Interaction between Optically-generated Charge-transfer states and Magnetized Charge-transfer States towards Magneto-electric Coupling

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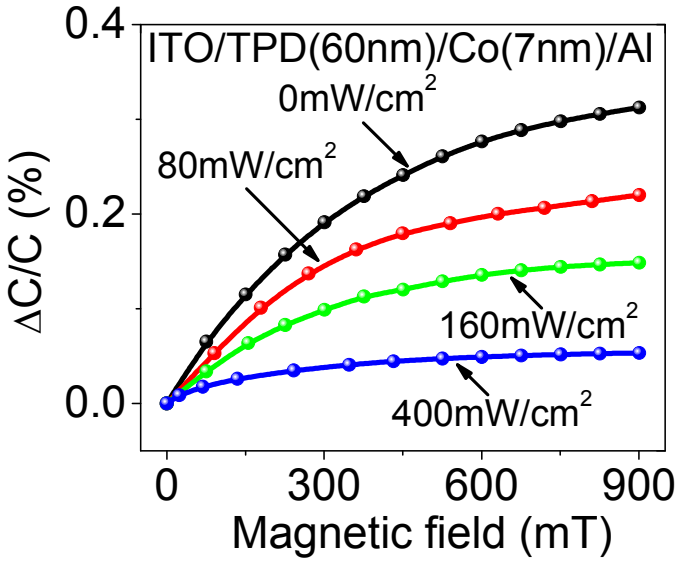

Figure S1, Magneto-dielectric from device of ITO/TPD(60nm)/Co(7nm)/Al under different photoexcitation intensities.

In Figure S1, the value of magneto-dielectric from device ITO/TPD(60nm)/Co(7nm)/Al decreases with increasing photoexcitation intensities. It is known that the magneto-dielectric originates from the magnetized CT states formed at the interface between TPD layer and Co layer. Consequently, the decreasing magneto-dielectric signal indicates that the density of magnetized CT states declines as the increasing photoexcitation intensities.

