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Supporting Information

Flexible and Stretchable Mechanoluminescent Fiber and Fabric

Jing Zhang†, Luke Bao†, Huiqing Lou, Jue Deng, Ao Chen, Yajie Hu, Zhitao Zhang, Xuemei Sun,and Huisheng Peng**

*State Key Laboratory of Molecular Engineering of Polymers, State Key Laboratory of Molecular Engineering of Polymers, Department of Macromolecular Science, and Laboratory of Advanced Materials, Fudan University, Shanghai 200438, China; *E-mail: sunxm@fudan.edu.cn, penghs@fudan.edu.cn.*

†These authors contributed equally to this work.

Experimental Section

Materials. ZnS:Cu (green) and ZnS:Mn (orange) phosphors were purchased from Shanghai Keyan Phosphor Technology Company Limited. The PDMS elastomer was obtained from Wacker (RT601). Silicone liquid (0.65 cSt) was purchased from Aladdin Reagent Incorporated. Ethanol (99.5 vol %) was obtained from Shanghai Zhenxing No. 1 Chemical Plant.

Preparation of the mechanoluminescent composite fiber. First, the elastic PDMS fibers were fabricated by injecting PDMS precursor, namely, a mixture of elastomer prepolymer and curing agent with a weight ratio of 9:1, into polytetrafluoroethylene tubes and then cured at 70 °C for 1 h. The mechanoluminescent material was composed of transitional metal-doped ZnS particles and PDMS precursor with a weight ratio of 7:3. Next, the mechanoluminescent active material was coated onto the surface of a PDMS fiber by dip coating and then cured at 70 °C for 0.5 h after the excess mixture was dripped off. Finally, the mechanoluminescent composite fiber was prepared after coating another layer of PDMS.

Preparation of the mechanoluminescent fabric based on mechanoluminescent ribbon. First, the elastic fabric was washed by deionized water and ethanol. Then, the PDMS precursor was diluted by silicone liquid with a volume ratio of 1:1. Next, the dilute PDMS precursor was coated onto the elastic fabric with a mask and then cured at 70 °C for 1.0 h. The ZnS/PDMS materials were prepared by mixing ZnS particles with PDMS precursor with a weight ratio of 7:3 under stirring and then put into a vacuum for 10 min to remove air bubbles. Afterwards, the ZnS/PDMS mixture was coated onto the PDMS-coated fabric with the same mask and cured at 70 °C for 0.5 h. Finally, coating another PDMS onto the fabric with a mask to obtain mechanoluminescent fabric.

Preparation of the mechanoluminescent fabric based on mechanoluminescent point. First, the elastic fabric was prepared by sparsely weaving PDMS fiber into a fabric. Then, the ZnS/PDMS mixture was selectively coating onto the intersections of elastic fabric and curing at 70 °C for 0.5 h. Finally, another PDMS layer was coated onto the mechanoluminescent point as the covering layer.

Mechanoluminescence characterization of the composite fiber. For quantificational investigation of the mechanoluminescence from the composite fiber, we designed a

stretching-releasing system to generate a periodic stress to the composite fiber (**Figure S5**). Both ends of the composite fiber were fastened to two fixtures and one of them was connected with a rotatable electric motor, while another was fixed in an immovable pedestal. The spectroradiometer with a detection spot size of 5.25 mm in diameter was vertically located in front of the composite fiber to collect luminescent data (**Figure S14**). As the detection spot size is larger than the diameter of composite fiber, the measured data are actually lower than the real luminescent intensities. Therefore, we calculated the measured data by dividing an area ratio given below

$$\frac{S_{fiber}}{S_{spot}} = \frac{2r\sqrt{R^2 - r^2} + 2R^2 \sin^{-1}\frac{r}{R}}{\pi R^2} \quad (1)$$

where S_{fiber}, S_{spot}, R and r correspond to the area of mechanoluminescent fiber, the area of detection spot, the radius of detection spot and the radius of mechanoluminescent fiber, respectively, as shown in **Figure S15**. During the study on the cycle performance, the composite fiber was first stretched to a high strain of 80% and then released to the original state at a frequency of 6 Hz by the stretching and releasing system. The mechanoluminescent intensity was measured every 18 s.

It should be noted that all the mechanoluminescence intensities of the composite fibers were measured in a dark room. The intensity of the surrounding light without fibers was measured to be lower than 0.02 cd/m^2, which was not enough to influence the trend of the data. In addition, all used intensity values of the mechanoluminescent fibers had been deducted the light noise from the measured values.

