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## Quantum cluster quasicrystals

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Quasicrystals remain among the most intriguing materials in physics and chemistry. Their structure results in many unusual properties, including anomalously low friction as well as poor electrical and thermal conductivity, but it also supports superconductivity, which shows that quantum effects in quasicrystals can be quite unique. We theoretically study superfluidity in a model quantum cluster quasicrystal. Using path-integral Monte Carlo simulations, we explore a two-dimensional ensemble of bosons with the Lifshitz-Petrich-Gaussian pair potential, finding that moderate quantum fluctuations do not destroy the dodecagonal quasicrystalline order. This quasicrystal is characterized by a small yet finite superfluidity, demonstrating that particle clustering combined with the local cogwheel structure can underpin superfluidity even in the almost classical regime. This type of distributed superfluidity may also be expected in certain open crystalline lattices. Large quantum fluctuations are shown to induce transitions to cluster solids, supersolids, and superfluids, which we characterize fully quantum mechanically.

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### I. INTRODUCTION

Despite the order-of-magnitude differences in the characteristic length and energy scales, solid-state and soft matter share many features and phenomena exemplified by the well-established analogy between smectic liquid crystals and superconductors [1,2] and the thriving field of electronic liquid-crystalline mesophases [3,4]. An interesting soft-matter effect with possible analogies at a much smaller scale—e.g., in Rydberg-excited Bose-Einstein condensates [5,6]—is clustering. In a good solvent, dilute polymers can be considered as extended finite-size objects, but if the concentration is large enough, they interpenetrate and overlap. The effective potential between them depends on architecture, functionalization, etc. [7], and in some cases, e.g., in amphiphilic dendrimers [8], it may promote ordered phases consisting of evenly spaced multiple-occupancy clusters [9,10]. Such clustering can be viewed as an instability resulting from a negative component of the Fourier transform of the potential [11].

The physical properties of cluster phases are controlled by their symmetry and by the morphology of the clusters, which may be spherical, cylindrical, sheetlike, inverted, or even bicontinuous [12]. Both symmetry and morphology are determined by an interparticle interaction, and a synthetic approach to generate a host of different phases including

cluster quasicrystals (QCs) is based on a simultaneous instability at two length scales [13–15]. The cluster phases should all exhibit some degree of activated hopping, and the ensuing finite diffusivity [16] is expected to be more prominent in the dodecagonal QCs where the neighboring clusters at the perimeter of the characteristic cogwheel-like patches are rather close to each other [14].

In a quantum system, such dynamics could well lead to novel types of supersolidity, implying the coexistence of (quasi)crystalline and superfluid behavior. To explore this possibility, we theoretically study a two-dimensional (2D) ensemble of bosons with a Lifshitz-Petrich-Gaussian pair interaction that produces a classical dodecagonal cluster QC.

We use path-integral Monte Carlo (PIMC) simulations to show that the QC remains stable if quantum fluctuations are not too large and that it supports local distributed superfluidity in clusters by a kind of percolating network. In these two respects, it departs from known supersolids [5,17–23] and superglasses [24–29]. We show that increased quantum fluctuations induce a series of phase transitions to cluster solids, supersolids, and superfluid phases. Our findings open different possibilities for weak quantum behavior characterized by local superfluidity, say, in cluster systems based on honeycomb or kagome lattices and their 3D analogs.

This paper is organized as follows: In Sec. II we introduce the model Hamiltonian, whereas Sec. III presents the properties of the dodecagonal QC structure when quantum fluctuations are taken into account. Findings and conclusions are outlined in Sec. IV.

