Autonomous topological time crystals and knotty molecular motors

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Abstract

We show that topology is a very effective tool, to construct classical Hamiltonian time crystals. For this we numerically analyze a general class of time crystalline Hamiltonians that are designed to model the dynamics of molecular closed strings. We demonstrate how the time crystalline qualities of a closed string are greatly enhanced when the string becomes knotted. The Hamiltonians that we investigate include a generalized Kratky–Porod wormlike chain model in combination with long range Coulomb and Lennard–Jones interactions. Such energy functions are commonplace in coarse grained molecular modeling. Thus we expect that physical realizations of Hamiltonian time crystals can be constructed in terms of knotted ring molecules.

Keywords: time crystals, Hamiltonian systems, knotted molecules

Supplementary material for this article is available online

(Some figures may appear in colour only in the online journal)

1. Introduction

The search for a time crystal [1–4] mainly focuses on physical systems that are out of equilibrium, and in particular subject to external periodic driving forces [5–13]. Experiments have confirmed that this kind of Floquet time crystals can be realized in non-equilibrium spin systems, including laser driven chains of trapped ytterbium ions [14] and diamond crystals that are doped with nitrogen impurities [15]. However, thus far there has been only meager interest in autonomous time crystals [16–18] in isolated equilibrium systems. In the particular case of classical autonomous Hamiltonian dynamics, a time crystal would be a time dependent periodic solution of Hamilton’s equation that is simultaneously a minimum of the energy. Initially it was thought that such time crystalline solutions do not exist [19, 20]. This follows because on a compact closed manifold the minimum of a Hamiltonian function H(q, p) is also its critical point, and Hamilton’s equation

\[ \frac{dq^a}{dt} = \frac{\partial H}{\partial p^a} \]  

\[ \frac{dp^a}{dt} = - \frac{\partial H}{\partial q^a} \]  

states that all critical points i.e. points where the derivatives of H vanish, are time independent. But there are easy ways to go around the argument: physical systems are often...
constrained, or restricted e.g. by conservation laws, and in such scenarios time-crystalline dynamics cannot be excluded by the preceding argument. As a simple heuristic example, consider the following function of two variables,

\[ H(p, q) = p^2 + q^2 \]  

(3)

On the surface \( p + q = 1 \)

(4)

the minimum of \( H \) at \( p = q = 1/2 \) does not coincide with its only critical point at \( p = q = 0 \): the condition (4) specifies a subspace, a line in \( \mathbb{R}^2 \), and the critical point of (3) is in the complement of this subspace. Recently, several examples of Hamiltonian time crystals have been constructed where an analogous situation occurs naturally in a physical context [16] and a general framework to systematically circumvent the No-Go theorems [19, 20] in an autonomous Hamiltonian context has been established [18]. See however [21–24] for related developments and remaining controversies.

Here we focus on the time crystals analyzed in [16]. They are examples of a general class of Hamiltonian free energy functions that describe the dynamics of piecewise linear closed strings. These energy functions are akin coarse grained models of molecular rings. A closed ring can have a complex entangled structure, in particular it can be knotted. We report that molecules appear to undergo autonomous swirling motion, crystalline molecular knots would undoubtedly considerably enhance activity to generators of molecular machines [25–32]. Time crystalline molecular knots would undoubtedly considerably enhance activity to generators of molecular machines [25–32].

2. Methods

The proper mathematical description [18] of a Hamiltonian time crystal builds on equivariant Morse theory [34–37] on a presymplectic manifold. But for the present purposes the following summary is sufficient: we consider Hamiltonian dynamics on a \( 2N \) dimensional phase space \( \mathcal{M} \) (a symplectic manifold). Darboux theorem states that we can always find position and momentum variables \( q^a, p^b \) (\( a = 1, \ldots, N \)) that are subject to the canonical Poisson bracket \( \{ q^a, p^b \} = \delta^{ab} \), and Hamilton’s equation (2), and since the minimum of the energy normally concurs with critical points of \( H(p, q) \) one concludes that there can not be any Hamiltonian time crystals [19, 20]. But as shown with the example (3) and (4) the situation becomes very different if there are additional conditions. This can occur whenever the Hamiltonian possesses a symmetry [16, 18]: Noether’s theorem states that a symmetry gives rise to conserved charges \( G_n(q, p) \)

\[ \{ H, G_n \} = \frac{dG_n}{dt} = 0. \]  

(5)

Their mutual Poisson brackets closes and coincides with the Lie algebra of the symmetry group,

\[ \{ G_n, G_\beta \} = f_{\alpha \beta}^\gamma G_\gamma \]  

(6)

The regular values of the conserved charges

\[ G_n(q, p) = g_n \]  

(7)

foliate the phase space \( \mathcal{M} \) into nonintersecting leaves; we label them by the values \( g_n \) and denote by \( \mathcal{M}_{g_n} \). Usually the union of these leaves covers the entire phase space \( \bigcup_{p} \mathcal{M}_{g_n} = \mathcal{M} \). But this does not always need to be the case [16, 18].

