Effective substrate potentials with quasicrystalline symmetry depend on the size of the adsorbed particles

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Abstract. We explore the effective potential landscapes that extended particles experience when adsorbed on the surface of quasicrystals. Commonly, these are solids with long-ranged order but no translational symmetry. The effective potentials significantly depend on the size of the adsorbed particles. We show how changing the particle radius changes the so-called local isomorphism class of the effective quasicrystalline pattern. This means effective potentials for different particle sizes cannot directly be mapped onto each other. Our theoretical predictions are confirmed by Monte Carlo simulations. The results are important for colloidal particles with different sizes that are subjected to laser fields with quasicrystalline symmetry as well as for systems where extended molecules are deposited onto the surface of metallic quasicrystals.

1 Introduction

Quasicrystals are solids that are not periodic but nevertheless possess long-ranged order [1]. Deposition of atoms or molecules on surfaces of quasicrystals can lead to monolayers with a large variety of complex quasicrystalline orderings (for reviews see, *e.g.*, [2–4]). A lot of different adatoms or deposited molecules were explored in recent years, including, *e.g.*, antimony and bismuth [5], copper [6], silicon [7], xenon [8], lead [9, 10], hydrocarbons [11, 12], or fullerenes [13, 14]. Similarly, vortices in type-II superconductors can be pinned onto a quasicrystalline pattern [15]. Furthermore, quasicrystalline monolayers can also be achieved in colloidal systems if a colloidal suspension that is confined to two dimensions is subjected to a laser field created by interfering laser beams that acts as an external potential [16–22]. If colloid-colloid interactions and the interaction with the external laser field are of similar strength so that they compete with each other, a huge variety of complex phases occur [17, 18, 20].

Here, we concentrate on the case where the external potential dominates and therefore laser fields with quasicrystalline symmetry enforce quasicrystalline order [16–18, 21]. Interestingly, even in the case of strong external potentials, colloidal particles can order in very different ways depending on their size as we will show in the following. Since the colloids integrate the external potential over their lateral extensions, they experience different effective

surface potentials and thereby order differently. Note that a comparable large variety of different types of quasicrystalline ordering is observed for molecules deposited on the surface of a quasicrystal. The effective substrate potential strongly depends on the size of the ad molecule even if the same substrate is employed. For example in case of adsorption on a Al-Ni-Co surface, different effective substrate potentials are obtained depending on whether methane or benzene are adsorbed as computed in [12]. Furthermore, different adatoms on i-AlPdMn, as discussed in [23], result in different orderings. In this article we will show that already for a simple quasicrystalline potential the so-called local isomorphism (LI) class of the induced quasicrystalline order depends on the size of the adsorbed particles. According to the definition of LI classes [24, 25], two quasicrystalline structures have the same LI class if and only if all finite subsets of one structure are included in the other one. For the patterns considered in this article, two structures usually are in the same LI class if they are equal up to a displacement while there is no straightforward mapping between two quasicrystals that belong to different LI classes. We will calculate the effective substrate potentials that extended particles experience. We explore how the type of quasicrystalline order can be predicted and characterized, although each particle size leads to another LI class. By changing the radius of the particles one passes through a continuous sequence of LI classes. In principle, LI classes can be characterized by their coefficients in a Fourier expansion. We will discuss how the amplitudes of the two major Fourier modes influence the

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style of the effective quasicrystalline patterns. Our results are confirmed by Monte Carlo simulations.

Note that in case of extended rod-shaped particles the effective substrate potential also depends on their orientations. This leads to even more complex patterns as demonstrated in Monte Carlo studies in refs. [26, 27] as well as experimentally in ref. [14]. However, for the sake of simplicity, in the following we will focus on circular particles such that the effective substrate potentials only depend on the locations of their centers of mass and their radii.

We explain the system and our simulations in sect. 2. In sect. 3 we calculate the patterns that are achieved for different particle radii. Our theoretical predictions for quasicrystalline substrates with ten-fold rotational symmetry are compared to the results of simulations in sect. 4 before we discuss predictions for quasicrystals with other rotational symmetries in sect. 5. Finally, we conclude in sect. 6.

2 System and simulation details

In the following, we first introduce the model system and explain the considered substrate potentials with quasicrystalline symmetry. Then we describe the details of the simulations in subsect. 2.2.