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## II. MODEL HAMILTONIAN FOR QUANTUM CLUSTER QUASICRYSTALS

We consider an ensemble of  $N$  two-dimensional bosons of mass  $m$  with a many-body Hamiltonian

$$\hat{H} = -\frac{\hbar^2}{2m} \sum_{i=1}^N \nabla_i^2 + \sum_{i<j}^N V(|\mathbf{r}_i - \mathbf{r}_j|), \quad (1)$$

where

$$V(r) = \exp(-\sigma^2 r^2/2) \sum_{k=0}^4 C_{2k} r^{2k} \quad (2)$$

is the Lifshitz-Petrich-Gaussian pair potential [14] and  $\mathbf{r}_i \equiv (x_i, y_i)$  is the position of the  $i$ th particle. If the parameters  $\sigma$  and  $C_{2k}$  are chosen such that its Fourier transform features two equal-depth negative minima and the ratio of the corresponding wave vectors is  $\sqrt{2 + \sqrt{3}} \approx 1.93$ , this potential leads to a dodecagonal QC pattern in a classical system [14]. We too use this particular set of parameters, focusing on quantum effects in the dodecagonal cluster QC. These effects depend on the magnitude of quantum fluctuations encoded by the de Boer parameter [30]

$$\Lambda = \sqrt{\frac{\hbar^2}{mr_0^2 V_0}}, \quad (3)$$

where  $V_0 = V(0)$  is the pair potential at  $r = 0$  and  $r_0$  is the characteristic length given by the inverse of the wave vector corresponding to the first minimum of the transform of  $V(r)$ .

## III. 12-FOLD QUANTUM CLUSTER QUASICRYSTAL

We employ PIMC simulations [31] based on the continuous-space worm algorithm [32] to find the equilibrium state of Eq. (1) at a fixed temperature and a fixed number of particles  $N$  (canonical ensemble), with  $N$  between 2048 and 8192. All simulations were carried out using periodic boundary conditions along both directions. In particular, we study the ensemble at temperatures around the range where the classical dodecagonal QC is stable, first at small  $\Lambda$ . Figure 1 shows the quantum dodecagonal QC for  $\Lambda = 0.1$ , reduced temperature  $t = k_B T/V_0 = 0.05$ , reduced density  $\rho r_0^2 = 0.8$ , and  $N = 8192$ . In this figure we focus on the semiclassical limit, i.e., on boltzmannons, where the zero-point motion due to quantum fluctuations is accounted for, whereas the world-line exchanges leading to superfluidity are initially excluded. Figure 1(a) shows a snapshot of the projection of world lines onto the  $xy$  plane obtained by tracing over the imaginary time evolution; this is a good representation of the square of the semiclassical many-body wave function [31]. In Fig. 1(a), the paths are essentially localized around the energy minima of the QC structure observed in the classical limit in Ref. [14] despite a somewhat larger reduced temperature (0.05 vs 0.03) and despite quantum fluctuations.

The similarity of the semiclassical and the classical QCs is further corroborated by the radial distribution functions  $g(r)$  [33] in Fig. 1(b), which are virtually identical except close to  $r = 0$ : The quantum  $g(r)$  is somewhat larger than the classical one, indicating increased local fluctuations of the

