

Supplementary Materials for

Programmable transparent organic luminescent tags

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Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/5/2/eaau7310/DC1)

Movie S1. Image writing using UV light.
Movie S2. Image reading using UV light.
Movie S3. Writing, reading, and erasing different patterns.
Movie S4. Image writing using UV light (original video).

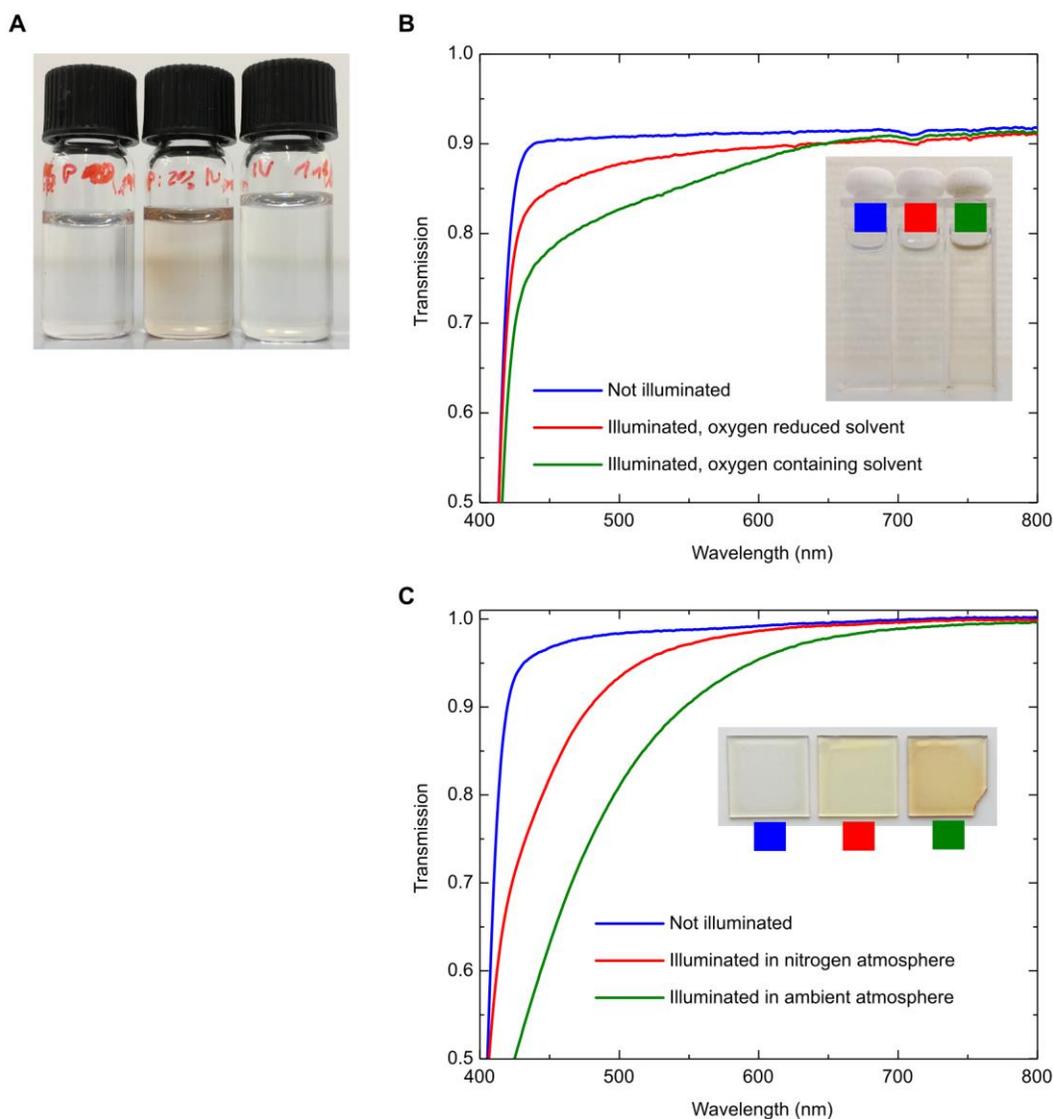


Fig. S1. Oxygen consumption effects in solutions and films containing PMMA and NPB.

