

# Quasiperiodicity and 2D topology in 1D charge-ordered materials

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**Abstract** – The mathematical description of 1D quasicrystals has recently been linked to that of 2D quantum Hall states. The topological classification of 1D quasicrystals and the corresponding interpretation of their observed charge transport have been widely discussed. We demonstrate the equivalence of both 1D quasicrystals and 2D quantum Hall states to a mean-field treatment of charge order. Using the fractal nature of the spectrum of charge-ordered states we consider incommensurate order as a limit of commensurate. The topological properties of both are identical, arising from a 2D parameter space of phase and wave vector, and fit into class A of the Tenfold Way. The topological nature of all the systems can be tested by measuring a quantized particle transport upon dragging the charge order.

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**Introduction.** – The existence of quasicrystals and the observation of the quantum Hall effect are two seemingly unrelated discoveries that revolutionized condensed-matter physics in the second half of the twentieth century. The discovery of quasicrystals began with the mathematical observation that solids need not involve periodic repetitions of unit cells, but can involve aperiodic tilings of two or more inequivalent cells [1]. Experimental verification in the form of a quasicrystalline Al-Mn alloy followed shortly after [2].

The observation and subsequent theoretical characterization of the quantum Hall effect, on the other hand, prompted the realization that phase transitions cannot all be characterized solely in terms of the continuous symmetries they break [3,4]. It led to a new, topological, classification scheme of all disordered quadratic fermion theories according to discrete symmetries: the Altland-Zirnbauer classification or “Tenfold Way” [5,6]. Work has begun on extending this scheme to include the point and space group symmetries present in crystalline matter [7–10].

Recently these two cornerstones of modern condensed-matter physics have been shown to be mathematically linked through the ubiquitous Harper equation [11]. This equation has long been known to govern the quantum Hall effect [12]. It was also recently shown to provide a description of 1D quasicrystals when used to describe electrons in a 1D lattice with an incommensurate periodically modulated potential [13]. Based on the equivalence of

the underlying mathematical structures, quantized transport properties analogous to the quantum Hall conductance were predicted for the quasicrystal, and observed in an experimental realization using optical waveguides [14].

In this letter, we add a further equivalence by applying the Harper equation to the description of charge order in (quasi-)1D materials. As in the case of the quasicrystals in optical waveguides, we find that a mean-field description of the charge-ordered state can be labelled by the same set of topological quantum numbers as the 2D quantum Hall effect. We further demonstrate that the incommensurate charge-ordered state indeed has a quasicrystalline character.

A question receiving much attention at the moment is whether quasicrystalline materials, being somewhere between order and disorder, fit into the Tenfold Way [14–19]. The waveguide experiments involve families of quasicrystals, resulting in a 2D parameter space [20]. The observed integer set of edge states is in agreement with the Tenfold Way’s prediction for a 2D parameter space provided the Cartan symmetry class is A, C, or D [5,6]. By interpreting quasicrystalline charge order as a limiting case of a sequence of different crystalline charge orders, we find that the topological properties are unchanged in taking the limit, and both cases sit in class A of the Altland-Zirnbauer scheme. We show that this topological classification of charge-ordered materials is robust beyond the mean field, and we propose an experimental test of

the topological character of charge-ordered 1D materials. This provides experimental access to a novel type of quantized adiabatic particle transport predicted in related systems [21].

### Incommensurate charge order and quasicrystals.

– Quasicrystals are defined to be quasiperiodic tilings of two (or more) inequivalent unit cells [1]. A quasiperiodic sequence contains no subset of finite length which can be tiled periodically to complete the set, but nor is it devoid of order. One way to see this in 1D is through the fact that a 1D quasicrystal can be generated from the projection of a 2D crystal [1]. As shown in fig. 1, a straight line can be drawn in a 2D lattice, in such a way that it hits exactly one lattice point. A second line is then drawn parallel to the first, going through the opposite corner of the same unit cell. Whenever a point of the 2D lattice falls between the lines, its projection onto the first line forms a site of the 1D quasicrystalline lattice. The projected cells form a never-repeating, quasiperiodic, sequence of two differently sized unit cells making up the 1D quasicrystal.

