Supplementary Informations:

# Controlling the spatial arrangement of organic magnetic anions adsorbed on epitaxial graphene on Ru(0001)

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Potential Energy Surface of the neutral dimers.

The potential energy surfaces (PESs) of the two neutral dimers, shown in Figure S1(a,b), present remarkable quantitative differences. On the one hand, the PES of [TCNQ]$_2$ exhibits a well defined minimum at d = 8.02 Å and Θ = 33.84° at which the two molecules bind with BE = -0.32 eV. At this minimum, the binding is due to the formation of saturated C$\cdots$H bonds between the two molecules. On the other hand, in the 2D PES of [F4-TCNQ]$_2$, a rather shallow minimum - BE = -0.09 eV - is present around the same position, d = 8.63 Å and Θ = 31.74°, indicating that, the binding between two neutral F4-TCNQ molecules is already considerably weaker that that between two neutral TCNQ molecules. At the PESs minima, the LUMOs of the two molecules (insets in Figure S1(a,b)) resemble the HOMOs of the doubly charged dimers shown in the insets of Fig. 5(b,d) of the main text. In particular, in the LUMO of [TCNQ]$_2$ the intermolecular region is completely node-less, whereas in [F4-TCNQ]$_2$ the same region presents a series of nodes. This contrasting character is a consequence of the different nodal structure of the LUMOs of the monomers (see Figure S2(c,d)), which results in a favorable overlap between the LUMOs of two neighboring TCNQ monomers, and to a less favorable overlap between those of two F4-TCNQ monomers. Electrons transferred to these two orbitals will thus behave differently: due to the favorable overlap, those occupying the LUMO of [TCNQ]$_2$ will be delocalized more efficiently across the two molecules than those occupying