Characterization. Photographs were taken using a digital camera (α6000, Nikon, Japan). Scanning electron microscopy (SEM) images and correspondisng energy dispersive spectrometer mapping images were recorded using a field-emission scanning electron microscopy (Ultra 55, Zeiss), operated at 3 kV and 20 kV. The luminescence intensity and CIE coordinates were measured with a spectroradiometer (PR-680, detection spot size of 5.25 mm). The stress and strain in the stretching and releasing cycles were measured from a Hengyi Table-Top universal testing instrument. Spectra were obtained using a miniature fiber optic spectrometer (Idealoptics PG2000-pro, China) installed in an optical microscopy (Olympus BX51, Japan). X-ray diffraction

analysis was conducted on an X-ray power diffractometer (Bruker, D8 ADVANCE) with filtered Cu Kα radiation.

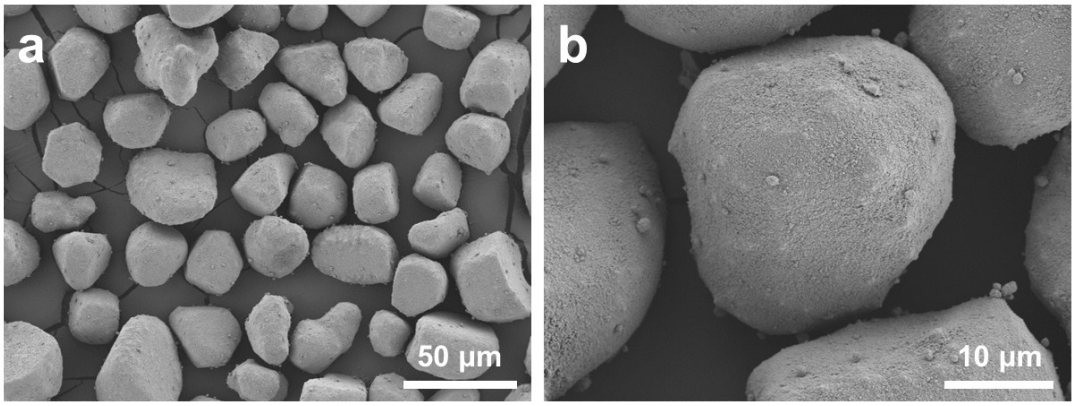

Figure S1. SEM images of ZnS:Cu particles at low (**a**) and high (**b**) magnifications.

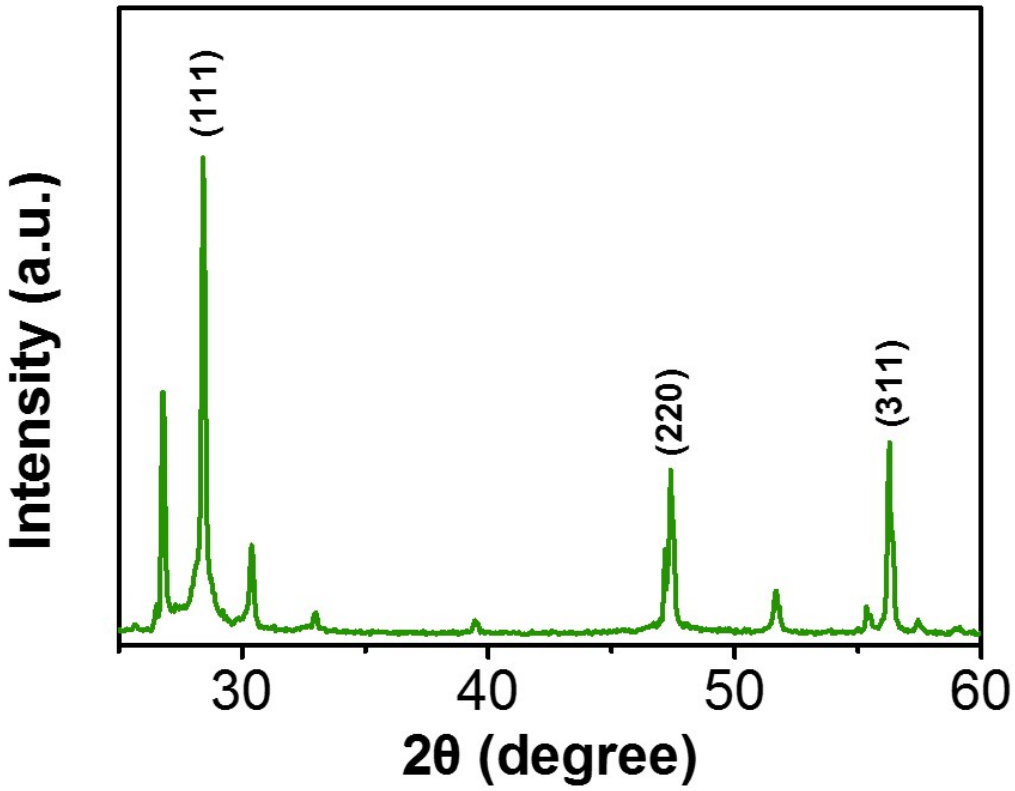

Figure S2. X-ray diffraction spectrum of ZnS:Cu particles.