A remnant of the 2D projection can be seen in the quasicrystal's diffraction pattern, which is generated by two different reciprocal lattice vectors [1]. By continuously shifting the set of parallel lines along one of the 2D lattice directions, a family of related quasicrystals is formed, all of which have the same diffraction pattern. The family is known as a local isomorphism class. Two quasicrystals are locally isomorphic if and only if every finite sequence of cells appearing in one also appears in the other [1].

A projection method similar to that used to define quasicrystals can be employed to generate the electron density modulations in (quasi-)1D charge-ordered materials. In such systems, chains of atoms spontaneously develop a modulation in their electronic density that does not coincide with the periodicity of the underlying atomic lattice. They include well-studied materials like NbSe<sub>3</sub>, KCP, and TTF-TCNQ, all of which contain weakly coupled 1D chains [22–26].

Specifically we consider a model 1D atomic lattice with spacing  $a$  and a modulation of charge density with period  $2\pi/Q \neq a$ . If the modulation period of the electron density  $2\pi/Q$  is not an integer multiple of the lattice spacing  $a$ , the charge order is said to be incommensurate. In this case, the sequence of atomic sites and positions of maximum electronic charge form a quasiperiodic, never-repeating, pattern. The combined sequence of sites and charge maxima can therefore be considered a quasicrystal. The situation can then be interpreted as arising from the projection of a second 1D lattice with lattice spacing  $na$ , where  $n$  is the lowest integer number larger than  $2\pi/Qa$ , rotated to a point where the projections of its sites onto the original line coincide with the maxima of the charge modulations (see fig. 1). As with the quasicrystal considered before, a family of locally isomorphic quasicrystalline charge-ordered states can be generated by

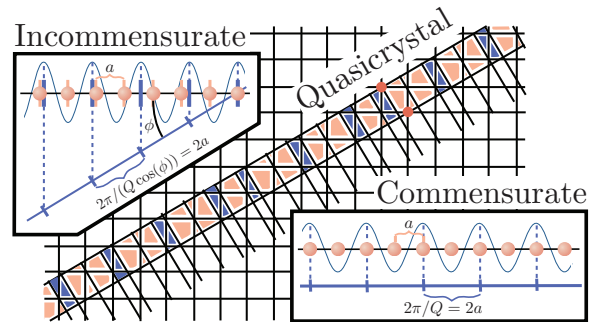


Fig. 1: (Colour on-line) Main panel: generating a 1D quasicrystal by projecting a 2D lattice. A pair of parallel lines is overlaid on the square lattice, in such a way that the lines hit only a single pair of lattice points on opposing corners of one of the unit cells. The lattice points between the lines are then projected, and form the sites of the 1D quasicrystal. The inequivalent cells are coloured red and blue for clarity. Top inset: generating an incommensurate charge-ordered state with period  $2\pi/Q$  on an atomic lattice with spacing  $a$ , by a similar projection method. In this case the sequence of atomic sites and maxima of the charge density modulation (red balls and blue tick marks) is quasiperiodic. Bottom inset: the same method also trivially generates commensurate charge order, when the angle between the two lines is rational (in this case zero).

continuously shifting the rotated line along the original lattice direction.

**Energy spectrum.** – The emergence of charge order can be described by a model of spinless fermions hopping on a 1D lattice, in the presence of nearest-neighbour density-density interactions (strength  $h$ ) [27]. At the mean-field level, the interactions can be decoupled by the introduction of the complex two-component order parameter:

$$\underline{\Delta}_Q(h) = 2h \sum_k (1, -\cos(k) \cos(Q/2))^T \langle \hat{c}_{k+Q}^\dagger \hat{c}_k \rangle. \quad (1)$$

Here  $Q$  connects two points on the Fermi surface of the 1D band structure. The upper and lower components correspond to the presence of site-centred and bond-centred charge order, respectively. In general, they will be coupled through the self-consistency condition for the mean-field expectation values. In the mean-field Hamiltonian the two components always appear in the combination

$$\Delta_Q(k, h) = (\cos(Q), \cos(k) \cos(Q/2)) \cdot \underline{\Delta}_Q(h). \quad (2)$$