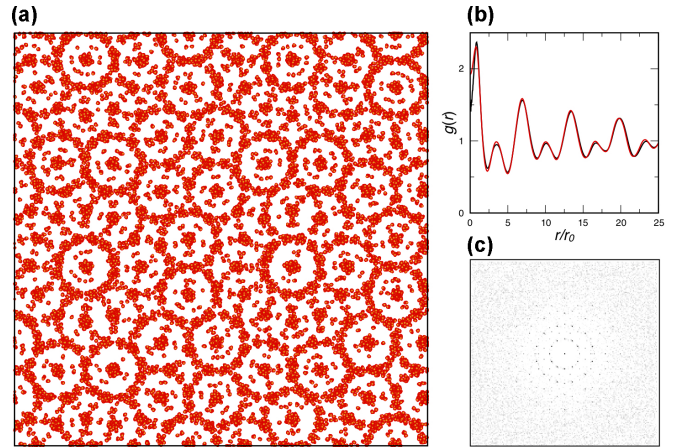


FIG. 1. Quantum QC in the semiclassical regime: (a) PIMC snapshot of the dodecagonal QC at  $\Lambda = 0.1$ , reduced temperature  $t = 0.05$ , and reduced density  $\rho r_0^2 = 0.8$ ; here,  $N = 8192$ . (b) Radial distribution function  $g(r)$  for  $\Lambda = 0.1$  (red line) and  $\Lambda = 0$  (black line). (c) Fourier transform of the positions of world lines in (a). The parameters of the interparticle potential in Eq. (2) are  $\sigma = 0.770\,746$ ,  $C_0 = 1$ ,  $C_2 = -1.094\,56$ ,  $C_4 = 0.439\,744$ ,  $C_6 = -0.049\,273\,9$ , and  $C_8 = 0.001\,831\,83$  as reported in Ref. [14].

particles [inset to Fig. 2(e)]. We note that the introduction of Bose-Einstein statistics further enhances this effect (see below).

The Fourier transform of the  $\Lambda = 0.1$ ,  $t = 0.05$  semiclassical dodecagonal QC [Fig. 1(c)] evidently has a 12-fold rotational symmetry. Figure 1(c) was obtained by taking the averaged position of each world line (centroid coordinates) in space. In terms of position, the peaks agree with the stronger inner peaks characterizing the classical counterpart of our QC [14] as well as with those seen experimentally in, e.g., a dendrimer-micelle QC [34]. The variations in intensity—and especially the presence of the diffuse outer ring—reflect the different form factor and thus a different intracluster structure, as also observed in  $g(r)$  at small  $r$ .

Full quantum effects combining fluctuations and bosonic statistics are investigated in Fig. 2. As the de Boer parameter is increased at  $t = 0.05$  and  $\rho r_0^2 = 0.8$ , the ensemble undergoes three transitions [Figs. 2(a)–2(d)]. At  $\Lambda \approx 0.12$ , the dodecagonal QC shown in Fig. 2(a) is replaced by a hexagonal cluster crystal [Fig. 2(b)]; the clusters are well defined and evidently larger than those in the QC, their spacing being the same as the radius of the dodecagonal wheels in the QC. Given that the pair potential features two local minima [14], the increase of cluster size and their rearrangement suggest that at the larger  $\Lambda$ , intracluster quantum fluctuations render the smaller-distance minimum less effective. The structural differences between the two phases readily show in the radial distribution functions [Figs. 2(e) and 2(f)]. The modulation of  $g(r)$  in the cluster solid is very prominent, virtually vanishing between nearest-neighbor clusters, whereas in the QC it is considerably smaller.

We now turn to the quantum properties of the QC and the cluster solid, first monitoring the frequency of cycles of permutations involving  $L$  bosons denoted by  $P(L)$ , with  $1 \leq L \leq N$ . The occurrence of long permutation cycles in the

