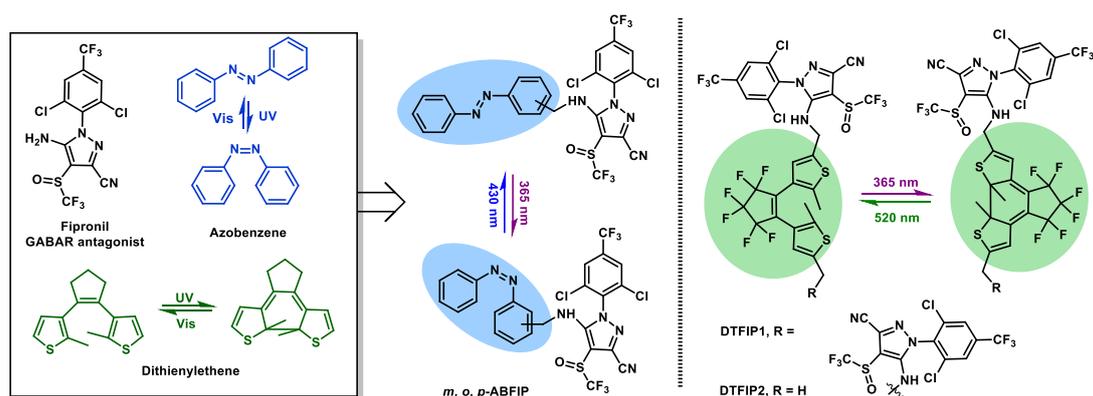
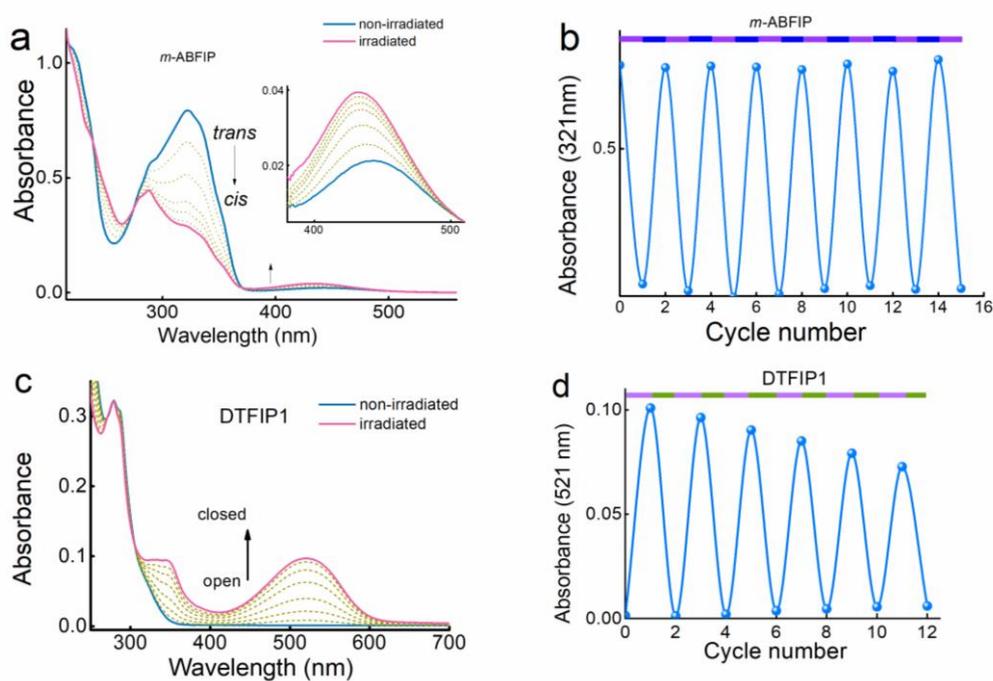