Using this definition, the mean-field Hamiltonian becomes

$$\hat{H} = \sum_{0 \leq k < 2\pi/a} \left\{ \frac{1}{2} \epsilon_k \hat{c}_k^\dagger \hat{c}_k + \Delta_Q \hat{c}_k^\dagger \hat{c}_{k+Q} + \text{H.c.} \right\}, \quad (3)$$

where  $\epsilon_k = 2t(1 - \cos(ka)) - \mu$  describes the bare band structure resulting from the hopping integral  $t$  and chemical potential  $\mu$ , and the order parameter is written as

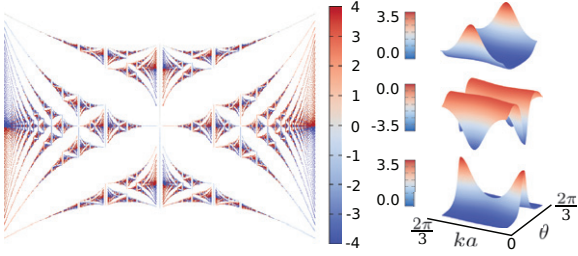


Fig. 2: (Colour on-line) Left: allowed energies (range  $[-4t, 4t]$ ) for all values of  $ka$  and  $\theta$  vs. filling fraction  $p/q \in (0, 1)$  with  $q \in [2, 50]$  (discarding noncoprime fractions), in the mean-field equation (4), with  $\underline{\Delta}_Q \rightarrow (t/\cos Q, 0)^T$ . The spectrum is almost identical to Hofstadter's, describing allowed electron energies in the integer quantum Hall effect [12]. The colour of each sub-band indicates the sum of Chern numbers  $C_1$  up to and including that sub-band (truncated at  $\pm 4$  for clarity). Right: the Berry curvatures for the sub-bands at  $1/3$  filling. The integrals over these surfaces (giving the  $C_1$  values) are 1,  $-2$ , and 1 in units of  $2\pi$ .

$\Delta_Q = |\Delta_Q| \exp(i\theta)$ . The phase  $\theta$  of the order parameter corresponds to the distance between one given atom and a particular maximum in the electronic charge distribution. Varying  $\theta$  thus corresponds to sliding the charge modulation along the atomic lattice, and in the incommensurate case this implies a continuous variation of the quasicrystalline charge order through the members of the local isomorphism class. If the chemical potential is tuned to a rational fraction  $p/q$  of the bare bandwidth  $4t$ , the charge order in the ground state of eq. (3) has period  $qa$ . Working in a reduced Brillouin zone of length  $2\pi/qa$ , the Hamiltonian can be rewritten as

$$\hat{H} = \sum_{0 \leq k < 2\pi/qa} \left( \hat{c}_{k+Q}^\dagger, \hat{c}_{k+2Q}^\dagger \dots \hat{c}_{k+qQ}^\dagger \right) H_k \begin{pmatrix} \hat{c}_{k+Q} \\ \hat{c}_{k+2Q} \\ \vdots \\ \hat{c}_{k+qQ} \end{pmatrix},$$

$$H_k = \begin{pmatrix} \epsilon_{k+Q} & \Delta_Q & 0 & \dots & 0 & \Delta_Q^* \\ \Delta_Q^* & \epsilon_{k+2Q} & \Delta_Q & 0 & \dots & 0 \\ 0 & \Delta_Q^* & \ddots & \ddots & & \vdots \\ \vdots & 0 & \ddots & & & 0 \\ 0 & \vdots & & & & \Delta_Q \\ \Delta_Q & 0 & \dots & 0 & \Delta_Q^* & \epsilon_{k+qQ} \end{pmatrix}. \quad (4)$$

In the presence of charge modulation with period  $qa$ , the off-diagonal elements in this Hamiltonian cause the energies to split into  $q$  sub-bands, separated by gaps of order  $|\Delta_Q|$  [27]. For the case  $\underline{\Delta}_Q \rightarrow (t/\cos Q, 0)^T$  eq. (4) becomes the matrix form of Harper's equation, applied by Hofstadter to the 2D electron gas in the presence of a magnetic field [12]. The plot showing all eigenvalues (allowed energies) for all possible values of  $ka$  and  $\theta$ , vs. filling

fraction, is known as Hofstadter's butterfly [28] (fig. 2). Notice that in contrast to Hofstadter's original picture, the colour coding in fig. 2 indicates that in the presence of an infinitesimal amount of bond-centred order, infinitesimally small gaps will appear halfway up the spectrum for fillings with even denominator.