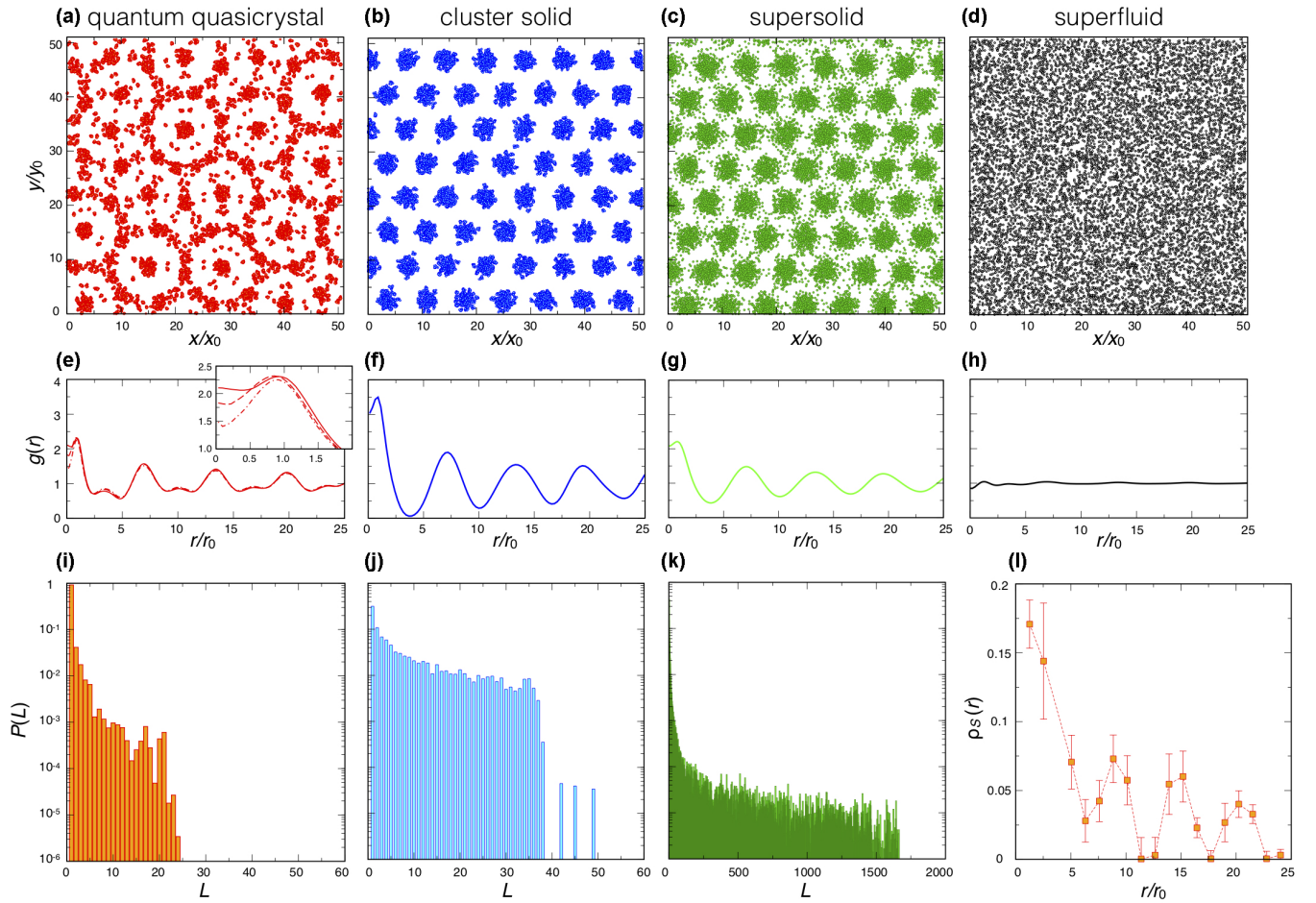


FIG. 2. Quantum quasicrystal and reentrant superfluidity: (a)–(d) Dodecagonal quantum QC, cluster crystal, cluster supersolid, and superfluid at  $t = 0.05$ ,  $\rho r_0^2 = 0.8$ , and  $\Lambda = 0.1, 0.141, 0.632$ , and  $1$ , respectively;  $N = 2048$ . (e)–(h) Radial distribution functions of the four phases in (a)–(d). Inset in (e): Distribution functions of QC for bosons (solid line), semiclassical boltzmannons (dashed line), and classical particles (dotted-dashed line). (i)–(k) Frequency of exchange cycles of length  $L$  in the QC, cluster solid, and supersolid in (a)–(c). (l) Superfluid density profile of the dodecagonal QC from (a).

histogram  $P(L)$  should be linked to the existence of a finite superfluid response throughout the system. In the  $\Lambda = 0.1$  QC, the distribution of  $P(L)$  stretches to  $L \approx 25$  [Fig. 2(i)]. Since the clusters contain about 18 particles, this implies a finite particle exchange between clusters, pointing to a local distributed superfluidity in this *quantum* QC phase. In the cluster solid [Fig. 2(j)], permutation cycles stay within single clusters, which contain about 36 particles.

