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# Crystals with a twist

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Nuclear physics experiments being constructed will create narrow beams of particles repelling each other with Coulomb forces but effectively attracted to a central axis, and cooled to such temperatures that they are expected to crystallize. A numerical search for ground states finds that particles lie within narrow shells, and within each shell on paths that twist about the central axis like lines around a barber pole. The system possesses an infinite set of ordered local-energy minima, and is probably a glass.

In 1986, Rahman and Schiffer[1] carried out molecular-dynamics simulations of particle beams under the conditions of nuclear physics experiments under construction at Darmstadt, Heidelberg, and Aarhus. The particles, which might be uranium nucleii with all electrons stripped away, circulate at $0.3c$ or more through alternating quadrupole fields that to first approximation produce a force on each particle simply proportional to its distance from a central axis.[2] It is hoped the beam will be brought down to a temperature, in its rest frame, less than 20 K. At this temperature, and at experimental densities, the beam should crystallize.[3] When Schiffer and Rahman studied this process, they found that the crystals are not bcc, which is the ground state of Coulomb particles in three dimensions,[4] but that the particles arrange themselves into sharp cylindrical shells, within which they form ordered triangular patterns.[5] The purpose of this Rapid Communication is to describe this structure in more detail and to find how it varies as a function of particle density. Despite the fact that the actual particle beams will be too hot to settle fully into a ground state, the question in principle remains, what should the lowest energy state of this system be?

The energy of $P$ classical particles repelling each other with Coulomb forces but attracted to the $z$ axis is

$$E=\tfrac{1}{2}\sum_{i\neq j}^{P}\frac{(Ze)^2}{|r_i-r_j|}+\tfrac{1}{2}\sum_{i}^{P}K\rho_i^2,$$

where $\mathbf{r}_i$ is the three-dimensional location of the $i$th particle, $\rho_i$ is the distance to the $z$ axis, $Ze$ is the charge of the particle, and $K$ gives the strength of the attractive force. By rescaling distance and energy, charge and force constants may be eliminated, so that one needs to only study the energy per particle

$$\mathcal{E}_d=\frac{1}{2P}\sum_{i\neq j}^{P}\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|}+\frac{1}{2P}\sum_{i}^{P}\rho_i^2. \tag{1}$$

It may seem that this energy leads to a unique ground state, but in fact to specify the problem sensibly, one must impose a density of particles per unit length and therefore consider a one-parameter family of structures.

To a first approximation, one may treat this collection of particles as a continuous distribution. Minimizing the energy (1), one finds that in this approximation charge density is constant within a radius $\rho_c=(2n)^{1/2}$, with $n=P/L$, $L$ the system length, outside of which it drops immediately to zero. The energy of this configuration diverges with length; specifying that the particles extend a distance $L$ along the $z$ axis leads to an energy per particle for the continuous problem $\mathcal{E}_c=n\ln(L/\rho_c)+3n/4$.

The only ground states of discrete particles that can be worked out exactly are at very low density, when particles simply lie evenly spaced along the $z$ axis. A stability analysis shows that as $n$ increases above

$$(4\textstyle\sum_{i=1,3,5\ldots}^{\infty}i^{-3})^{-1/3}=0.619\ldots,$$

particles begin to rise off the $z$ axis. The initial instability leaves all particles in a plane that includes the $z$ axis, with particles alternately zig-zagging up and down by a constant amount in the plane. Another description of this state is that it is a helix with a period that is exactly twice the interparticle spacing. Numerical evaluation of (1) shows that at a density of $n\approx 0.841$, the period of the helix divided by the interparticle spacing begins slowly and continuously to increase. This is the first density for which the ground state is not periodic, although the state is invariant under simultaneous translation along the $z$ axis and rotation about it by an appropriate angle. For densities $n\leq 2.86$, all particles lie in a single shell. At slightly larger densities, the ground state consists of an outer shell with a line of particles threading through the center at $r=0$. At approximately $n\approx 3.6$ the particles on that central line rise up off the axis, twist, and produce a second shell. This process whereby successive shells appear as density increases appears to continue indefinitely.