In this article we show how topological conditions can restrict and prevent the collection of available \( \{ \mathcal{M}_{g_n} \} \) from properly covering the entire physically relevant phase space. Whenever this happens, in such a way that the critical points of the Hamiltonian function \( H(q, p) \) are located in the complement of \( \bigcup_{p} \mathcal{M}_{g_n} \) in the phase space \( \mathcal{M} \), the minimum value of \( H \) on the level surfaces (7) no longer concurs with its critical point set. As a consequence a time crystal can form; examples where this takes place have already been presented in [16, 18] and here we present further explicit examples.

The Hamiltonian time crystals that we shall construct are designed to model discrete, piecewise linear closed strings that are made of \( N \) atoms. The atoms are pointlike interaction centers that define the vertices \( x_i \) (\( i = 1, \ldots, N \)) of the closed string; in the following we use the cyclic convention \( x_{N+1} = x_1 \). The links \( n_i = x_{i+1} - x_i \) between the atoms describe the covalent bonds. Since global rotation and translation symmetry is unbroken, the link variables \( n_i \) serve as the dynamical coordinates. We take their lengths \( |n_i| \) to be fixed, like covalent bond lengths usually are in a coarse grained description, at least in average over a sufficiently long time scale; for example, the fluctuation in a C–C covalent bond length is less than 1 pm, and the time scale for its oscillation is around 10 fs. For convenience we set all \( |n_i| = 1 \) in the sequel. We adopt the Lie–Poisson brackets [16]

\[ \{ n^a_i, n^b_j \} = -\epsilon^{abc} \delta_{ij} n^c_i \]  

(8)

since they preserve the bond lengths \( \{ n_i, n_k \cdot n_k \} = 0 \) for all pairs \( (i, k) \) independently of the Hamiltonian details. In fact, the brackets (8) are designed to uniformly generate any kind of local molecular motion of the discrete string, except for stretching and shrinking of the links. Note that we can reduce (8) to the standard canonical brackets of momenta and
coordinates, like those that appear in (2), simply by setting $p = n_3$ and $q = \tan^{-1}(n_2/n_1)$. But we prefer the Lie–Poisson format since the $n_i$ are most convenient variables to describe the geometry of the discrete string.

To build a time crystalline Hamiltonian $H(n)$ we start and introduce

$$H_1 + H_2 = a \sum_{i=1}^{N} n_i \cdot n_{i+1} + b \sum_{i=1}^{N} n_i \cdot (n_{i+1} \times n_{i-1}).$$  \hspace{1cm} (9)

We note that $H_1$ is akin the Kratky–Porod (worm-like-chain) energy of bending [38] that is widely used in studies of molecular chains [39]. The second term $H_2$ extends it by inclusion of twisting [16].

For more realistic molecular modeling [39] we also include long distance interactions between the atoms such as the electromagnetic Coulomb potential and the Lennard–Jones potential. The latter comprises both the attractive van der Waals and the repulsive Pauli exclusion interactions, but for clarity of presentation we only include the Pauli repulsion, in addition of Coulomb potential. Thus we can add to the Hamiltonian (9)

$$U(x_1, \ldots, x_N) = \frac{1}{2} \sum_{i,j=1}^{N} \frac{e_i e_j}{|x_i - x_j|} + \frac{1}{2} \sum_{i,j=1}^{N} \left( \frac{r_{\text{min}}}{|x_i - x_j|} \right)^{12}$$

$$\times (i \neq j)$$

(10)

where $e_i$ is the charge at the vertex $x_i$ and $r_{\text{min}}$ characterizes the extent of the Pauli exclusion that prevents string crossing; in the case of actual molecules covalent bonds do not cross each other. The potentials (10) can also be expressed in terms of the vectors $n_i$, and since our string is closed we introduce the following symmetrized description (with $i > j$)

$$x_i - x_j = \frac{1}{2}(n_j + \cdots + n_{i-1} - n_i - \cdots - n_{j-1}).$$

(11)