The competition between the tendency of bosons to delocalize at low temperatures and the structure of the QC and cluster crystal is also reflected in the superfluid density. In the PIMC framework, the superfluid density  $\rho_s$  is evaluated by applying the linear response theory to address the response of the boundary motion of the ensemble. In this study, superfluidity was sampled by using the expression  $\rho_s = (4\rho m^2)/(\hbar^2 \beta I_{cl})(A^2)$ , where  $\beta = 1/k_B T$  and  $I_{cl}$  is the classical moment of inertia of the particles calculated with respect to the axis perpendicular to the  $xy$  plane. In the context of the path-integral formalism, the estimator  $A$  then gives the world-line area of closed particle trajectories projected onto the  $xy$  plane. Likewise, the local contribution to the superfluid density  $\rho_s(r)$  [35,36] is obtained by sampling the

radial dependence of the local area operator  $A \cdot A(r)$  and the corresponding local moment of inertia  $I_{cl}(r)$ . In a true periodic structure [see, for instance, the supersolid phase in Fig. 2(c)] the evaluation of  $\rho_s$  using the area estimator techniques gives results that are fully consistent with the well-known “winding number” estimator [37].

In the QC in Fig. 2(a),  $\rho_s$  is small but finite, the fraction of superfluid particles being about 1%–2% consistent with the measured exchange cycles and a picture of weak distributed superfluidity. On the other hand, the global superfluid signal is completely suppressed in both QC and cluster crystal.

Figure 2(l) shows the superfluid density profile in the QC, which is evidently nonuniform. By comparing the profile with the snapshot and  $g(r)$  in Figs. 2(a) and 2(e), respectively, we see that  $\rho_s(r)$  is small but non-negligible both in the central clusters of the dodecagonal wheels and around their perimeter. In fact, the local superfluid signal is nonzero in accordance with the structure of Fig. 2(a). Consistent with the quantum-mechanical exchanges shown in Fig. 2(j), in the cluster crystal  $\rho_s(r)$  should be finite (about unity as  $t \rightarrow 0$ ) inside each cluster and zero otherwise. Given the size of our system, these results are not affected by finite-size effects. We

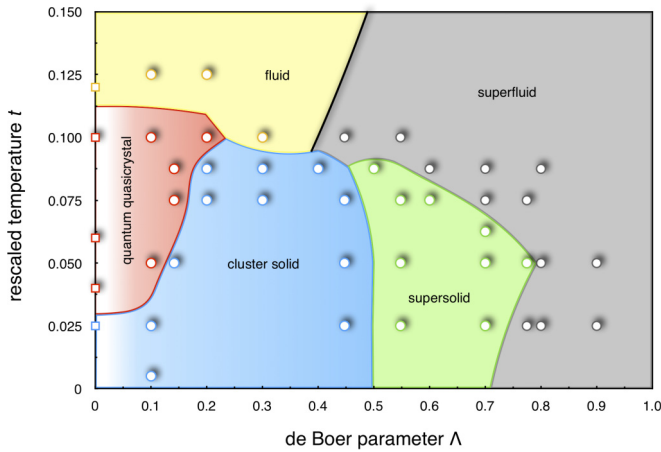


FIG. 3. Phase diagram of 2D Lifshitz-Petrich-Gaussian bosons featuring the dodecagonal quantum QC, supersolid, and superfluid as well as a hexagonal cluster solid and a normal fluid phase. Circles represent the  $N = 2048$  data points analyzed; the indicated phase boundaries are approximate. The thick black line depicts the Berezinskii-Kosterlitz-Thouless transition  $t_{\text{BKT}} \approx \pi \bar{\rho}_s r_0^2 \Lambda^2 / 2$  ( $\bar{\rho}_s$  being the superfluid density at  $t_{\text{BKT}}$ ) between the normal fluid (yellow) and the superfluid phase (gray).

note that a full finite-size scaling of  $\rho_s$  is not possible here, as increasing the already very large  $N$  makes computations exceedingly long at sufficiently low  $T$ , while decreasing  $N$  changes the QC structure in favor of a  $\sigma$ -type one. We find a comparable superfluid signal in this latter case.

