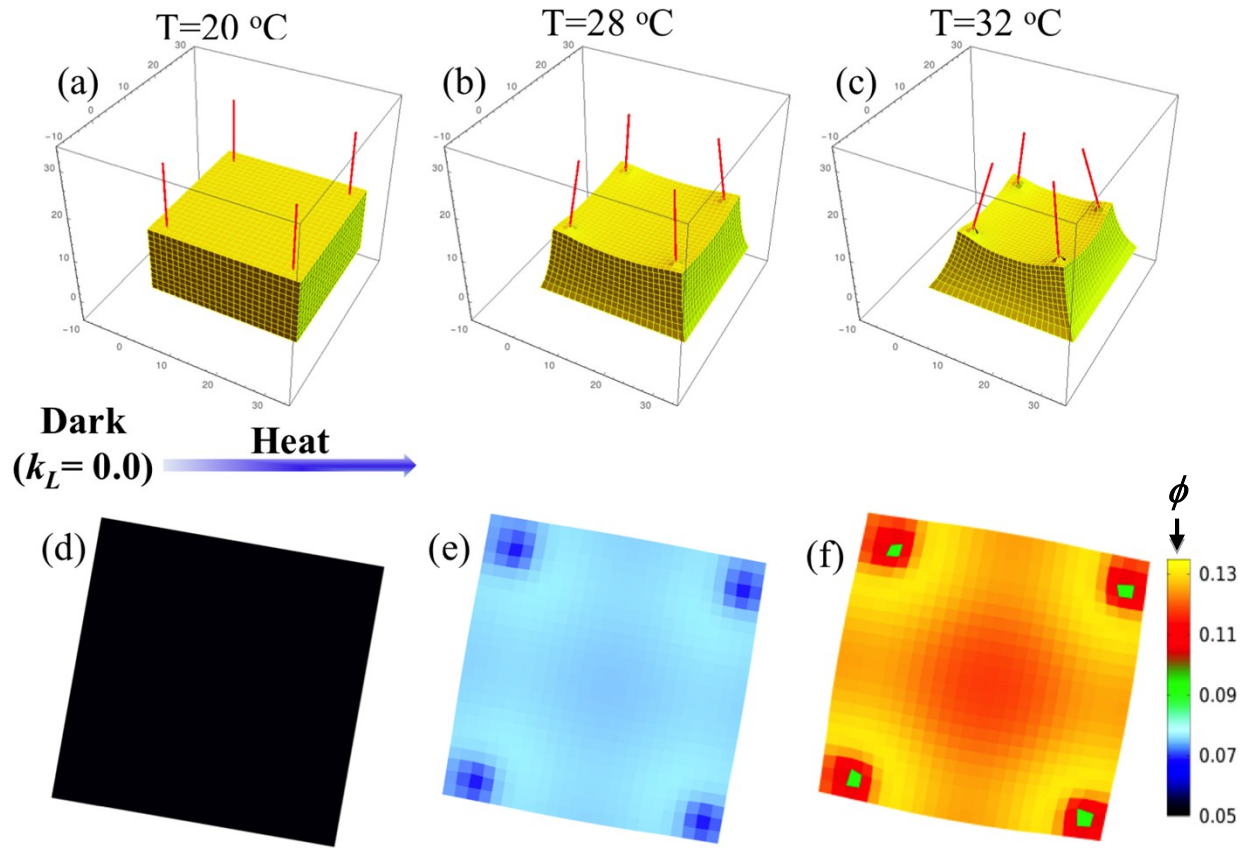


**Figure S5:** (a) The gel-fiber composite response to light ( $k_L = 0.5$ ) at  $T = 20^{\circ}\text{C}$  for the size of SP-functionalized region  $R = 5$ . In (b) and (c), the sample is heated to  $T = 28^{\circ}\text{C}$  and  $T = 32^{\circ}\text{C}$ , respectively, under light ( $k_L = 0.5$ ). (d-f) The polymer volume fraction,  $\phi$ , within the top layer of the gel, for the cases shown in (a-c). The fibers bend towards more collapsed region (high polymer volume fraction).