Thus a combination of (9) and (10) determines a Hamiltonian function $H$ that only depends on the vectors $n_i$, and the Lie–Poisson bracket (8) gives us the following Hamilton’s equation,

$$\frac{\partial n_i}{\partial t} = \{n_i, H\} = -n_i \times \frac{\partial H}{\partial n_i}. $$

(12)

For the conserved charges (5), we observe that the end-to-end distance is conserved,

$$\left\{ H, \sum_{i=1}^{N} n_i \right\} = 0$$

where $H$ is any combination of (9) and (10). As a consequence an initially closed string remains closed during the evolution (12) and the relations (6) and (7) become

$$G = \sum_{i} n_i = x_{N+1} - x_1 \equiv 0$$

(13)

$$\left\{ G^a, G^b \right\} = -\epsilon^{abc} G^c.$$ \hspace{1cm} (14)

Accordingly we have the general set-up of time crystals [18] and we can proceed to search for a time crystalline solution of Hamilton’s equation (12).

The Lagrange multiplier theorem [40] ensures that when the conserved quantity $G$ has the value (13) that specifies a closed string, the minimum value of the Hamiltonian $H(n)$ is a critical point $(n_{cr}, \lambda_{cr})$ of the following extended Hamiltonian function $H_{\lambda}$:

$$H_{\lambda} = H + \lambda \cdot G$$

(15)

$$\frac{\partial H}{\partial n_{p,cr}} = -\lambda_{cr} \quad \& \quad G(n_{cr}) = 0.$$ \hspace{1cm} (16)

Whenever $\lambda_{cr} \neq 0$ we then have a time crystal i.e. a time dependent minimum energy solution of (12) with initial condition $n_i(0) = n_i(0)$. For this critical point configuration of (15) we use (16) to write the equation (12) for the time crystal as follows:

$$\frac{\partial n_i}{\partial t} = -\lambda_{cr} \times n_i \quad \text{with} \quad n_i(t = 0) = n_{cr}.$$ \hspace{1cm} (17)

The critical value $\lambda_{cr}$ is clearly time independent so that the equation (17) describes uniform rotation of a time crystalline closed string, with angular velocity determined by $\lambda_{cr}$.\hspace{1cm}

To explicitly construct a time crystal we need to find the critical values $n_{cr}$ and $\lambda_{cr}$. The structure of (12) makes this straightforward, as we only need to find the large $-t$ limit of

$$\frac{\partial n_i}{\partial t} = -n_i \times \frac{\partial H}{\partial n_i} + \mu n_i \times \left( n_i \times \frac{\partial H}{\partial n_i} \right)$$

(18)

where $\mu > 0$. This follows since (18) gives us

$$\frac{\partial H_{\lambda}}{\partial t} = -\mu \left[ 1 + \frac{\mu}{\lambda_{cr}} \sum_{i=1}^{N} \left| \frac{\partial n_i}{\partial t} \right| \right]^2$$

(19)

so that the time evolution (18) proceeds toward decreasing values of $H_{\lambda}$ and the flow continues until the critical values $(n_{cr}, \lambda_{cr})$ are reached, in the $t \rightarrow \infty$ limit. Whenever more than one solution is found, for a time crystal we need to choose the one with smallest value of $H$.

We have employed (18) to investigate how the knotness of the string affects the existence of a time crystalline energy minimum. For this we have compared the minimum energy configurations that we obtain by numerical simulation of (18), in the case of an unknotted string and a knotted string that supports a trefoil knot. In our simulations we start from some arbitrarily chosen initial configuration, with the given knot topology, and follow its evolution toward energy minimum along the trajectory (18). In order to confirm that we indeed have located the absolute minimum of the energy, and not just a local one, we repeat the simulation by varying the initial configuration.