2.1 Substrates with quasicrystalline symmetry

First, we consider a substrate for point particles. Interference patterns of N laser beams have been employed in experiments as an external potential that can be used to manipulate the ordering in a two-dimensional colloidal system [16, 17, 19]. Note that the surface of many quasicrystals consists of similar patterns (see, *e.g.*, [2]). For point particles the potential created by N interfering laser beams is [28, 29]

$$V(\mathbf{r}) = -\frac{V_0}{N^2} \sum_{j=0}^{N-1} \sum_{k=0}^{N-1} \cos[(\mathbf{G}_j - \mathbf{G}_k) \cdot \mathbf{r} + \phi_j - \phi_k] \quad (1)$$

(see also background in fig. 1). The lattice vectors $\mathbf{G}_j = [2\pi \cos(2\pi j/N)/a_V, 2\pi \sin(2\pi j/N)/a_V]$ are obtained by projecting the wave vectors of the laser beams onto the xy -plane, where the length scale of the potential is given by a_V . The phases of the laser beams are denoted by ϕ_j . In the following we choose $\phi_j = 0$ for all beams. Changing one or more phase would correspond to a combination of a phonic and a phasonic displacement [30]. Phonons and phasons both preserve the LI class in the limit of long wavelengths [25]. The behavior of colloids in laser fields with changing phasonic displacements have already been studied for decagonal quasicrystals in [22, 32] and for other two-dimensional quasicrystals in [31, 33]. For a recent discussion how the phasonic degrees of freedom differ from modes that change the LI class, see also [34–36].

In order to model the adsorption of particles with non-zero radius R and center of mass $\mathbf{r}_0$ on a such a quasicrystalline substrate, we calculate the effective substrate

Fig. 1. We consider a two-dimensional substrate with quasicrystalline symmetry (here with ten-fold rotational symmetry as obtained by eq. (1) for $N = 5$). The effective potential acting on a circular particle with radius R is obtained by integrating the substrate for point particles over the area that is covered by the extended particle. The resulting patterns depend on the ratio of the radius R and the length scale of the substrate a_V .

potential $V_R(\mathbf{r}_0)$ by integrating over the particle area

$$V_R(\mathbf{r}_0) = \frac{1}{\pi R^2} \int_0^R r dr \int_0^{2\pi} d\varphi V(\mathbf{r}_0 + r\mathbf{e}_r(\varphi)), \quad (2)$$

where $\mathbf{e}_r(\varphi) = [\cos(\varphi), \sin(\varphi)]$. This potential will be employed for our theoretical analyses as well as in our simulations.

2.2 Simulation details

For the case of ten-fold rotational symmetry realized by $N = 5$ laser beams, we perform Monte Carlo simulations of a two-dimensional hard disc systems under the influence of the effective potential given by eq. (2). We choose the substrate strength $V_0 = 100k_B T$, such that we observe quasicrystalline order of the disks for all radii R of the particles and all densities that we study. Usually we consider area fractions around 0.4. We checked in additional simulations that changes of the area fractions within the range from 0.3 to 0.6 have no substantial effect on the particle ordering.

We consider periodic boundary conditions. In order to minimize the discontinuities of the potential at the boundaries of the box, a so-called rational approximant [37] of the potential is implemented. Accordingly, the edge lengths L_x and L_y of the simulation box are chosen such that $L_x/a_V = 2n$ and $L_y/a_V = m/\sin(\pi/5)$, where n and m are Fibonacci numbers, *i.e.*, $(n, m) \in \{(8, 13), (13, 21), (21, 34), (34, 55), \dots\}$. In order to achieve the desired area fraction, a particle number between 1000 and 2250 is used.

We run at least ten independent simulations for each disc radius. In every run the system is equilibrated during 10^5 Monte Carlo steps and we subsequently evaluate the positions of the particles from 800 configurations taken every 500 steps.