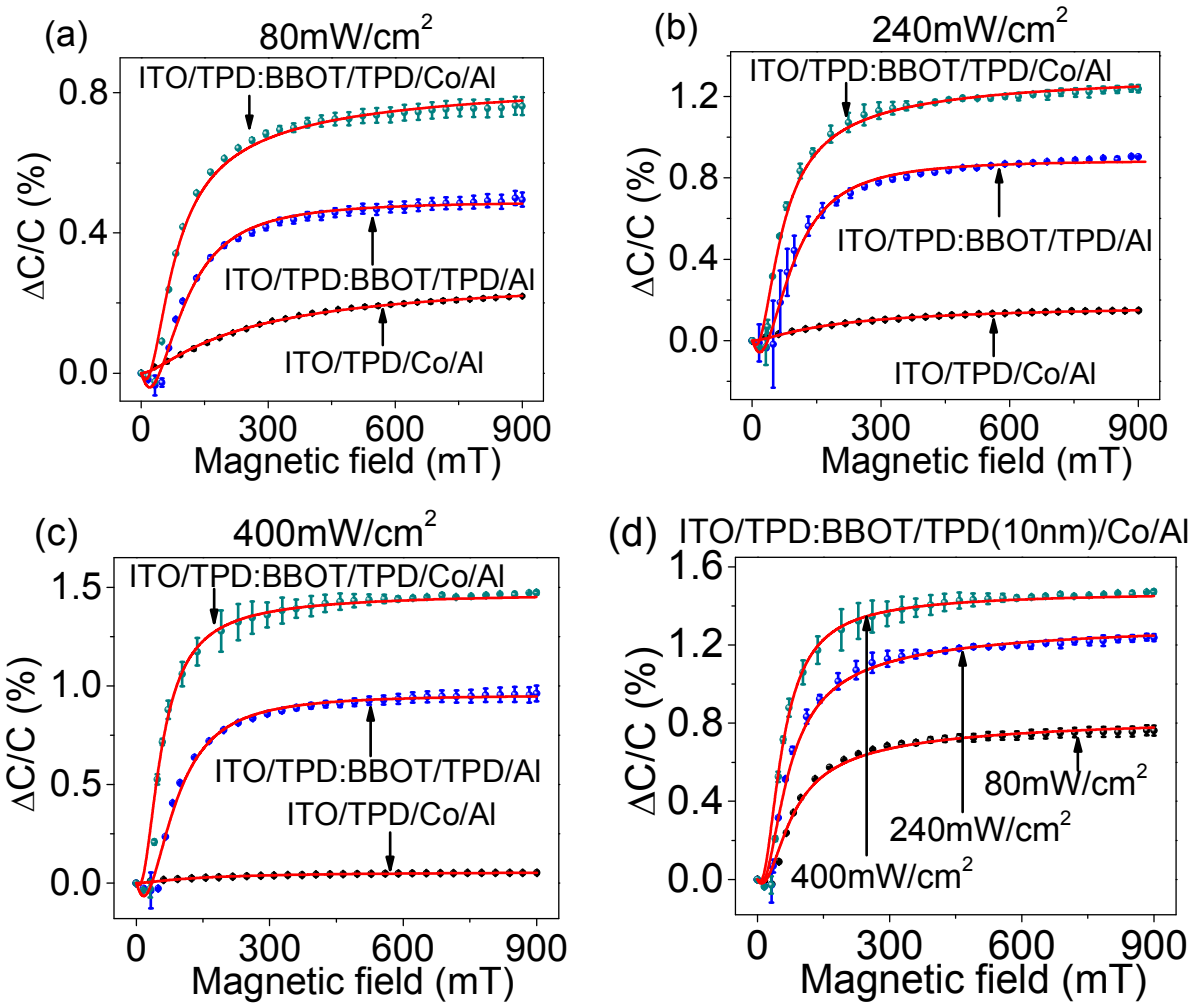

Figure S2, Magneto-dielectric from devices of ITO/TPD/Co/Al, ITO/TPD:BBOT/TPD/Al and ITO/TPD:BBOT/TPD/Co/Al under different photoexcitation intensities: (a) 80mW/cm^2 (b) 240mW/cm^2 and (c) 400mW/cm^2. (d) Magneto-dielectrics from the device ITO/TPD:BBOT/TPD(10nm)/Co/Al under different photoexcitation densities. Red solid lines are the fitting curves.

Magneto-dielectric from magnetized CT states is fitted with Equation (1):

$$MFC_{M-CTs} = \alpha \frac{B^2}{(B + B_0)^2} \quad (1)$$

Magneto-dielectric from optical CT states is fitted with Equation (2):

$$MFC_{P-CTs} = \left(\beta \frac{B^2}{(B+B_1)^2} + b \frac{B^2}{B^2 + {B_2}^2}\right) \quad (2)$$

Magneto-dielectric from device including interaction between magnetized CT states and optical CT states is fitted with Equation (3)

$$MFC_{Total} = MFC_{P-CTs} + MFC_{M-CTs} + MFC_{Interaction}$$

$$= \left(\beta \frac{B^2}{(B+B_1)^2} + b \frac{B^2}{B^2 + {B_2}^2}\right) + \alpha \frac{B^2}{(B+B_0)^2} + c * b \frac{B^2}{B^2 + {B_2}^2} * \alpha \frac{B^2}{(B+B_0)^2} \quad (3)$$

In Figure S2, the red lines are fitting curves by Equations (1), (2) and (3) with parameters in Table SI.

Table SI, Parameters used for fitting magneto-dielectric in Figure S2.

	α	β	b	c	B_0	B_1	B_2
80mW/cm^2	0.00284	-0.00139	0.00677	13.14961	120.79591	9.02418	68.23839
240mW/cm^2	0.00184	-0.00232	0.01145	109.82208	99.25493	9.02418	59.13854
400mW/cm^2	0.00061	-0.00281	0.01545	164.43385	73.24778	9.02418	50.66152