Unfortunately, gradient descent with unlimited long range interactions can be somewhat slowly convergent, and the computer time needed to locate the energy minimum with a very high precision that is needed here, grows rapidly with the number of vertices $N$. Since we also need to verify that our final configuration is a global minimum, not just a local one, the computer time becomes substantial. For that reason, we only describe the results for $N = 12$ vertices; for less than $N = 12$ it becomes hard to form a trefoil, and we have confirmed that our results are not affected when the value of $N$ starts increasing.
3. Results

We start by describing our results, in the case of unknotted strings. In this case we confirm that generically, there are no time crystalline solutions:

We first consider the evolution (18) of a random unknotted initial configuration, in the case of the Hamiltonian $H_1$ with $a > 0$. The flow (18) terminates when all $\mathbf{n}_i \cdot \mathbf{n}_{i+1} = -1$ which is also the absolute minimum energy configuration in this case. The critical value $\lambda_{cr}$ that we evaluate from (15) and (16) vanishes, and when we substitute the minimum energy configuration in the pertinent version of (12) we confirm that there is no motion. Thus there is no unknotted time crystal for $H_1$ with $a > 0$.

When $a < 0$ the unknotted energy minimum of the Hamiltonian $H_1$ is a time independent regular planar dodecagon; $\lambda_{cr} = 0$ and there is no unknotted time crystal in this case either.

The regular dodecagon is also the unknotted, time independent minimum energy configuration of the Coulomb potential in (10) when the $e_i$ are positive.

In the case of $H_2$ we already know from [16] that for $N = 3$ this Hamiltonian gives rise to a time crystal in the shape of an equilateral triangle; for $N = 4$ it supports a time crystal that relates to the tetragonal disphenoid. In the present case, with $N = 12$, we find that the flow (18) terminates in the jagged unknot shown in figure 1(a). In this minimum energy configuration the vectors $\mathbf{n}_i$ and $\partial H / \partial \mathbf{n}_i$ acquire their maximally antiparallel orientation that is available for $N = 12$ vertices; we compute

$$\arccos \left( \frac{\mathbf{n}_i \cdot \partial H / \partial \mathbf{n}_i}{\sqrt{\partial H / \partial \mathbf{n}_i \cdot \partial H / \partial \mathbf{n}_i}} \right) \approx 3.119 \text{ (rad)}. \quad (20)$$

The structure is the only time crystalline unknot we have been able to construct, in the present study. The time crystal rotates around its four-fold symmetry axis, but with the very small angular velocity $\omega \approx 0.0158$. More generally, when the (even) number of vertices $N$ increases the angles (20) approach the value $\pi$, where the time crystal dissolves and the ensuing unknot does not move.

Next, we proceed to knotted strings, with the topology of a trefoil knot. In this case the generic result is a time crystal, and we now describe three representative time crystal solutions:

We first construct a representative initial ansatz trefoil for the flow equation (18); we have confirmed that our results are independent of the initial structure we use, by deforming it. For the ansatz, we start with the continuum trefoil (with $s \in (0, 2\pi)$)

$$x_1(s) = L \cdot [\cos(s) - A \cos(2s)] \quad (21)$$

$$x_2(s) = L \cdot [\sin(s) + A \sin(2s)] \quad (22)$$

$$x_3(s) = \pm L \cdot \sqrt{1 + A^2} \sin(3s). \quad (23)$$

Here $L$ and $A$ are parameters and $\pm$ determines whether the trefoil is left-handed (+) or right-handed (−). This trefoil has a high level of three-fold symmetry, for example each of the three coordinates has the equal radius of gyration value $R_g = L\sqrt{1 + A^2}$.

To discretize (21)–(23) for $N = 12$, we first divide it into three segments, all with an equal parameter length $\Delta s = 2\pi/3$. We then divide each of these three segments into four subsegments, all with an equal length in space for $N = 12$ vertices. We set $A = 2$ and when we choose $L = 0.340$ each segment has a unit length, and the three space coordinates $(x_1, x_2, x_3)$ have the radius of gyration

$$R_g^{(i)} = \sqrt{\frac{1}{N} \sum_{n=1}^{N} (x_i(n) - \bar{x}_i)^2} \quad (24)$$

values $(0.722, 0.722, 0.715)$; here $\bar{x}_i$ is the average of the $x_i(n)$. This constitutes a representative initial trefoil ansatz that we use in the flow equation (18).

As a first example, we consider a Hamiltonian that is a linear combination of the bending rigidity $H_1$ in (9) and the Pauli exclusion in (10), with the parameter values $a = 1/4$ and $r_{min} = 3/4$. The flow (18) terminates to a minimal energy trefoil configuration $\mathbf{n}_{cr}$ with radius of gyration values $0.710, 0.710, 0.874$; note that the initial ansatz trefoil is slightly oblute in the $x_3$ direction, but the minimal energy trefoil $\mathbf{n}_{cr}$ is slightly prolate. We use the minimal energy configuration $\mathbf{n}_{cr}$ as an initial condition in (12) and confirm that we have a time crystal that rotates around the $x_3$ axis with angular velocity $\omega \approx 0.619$ in our units; the direction of rotation depends on the sign of $x_3$ in (23). Note that the value of $\omega$ is much higher than in the case of the unknotted time crystal that we reported.