The numerical evaluation of the sum (1) is not completely straightforward since particles interact over large distances, and restricting the sum even to several thousand particles produces edge effects that are enough to mask the true ground state. One possible solution is to allow particles to move freely within a cylindrical section of length $l$ and then to duplicate the cell periodically along the cell axis. Ewald summation allows one to compute such sums numerically. Rahman and Schiffer use the standard summation formulas for three dimensions[4] so that their cell repeats periodically not only along the cylinder axis, but in two directions perpendicular to it as well. It is preferable to work out the sums for the case in which the cell repeats only along the cell axis. However, periodic repetition of a unit cell along the cell axis is not the most general symmetry one can use. A more general

 

case is one in which unit cells do not simply repeat periodically, but twist around the $z$ axis at a fixed angle $\theta_t$ while they repeat as well. If we begin with a single point at $z=0$, $r=1$, $\theta=0$ in cylindrical coordinates, then specifying a cell length $l$ and twist angle $\theta_t$ produces the helical collection of points $r_j=1$, $\theta_j=j\theta_t$, and $z_j=jl$ for all $j$.

As the number of particles $P$ becomes infinite, the sum (1) diverges logarithmically, so the energy of the continuous system should be subtracted before allowing $P\to\infty$. The Ewald summation formula for a given number of particles per unit length $n$, when cells are repeated at distance $l$ while being twisted through angle $\theta_t$, is

$$\mathcal{E}=\mathcal{E}_d-\mathcal{E}_c=-g/(\pi)^{1/2}+\tfrac{1}{2}n\gamma+n\ln(g\rho_c)-3n/4$$
$$+\frac{1}{2nl}\sum_{i,j\in d}\left[\sum_{n=-\infty}^{\infty}\frac{(1-\delta_{n0}\delta_{ij})Q(0.5,g^2\mathcal{R}_n^2(i,j))}{\mathcal{R}_n(i,j)}+\frac{2}{l}\sum_{m=-\infty}^{\infty}T_m^a(i,j)\cos[\theta_t \bmod(2\pi)z_{ij}/l-m\theta_{ij}]\right.$$
$$\left.-l^{-1}\{E_1(g^2(r_i^2+r_j^2))+\ln[g^2(r_i^2+r_j^2)]+\gamma\}\right], \quad (2)$$

where one sums over the particles in the unit cell, describing their locations in cylindrical coordinates, $\theta_{ij}=\theta_i-\theta_j$, $z_{ij}=z_i-z_j$,

$$\mathcal{R}_n(i,j)=[r_i^2+r_j^2-2r_ir_j\cos(n\theta_t+\theta_{ij})+(z_{ij}+nl)^2]^{1/2},$$

$$T_m^a(i,j)=\int_0^g\frac{d\rho}{\rho}\exp[-\rho^2(r_i^2+r_j^2)-(m\theta_t)^2/4l^2\rho^2]\times[I_m(2\rho^2r_ir_j)-\delta_{m0}],$$

with $\gamma$ Euler's constant, $E_1$ an exponential integral, $I_m$ a Bessel function, and $Q$ an incomplete gamma function.[6] The parameter $g$ may be chosen somewhat freely to speed convergence of the sums; however, one must have $g<\min\{\pi/[(20)^{1/2}l],n\}$ to obtain accuracy of one part in $10^{10}$ or else a set of terms I have not written down must be included. By also calculating the derivatives of this expression, and evaluating them numerically to one part in $10^{10}$, I have conducted a search for ground states, using the conjugate gradient algorithm of Press *et al.*[7] A typical result appears in Fig. 1. Two striking features of these structures have already been reported by Rahman and Schiffer; that particles sit in narrow shells, and that they lie on helices.

When the ground state consists only of a single shell, it is highly symmetrical since its appearance is invariant irrespective of which particle is at the center of one's field of view. To characterize this symmetry, imagine taking a shell, slicing it down the side, and pressing it flat; that is, plotting its particles in the $z\theta$ plane. Normally, periodic structures in the plane with a trivial unit cell are described by an arbitrary pair of two-component vectors, **a** and **b**.