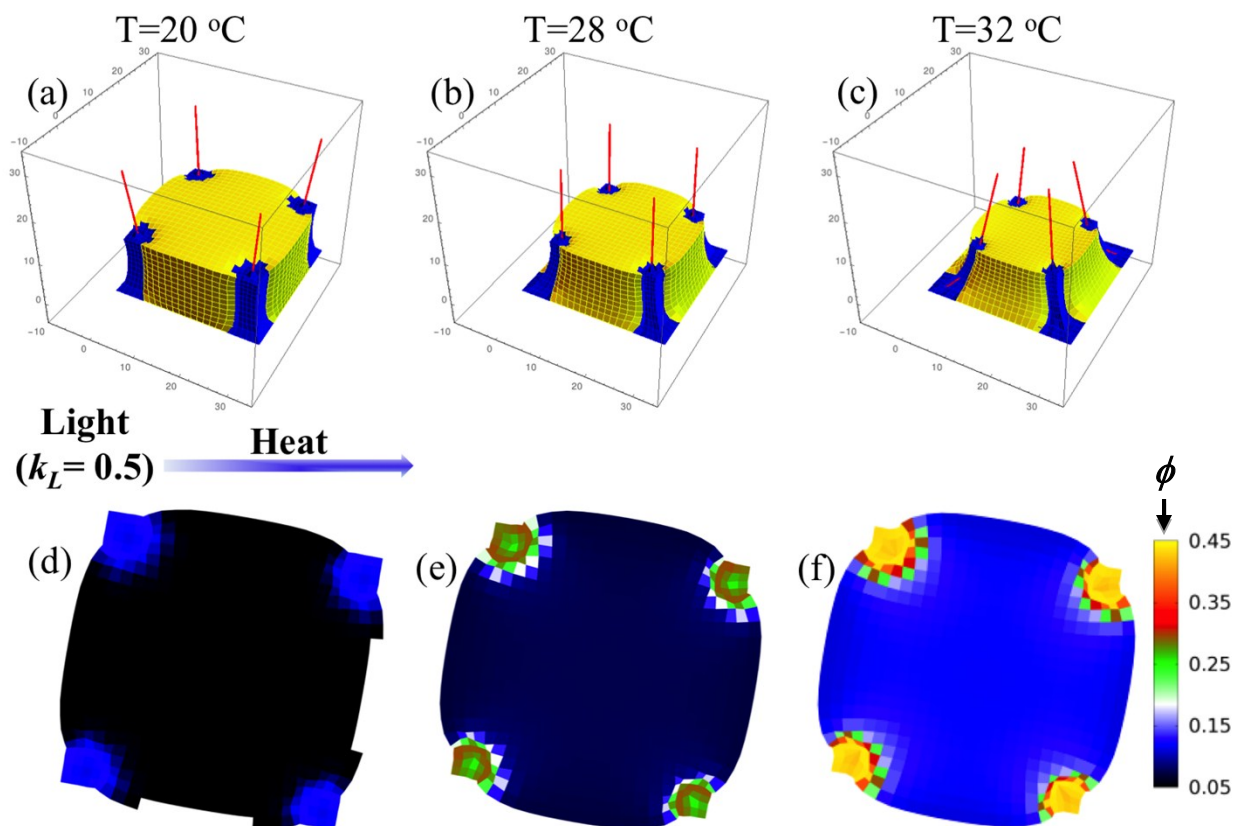
**Figure S6:** Time evolution of  $r_{tip}$  for the samples shown in Figs. S4-S5. (a) Shows the effect of heat for non-illuminated ( $k_L = 0.0$ ) samples and (b) on the illuminated ( $k_L = 0.5$ ) samples. The solid black line in (a-b), marks the equilibrium position of the fiber tips in the reference case (i.e.,  $T = 20^\circ\text{C}$  and  $k_L = 0.0$ ). (c) The temperature dependence of  $r_{tip}$  after the sample has reached the equilibrium under the light (red curve;  $k_L = 0.5$ ) and in the dark (black curve;  $k_L = 0.0$ ). The sample illustrates qualitatively similar monotonic behavior as in Fig. 11c.