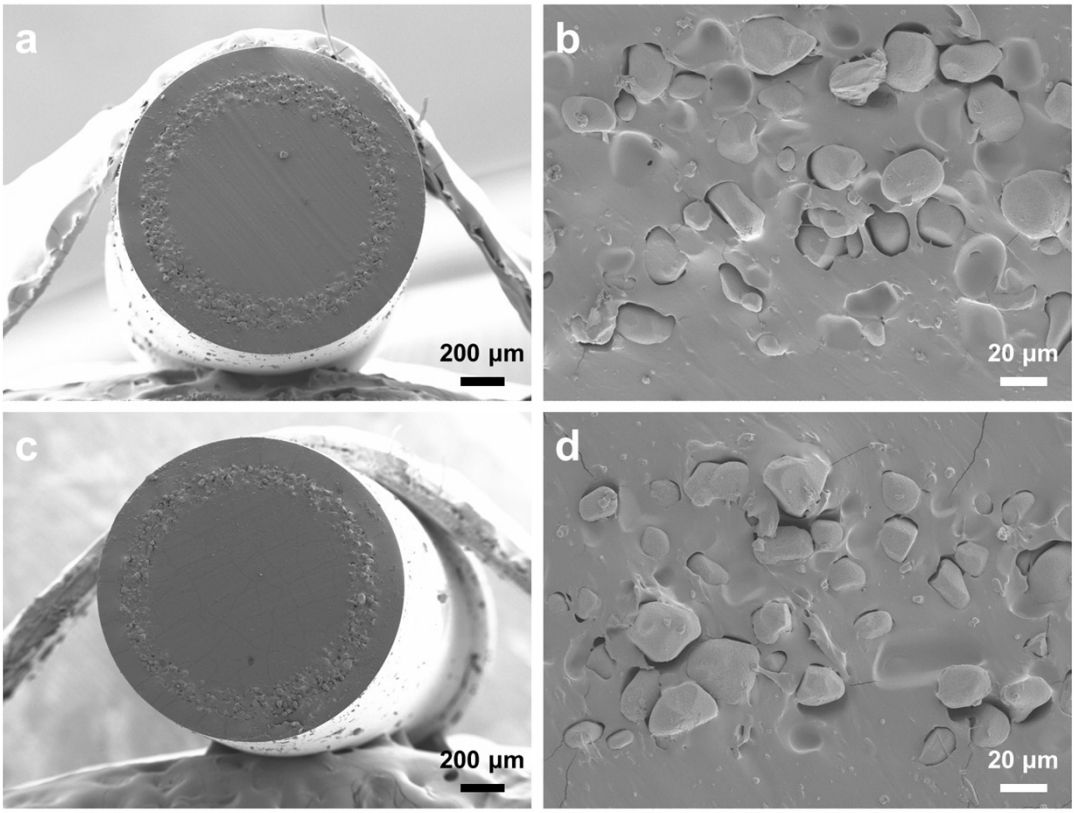

Figure S3. Cross-sectional view of the composite fiber before (**a**, **b**) and after (**c**, **d**) 10,000 stretching and releasing cycles.

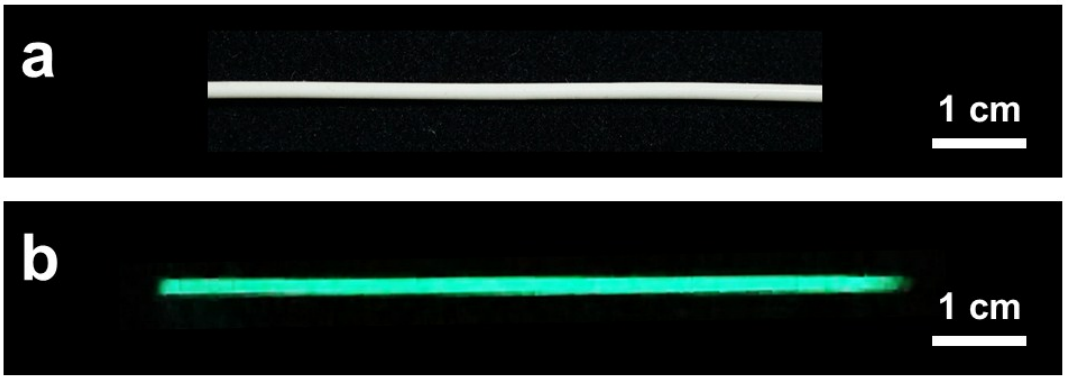

Figure S4. (**a**) Photograph of a mechanoluminescent composite fiber. (**b**) Photograph of mechanoluminescence of the composite fiber in (**a**) under stretching.

