Tunable chirality sorting by external noise in achiral periodic potential

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Due to the excellent properties of assembled mesoscopic chiral structures beyond their achiral counterparts or those at the single-particle level, sorting methods for such chiral structures are very demanded. Here, we propose a mechanism utilizing the constructive role of external noise to attain a tunable sorting of chiral particles in an achiral periodic potential. By adjusting the intensity of the external noise, an optimal chirality sorting with complete chirality-separation and a rollover of chirality selectivity are observed. Detailed analysis reveals that there are a direct transition and a multi-step transition between moving paths of chiral particles, and the dominant transition depends essentially on the level of the external noise. Such a mechanism provides a practicable way to control the moving direction of the chiral particles, by which several kinds of enantiomorphs with different chirality degree can also be completely separated at the same time by solely adjusting the external noise. The broad application of noise-tuned chirality sorting for other types of periodic potentials or correlated noise is also demonstrated by intensive simulations. Since the external noise is independent on the internal properties of the system and can be conveniently controlled, our method provides a promising routine for tunable chirality sorting.

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Chirality is a property of mirror asymmetry essential in several branches of science, especially for biomolecules to be of biological activity. Very recently, explosive attention has been paid to mesoscopic chiral structures assembled from micro- or meso-blocks. Such assembled chiral structures are shown to be of very special optical, electric and magnetic responses rather than their achiral counterparts or that at the single-particle level. Since the formed products may be a mixture of enantiomorphs, techniques that can efficiently sort such assembled chiral structures are then received great research interest.

So far, several types of chirality-sorting methods have been proposed to this purpose. For the first type, a system of intrinsic chirality along the separation direction can be utilized to sort chiral structures within it, including asymmetric shear flows, helical flow fields, optical fields, some materials or structures. For the second type, periodic potentials without intrinsic chirality provide another possible ways for chirality sorting. The pioneer work of this type is done by de Gennes, where a macroscopic chiral crystal is found to glide in a direction differing slightly from the axis of maximum slope when it is slipping over an inclined solid support. For separation of smaller chiral objects such as macromolecules or assembled mesoscopic structures where thermal fluctuation is nonnegligible, de Gennes argued that the fluctuation would destroy this effect. Similarly in periodic potentials, other chirality sorting methods against thermal noise have also been proposed. Besides of the internal thermal noise, external noises will also been brought in by the applied periodic potentials. Notice that, while the thermal noise usually hinders the efficiency of chirality separation in achiral potentials in the aforementioned methods, it has been reported that external noises may play a constructive role in many nonlinear dynamical systems. It is then very interesting to explore whether the external noise can be utilized to provide enhancement or even new routines for chirality sorting.

In this Letter, we report a tunable chirality sorting induced by the external noise in an achiral periodic potential. Special attention is paid to the effect of the external noise on the moving of the chiral particles, where an optimal chirality sorting with complete chirality-separation and a rollover of chirality selectivity are observed. Interestingly, noise-dependent transitions between moving paths of the chiral particles are revealed, via which the moving direction of the chiral particles and consequently the chirality sorting can be well tuned by solely adjusting the external noise. The way to completely separate several kinds of enantiomorphs with different chirality degree at the same time is further realized based on the same mechanism.

We consider a mesoscopic 2-dimensional chiral structure as an equilateral trilateral particle assembled by three rigidly coupled colloids located at \( r_i, i = 1, 2, 3 \) (Fig.1). The chirality of the particle is realized by setting the three colloids to be of different sizes, which consequently results in different friction coefficients \( \gamma_i \) for each colloid. Taking \( \gamma_1 < \gamma_2 < \gamma_3 \), we consider a particle is of ( + ) chirality if the 1st, 2nd and 3rd colloids are arranged anticlockwise as shown in the left of Fig.1 and is of ( - ) chirality for clockwise arrangement as in the right. The state of such a rigid particle can then be described by the position of its friction center \( \mathbf{R} = \sum_{i=1}^{3} \gamma_i \mathbf{r}_i / \sum_{i=1}^{3} \gamma_i \), and by the orientation angle \( \phi \) between the vector \( \mathbf{r}_1 - \mathbf{R} \) and the X axis. The position of each colloid can be determined by