3 Identifying the accessible LI classes

For the following calculations it is convenient to rewrite the potential for point-like particles in eq. (1). As illustrated in fig. 2 for the decagonal case (*e.g.*, $N = 5$), instead of a double sum over pairs of projected wave vectors $\mathbf{G}_j$ the potential can be expressed as a sum over $\lfloor N/2 \rfloor + 1$ terms that are related to differences of the projected wave vectors, *i.e.*

$$V(\mathbf{r}_0) = -\frac{V_0}{N^2} \sum_{j=0}^{\lfloor N/2 \rfloor} A_{j,N} \sum_{k=0}^{N-1} \cos[\Delta\mathbf{G}_{jk} \cdot \mathbf{r}_0 + \Delta\phi_{jk}], \quad (3)$$

where $\lfloor N/2 \rfloor$ denotes the largest integer that is smaller or equal to $N/2$, $\Delta\mathbf{G}_{jk} = \mathbf{G}_k - \mathbf{G}_{k+j}$, and $\Delta\phi_{jk} = \phi_k - \phi_{k+j}$. Since all contributions occur twice except for $j = 0$ or $j = N/2$ the prefactor $A_{j,N}$ is $A_{j,N} = 1$ in case of $j = 0$ or $j = N/2$ and $A_{j,N} = 2$ otherwise. Note, the term for $j = N/2$ only occurs if N is even. Furthermore, since $\Delta\mathbf{G}_{0k} = \mathbf{0}$ the contribution for $j = 0$ leads to a constant term, *e.g.*, $A_{0,N}N = N$ for $\Delta\phi_{0k} = 0$. All other $\lfloor N/2 \rfloor$ terms are related to stars of vectors $\Delta\mathbf{G}_{jk}$, *e.g.*, there are two stars of vectors for the case $N = 5$ as shown in fig. 2b.

We insert the rewritten formula for the laser potential of point particles into eq. (2) and obtain

$$V_R(\mathbf{r}_0) = -\frac{V_0}{\pi R^2 N^2} \int_0^R r dr \int_0^{2\pi} d\varphi \sum_{j=0}^{\lfloor N/2 \rfloor} A_{j,N} \times \sum_{k=0}^{N-1} \cos[\Delta\mathbf{G}_{jk} \cdot (\mathbf{r}_0 + r\mathbf{e}_r(\varphi)) + \Delta\phi_{jk}]. \quad (4)$$

Conversions of the integrand lead to

$$\begin{aligned} & \int_0^R r dr \int_0^{2\pi} d\varphi \cos[\Delta\mathbf{G}_{jk} \cdot (\mathbf{r}_0 + r\mathbf{e}_r(\varphi)) + \Delta\phi_{jk}] \\ &= \frac{1}{2} \exp[i\Delta\mathbf{G}_{jk} \cdot \mathbf{r}_0 + \Delta\phi_{jk}] \\ & \times \int_0^R r dr \int_0^{2\pi} d\varphi \exp[i\Delta\mathbf{G}_{jk} \cdot r\mathbf{e}_r(\varphi)] \\ &+ \frac{1}{2} \exp[-i\Delta\mathbf{G}_{jk} \cdot \mathbf{r}_0 - \Delta\phi_{jk}] \\ & \times \int_0^R r dr \int_0^{2\pi} d\varphi \exp[-i\Delta\mathbf{G}_{jk} \cdot r\mathbf{e}_r(\varphi)] \\ &= \cos[\Delta\mathbf{G}_{jk} \cdot \mathbf{r}_0 + \Delta\phi_{jk}] \pi R^2 I_R(\Delta\mathbf{G}_{jk}), \end{aligned} \quad (5)$$

where

$$I_R(\Delta\mathbf{G}_{jk}) = \frac{1}{\pi R^2} \int_0^R r dr \int_0^{2\pi} d\varphi \exp[ir\Delta\mathbf{G}_{jk} \cdot \mathbf{e}_r(\varphi)]. \quad (6)$$

Fig. 2. (a) Star of projected wave vectors $\mathbf{G}_j$ (black arrows) for a laser interference pattern created by $N = 5$ beams. The blue and red lines indicate the differences $\Delta\mathbf{G}_{1j} = \mathbf{G}_j - \mathbf{G}_{j+1}$ of neighboring and the differences $\Delta\mathbf{G}_{2j} = \mathbf{G}_j - \mathbf{G}_{j+2}$ of next to neighboring wave vectors, respectively. (b) The difference vectors $\Delta\mathbf{G}_{1j}$ and $\Delta\mathbf{G}_{2j}$ can be rearranged as two stars. As shown in eq. (3) the laser interference patterns can be rewritten as a sum over a term only depending on $\Delta\mathbf{G}_{1j}$ (blue star) and a term only depending on $\Delta\mathbf{G}_{2j}$ (red star). The LI-class of a quasicrystalline pattern depends on how the two terms related to the two stars are weighted relative to each other.