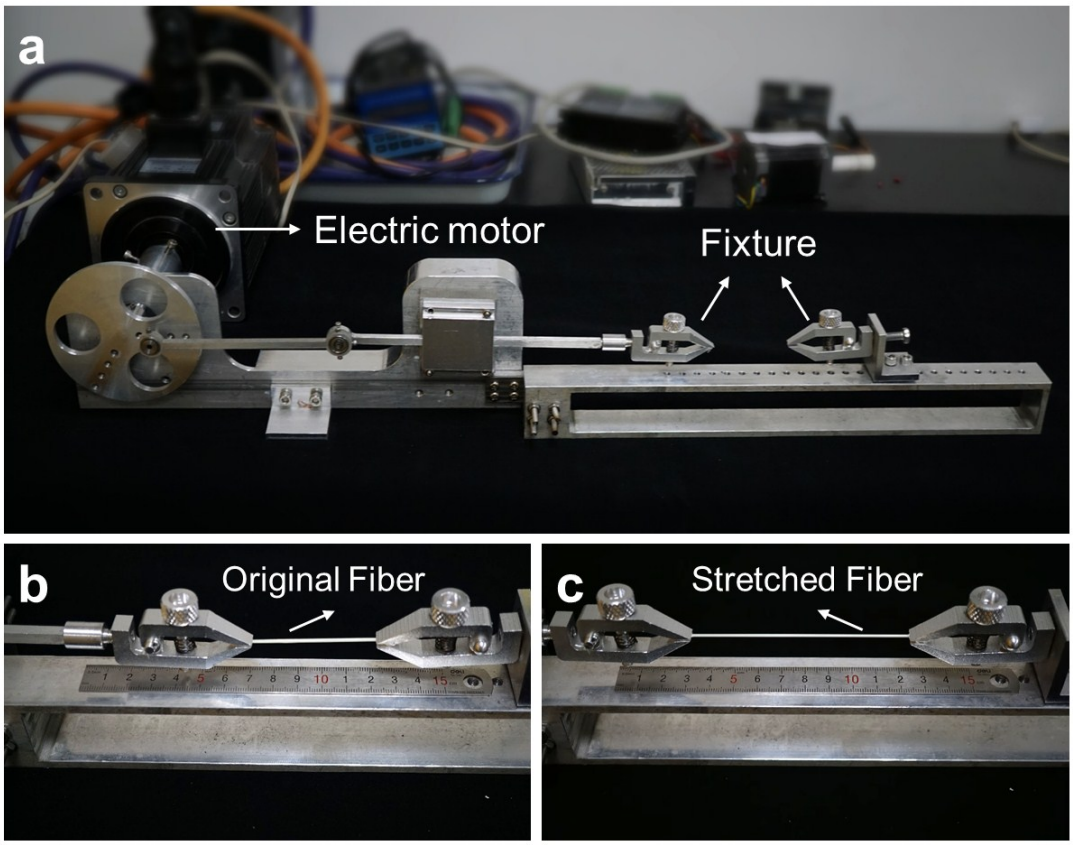

Figure S5. (**a**) Photograph of the designed stretching and releasing system. (**b**, **c**) Photographs of composite fibers before (**b**) and after (**c**) stretching.