the LUMO of [F4-TCNQ]$_2$. Indeed, moving from the neutral monomer to the neutral dimer, the increase in the EA due to the delocalization the an additional electron, ΔEA = EA$_{\text{dimer}}$ - EA$_{\text{monomer}}$, is -0.50 eV for [TCNQ]$_2$, and -0.47 eV for [F4-TCNQ]$_2$ dimer, *i.e.*, the change is $\sim$30 meV larger in the former that in the latter.
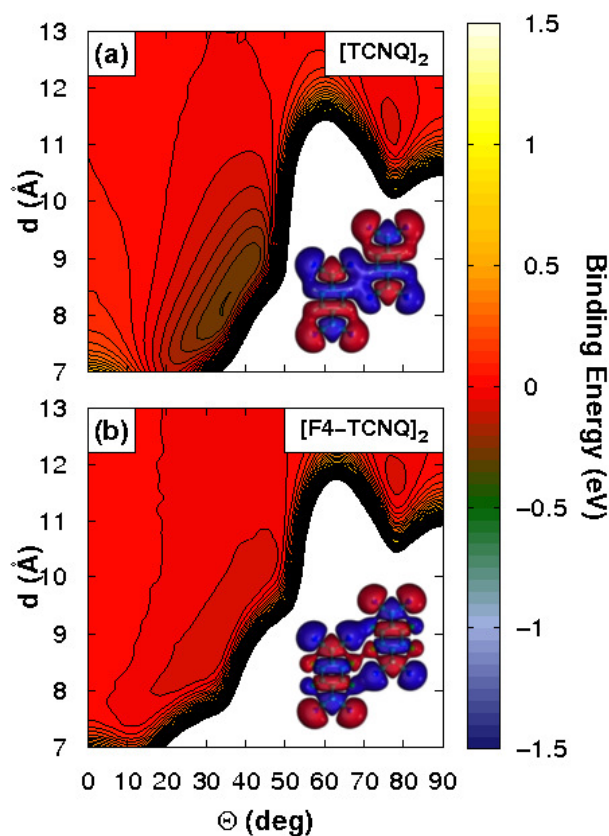
Figure S1: 2D potential energy surfaces (PES) of [TCNQ]$_2$ (a) and [F4-TCNQ]$_2$ (b). The separation between each isodensity curve is 5.0×10^{-2} eV. The inset in (a,b) show the LUMO of the molecule at the PES minimum, with the wavefunction colored according to its negative (red) and positive (blue) sign. Carbon, nitrogen, fluorine and hydrogen atoms are shown in cyan, blue, green and white, respectively.
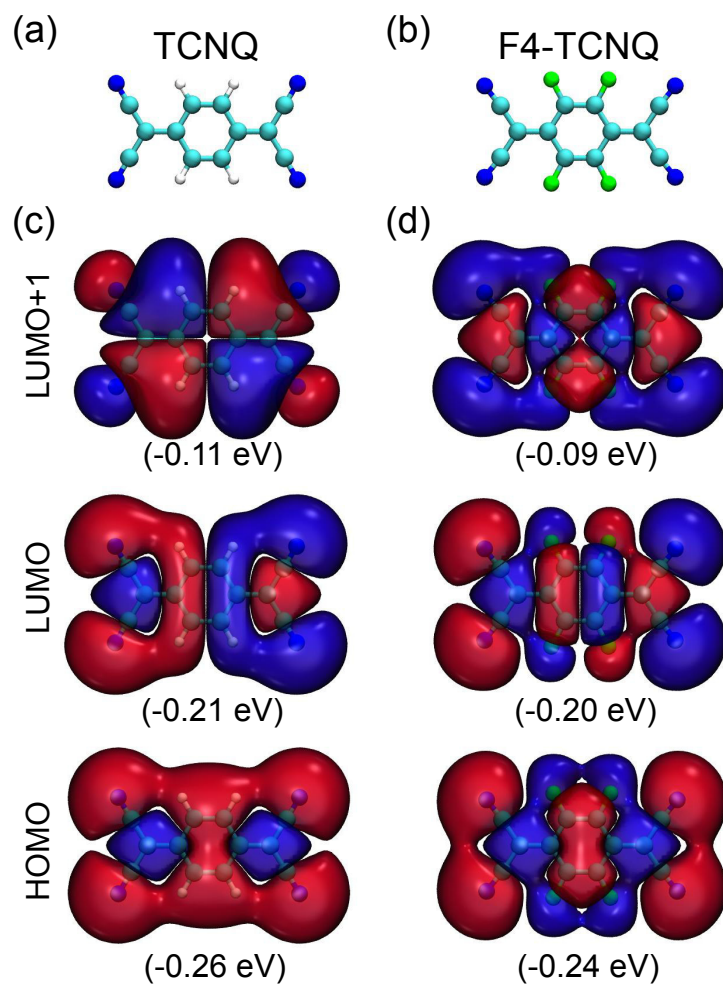
Figure S2: Structure and frontier molecular orbitals of neutral TCNQ (a,c) and neutral F4-TCNQ (b,d) in the gas-phase. In (c,d) the wavefunction is colored according to its negative (red) and positive (blue) sign. The number in parenthesis indicate the position in energy of each orbital with respect to the *vacuum* level. Carbon, nitrogen, fluorine and hydrogen atoms are shown in cyan, blue, green and white, respectively.

Table S1: Selected quantities calculated for the TCNQ and F4-TCNQ monomers and dimers, using the PBE and the B3LYP exchange correlation functionals. EA: electron affinity, BE: binding energy.

	PBE	B3LYP	PBE	B3LYP
	TCNQ monomer		**F4-TCNQ monomer**	
1st $EA_{neutral}$	-3.67	-3.75	-4.10	-4.28
1st $EA_{singly\ charged}$	0.48	0.42	0.04	-0.09
	TCNQ dimer		**F4-TCNQ dimer**	
BE (eV)	-0.35	-0.45	-0.12	-0.12
Θ (deg)	33.67	33.59	31.60	31.64
d (Å)	7.98	7.92	8.55	8.49
1st $EA_{neutral}$ (eV)	-4.18	-4.04	-4.58	-4.53