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\( \mathbf{r}_i = \mathbf{R} + \mathbf{q}_i(\phi) = \mathbf{R} + \mathbf{O}(\phi)\mathbf{q}_i^{(0)} \). Here, \( \mathbf{q}_i = \mathbf{r}_i - \mathbf{R} \) is the vector pointing from \( \mathbf{R} \) to the \( i \)th colloid, \( \mathbf{q}_i^{(0)} \) denotes its value in a reference configuration with \( \phi = 0 \), and \( \mathbf{O}(\phi) \) is a rotation matrix whose elements are \( O_{11} = O_{22} = \cos(\phi) \) and \( O_{21} = -O_{12} = \sin(\phi) \). The particle obeys the coupled Langevin equations as follows,

\[
\frac{d\mathbf{r}(t)}{dt} = \sum_{i=1}^{3} \mathbf{F}(\mathbf{r}_i) + \sum_{i=1}^{3} \gamma_i \mathbf{v}^f(\mathbf{r}_i) + \xi(t) + \zeta(t). \tag{1}
\]

\[
\frac{d\phi(t)}{dt} = \mathbf{e}_z \cdot \sum_{i=1}^{3} \mathbf{q}_i(\phi) \times [\mathbf{F}(\mathbf{r}_i) + \gamma_i \mathbf{v}^f(\mathbf{r}_i)] + \zeta(t). \tag{2}
\]

In the presence of a static external periodic potential, the particle is affected by a force \( \mathbf{F}(\mathbf{r}_i) \) on each colloid. Here, the potential is taken as a superposition of three standing waves \( U(\mathbf{r})/\gamma_i = C \sum_{j=1}^{3} c_j \cos(K_k \mathbf{j}_k \cdot \mathbf{r} + \delta_j) \) [23, 24], where \( \mathbf{r} \) is a given position, \( C \) shows the potential strength, \( K \) denotes the inverse spatial scale, \( \mathbf{j}_k \) is a unit vector, \( c_i \) is the ratio of each standing wave (the sum of \( c_i^2 \) must be 1), and \( \delta_i \) denotes the phase offset. Such a potential may be realized by an external electric or optical field [24]. We fix \( c_1 = 0.256, c_2 = c_3 = 0.683, \mathbf{k}_1 = (1, 0), \mathbf{k}_2 = (-0.5, \sqrt{3}/2), \mathbf{k}_3 = (0.5, \sqrt{3}/2) \), \( \delta_1 = 0, \delta_2 = 2.49 \), and \( \delta_3 = 3.79 \), so that the two enantiomorphs cannot be separated. For \( \gamma_1 < \gamma_2 < \gamma_3 \) on an equilateral triangle’s vertexes \( \mathbf{r}_i \), \( \mathbf{R} \) is the center of friction and \( \phi \) is the angle between the vector \( \mathbf{r}_i - \mathbf{R} \) and the \( X \) axis. The (–) particle differs only in the sequence of \( \gamma_i \).

![FIG. 1: Schematic of the chiral particles driven by a fluid field \( \mathbf{v}^f \) parallel to the \( X \) axis (gray arrows) through an achiral periodic potential \( U(\mathbf{r}) \) (colored background). The (+) particle consists of three colloids arranged anticlockwise with \( \gamma_1 < \gamma_2 < \gamma_3 \).](image)

The potential is \( \mathbf{F}(\mathbf{r}_i) = -\gamma_i \mathbf{K} \sum_{j=1}^{3} c_j \mathbf{k}_j \sin(K_k \mathbf{j}_k \cdot \mathbf{r}_i + \delta_j) \).

In Eq. (1), \( \mathbf{v}^f(\mathbf{r}_i) \) denotes a velocity field of a fluid which provides a driven force for particle movement. In order to be consistent with the symmetric form of the potential field, the particle is driven by a flowing fluid moving in the positive direction of the \( X \) axis at a speed of \( v_0 \), i.e., \( \mathbf{v}^f(\mathbf{r}) = v_0 \mathbf{e}_x \) where \( \mathbf{e}_x = (1, 0) \) is the unit vector along the \( X \) axis. Moreover, the random forces caused by the thermal fluctuation and external noise should be taken into account, which are realized by adding the last two terms \( \xi(t) \) and \( \zeta(t) \) to the right side of Eq. (1). The first one is an independent Gaussian white noise satisfying the fluctuation-dissipation relation \( \langle \xi_\mu(t) \xi_\nu(t') \rangle = 2k_B T \delta(t - t')\delta_{\mu\nu}/\sum_i \gamma_i \), where the subscript \( \mu(\nu) \) denotes the component along the \( X (Y) \) axis, \( k_B \) is the Boltzmann constant, \( T \) denotes the temperature. The second one is an external noise with \( \langle \zeta_\mu(t) \zeta_\nu(t') \rangle = 2D_e \delta(t - t')\delta_{\mu\nu} \), whose intensity \( D_e \) can be tuned, externally.