## Figures



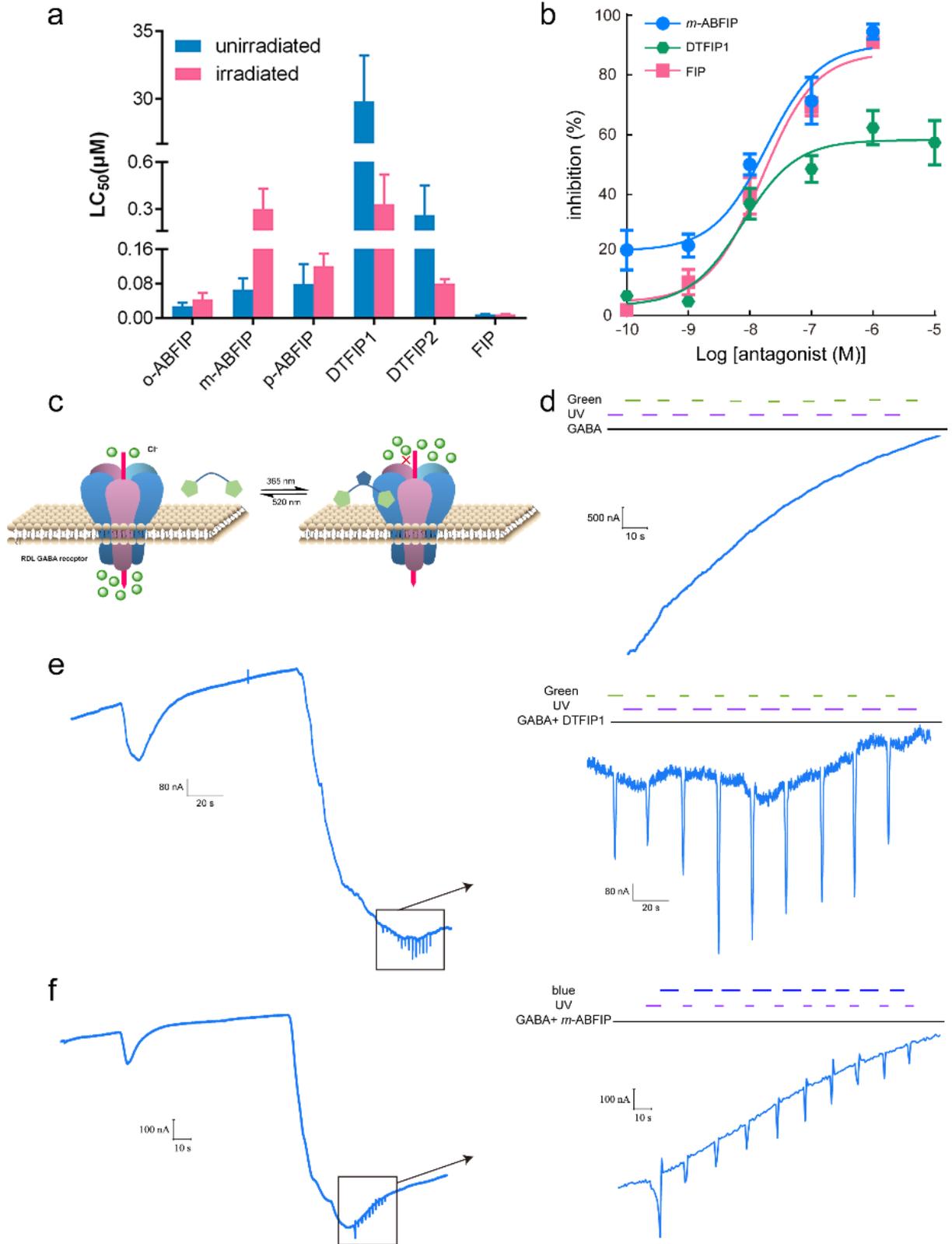
**Figure 1. Design concept of photoswitchable azobenzene-fipronils (ABFIPs) and dithienylethene-fipronils (DTFIPs) as insect ionotropic GABAR ligands.**

Note, ABFIPs are obtained by exterior modification of fipronil by azobenzene. DTFIPs are obtained by linking fipronil molecule with dithienylethene.