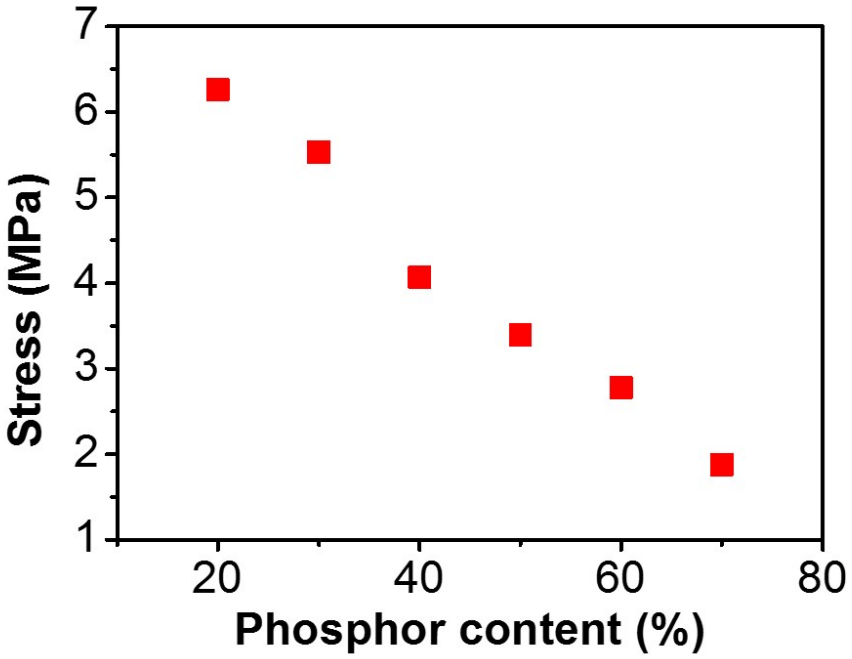

Figure S6. Variation of the tensile strength of the composite fiber with phosphor contents from 20% to 70%.

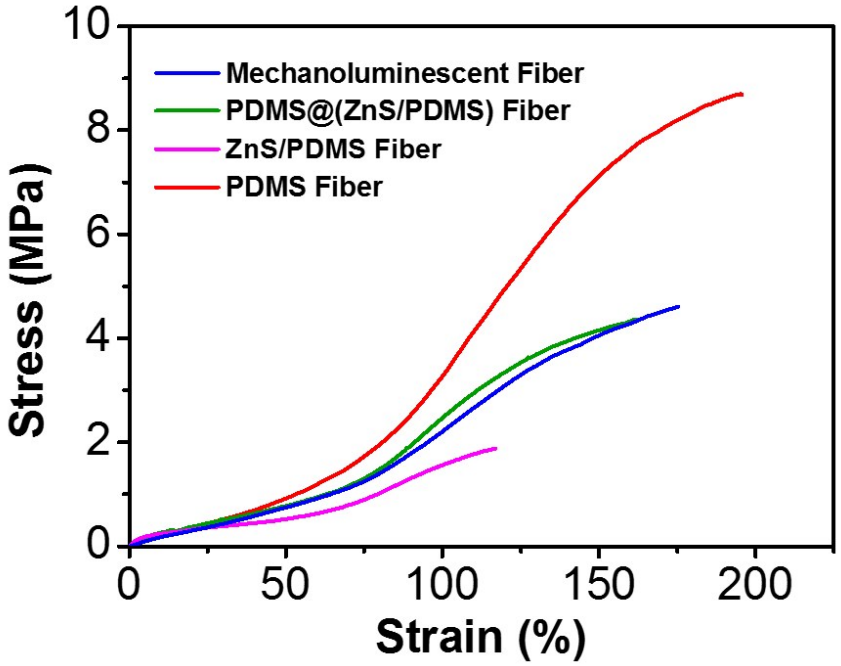

Figure S7. Stress-strain curves of the PDMS fiber, ZnS/PDMS fiber, PDMS@(ZnS/PDMS) fiber (mechanoluminescent fiber without the outer PDMS layer) and mechanoluminescent composite fiber.