Besides of the translation movement, the periodic potential and the fluid field can also lead to rotation of the chiral particle. The dynamical equation of the angle \( \phi \) is described by Eq. (2), where the scalar \( |\mathbf{q}_i| \) is the mode of \( \mathbf{q}_i \). The two terms in braces in the right side of Eq. (2) represent torques exerted by \( \mathbf{F} \) and \( \gamma_i \mathbf{v}^f \), respectively, and the last term \( \xi_\phi(t) \) is the rotational fluctuation satisfying \( \langle \xi_\phi(t) \xi_\phi(t') \rangle = 2k_B T \delta(t - t')/\sum_i \gamma_i |\mathbf{q}_i|^2 \).

In simulations, parameters are rescaled by the side length of the equilateral trilateral particle, the friction coefficient of the smallest colloid in the particle and the speed of the fluid field so that \( d = 1, \gamma_1 = 1 \) and \( v_0 = 1 \). We fix the inverse spatial scale \( K = 0.3 \), the potential strength \( C = 6.67 \) and \( k_B T = 10^{-5} \). For consistence, all of the following results are obtained from particles running for a long time \( t = 10^6 \) with \( 2 \times 10^4 \) randomly choosing initial states near the origin.

Firstly, we investigate how the external noise influence chirality sorting of particles with \( \gamma_2 = 1.5 \) and \( \gamma_3 = 2.0 \). Typical trajectories for \( D_e = 0, 1.0 \times 10^{-3} \) and \( 10.0 \) are shown in Fig. 2(a), (b) and (c), respectively. For \( D_e = 0 \) (Fig. 2(a)), both of the two enantiomorphs can move along two different paths, one of which departs from the \( X \) axis with a positive projected direction along the \( Y \) axis (named as the positive path, movie S1), and the other with a negative one (the negative path, movie S2). Nevertheless, the trajectories for both of the enantiomorphs along each path are entangled with each other, so that the two enantiomorphs cannot be separated. For \( D_e = 1.0 \times 10^{-3} \) (Fig. 2(b)), it is quite interesting that all of the (+) particles move along the positive path, while all of the (–) particles along the negative path, resulting in two well separated clusters of particles. For \( D_e = 10.0 \), (+) and (–) particles are mixed once again as shown in Fig. 2(c). Unlike the case for \( D_e = 0 \) where two moving paths are distinct, these paths converge to be the one along the \( X \) axis.

In order to quantitatively describe the relationship between the chirality sorting and the external noise, an order parameter \( S \) is defined to measure the chirality selectivity as

\[
S = \int_0^\infty P_{(+)}(Y) dY - \int_0^\infty P_{(–)}(Y) dY, \tag{3}
\]

where \( P_{(+)}(Y) \) denotes the distribution of the final position for the (+) particle projected on the \( Y \) axis. For \( S > 0(S < 0) \), (+) particles distribute more(less) upon...
FIG. 2: External noise-induced chirality sorting and rollover of chirality selectivity. Typical trajectories of the (±) particles for (a) $D_e = 0$, (b) $1.0 \times 10^{-3}$ and (c) $10.0$. (d) Chirality selectivity $S$ as a function of the external noise intensity $D_e$ for $\gamma_2 = 1.5$, $\gamma_3 = 2.0$ and $\gamma_2 = 2.0$, $\gamma_3 = 3.0$.

- $X$ axis than $(-)$ particles. $S = 0$ means that there is no chirality sorting can be observed, while $S = \pm 1$ indicates that the two enantiomorphs can be separated, completely. The obtained $S$ as a function of $D_e$ is presented in Fig. 2(d) by the black line. For small $D_e$, e.g., $D_e < 5.0 \times 10^{-6}$, $S$ is nearly 0. As $D_e$ increases to be of moderate values, $S$ increases rapidly to 1 and maintains $S = 1$ for a wide range of $D_e$. For large enough $D_e$, e.g., $D_e > 1.0$, $S$ will drop back to nearly 0 again. These observations demonstrate clearly that, chirality selectivity depends non-monotonically on the intensity of the external noise, and the external noise can induce an optimal chirality sorting with 100% selectivity in achiral potential field.