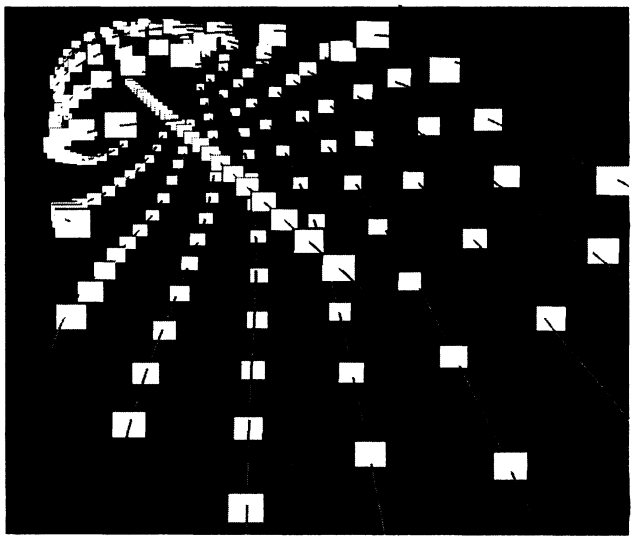

FIG. 1. A rich set of helical crystals forms when classical charged particles repel each other but are attracted to a central line. Here an example is shown with two shells at the moderately low density of $n=3$. This crystal sits at a local energy minimum $3\times10^{-3}$ in energy above the ground state. Notice that because of the perturbing influence of the central line of charge, particles in the outer shell are not all at the same radius, but describe secondary helices around the lines which have been drawn to guide the eye.

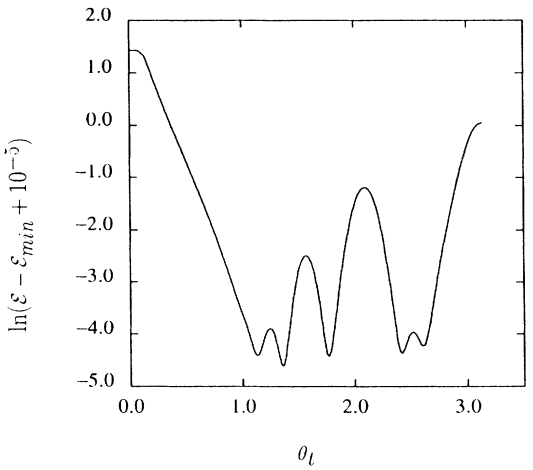


FIG. 2. If a single particle is placed in a unit cell, allowed to minimize energy as a function of distance from the central axis, but forced to twist at angle $\theta_t$, there will be several nearly degenerate minima as a function of $\theta_t$. $\mathcal{E}_{\min}$ is simply the minimum value of $\mathcal{E}$ as a function of $\theta_t$. The graph above was created at $n=3$, but the number of minima increases rapidly with density. At $n=10$, the same calculation would give 29 minima.

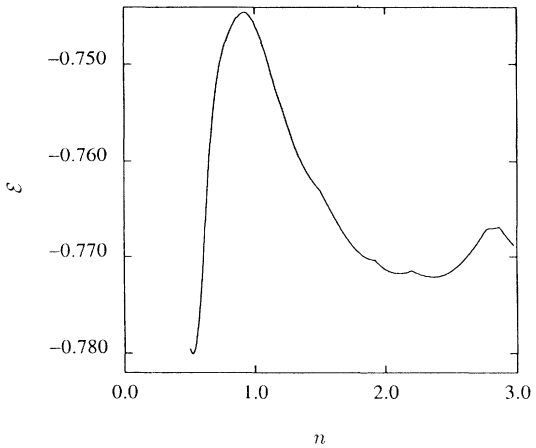


FIG. 3. This upper bound to ground-state energy was created by placing between one and eight particles in a unit cell, starting from various initial conditions, and searching for local energy minima while scanning slowly up and down in density. Each cusp corresponds to a change in helical symmetry.

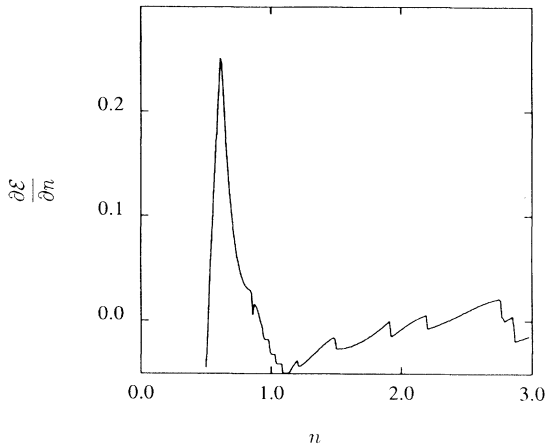


FIG. 4. The derivative of energy as a function of density is plotted up to $n=3$ so as to make the changes of symmetry more evident.