At even larger  $\Lambda$ , fluctuations become even more prominent and at  $\Lambda \approx 0.54$  they lead to the transition from the cluster solid to the supersolid. In the latter, the diameter of the clusters is larger than in the cluster solid whereas the lattice spacing remains the same. This facilitates delocalization and long exchanges of particles hinted at by the many particles seen between the clusters in the  $\Lambda = 0.632$  snapshot in Fig. 2(c) and proven by the distribution of the exchange cycles in Fig. 2(k), which includes cycles with over 1500 bosons in an ensemble of  $N = 2048$  particles. The superfluid fraction of the supersolid in Fig. 2(c) is less than unity, amounting to  $\rho_s \approx 0.46$ , as expected for a spatially modulated superfluid [17,38–40], and remains almost unchanged upon cooling. Particle delocalization is further seen in  $g(r)$  in Fig. 2(g) where the maxima and minima are at the same positions as in the cluster solid but are much less prominent. At  $\Lambda \approx 0.8$ , the strong world-line delocalization turns the ensemble into a homogeneous superfluid where the superfluid fraction is 1. Figure 2(d) shows a snapshot of this phase at  $\Lambda = 1$ , and the corresponding radial distribution function in Fig. 2(h) is almost featureless.

The phase diagram of the  $N = 2048$  ensemble at a density of  $\rho r_0^2 = 0.8$  is shown in Fig. 3. The large- $\Lambda$  region is occupied by the superfluid whereas the large- $t$  region belongs to the normal fluid. The region stretching roughly to  $t = 0.1$  and  $\Lambda = 0.7$  is divided among three solid phases, with the normal dodecagonal QC present only at vanishing  $\Lambda$ s. The

quantum QC/cluster solid/supersolid/superfluid sequence is representative of  $t$ 's between about 0.03 and 0.1, whereas at  $t$ 's below 0.03 the quantum QC phase is absent. Interestingly, at low  $t$  and for  $\Lambda \lesssim 0.2$ , quantum fluctuations do not stabilize a QC but rather strengthen the occurrence of the cluster crystal, in agreement with the classical case [14].

#### IV. CONCLUSIONS

This phase diagram shows the reentrant nature of superfluidity in our system. With a proper structural support—here, the dodecagonal QC—superfluid behavior can be extended down to small values of the de Boer parameter, albeit in a fraction of particles rather than globally as in the large- $\Lambda$  superfluid phase. There may exist other non-close-packed 2D lattices that could host distributed superfluidity, say, a honeycomb or kagome lattice with vertex figures 6.6.6 and  $(3.6)^2$ , respectively. Figure 3 is also important because it provides additional insight into the mechanism of the structure formation in pair potentials with equal-depth-minima transforms, showing that the temperature range where the desired structure is stable is reasonably broad but that at large and very small  $t$ 's it is replaced by the fluid and the energy-minimizing phase, respectively. Finally, our phase diagram was computed at a fixed particle density. Given that in the classical repulsive coreless cluster-forming systems the phase sequence is qualitatively the same at all densities [9,41], we expect that at a somewhat larger or smaller density our phase diagram too is simply rescaled but otherwise unaltered.

Our quantum QC is a self-assembled phase with local and distributed superfluidity close to the classical regime, which contributes to the advances at the crossroad between quasiperiodic systems and quantum phenomena illustrated by, e.g., topological states in quasicrystals [42], the Dirac electrons in dodecagonal graphene [43], and time quasicrystals [44]. Our results emphasize that this complex behavior can result solely from pair interactions, and it would be interesting to search for it in other classes of two-length-scale soft-core potentials [13,45] as well as in experiments. The recent observation of self-assembled supersolid behavior in 1D with dipolar magnetic atoms [22,23,46], similar to that predicted in cluster-forming interactions, raises the question of whether self-assembled QC behavior may be engineered in such systems—possibly aided by structured optical potentials such as those used in cold-atom experiments where superfluidity is furnished by a Bose-Einstein condensate trapped in a laser-generated lattice [47,48], or even a condensate featuring a Rashba spin-orbit coupling combined with dipolar interactions [49].

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