Then, we are interested in how the external noise influence chirality sorting of particles with other arrangement of values for $\gamma_i$s. The arrangement can be described by another order parameter $\Delta \gamma \equiv \gamma_3 - \gamma_2 = \gamma_2 - \gamma_1$ by setting the difference between two consequent $\gamma_i$s to be the same. Such an order parameter can also be considered as a measurement of the chirality degree for chiral particles. For $\Delta \gamma = 1.0$, i.e., $\gamma_2 = 2.0$ and $\gamma_3 = 3.0$, dependence of chirality selectivity $S$ on the external noise intensity $D_e$ is plotted in Fig. 2(d) by the red line. Similar to the one for $\Delta \gamma = 0.5$, $S$ increases to 0 for $D_e < 5.0 \times 10^{-6}$. Nevertheless, $S$ decreases quickly to $S = -1$ as $D_e$ increases to be slightly larger than $5.0 \times 10^{-6}$, indicating a chirality sorting with all of the (+) particles only distributed below the $X$ axis, in contrast to the situation for $\Delta \gamma = 0.5$. Remarkably, by further increasing $D_e$, a rollover of chirality sorting is observed, i.e., $S$ increases rapidly from $-1$ to $1$, for $D_e > 1.0 \times 10^{-4}$. In both of the parameter regions where $|S| = 1$, the chiral particles are separated completely while moving path of the $(+)(-)$ particles changes from negative(positive) one for $S = -1$ to positive(negative) one for $S = 1$. As $D_e$ increases to be large enough, $S$ drops back to 0 again. In short, besides of the optimal chirality sorting, a rollover of the chirality sorting can also be induced by the external noise.

In order to reveal the underlying mechanism for the interesting observations aforementioned, we now try to figure out the transition process between moving paths for chiral particles with different parameters. As depicted in Fig. 3(a) and (b), the (+) particle always oscillates its $\phi$ below 0 periodically along the positive path (the blue lines), and upon 0 along the negative one (the red lines). The $(-)$ particle oscillates similarly as the $(+)$ one except that the sign of $\phi$ is opposite. Interestingly, two types of noise-induced reversible transitions between these two paths can be observed for $D_e > 0$ (Fig. 3(a) and (b)). For the first type transition indicated by the black lines (named as the direct transition), the particle jumps from one moving path to the other directly (movie S3 and S4). For the second type indicated by the green lines (the multi-step transition), the particle from one moving path jumps firstly to a state with opposite sign of $\phi$ while the moving direction keeps the same as the one before jumping, then changes its moving direction oppositely to move along the other moving path (movie S5 and S6). Furthermore, we find that the $(+)(-)$ particle prefers to move along the negative(positive) path via the direct transition, while tends to move along the positive(negative) path via the multi-step transition. Then, these two transitions will determine how long the particle stays in the two different paths, and then where the particle finally reaches.

The probability $\rho$ of how long the $(+)$ particle stays in the positive and negative paths is plotted in Fig. 3(c) for $\Delta \gamma = 0.5$. For small $D_e$ such as $D_e = 0$ (green bars), transitions can hardly be observed and the particle can move along both of the paths with equal probabilities. For $D_e = 1.0 \times 10^{-3}$ (red bars), both of the transitions can be induced by the external noise, while the multi-step transition occurs more frequently than the direct one. As a result, the $(+)$ particle distributes much longer on the positive path than the negative one, leading to a chirality selectivity $S = 1$ as shown in Fig. 2(d). For $D_e = 10.0$, the large enough external noise even
merges the two moving paths to be the same one along the \( X \) axis, and the particle can be viewed as distributing equally along the two paths again. In other words, the noise-induced multi-step transition of the particle moving paths is the very reason of the observed optimal chirality sorting for \( \Delta \gamma = 0.5 \).

The rollover of the chirality sorting can also be understood based on the same picture. For \( \Delta \gamma = 1.0 \), there are also two similar moving paths and two path transitions as those for \( \Delta \gamma = 0.5 \). The probability \( P \) for the \((+)\) particle with \( D_e = 0, 2.0 \times 10^{-5}, 7.0 \times 10^{-5}, 1.0 \times 10^{-3} \) and \( 10.0 \) is presented in Fig.4(d). Similar to \( \Delta \gamma = 0.5 \), the \((+)\) particle stays equally along the positive and negative paths for small \( D_e = 0 \) (green bars) or large \( D_e = 10.0 \) (magenta bars) external noise and prefers one of these two paths for moderate noise, which results in optimal chirality sorting. For \( D_e = 2.0 \times 10^{-5} \) (red bars), it is observed that only the direct transition occurs. Thus, almost all of the \((+)\) particle stays in the negative path, leading to a type of chirality sorting with \( S = -1 \). As \( D_e \) increases to be \( D_e = 1.0 \times 10^{-3} \), the multi-step transition emerges and dominates the path transition process. Thus, the \((+)\) particle turns to prefer the positive path, resulting in another type of chirality sorting with \( S = 1 \). In between, there is a mixed state for \( D_e = 7.0 \times 10^{-5} \) as depicted by the nearly equal-height cyan bars, where chiral particles cannot be separated. It is then the noise-dependent changing of dominant transition leads to the rollover of the chirality sorting.