**Topological classification.** – The Tenfold Way classifies the allowed topologies of all gapped, disordered, quadratic fermion theories into ten classes based on the dimension of the parameter space and the behaviour under charge conjugation  $\mathcal{C}$ , time reversal  $\mathcal{T}$ , and the combined operator  $\mathcal{S} = \mathcal{C}\mathcal{T}$ . The classes take labels from the Killing-Cartan classification of Lie groups [5]. A measurable consequence of a nontrivial topology is the existence of edge states robust to disorder, characterized by a nonzero Chern number  $C_1$  [4].

By inspecting the form of  $H_k$  in eq. (4) we can identify the symmetry class of the model according to the Tenfold Way. The original classification was developed for translationally invariant systems, and extensions to crystalline systems have only recently been formulated [9,10]. In 1D, however, the original formulation receives little complication, since there are only two space groups (trivial and dihedral). The mirror symmetry of the dihedral case is dealt with elsewhere [7,8], and is not relevant for the case of charge-ordered materials, as the presence of a nonzero  $\theta$  will spontaneously break any mirror symmetry of the atomic lattice. We can thus classify the 1D charge-ordered state directly according to the Tenfold Way.

The chiral cases AIII, BDI, and CII do not apply to eq. (4), as they require zeroes along the diagonal of  $H_k$ . The Bogoliubov-de Gennes cases C, D, DI, and DIII can likewise be seen to be inapplicable, since the presence of scatterings  $k \rightarrow k \pm Q$  between reduced Brillouin zones rules out the possibility of expressing  $H_k$  as a  $2 \times 2$  matrix, except in the special case of half-filling<sup>1</sup>. Within the remaining three classes, the fact that  $H_k$  is necessarily complex directly implies that 1D charge-ordered states fall into the symmetry-free class, A.

In class A, all systems with an odd-dimensional parameter space are topologically trivial, whereas systems with even-dimensional parameter spaces may have any integer number of edge states. In cases where a generalized linear response function can be formed, the corresponding susceptibility will then be precisely quantized [4–6,29]. In the case of charge order, this is manifested in a quantization of the conductance. This may be understood by realizing that the phase of the order parameter,  $\theta$ , which signifies the position of the charge modulation relative to the atomic lattice, may be varied in the mean-field solution of eq. (4). The band structure can thus be drawn within a 2D parameter space spanned by  $\theta$  and  $ka$ , as indicated on the right of fig. 2 for the case  $p/q = 1/3$ .

<sup>1</sup>Notice that precisely at half-filling there is an additional  $\mathcal{C}$  (particle-hole) symmetry, with  $\mathcal{C}^2 = -1$ , which places the Hamiltonian at that filling fraction in class C.

Each of these 2D sub-bands can be assigned a topological quantum number, the Chern number  $C_1$ , by integrating its Berry curvature over the 2D space. In this specific problem,  $C_1$  can also be found by solving a Diophantine equation [4,28]. The quantized conductance is the sum of the Chern numbers for all occupied sub-bands [4]. In the quantum Hall effect, the Hall conductivity  $\sigma_{xy}$  is given by  $(e^2/2\pi\hbar)C_1$ . In the present case, the response function associated with the topological invariant is instead a “quantized adiabatic particle transport” [21,30,31]: adiabatically cycling  $\theta$  through a full period (sliding the charge density modulation through a single wavelength) results in the transfer of an integer number  $C_1$  of electrons across the length of the 1D chain.

Notice that the symmetry-free class A, into which the charge-ordered states fall, also signifies that  $\mathcal{C}$ ,  $\mathcal{T}$ , and  $\mathcal{S}$  symmetries are broken. For any filling other than one-half, the breaking of  $\mathcal{C}$ , or particle-hole symmetry, is clear. To see that time reversal symmetry  $\mathcal{T}$  is also broken, consider the order parameter  $\Delta_Q$  as defined by eqs. (1) and (2). In the presence of a complex order parameter, the expectation value  $\langle \hat{c}_{k+Q}^\dagger \hat{c}_k \rangle$  signifies a current in reciprocal space, which changes sign under time inversion. Notice that this current arises from the scattering of electrons around the torus of the reduced Brillouin zone. Time reversal symmetry  $\mathcal{T}$  is thus broken for any nonzero value of  $\theta$ , even though there is no equivalent real-space current due to interference between scatterings by  $\pm Q$ . Note that a nonzero  $\theta$  also implies the existence of an electrical polarization in real space [32,33]. The definition of the polarization, however, is ambiguous unless a cut is introduced in the system, at which point the crystal as a whole gains charges on either end [32,33].