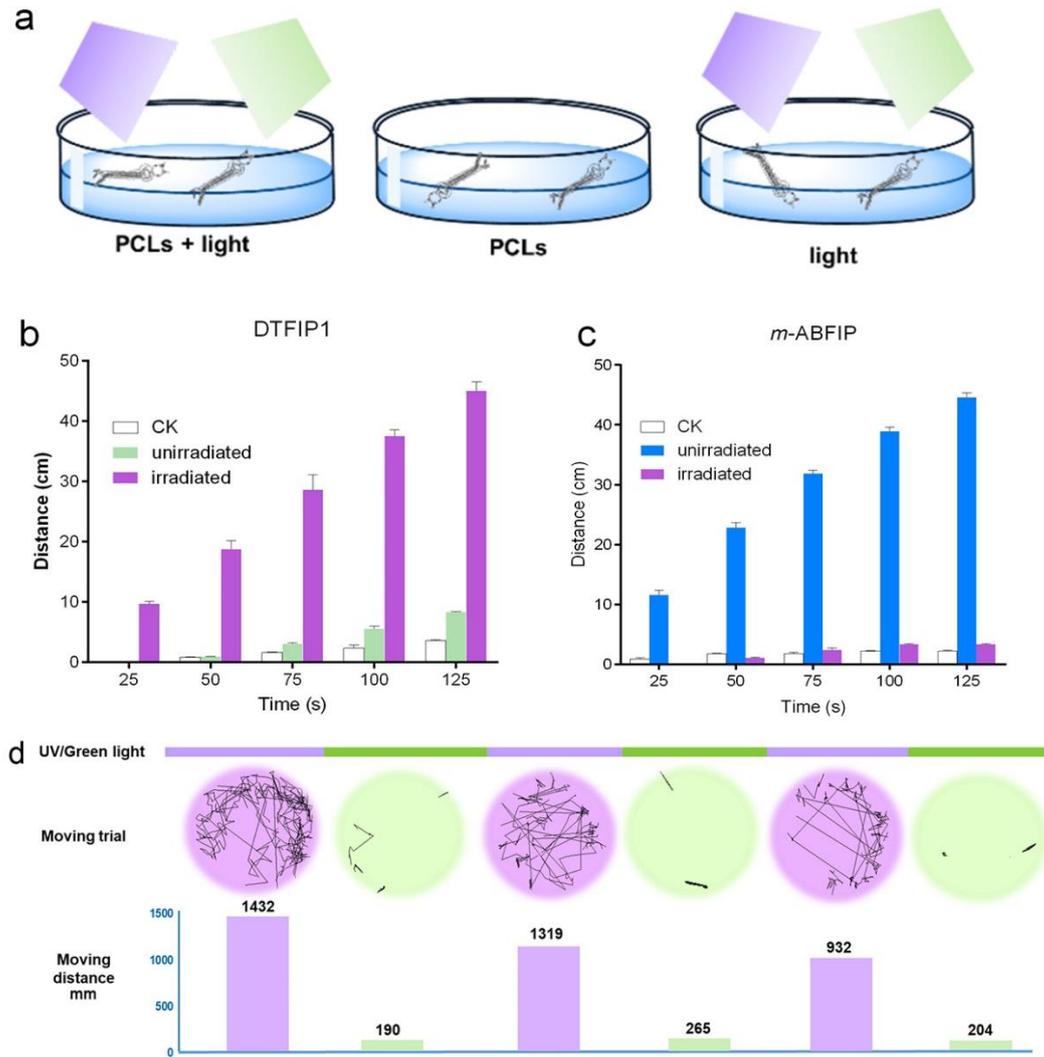
**Figure 2. Photophysical properties of ABFIPs and DTFIPs.** (a) UV-Vis absorbance spectra evolution of *m*-ABFIP (0.02  $\mu$ M in acetonitrile) by irradiation with 365 nm light. (b) Cycle performance of *m*-ABFIP, change in absorption at 321 nm during alternative irradiation of a solution (0.02  $\mu$ M in acetonitrile) with 365 nm light for 1 min and 430 nm light for 1 min. (c) UV-Vis absorbance spectra evolution of DTFIP1 (0.02  $\mu$ M in acetonitrile) by irradiation with 320 nm light. (d) Cycle performance of DTFIP1, change in absorption at 322 nm during alternative irradiation of a solution (0.02  $\mu$ M in acetonitrile) with 365 nm light for 1 min and 520 nm light for 1 min.

a solution (0.02  $\mu\text{M}$  in acetonitrile) with 365 nm light for 1 min and 520 nm light for 2 min.



**Figure 3. DIFIT1 and *m*-ABFIP enable optical control over ionotropic GABAR. (a) *In vivo* activity of PCLs in mosquito larvae indicating lethal concentrations killing 50% of the population**

(LC<sub>50</sub>) before and after irradiation. **(b)** Dose-response curve for inhibiting the normalized GABA current in homomeric *LsRDL*-type GABA receptors overexpressed in *Xenopus laevis* oocytes (IC<sub>50</sub>, 7.8 nM for *m*-ABFIP and 18.5 nM for DTFIP1, n = 6). **(c)** Schematic illustration of photoisomerization of DTFIP1 upon irradiation stimulating reversible opening and closing of the chloride channel. **(d)** Current recordings of sole oocytes upon irradiation with UV and green light. **(e)** Real-time membrane current monitoring of *Xenopus laevis* oocytes expressing homomeric *LsRDL* GABA receptors using the TEVC configuration. Inward current is elicited by 3 μM GABA and 1 μM DTFIP1. Cycles of current increase and decrease were obtained by alternative irradiation of oocytes with UV (λ= 365 nm) and green light (λ= 520 nm) for 1 s each. **(f)** Photocurrent change recordings on oocytes treated with 3 μM GABA and 1 μM *m*-ABFIP by alternative irradiation with UV (λ= 365 nm) and blue light (λ= 430 nm) for 1 s each. Experiments were repeated independently six times using different oocytes.



**Figure 4. Optical control behavioral responses of mosquito larvae.** (a) Schematic illustration of the behavioral experiments. Left: two mosquito larvae treated with PCL in petri dishes were alternatively irradiated with UV and green light. Middle and right: control experiments for larvae treated with PCL or light (5% DMSO aqueous solution) alone. (b, c) Moving distance of mosquito larvae treated with (b) DTFIP1 (5  $\mu$ M) and (c) *m*-ABFIP (0.6  $\mu$ M), with or without UV light irradiation (n=3). (d) Moving trajectory and moving distance of larvae treated with DTFIP1 (5  $\mu$ M) after 2 min alternative irradiation with UV (320 nm) and green light (520 nm) (n = 6).