(A) Solutions containing (from left to right) PMMA, PMMA:NPB (2 wt%) and NPB in anisole shown after illumination with UV light. Only PMMA:NPB (2 wt%) solution changes its transmission and color. Yellowing is a commonly known indication for polymer oxidation. (B) Transmission of vials containing PMMA:NPB (2 wt%) in anisole. In contrast to a freshly prepared solution (blue), the same solution (green) shows an increase in absorption in the blue wavelength regime after 30 minutes of UV illumination of 7.0 mWcm^{-2} . The effect is significantly smaller in solution with a reduced amount of oxygen (red in comparison to green). This shows the interaction of PMMA, NPB and oxygen in oxidizing the polymer by UV absorption of NPB and singlet oxygen generation through NPB-triplet quenching. (C)

Transmission of drop casted films containing PMMA:NPB (2 wt%). Without previous illumination the thick film is almost transparent in the visible. After 180 minutes of intense UV illumination in nitrogen (red) and ambient atmosphere (green), respectively, transmission of both samples has changed as it did in solution. Yellowing is significantly stronger in the film which was illuminated in the presence of oxygen. This agrees with the results of the solution measurements showing oxidation of PMMA by singlet oxygen. Photo credit: Max Gmelch, Dresden Integrated Center for Applied Physics and Photonic Materials.

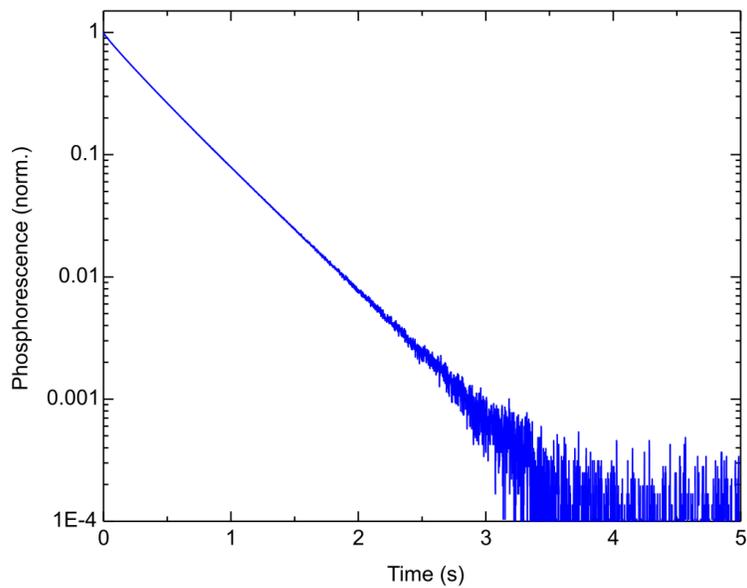


Fig. S2. Phosphorescence decay of PMMA:NPB (2 wt %) covered by an oxygen barrier.

Fitting with a double exponential decay curve provides an average phosphorescence lifetime of 406 ms.

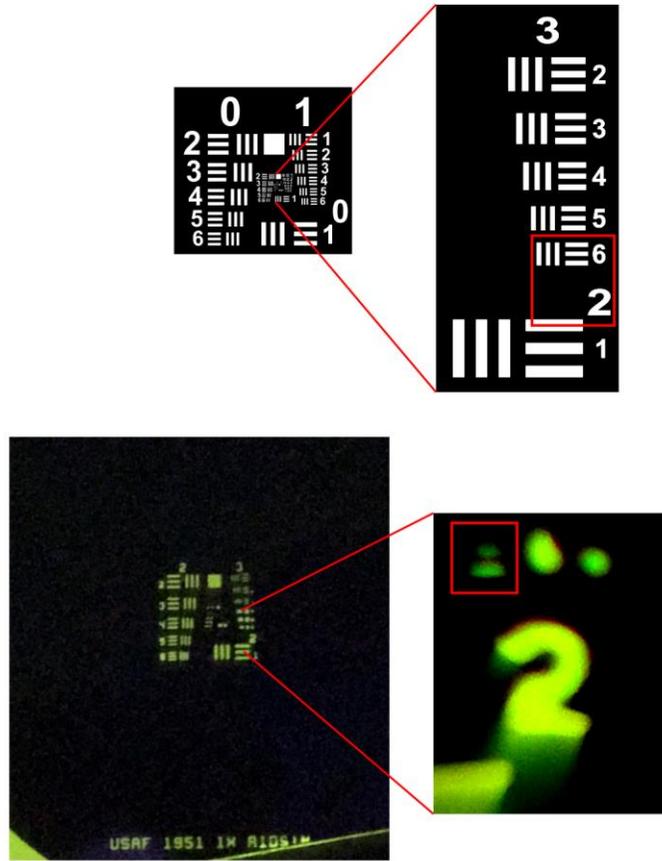


Fig. S3. USAF 1951 test target and achieved maximum resolution. The maximum achieved resolution was line separation in USAF 1951 group 3, element 6. This corresponds to a resolution of 724 dpi. Photo credit: Max Gmelch, Dresden Integrated Center for Applied Physics and Photonic Materials.