**Topology of quasicrystals.** – So far we only considered the topological classification of commensurately charge-ordered states, since eq. (4) only allows solutions for commensurate phases to appear in fig. 2. Nevertheless we can infer certain properties of the incommensurate states from the fractal structure of the spectrum. A close inspection of the underside of the lower left wing, for example, reveals that summing the Chern numbers up to that gap yields  $C_1 = 1$  for any rational filling fraction. In fact, this global structure of the fractal is made robust by Bellissard’s “gap labelling theorem”, which shows both that the gaps at irrational filling fractions can each be assigned a unique number equal to the sum of Chern numbers, and that these labels are robust under perturbations including a shift of filling fraction [34,35]. The global structure of the fractal therefore guarantees that any incommensurate state at irrational filling fraction  $\eta$  has the same nonzero sum of Chern numbers as the state with commensurate order at rational filling  $p/q = \eta - \epsilon$ , with  $\epsilon \rightarrow 0$ . Importantly, the arguments placing our commensurate problem in the symmetry-free class A are unaffected by the limiting procedure, and hence we conclude that the incommensurate charge-ordered states fall into the same class.

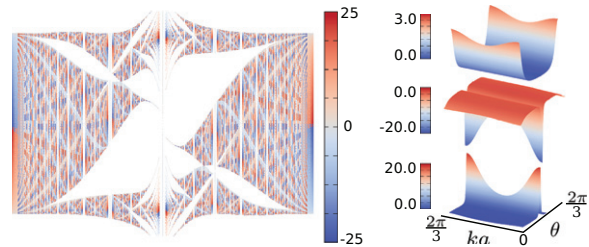


Fig. 3: (Colour on-line) Left: the allowed energies (scale truncated at  $[-2.5t, 2.5t]$ ) as a function of filling fraction  $p/q \in (0, 1)$ ,  $q \in [2, 50]$  for the self-consistent solution of eq. (3) with  $h = t$ . Each sub-band is adiabatically connected to the corresponding sub-band in fig. 2. This implies that the sums of Chern numbers of filled sub-bands, indicated by the colour scale, are the same. Right: the Berry curvatures for the three sub-bands at  $1/3$  filling. Although the shapes are different from those in fig. 2, the integrals over them remain  $1, -2, 1$  in units of  $2\pi$ .

Since the incommensurate states represent quasicrystalline charge order, this result implies that 1D quasicrystalline materials such as the ones studied in the recent optical waveguide experiments also belong to class A [14,20]. This fact, alongside the observation of quantized transport in these materials, confirms that the relevant parameter space in the waveguide experiments is 2D [20]. As in the case of commensurate charge order, the parameter space is spanned by  $ka$  and  $\theta$ , where  $\theta$  is the parameter which labels different members of the local isomorphism class. Additionally, the assignment of 1D quasicrystals to class A implies they too have broken time reversal symmetry  $\mathcal{T}$ . Although there is no obvious way in which quasicrystalline materials break this symmetry, the interpretation of incommensurate charge order as a quasicrystal makes clear that, as in the commensurate case, it is due to the presence of a complex-valued order parameter and the corresponding current in reciprocal space.

**Measuring quantized transport.** – In the analysis of the spectrum of eq. (4) we imposed a fixed value of  $\Delta_Q(h)$ , and did not take into account the self-consistency condition of eq. (1). Although the self-consistency requirement can affect both the sizes of the gaps in fig. 2 and the mirror symmetry about  $Q = \frac{1}{2}$ , the topology of the sub-bands should not be affected, as the gaps remain open for any nonzero  $h$ . This implies that the Chern numbers characterizing the contribution of each sub-band to the adiabatic transport will be protected. Solving eq. (3) self-consistently, we obtain the allowed energies and Chern numbers displayed in fig. 3 [4,28]. As expected, the values of  $C_1$  associated with each sub-band are unaltered by deformations of the gap sizes. It should be noted that solitons, higher harmonics, and other nonlinear effects can similarly affect the quantitative shape of the spectrum, but again this will not alter the topologies of the bands [36,37].