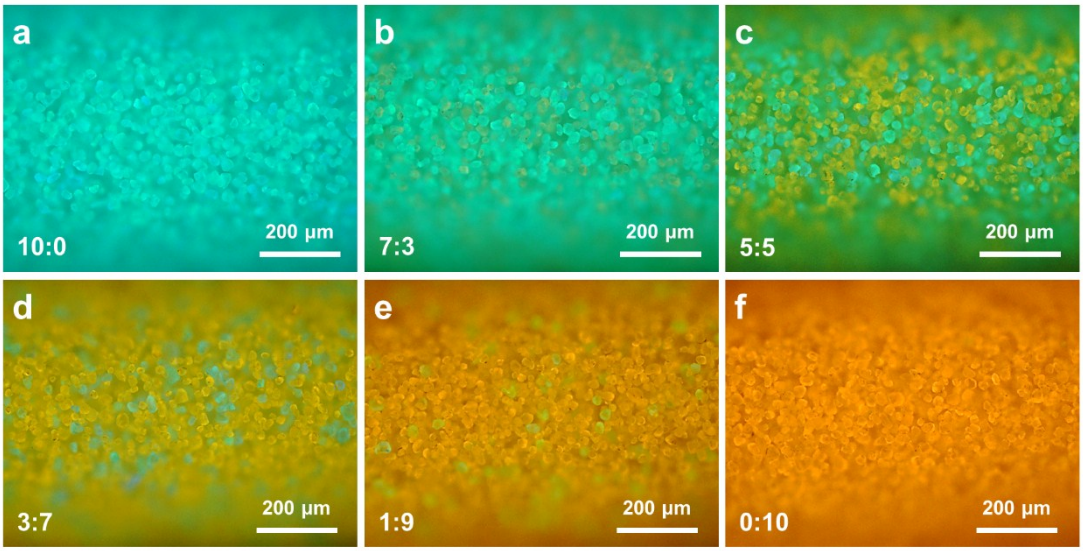

Figure S8. Optical microscope images of the mechanoluminescent fibers with two ZnS particles at weight ratios (green:orange) of 10:0, 7:3, 5:5, 3:7, 1:9 and 0:10.

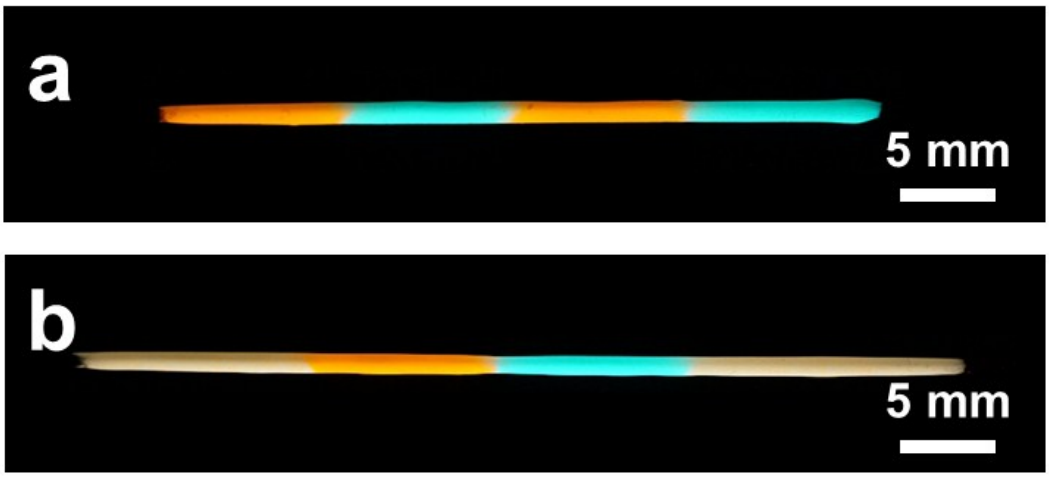

Figure S9. Photographs of the bicolored (**a**) and tricolored (**b**) mechanoluminescent fibers under UV irradiation.

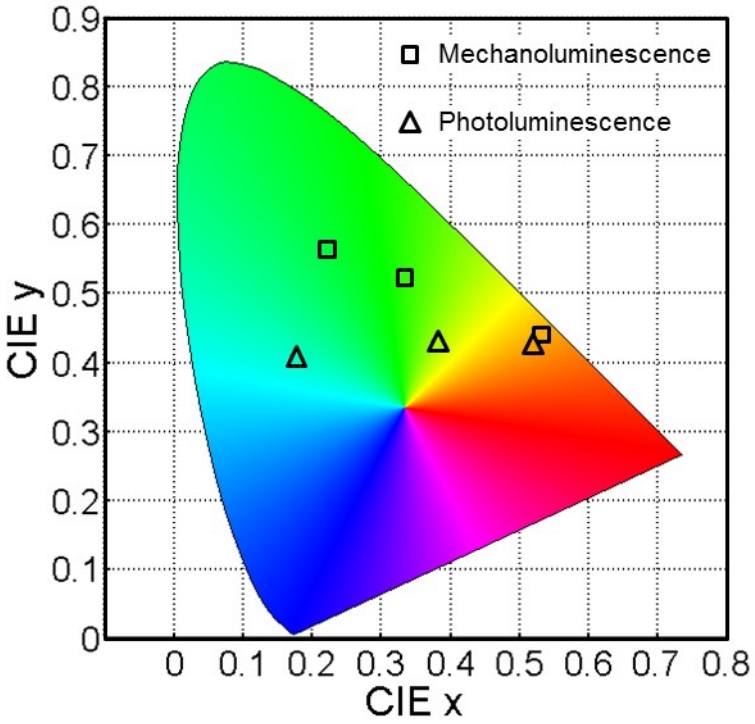

Figure S10. The CIE coordinate (x, y) values of mechanoluminescence and photoluminescence from the different colored composite fibers.

