Supporting Information

Exotic Property of Azobenzene Sulfonic Photoalignment Material Based on Relative Humidity

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Figure S1. The writing dynamics of the photoalignment films with different thickness. The film thickness could vary from 5 nm to 50 nm by adjusting the concentration of DMF solution. (i) The *in-situ* phase retardation change; (ii) Normalized phase retardation for reorientation speed comparison at different relative humidity: (a) 20%RH, (b) 60%RH. At low relative humidity (20%RH), the total phase retardation is low, indicating fewer azo molecules are photoaligned. However, the thicker film has higher reorientation speed. It is probably because that the azo molecules are densely packed at low humidity, especially close to the substrate, where the azo molecules are hard to be photoaligned. The pinning effect due to the substrate becomes smaller as the film gets thicker and therefore the molecules are easier to be reoriented. At optimal relative humidity (60%RH), the reorientation speed is the same for the films thinner than ~30 nm. The water molecules are inserted to provide more free volume for reorientation and the pinning effect from the glass substrate gets ignorable. As the film gets thicker, more actinic light gets absorbed and the reorientation speed becomes slower above the critical thickness.
Figure S2. Ordering comparison of the photoalignment films induced by 1 J/cm² and 3 J/cm² 365 nm LPUV at different relative humidity. (a) Phase retardation and (b) order parameter of the SD1 film. The incident power of the LPUV light at the place of SD1 film is 5 mW/cm². Different colors correspond to different humidity regions for photoalignment as in Figure 3.

Figure S3. The film alignment before and after LPUV excitation with increasing humidity. The SD1 film is written by LPUV at 20%RH. After LPUV is turned off, no further phase retardation changes at 20%RH. Then the relative humidity increases to
80% without light excitation, where dramatic phase retardation increment is observed. The grey area indicates where the LPUV is off.

**Figure S4.** The aligned SD1 film (50 nm, spin-coated in DMF) observed under depolarized optical microscope with the alignment direction (a) parallel and (b) 45° to the polarizer. The crossed white arrows indicate crossed polarizers and the yellow arrow indicate the alignment direction.