Real materials cannot maintain the infinitely fine structure of a fractal band. Instead, the gaps separating consecutive sub-bands will be closed when they become

of the order of the effective energy scale set by local impurities, disorder, or lattice defects, and the Chern numbers of the two merging bands are added together as mandated by the gap labelling theorem. Large enough gaps, however, may be expected to survive. For the corresponding filling fractions, the sum of Chern numbers remains nonzero, even for incommensurate charge order in the presence of defects and disorder. The quantized adiabatic particle transport associated with the topology and quasicrystalline nature of incommensurate charge order should therefore be present in experimentally accessible charge-ordered materials.

Measuring the quantized transport is complicated, however, by the fact that impurities in real materials can pin the charge order in place, inhibiting its freedom to slide along the atomic lattice. It is well established that an applied voltage may overcome the pinning potential in incommensurately charge-ordered materials, allowing them to conduct even in the presence of pinning centres [27,38–40]. The current produced includes both the adiabatic particle transport originating in the band topology, and the effects of nonadiabatic band-mixing. Since we are here only interested in the adiabatic transport, we propose an alternative way of accessing the quantized particle transport in both commensurate and incommensurate charge-ordered states, based on the use of atomic condensates in optical lattices. Such systems are more readily controllable and defect-free than 1D chains in real materials. Indeed, the Harper equation was recently realized in a cold-atom setup [41]. The charge density modulations in such systems can be intentionally locked into place either using the confinement potential or by simulating a strong impurity on a single site. Provided that this intentional pinning does not disrupt the large-scale gap structure, the topology of the sub-bands remains unaffected. By manipulating the impurity location or confinement potential, the charge density modulation can then be dragged along the optical lattice, continuously cycling the value of  $\theta$ . As the corresponding charge-ordered pattern moves through one wavelength, an integer number of atoms will be transported across the length of the optical lattice. Again, the integer is the sum of Chern numbers of occupied sub-bands.

**Conclusions.** – In this letter we have shown the equivalence between families of locally isomorphic 1D quasicrystals and a mean-field model of incommensurate charge order in 1D materials. Both fall into class A of the Altland-Zirnbauer classification of topological phases, with broken  $\mathcal{C}$ ,  $\mathcal{T}$ , and  $\mathcal{S}$  symmetries. Having a two-dimensional parameter space, they exhibit an integer set of possible edge states, in agreement with recent results in optical-waveguide experiments [13,14]. The gap labelling theorem further allows us to describe the global structure of the fractal pattern of allowed energies [34,35], and hence to interpret incommensurate charge-ordered materials as the limit of a series of commensurate phases. Importantly, the

space group of the commensurate case is trivial, meaning no additional restrictions to the Tenfold Way need to be introduced, and the limiting procedure preserves the topological class. It is thus shown that the Chern numbers, describing both the edge states and the quantized transport properties of all charge-ordered phases, arise from an integral of the Berry curvature over the 2D space generated by  $k$  and the phase  $\theta$  labelling different members of the local isomorphism class. The topological properties of the system survive in the self-consistent solution, and are robust against the inclusion of weak fluctuations, defects, and impurities. The large-scale structure of the Hofstadter spectrum guarantees the possibility of quantized adiabatic particle transport in any charge-ordered material, as long as the gap size is nonzero.

Finally, we propose an experimental test of the topological properties of charge-ordered materials by exploiting the possibility of directly controlling the phase of a charge density modulation in atomic condensates in optical lattices. Dragging the modulation along a single wavelength will lead to the transfer of a quantized number of atoms across the system, providing a standard of conductance in cold-atom setups. Through the equivalence between incommensurately charge-ordered states and quasicrystals we see that the results of this experiment give a re-interpretation to previous experiments in optical waveguides [14].

Charge-ordered materials unite a range of seemingly distinct physical systems, including forming a bridge between 1D quasicrystals and 2D topological phases. In this way they provide an intuitive, naturally occurring, physical system in which the consequences of topological order can be straightforwardly realized and understood.

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