To explore fully how parameters affect chirality sorting, a separation-phase diagram in the \( D_e - \Delta \gamma \) plane is obtained by extensive simulations (Fig.4). Except for the optimal chirality sorting and its rollover, a new type of rollover occurs for \( \Delta \gamma \) larger than \( 2.5 \). Taking \( \Delta \gamma = 6.0 \) as an example, with increasing \( D_e \), a rollover from \( S = 1 \) to \( S = -1 \) occurs, in contrast to the one for \( \Delta \gamma = 1.0 \) mentioned above. Such a new type of rollover behavior can be observed for very large \( \Delta \gamma \) (even for \( \Delta \gamma > 100 \)). Notice that, in the region around \( \Delta \gamma = 2.5 \), \( S \) seems to be 0 for all \( D_e \). In fact, rollover of \( S \) can also be observed in such a region (Fig.S1 in the supplemental information) except that the optimal value of \( |S| \) is smaller. Despite of such quantitative difference, the involved mechanisms in this region is qualitatively the same as that in other regions.

With the separation-phase diagram, it is possible for us to tune the chirality sorting conveniently by changing the external noise. Firstly, the moving directions of the chiral particles can be tuned by \( D_e \) solely. The noise-controlled moving directions of two enantiomorphs can be described by relative angle \( \Delta \theta \) between the vectors from the origin to the averaged position of the \((\pm)\) particle. \( \Delta \theta \) as a function of \( D_e \) for \( \Delta \gamma = 1.0 \) is presented in Fig.5(a) as an example. With increasing \( D_e \), \( \Delta \theta \) first drops from 0 to \(-0.83\), then increases to 0.22, and final gets back to 0. Thus, by adjusting the value of the external noise \( D_e \), we can not only get complete chirality sorting, but also control the moving directions of the two enantiomorphs. Secondly, several kinds of enantiomorphs with different \( \Delta \gamma \) can also be completely separated at the same time for an appropriate external noise. For example, the distribution \( P(Y) \) of the projected position on the \( Y \) axis for four different chiral particles (two kinds of enantiomorphs with \( \Delta \gamma = 0.6 \) or 1.2) is shown in Fig.5(b) for \( D_e = 1.0 \times 10^{-4} \). Clearly, all of these four kinds of particles move along different directions and fall in different areas on the \( Y \) axis without any overlap, indicating that all these four kinds of particles can be separated perfectly.

To broaden the application of the noise-tuned chirality sorting, chirality sorting in different types of periodic potentials are also investigated by intensive simulations. It is found that, the complete chirality sorting can still be observed if the potential is periodic both along the \( X \) axis (the flowing direction) and along the \( Y \) axis (vertical to the flowing direction) while other details of the potential such as the shape seems to be not relevant. Noise-tuned chirality sorting for two other types of periodic potentials can be found in Fig.S2 in the supplemental information. To further validate the concept of noise-tuned chirality sorting in systems of colored noise, similar simulations
are performed by replacing the external white noise in our model by external time-correlated or space-correlated noise (details can be found in the supplement information). Quite interestingly, an optimal chirality for small $\Delta \gamma$ (0.5) and a rollover of chirality sorting for larger $\Delta \gamma$ (1.0) can be observed, too (Fig.S3 in the SI). Since the finite time(space) correlation length of the external noise ($\gamma(0.5)$ and $\gamma(1.0)$) can be observed, too (Fig.S3 in the SI). Since the finite time(space) correlation length of the external noise offers a rich parameter space for chirality sorting, it may deserve a systematic investigate in future works.

In summary, we found that the external noise can induce transition between moving paths of the chiral particles in a periodic potential, via which an optimal chirality sorting with 100% selectivity and a rollover of chirality selectivity are resulted. The major advantage of this method compared to many other separation concepts is the tunable chirality sorting as well as the controllable moving direction of chiral particles by utilizing the constructive influence of the external noise, e.g., for enantiomorphs with quite different degree of chirality, we can get complete sorting simultaneously just by simply adapting the external noise in the same periodic potential. The broad application of noise-tuned chirality sorting for other types of periodic potentials or correlated noise is also demonstrated by intensive simulations. Since the external noise is independent on the internal properties of the systems and can be conveniently controlled, our method provides a promising routine for tunable chirality sorting, and may open new perspectives on both theoretical and experimental investigations of chirality sorting for assembled chiral structures in future.

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