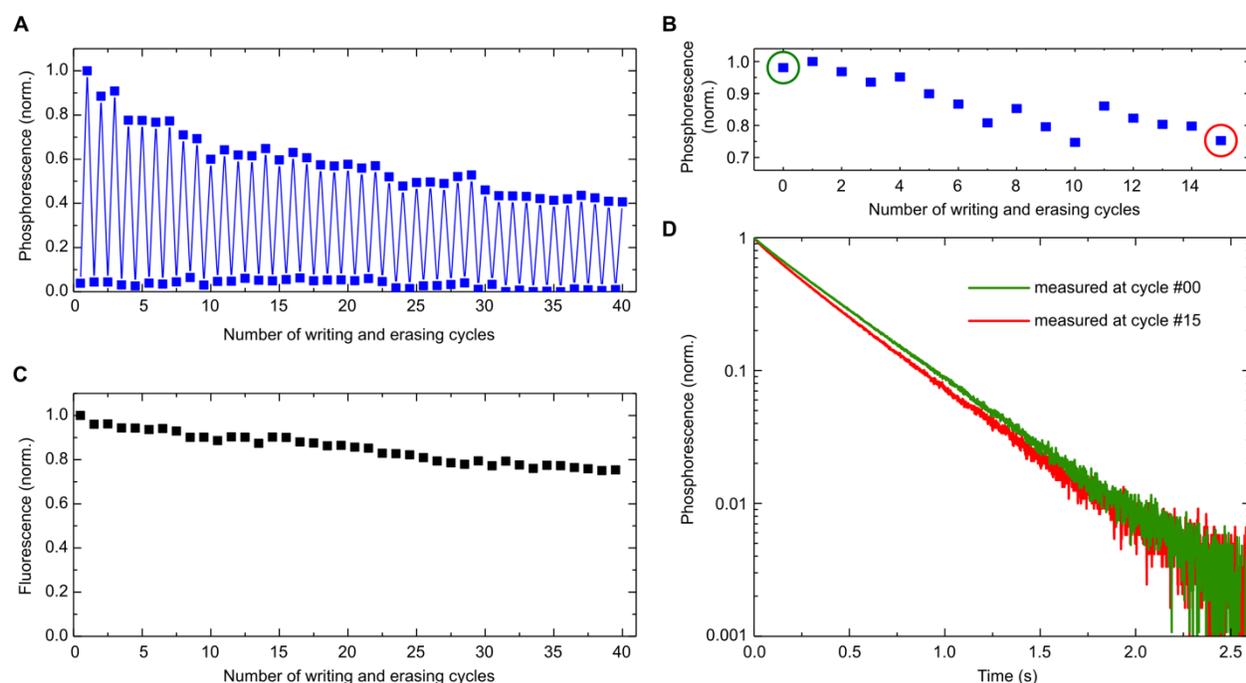


Fig. S4. Writing and erasing cycles and resulting degradation processes. (A) Normalized phosphorescence intensity after writing and erasing processes for 40 cycles. Writing was achieved by UV excitation of the sample until full phosphorescence intensity. Each erasing iteration consisted of IR illumination for 30 s and subsequent cooling down to room temperature before measuring. The phosphorescence intensity decreases with increasing cycle number down to 40% after 40 cycles. (B) Normalized activated phosphorescence intensity after writing and erasing processes for 15 cycles. Each erasing iteration consisted of heating on a hotplate for 60 s at 95°C and subsequent cooling down to room temperature before measuring. The phosphorescence intensity decreases with increasing cycle number down to 75% after 15 cycles. The green and red circles indicate the points in time for (D). (C) Fluorescence intensity in each cycle of (A). The decrease after 40 cycles is down to 80%. Therefore, only one part of the phosphorescence decrease can be explained by photo bleaching and emitter degradation. (D) The other part can be attributed to the degrading of PMMA next to the emitting molecules caused by singlet oxygen. This results in a reduced rigidity and therefore an increase in non-radiative depopulation rates of the triplet state. In consequence, triplet states are quenched either completely or partly. While full quenching of the phosphorescence of one molecule is not measurable, partly quenching is observed in a decrease of triplet state lifetime from 418 ms (green) to 393 ms (red) from before to after 15 cycles.