With $\Delta\mathbf{G}_{jk} \cdot \mathbf{e}_r(\varphi) = |\Delta\mathbf{G}_{jk}| \cos[\alpha_{jk} - \varphi] = |\Delta\mathbf{G}_{jk}| \cos \vartheta_{jk}$, where α_{jk} denotes the direction of $\Delta\mathbf{G}_{jk}$ and $\vartheta_{jk} = \alpha_{jk} - \varphi$, eq. (6) becomes

$$I_R(\Delta\mathbf{G}_{jk}) = \frac{1}{\pi R^2} \int_0^R r dr \int_0^{2\pi} d\vartheta_{jk} \exp[ir|\Delta\mathbf{G}_{jk}| \cos \vartheta_{jk}]. \quad (7)$$

Note that $|\Delta\mathbf{G}_{jk}|$ is the same for all k due to the symmetric arrangement of the vectors and therefore we will denote it by $\Delta G(j)$. Furthermore, due to the periodicity of $\cos \vartheta_{jk}$ and the integration over a full period, the integration over ϑ_{jk} can be replaced by an integration over ϑ for all k and j . Since $I_R(\Delta G(j)R) = I_R(\Delta G(j))$. Upon substituting $\omega_j = \Delta G(j)r$ and $\Omega_j = \Delta G(j)R$ as well as incorporating Bessel functions of the first kind $J_n(x)$, the integration is finally carried out to yield

$$\begin{aligned} I(\Delta G(j)R) &= I(\Omega_j) \\ &= \frac{1}{\pi \Omega_j^2} \int_0^{\Omega_j} d\omega_j \omega_j \int_0^{2\pi} d\vartheta \exp[i\omega_j \cos \vartheta] \\ &= \frac{2}{\Omega_j} J_1(\Omega_j). \end{aligned} \quad (8)$$

Therefore, we obtain for the effective potential for extended particles in eq. (4)

$$V_R(\mathbf{r}_0) = -\frac{V_0}{N^2} \sum_{j=0}^{\lfloor N/2 \rfloor} I(\Delta G(j)R) \times A_{j,N} \sum_{k=0}^{N-1} \cos[\Delta\mathbf{G}_{jk} \cdot \mathbf{r}_0 + \Delta\phi_{jk}]. \quad (9)$$

In principle, the formula looks similar to the potential of point particles as given in eq. (3). However, while in eq. (3)

all stars of vectors are weighted equally, in the effective potential for extended particles in eq. (9) the contributions of different stars contain an additional weight function that depends on the radius R of the particles and the length of the vectors in the corresponding star. Especially the sign of the weight function may be positive or negative, which can give rise to significant variations of the effective potential.

For our calculations we considered a substrate potential that can be realized by interfering laser beams. Note that for other substrate potentials the calculation works similarly. The substrate potential for point particles can be written as sum over stars of wave vectors. An integration leads to the effective potential for extended particles. It contains a weight function for each star that depends on the size of the particles.

Before we discuss the results of our calculations for the case of decagonal quasicrystals in more detail, we comment on the change of the LI class due to changes of the size of the particles. Since weight functions are also present in the expansion of the free energy of a quasicrystal (or a crystal in general) and since the weight functions depend on the radius of the particles, the free energy changes when the size of the particles is changed. Since a change of the free energy always implies that the LI class is changed as well [25], the LI class indeed depends on the size of the particles.

4 Results for decagonal quasicrystals

In this section, we consider the interference pattern created by $N = 5$ beams as our substrate for point-like particles (see fig. 3a). In fig. 3 we have assembled a collection of the calculated effective substrate potentials that act on extended particles for different radii R (figures in the middle rows) as well as plots from Monte Carlo simulations of the corresponding colloid systems (figures in bottom rows). Since the strength of the substrate potential is chosen to be very strong in the simulations, we are able to observe exactly the type of quasicrystalline order that is expected from the calculations. The top figures in fig. 3 show the weight functions $I(\Omega_j)$ as determined in eq. (8) for the two stars that occur in the considered decagonal potential. The two values of $\Omega_j = \Delta G(j)R$ for the specified particle size are marked by circles and the colors of the circles indicate the corresponding star as drawn in fig. 2b.