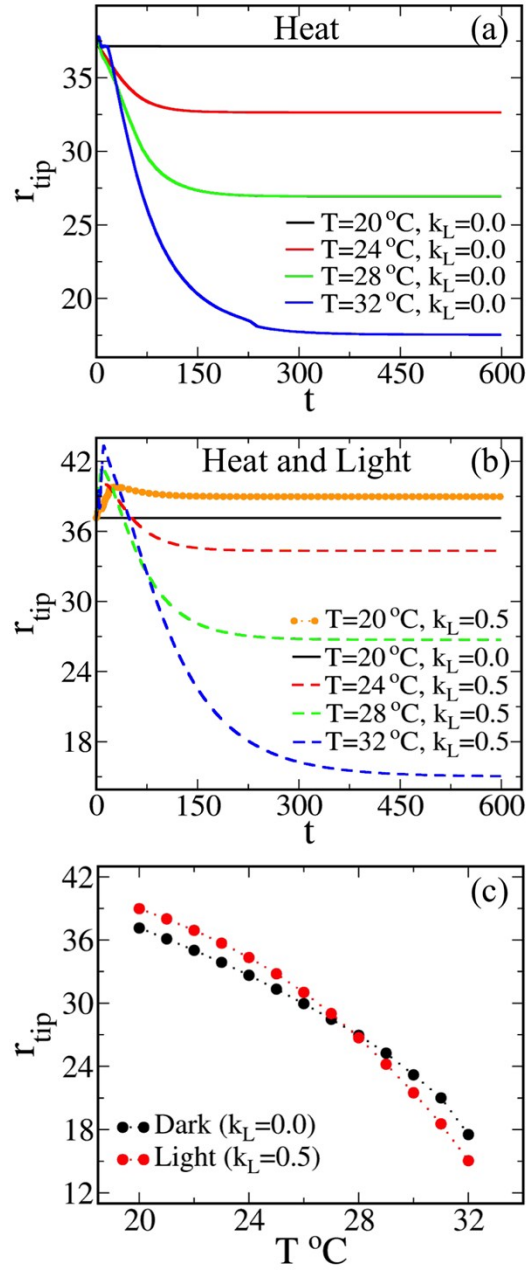


**Figure S7:** (a) Initial setup of the gel-fiber composite with four fibers arranged at the edges with separation between the tips being equal to  $r_{tip} = 28$  elements at  $T = 20^\circ\text{C}$ . The gel sample size is  $25 \times 25 \times 12$ , and the length of the fiber extending out of the gel is 14 dimensionless unit. (b-c) The response of the composite to heat (at  $T = 28^\circ\text{C}$  and  $T = 32^\circ\text{C}$ ) in the dark ( $k_L = 0.0$ ). (d-f) The polymer volume fraction,  $\phi$ , within the top layer of the gel, for the cases shown in (a-c).