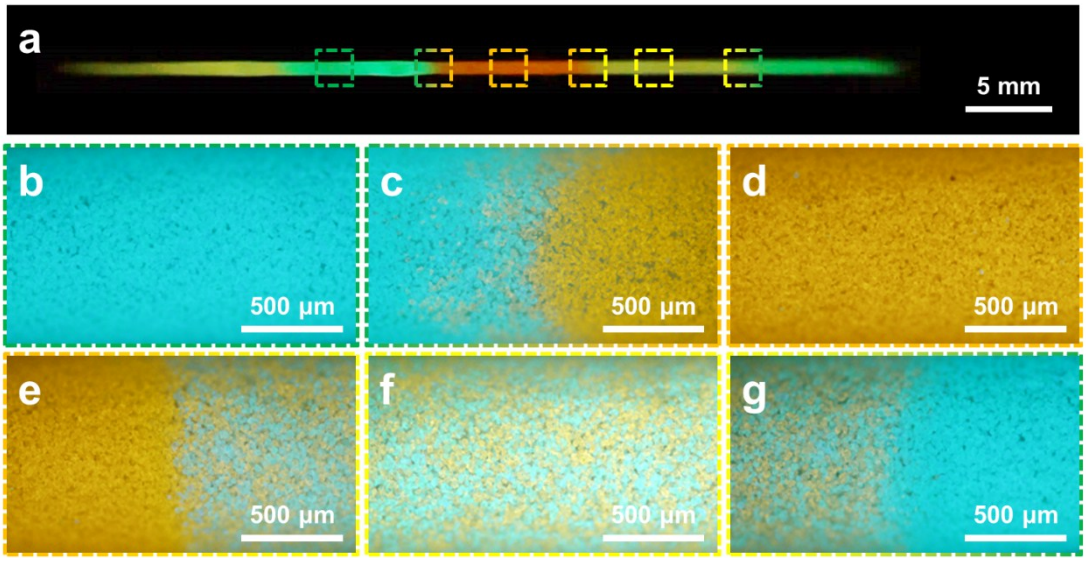

Figure S11. (**a**) Photograph of a tricolored mechanoluminescent fiber. (**b**-**g**) Optical microscope images of a tricolored mechanoluminescent fiber in (**a**).

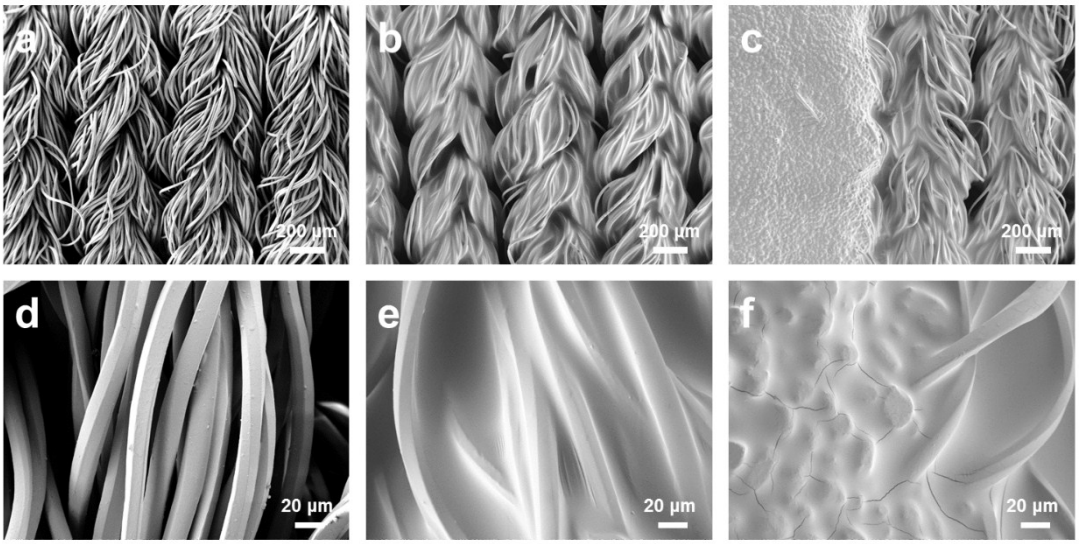

Figure S12. SEM images of elastic fabric (**a**, **d**), PDMS coated fabrics (**b**, **e**) and ZnS/PDMS coated fabric (**c**, **f**) at low and high magnifications.

