One-Dimensional Quasicrystals from Incommensurate Charge Order
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To bring to the fore the differences between the various charge-ordered phases discussed in the main text, it is convenient to express the modulation on top of the average charge modulation in each state as:

$$\rho(x) = \psi \cos(K \cdot x - \delta(x)).$$  \hspace{1cm} (1)

Here, $K$ is a wave vector commensurate with the lattice, which we will set to $K = \frac{2\pi}{3a}$ from here on. The commensurate charge density wave (CCDW), incommensurate charge density wave (ICDW), discommensuration state (DC), and quasicrystalline state (QC) can then be distinguished by considering the distinct behaviour of the phase $\delta(x)$ in each. This is shown in Fig. 1 below.

**FIG. 1:** (color online) Possible charge orderings discussed in the main text. For all cases the phase $\delta(x)$ defined in Eq. (1) is shown divided by $\pi$, as a function of position in units of the lattice spacing $a$. With these conventions, the commensurate phases are represented by straight lines with rational values for their slopes. The incommensurate phase is a straight line with an irrational slope. The discommensuration phase consists of horizontal sections connected by a regular, periodic array of discommensurations, which are vertical lines of height $2/3$. The spacing of the discommensurations is such that the phase returns to the dashed incommensurate line at every discommensuration. Finally, the quasicrystal consists of a quasiperiodic sequence of horizontal (period 3) sections of width 3, and sections at the slope corresponding to commensurate order with period 4, with width 4. The aperiodic sequence can be constructed by always choosing the section whose end-point stays closest to the dashed incommensurate line.
For the CCDW, $\delta(x)$ is zero. For the ideal ICDW, $\delta(x)$ is the straight line $\delta(x) = (Q - K)x$, with $Q$ the incommensurate wave vector. The discommensuration phase is given by sections of zero slope connected by vertical steps at regular intervals. The height of each step is such that the density wave jumps forward by precisely one lattice spacing. For $K = 2\pi/(3a)$, the step height is thus $2\pi/3$. The width of the horizontal sections is such that the average slope of the entire stepped structure is equal to $Q$. The quasicrystal also has average slope $Q$, but consists of horizontal sections separated by sections with slope $\delta(x) = (K' - K)x$, where $K'$ is a second commensurate wave vector: in this case $2\pi/(4a)$. The sections of zero and non-zero slope in this phase form a quasiperiodic sequence.

The DC phase often arises in real materials as the most energetically-favourable way of combining electronic density modulations with a lattice coupling [1]. In practice, the discommensurations in these materials will not be perfectly sharp. Instead, they broaden slightly and the connection in $\delta(x)$ with the neighbouring horizontal regions is made smooth. We follow McMillan in accounting for this using a Fourier expansion with coefficients, controlling the discommensuration width, selected so as to minimize the free energy of the DC state. The QC phase, on the other hand, is created by starting from an ideal ICDW and then shifting the peaks of the electronic density modulation directly onto the nearest atomic position. This creates a quasiperiodic sequence consisting of sections with one of two well-defined commensurate wave vectors.

The QC phase and ICDW both have quasiperiodic elements. For the ICDW, the sequence of atomic positions and charge maxima encountered as the material is traversed is quasiperiodic. This type of ‘quasiperiodicity’, however, is generic to any superposition of two incommensurate structures, including even things like two parallel picket fences seen from a distance. In contrast, the QC phase is quasiperiodic in each of its components individually. Moreover, it also consists of precisely two unit cells. Each piece of period four within the QC state is locally indistinguishable from any other piece of period four. In the ICDW, the atomic neighbourhood of each peak in the electronic structure is perfectly unique. In that sense, it contains infinitely many unit cells, rather than two. We therefore classify the QC state as a true one-dimensional quasicrystal, in contrast to the ICDW, which is only a generic quasiperiodic sequence of charge maxima and atoms.

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