In our case, since points at $\theta=0$ and $\theta=2\pi$ must be identified, **a** and **b** must satisfy the requirement that $ia_\theta+jb_\theta=2\pi$; $ia_z+jb_z=0$ for some integers $i$ and $j$. If $i$ or $j$ is 0 then the shell is periodic along the $z$ axis. Otherwise, particles lie on helices of periods $2\pi a_z/a_\theta$ and $2\pi b_z/b_\theta$. When the ground state contains more than one shell, the helical symmetry is only approximate, as shown in Fig. 1.

It should be clear that a search for ground states of the sort being described here is not at all guaranteed to succeed. The states one can examine are all of a particular symmetry, although there is no guarantee that the true ground state should have that symmetry. In addition, a conjugate gradient algorithm always converges to local minima without looking to see if a better minimum might lie over a nearby peak. In fact, the energies of these crystals vary in a complicated way as a function of the helical twist angle $\theta_t$. Suppose that we place one particle in a unit cell and allow it to minimize the energy (2) but constrain the twist angle $\theta_t$. The result is shown as a function of $\theta_t$ in Fig. 2. These peaks and valleys form as particles at different distances come into registry as a result of the twisting.

The best upper bound to the ground-state energy I was able to find, with between one and eight particles per unit cell, is presented in Fig. 3. Each cusp in the curve represents a transition to a state of new symmetry, brought out in Fig. 4. For some ranges of densities, the ground state seems to be given by a twist angle that is a simple fraction of $\pi$. For other ranges of densities, the twist angle changes continuously with density. At the highest density I have investigated systematically, $n=3$, the ground state consists of five particles in an outer shell with a single particle sitting on the $z$ axis threading through the center. The energy behaves in a complicated way even before the crystal develops many shells. The study has not been carried systematically above $n=3$ since there is no reason to suppose that multiple shells will share a single helical symmetry.

The results obtained here indicate that for densities above 0.619 the system possesses a countably infinite set of local energy minima. Recall that shells with helical symmetry are indexed by two real numbers and two integers. One of the reals is determined by the density and the second corresponds to the twist angle $\theta_t$. As shown in Fig. 3, shells have a number of energy minima as a function of $\theta_t$, some of which differ in energy only by one part in $10^4$. Thus local minima are indexed by these values of $\theta_t$ and two integers.[8]

There is no obvious sense in which twisted crystals are frustrated. Nonetheless it is likely that they are glassy because of the many nearly degenerate energy minima; the low-energy states of the system would consist of finite segments of the cylinder possessing various helical symmetries separated from each other by grain boundaries. I have been unable to find the thermodynamic behavior of the crystals numerically,[9] but expect that it could be studied in an experimental system built of small charged spheres. In this way the menagerie of twisted crystals could be viewed with much greater ease than in the relativistic particle beam that inspired them.

I would like to thank L. Kadanoff for originally suggesting this problem, and J. Schiffer for telling me about the details. The computers on which most of the work was carried out were purchased with start-up funds from the University of Texas; Cray time for abortive Monte Carlo and Langevin equation studies was provided by the Center for High Performance Computing at the University of Texas.

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[5]This has also been noted by H. Totsuji and J. Barrat, Phys. Rev. Lett. **60**, 2484 (1988); D. Habs (unpublished).

[6]W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes* (Cambridge Univ. Press, Cambridge, 1988), p. 160.

[7]W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, Ref. 6, p. 301.

[8]In order to see if a given state is a true local minimum, one can create an exact replica of it with a doubled unit cell, doubling the number of particles one works with, to see if it can relax further. A helical state for which $\theta_t$ is not optimal will sometimes twist apart into a state with grain boundaries when given this extra freedom, but if $\theta_t$ is chosen to give a local energy minimum I have not seen this happen.

[9]I tried unsuccessfully to measure the specific heat of this system as a function of temperature for a few densities between 1 and 3, using both Monte Carlo and Langevin equation techniques. Monte Carlo was especially unsuccessful since motions such as twists of the crystal, which we have seen may be energetically favorable, are essentially forbidden if only one particle can move at a time. Use of a Langevin equation cured this problem, but even using a fully vectorized routine on a Cray, it proved impossible to obtain meaningful measurements; fluctuations were too large. Even defining a melting temperature for this system might be difficult, as when a crystal has more than one shell, different shells may well melt at different temperatures.