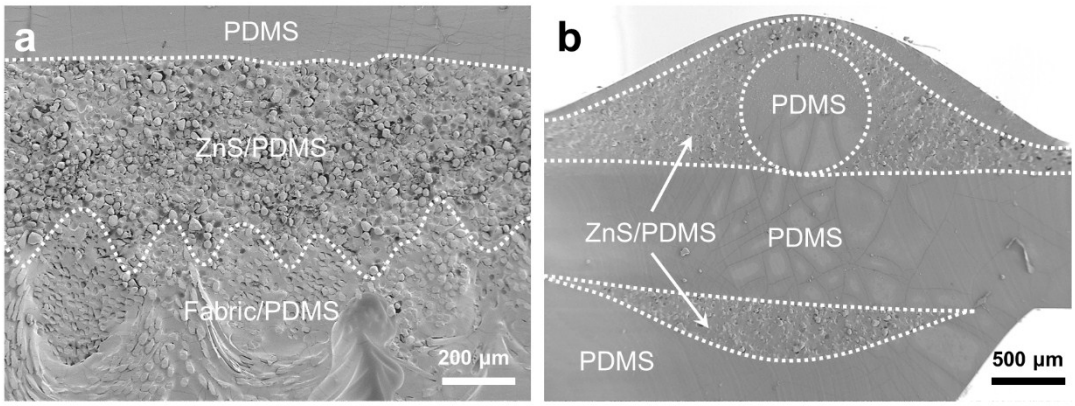

Figure S13. SEM images of the mechanoluminescent ribbon (**a**) and point (**b**) in cross-sectional view.

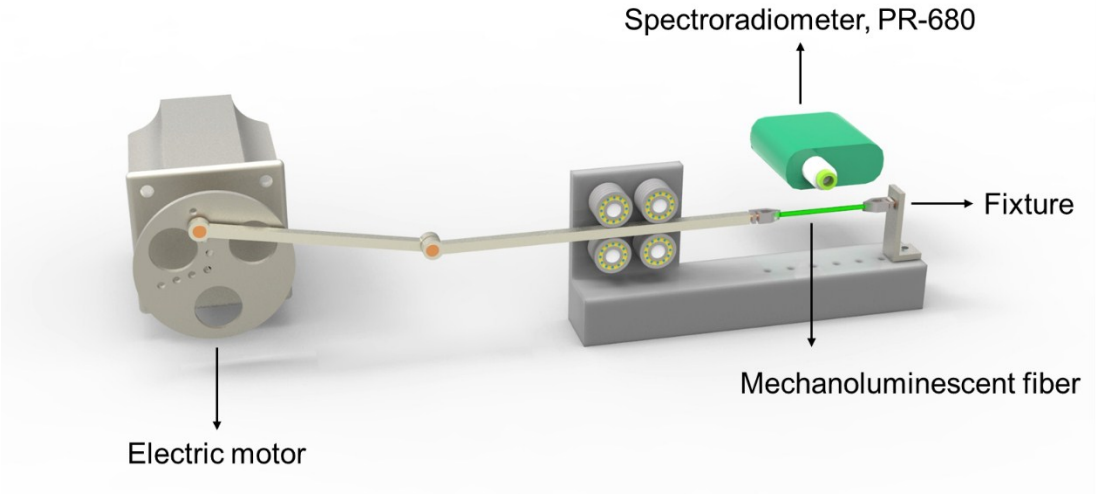

Figure S14. Schematic illustration to the mechanoluminescence detection system.

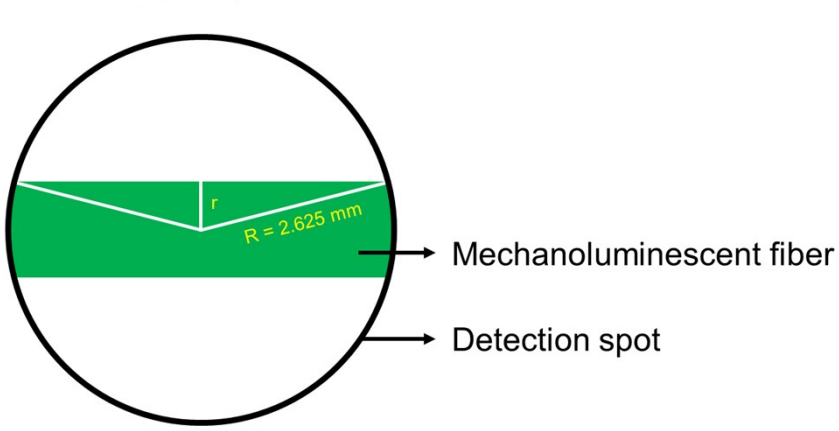

Figure S15. Schematic illustration to the detection area of the spectroradiometer.