As a second example we consider a sum of the twist-bend coupling $H_2$ in (9) and the Pauli exclusion in (10), with parameter values $b = 1/4$ and $r_{min} = 3/4$. Now the flow (18) terminates at a minimal energy prolote trefoil $\mathbf{n}_{cr}$, with radius of gyration values $(0.715, 0.715, 0.875)$. When we use the $\mathbf{n}_{cr}$ as
initial condition in (12) we find that this trefoil is a time crystal that rotates around the $x_3$ axis, with a clearly higher angular velocity $\omega \approx 1.046$.

In the third example the Hamiltonian is a sum of the Coulomb interaction and the Pauli exclusion (10), with parameters $e_1 = 1$ and $r_{\text{min}} = 3/4$. The flow (18) terminates at a prolate trefoil $n_{\text{cr}}$ with radius of gyration values $(0.717, 0.717, 0.889)$. We again have a time crystal solution of (12), now with an even higher angular velocity $\omega \approx 1.571$.

Notably, all the three time crystalline trefoils have a very similar slightly prolate shape: all the mutual root-mean-square distances between the trefoils are less that 0.01. In figure 1(b) we illustrate the third time crystalline trefoil, as an example. We also depict the way it rotates, and in the supplementary material (https://stacks.iop.org/JPCM/33/015702/mmedia) we have a movie that shows this; there is also a movie that shows how this time crystal is reached, by the flow equation (19).

4. Discussion

Even though we have only simulated molecular rings that have the topology of an unknot or a trefoil knot, the relation between knottness and the time crystal state that we have revealed is very general. This is because the critical point sets $\{x_{\text{cr}}\}$ of our Hamiltonian functions always pertain to a definite string conformation, with a definite knotted topology. But the topology of a generic knot is in general different from that of the critical point of the Hamiltonian. At the level of equations, whenever the Lagrange multiplier $\lambda_0$ is non-vanishing, there is mismatch between the topology of a knot and that of the critical string, and this mismatch causes a frustration that drives the time crystal.

For example, we have noted that for $a > 0$ the Hamiltonian $H_1$ acquires an absolute minimum at the critical point with all $n_r \cdot n_{r+1} = -1$. For this, the stationary solution of (12) has the topology of an unknot so that there is no unknotted time crystal. But whenever the structure forms a knot, there is always a conflict between the topology of the knot and the geometry of the fixed point set, and provided string self-crossing is suppressed we expect to have a time crystal. Similarly, when $a < 0$ the energy minimum of the Hamiltonian $H_1$ defines an unknotted regular planar dodecagon. There is no unknotted time crystal in this case, while any knotted structure is in a time crystalline state. The regular dodecagon is also the unknotted knotted time crystal in this case, while any knotted structure is in a time crystalline state. The regular dodecagon is also the unknotted minimum energy configuration of the Coulomb potential in (10) when the $e_i$ are positive, and again we have found that there is no unknotted time crystal solution of (12). Thus, in all our examples the existence and stability of our knotted time crystal solution is entirely due to a conflict between the topology of the string that is described by the critical point of the Hamiltonian, and the topology of the time crystalline knotted string.

5. Summary

Our simulation results establish that topology can be an important factor, that can cause a physical system, either autonomous as here or driven as in [41], to become time crystalline. The examples that we have constructed are coarse grained molecular rings, with the topology of a trefoil knot. But we have presented general arguments why we are confident that our results extend to more general knotted topologies. Moreover, the Hamiltonians that we have studied are very universal, they are employed widely in coarse grained, effective theory descriptions of (bio)polymers and molecular chains. Thus we expect that actual physical realizations of topologically driven time crystals can be found in terms of knotted molecules. To identify promising candidates, in the case molecular structures, more realistic all-atom simulations should be performed. However, the computational challenges are enormous, as the presence of long distance interactions with no distance cut-off makes the search of a minimum energy configuration NP hard. Once actual material examples have been identified and experimentally constructed, we expect that topologically stable molecular time crystals will find many applications [6].

The data that support our findings are available from the corresponding author upon request.

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