Supplemental Materials for "Magnetically Switchable Light-Matter Interaction in the Two-Dimensional Magnet CrI₃"

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Supplementary Fig. 1. Band structure of the bilayer CrI_3 for the (a) AFM phase with SOC, (b) AFM without including SOC and (c) FM phase with SOC. In both (b) and (c), the **k** to $-\mathbf{k}$ symmetry is preserved in the band structure. In contrast, momentum inversion symmetry is broken in (a).



Supplementary Fig. 2. The photoconductivity contributions from (a) the three-band and (b) twoband processes. Distribution of the photoconductivity σ_{xx}^x ($\hbar\omega = 1.2 \text{ eV}$) in the first Brillouin zone, the hexagonal area. Note that the colorbar of the three-band contribution is two orders in magnitude smaller than the two-band one. The momentum inversion symmetry is broken in both (a) and (b). The three-band distribution is relatively uniform in the Brillouin zone, because it has no energy selection rule.



Supplementary Fig. 3. The photoconductivity for different relaxation time (a) $\hbar/\tau = 5$ meV, (b) 1 meV (same as Fig. 2a) and (c) 0.01 meV. The photoconductivity scales linearly with τ .



Supplementary Fig. 4. The photoconductivity σ_{xx}^x in different electric field applied for the (a) AFM and (b) FM phases. The electric field E is in unit of V/*a.u.* (1 *a.u.* = 0.53 Å).