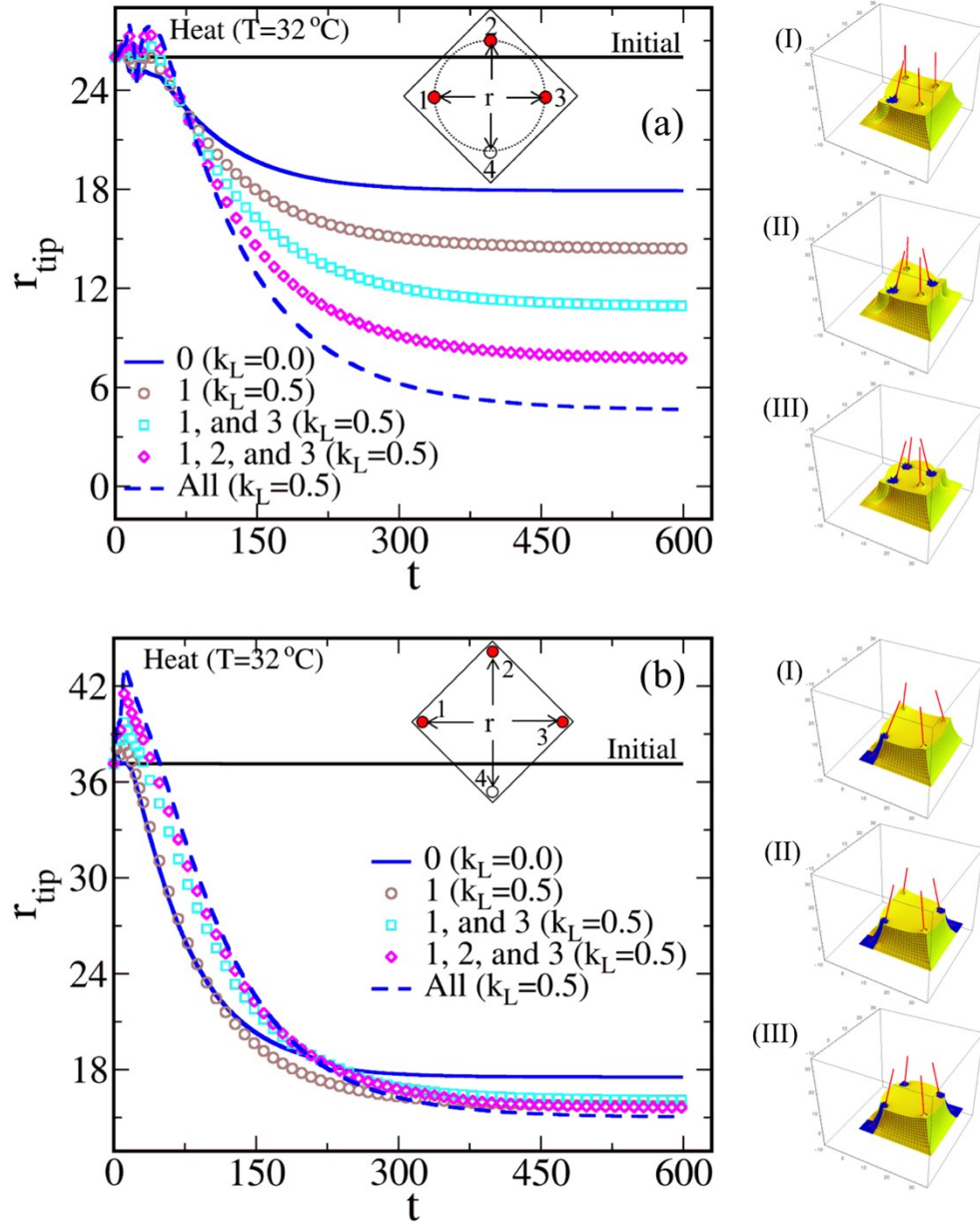




**Figure S8:** (a) Response to light ( $k_L = 0.5$ ) at  $T = 20^\circ\text{C}$  for the size of SP-functionalized region  $R = 5$  where fibers bend away from each other. In (b) and (c), the sample is heated to  $T = 28^\circ\text{C}$  and  $T = 32^\circ\text{C}$ , respectively, under light ( $k_L = 0.5$ ) where fibers bend towards each other. (d-f) The polymer volume fraction,  $\phi$ , within the top layer of the gel, for the cases shown in (a-c).



**Figure S9:** Time evolution of  $r_{tip}$  for the samples shown in Figs. S7-S8. (a-b) The effect of heat for non-illuminated ( $k_L = 0.0$ , in (a)) and illuminated ( $k_L = 0.5$ , in (b)) samples. The solid black line in (a-b) marks the initial equilibrium position of the fiber tips at  $T = 20^\circ C$  and  $k_L = 0.0$ . (c) The temperature dependence of  $r_{tip}$  after the sample has reached the equilibrium under the light (red curve;  $k_L = 0.5$ ) and in the dark (black curve;  $k_L = 0.0$ ). Both curves illustrate qualitatively similar monotonic behavior.



**Figure S10:** Time evolution of  $r_{tip}$  when only few selected posts (as indicated in the legend) are illuminated compared with no illuminated posts (solid blue line), and all illuminated posts (dashed blue line). Here, we set  $T = 32^\circ\text{C}$ . (a) Composite with four fibers arranged diagonally with separation between the tips being equal to  $r_{tip} = 20$  elements. (b) Composite with four fibers arranged at the edges with initial separation between the tips  $r_{tip} = 28$  elements. Following posts are illuminated in (a) and (b): (I) 1, (II) 1 and 3, (III) 1, 2 and 3. The red circles in the insets of (a) and (b) show the numbering of the illuminated posts.