The extended particles integrate over large variations in the potential landscape and thereby the contrast decreases for increasing radius R . In our calculations, the decrease of contrast is respected by the decay of $I(\Omega_j)$ with increasing $\Omega_j = \Delta G(j)R$. Note that for the effective potential patterns shown in fig. 3 the color code is chosen independently in each figure in order to enhance visibility such that the decrease of contrast is not visible. However, in the simulated particle positions the patterns for large R (fig. 3e,f) are more blurred than for small R . In general, due to the decreasing contrast in the effective potentials, the particle-particle interactions become more dominant

with increasing R , unless the laser intensity is increased accordingly to counter balance this effect. In the insets in fig. 3 we present Fourier analyses of the simulation results. The blue and the red vertical lines mark the Fourier modes that correspond to the blue and red vectors shown in fig. 2, respectively. For these modes, the Fourier amplitudes and their signs for these modes are in qualitative agreement with the theoretical predictions.

Interestingly, similar quasicrystalline patterns can occur for different radii R . For example, fig. 3f is similar to fig. 3d if the colors are inverted, *i.e.*, maxima are replaced by minima and vice versa. Furthermore, there are similarities between fig. 3e and c, which become clear again when the color code is inverted and this time also by rescaling the characteristic length scales of the patterns. Such similarities are caused by the special functional form of $I(\Omega_j)$. In case of figs. 3d and f, the weight function causes the contribution of one star to be weighted positive while the other star is weighted negative. The color inversion is related to the opposite signs of these weights. In case of figs. 3c and e one of the two weights is zero. The other weight values differ in sign. Furthermore, in fig. 3c the term of the blue star has a non-zero weight, while in fig. 3e the weight of the red star is non-zero. Therefore, the length scales of the two patterns are given either by the vectors of the blue star or the vectors of the red star, which explains the necessary scaling to match both patterns.

Due to the oscillations in $I(\Omega_j)$ similar patterns repeat (up to a difference of the contrast) if the radius R is further increased. Considering just the two essential weight values $I(\Omega_j)$ and especially their sign enables a fast prediction of what type of patterns can be expected. Note, however, that the type of the realized LI class depends on the exact values of the weights.

In this section we considered the effective substrate potentials that are obtained if the interference pattern created by $N = 5$ laser beams is considered as starting pattern. In the next section we discuss further examples with other rotational symmetries.

5 Predictions for other quasicrystalline symmetries

The calculations presented in sect. 3 are valid for interference patterns with an arbitrary number of laser beams. Furthermore, similar calculations are possible for all quasicrystalline patterns (or even all crystalline patterns) with perfect rotational symmetry because they can be expanded in terms of contributions connected to stars of lattice vectors. In addition to the results for $N = 5$ laser beams shown in the previous section, we present additional examples for $N = 6$, $N = 8$, and $N = 12$ laser beams in the following.

In fig. 4a, d, and g examples of effective substrate potentials for different particle radii in case of the periodic interference pattern of $N = 6$ beams are shown. In such a two-dimensional periodic structure all lattice vectors are sums of integer multiples of two basis vectors and therefore the stars of lattice vectors considered in the expansion

Fig. 3. The ordering of particles in the interference pattern created by $N = 5$ laser beams depends on their radius R : (a) point particles, (b-f) increasing particle radius, where R is given in units of the length scale $a_V = 2\pi/G_j$ of the potential. In the top rows of all figures the weight function $I(\Omega_j)$ is shown. The actual weights of the two contributing stars are marked by red and blue circles using the same colors as in fig. 2b. The middle rows of all figures show the calculated effective substrate potential for particles of the specified size. Blue areas denote minima and red areas maxima of the potential energy. Note that the color code is chosen for each figure individually in order to achieve the best contrast. The bottom rows of all figures display particle positions determined in Monte Carlo simulations of hard disks of the corresponding sizes. The simulation data is shown for ten independent runs with 800 configurations per run. The insets show the radial dependence of the Fourier transformed density distributions $\tilde{\rho}(g)$ obtained from the simulations, averaged over all directions and divided by the mean density $\bar{\rho}$. The peaks marked by the blue and red line denote the contributions of the Fourier modes associated with the blue and red vectors depicted in fig. 2.

