Two Dimensional Crystals of Ferromagnetic [MnCr(oxalate)<sub>3</sub>]<sup>-</sup> Layer Alternately Stacked with Supramolecular Cations of (2-(*x*-phenyl)ethan-1-aminium)<sup>+</sup>[18]crown-6 (x = H, *o*-fluoro, *m*-fluoro, and *p*-fluoro)

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The magnetoelectric (ME) effect in multiferroics is a prominent cross correlation phenomenon, in which the electric field controls the magnetization, and the magnetic field rules the electric polarization. A strategy for inducing ME effect in hybrid crystals is that enhancing the correlation between two relatively independent units, which rule the electric or magnetic order respectively.<sup>[1]</sup>

Here we introduced the supramolecular cations of (2-(x-phenyl)ethane -1aminum)<sup>+</sup>[18]crown- 6 (x = H, o-fluoro, m-fluoro, and *p*-fluoro) as a possible ferroelectric moiety between ferromagnetic honeycomb layer of [MnCr(oxalate)<sub>3</sub>]<sup>-</sup> to obtained four hybrid crystals 1-4. As shown in Figure 1, crystals exhibited layered structures, 2D where cations of ((2phenyl)ethane-1aminium)<sup>+</sup> in 1 and (2-(ofluorophenyl)ethan-1-

in

2

partly

 $aminium)^+$ 

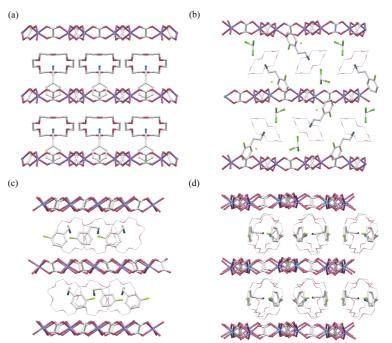


Figure 1. Crystal structures of (a) **1**, (b) **2**, (c) **3**, and (d) **4** 

injected into the layer of [MnCr(oxalate)<sub>3</sub>]. The presence of fluorine substituents on *m*- and *p*-positions hindered cations intersperse [MnCr(oxalate)<sub>3</sub>]<sup>-</sup> layer in crystal **3** and **4**, resulting in relatively independent layered structures. The crystal structures, dielectric, and magnetic properties of **1-4** will be discussed in detail.

1) Lee J H, Fang L, Vlahos E, et al. Nature, 2010, 466, 7309.