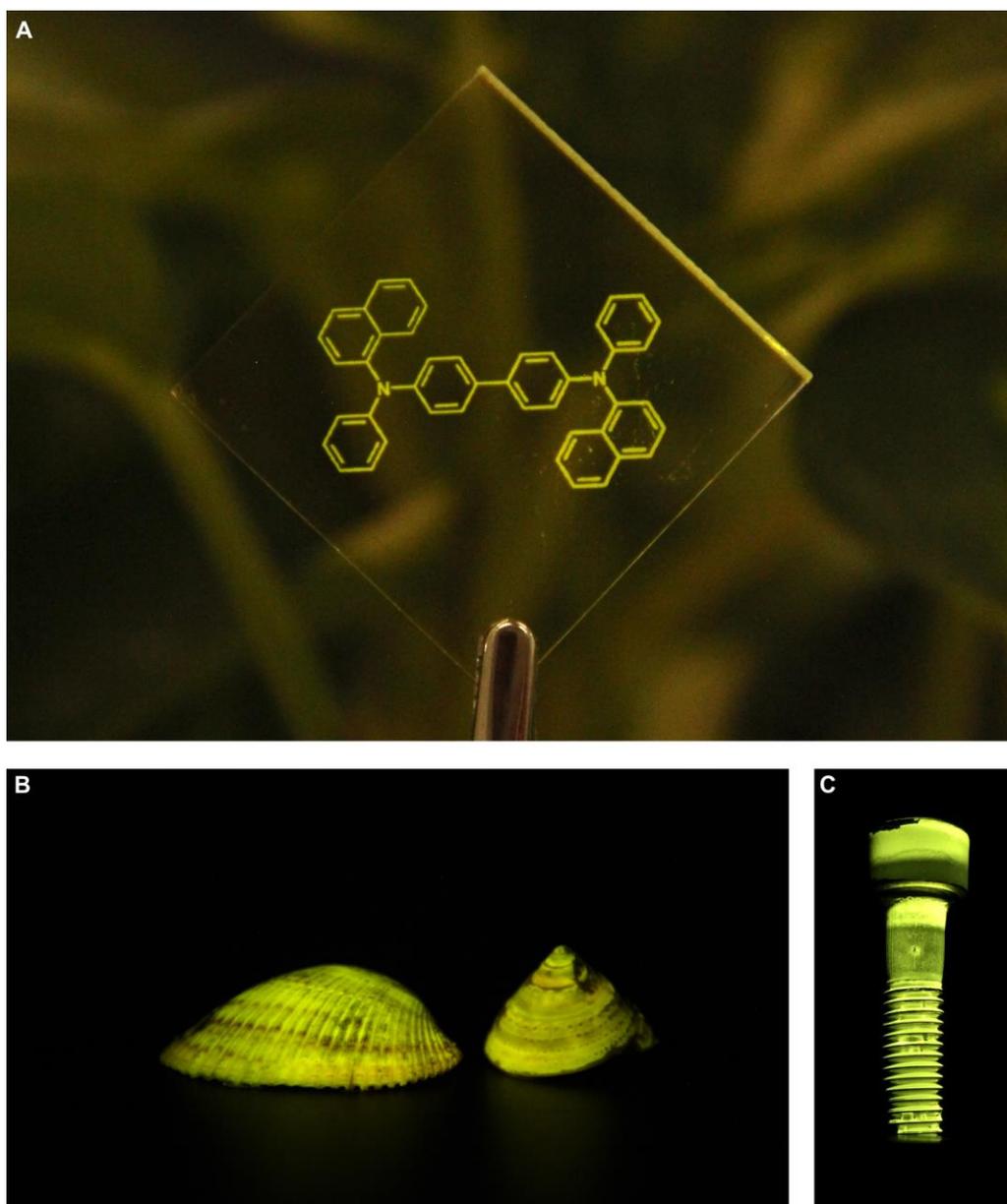


Fig. S5. Different coating methods on different substrate materials. (A) Spin coating on glass. The image shows the structure of the emitting molecule NPB, written into the layer by mask illumination. Photo credit: Felix Fries, Dresden Integrated Center for Applied Physics and Photonic Materials. (B) Spray coating on shells. (C) Dip coating on stainless steel. Photo credit: Max Gmelch, Dresden Integrated Center for Applied Physics and Photonic Materials.

Movie S1. Image writing using UV light. By masked UV illumination of the sample, a phosphorescent pattern is printed into the transparent sample. Here, pulsed excitation with a frequency of 1 Hz was used. When taking only one video frame during the off-time of the excitation, the emergence of the phosphorescence is clearly visible. Hereby, the time delay of each frame to the corresponding switch-off was held constant. The playback speed is accelerated by a factor of 10. Movie S4 shows the same recording but without fluorescence frames omitted.

Movie S2. Image reading using UV light. After having finished the writing process, the mask is removed. Subsequent UV illumination of the whole sample only gives rise to phosphorescence at the activated area. Again, pulsed excitation with a frequency of 1 Hz was used.

Movie S3. Writing, reading, and erasing different patterns. A whole cycle containing writing, reading, erasing and subsequent writing of a different pattern is shown.

Movie S4. Image writing using UV light (original video). Here the raw data of Movie S1 are shown. Using masked UV illumination, a phosphorescent pattern is written into the sample. Pulsed excitation with 1 Hz leads to alternating observation of fluorescence and emerging phosphorescence. The playback speed is accelerated by a factor of 10.