Fig. 4. Effective substrate potentials for particles with different radii R in laser fields created by $N = 6$ (left columns), $N = 8$ (center column), or $N = 12$ (right column) interfering laser beams.

used in sect. 3 are closely related. In fact, for the case $N = 6$ all lattice vectors of the second star are obtained as sums of two lattice vectors of the first star. As a consequence, by different weights the contrast of the effective pattern might change and the pattern can even be inverted. However, the position of extrema does not change. Note that the change of the LI class connected to the inversion that is visible in figs. 4d and g is similar to one of the modes that recently were described for periodic and aperiodic quantum crystals [38]. It was claimed that since the modes are almost gapless they correspond to phasons that only occur in quantum systems [38]. However, obviously these modes are not unique to quantum systems and since they do not preserve the LI class they are not phasons [34–36].

Quasicrystals can be characterized by their rank, *i.e.*, the number of basis vectors needed to span the set of all lattice vectors (see, *e.g.*, [39]). While two-dimensional periodic crystals possess two basis vectors and therefore are rank-two structures, the rank of most two-dimensional quasicrystalline patterns that were observed in nature so far is four. Furthermore, rank-four quasicrystals also occur for much weaker substrate potentials than quasicrystalline order of larger rank [19, 21]. Besides the decagonal quasicrystal that we considered in sect. 4, the interference

patterns with $N = 8$ and $N = 12$ also possess rank four. Therefore, we study quasicrystalline substrates with these symmetries in the middle and right column of fig. 4. In principle, when increasing the radius, we find a similar sequence of effective substrate potentials as for the decagonal quasicrystals. This is due to the fact, that all rank-four quasicrystals possess two incommensurate length scales per direction. The change of contrast and the inversion also occur in periodic structures. Furthermore, in all quasicrystalline patterns of rank four but not in the periodic rank-two structures we observe that the dominating length scale can change when particle size increases. For example figs. 4e and h look similar to the case of figs. 3c and e.

6 Conclusion

By employing analytic calculations and simulations, we have determined how the effective substrate potential acting on extended particles depends on the size of these particles. Our considerations are not limited to colloidal particles in laser fields but also apply to molecules that are adsorbed on the surfaces of quasicrystals. The quasicrystalline substrate potential can be expanded into plane

waves that are grouped into stars of lattice vectors. We have shown that the effective potentials depend on the particle sizes, because each star of lattice vectors carries its own weight factor. Furthermore, the LI class of an effective pattern changes if the radius of the particles is changed. However, despite the change of the LI class, similar effective potentials might occur for different particle radii. For example, patterns with different contrast and inverted potential landscape exist and in the case of quasicrystalline structures they even possess different governing length scales. So, they have to be inflated or deflated to align them. In our study we considered structures of rank two and four but we expect similar effective patterns in case of quasicrystals with larger rank. Then, even more incommensurate length scales exist and as a consequence similar patterns probably occur for all of these lengths scales.

Our study demonstrates that for one quasicrystalline substrate already a large variety of effective patterns can be achieved if the adsorption of particles of different sizes is considered. Though even the LI class of all these effective potentials are different, determining the weight function and the actual weights for a specific particle size might help to predict, connect, and properly characterize all these patterns.

Since the effective substrate potential depends on the size of colloids, the dynamics of the particles is affected as well. As a consequence, phenomena like directional locking of colloids on quasicrystalline substrates [40, 41] or the colloidal motion driven by phasonic drifts [22, 32] might be employed in order to separate colloidal particles by their size.

A question for future research is how the LI class can be controlled in case of intrinsic quasicrystals. Even isotropically interacting colloidal particles are known to be able to self-assemble into quasicrystalline structures [42–44] or cluster quasicrystals [45]. It would be interesting to find out how patterns with a specific LI class can be obtained in a well-controlled way either by energetically favoring specific stars of lattice vectors or dynamically by choosing the appropriate growth mode [46].

Author contribution statement

F.R. performed the Monte Carlo simulations, M.Sa. developed the theoretical analysis, H.S. and M.Schm. designed the studies. All authors wrote the manuscript together.

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