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Non-Overlapping Arrangement of Identical Objects: An insight for molecular close packing†

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In this study, we investigate the arrangement of identical objects to achieve non-overlapping configurations, a fundamental challenge across various scientific and engineering disciplines, particularly in the context of molecular close-packing. We introduce the concept of "Maximal Boundary Distance", defined as the largest distance between extreme points of an object in a given direction. This length ensures that aligned objects touch without overlapping. Our approach is validated through mathematical proofs and illustrated with both, two-dimensional and three-dimensional examples, emphasizing applications in molecular systems. We further explore the practical implications of this for the packing of molecular crystals by identifying optimal lattice constants. The stability of these configurations is further analyzed using the Density Functional Tight Binding method. This work not only advances our understanding of object arrangement but also has significant implications for the strategic positioning of molecules in close-packed structures.

1 Introduction

Arranging objects within a given space to achieve packed configurations is a fundamental challenge with wide-ranging applications across disciplines. From the physicochemical point of view, this is related to what is often referred to as "packing" or "close-packing".[1] While the principles of close-packing have been extensively explored, the precise conditions for achieving non-overlapping arrangements of objects systematically have remained a subject of ongoing investigation.[2–4]

In many contexts, ranging from architecture and design to material science and computer graphics, the ability to place identical objects in a manner that ensures they neither intersect nor overlap is of paramount importance.[5,6] Packed arrangements play an essential role in maintaining structural integrity and facilitating efficient use of space.[7] Yet, the theoretical and methodological conditions required to achieve this objective for two and three-dimensional objects are far from complete. Arranging objects efficiently within confined spaces presents a significant computational challenge that spans multiple disciplines.[8] Historically, this problem has been tackled through various methodologies, each contributing unique insights into space optimization. The paper by Iori *et al.* discusses exact solution techniques for two-dimensional cutting and packing problems, providing foundational methods that inform current practices in object arrangement.[9] In more specialized applications such as logistics and routing, Wei *et al.* demonstrate the integration of routing and packing problems, using advanced heuristics to address the capacitated vehicle routing problem with two-dimensional loading constraints. Their approach highlights the practical implications of combining computational efficiency with real-world logistical needs.[10] Further expanding the scope of computational design, Wang *et al.* review state-of-the-art techniques in the assembly of rigid parts. Their work emphasizes the importance of precision and stability in the computational design of assemblies, offering insights applicable to both industrial and more theoretically driven pursuits.[11] Each of these studies contributes to a layered understanding of how computational methods have evolved and how they continue to influence practical and theoretical approaches to object accommodation and space optimization across various fields.

This paper is devoted to finding a solution to the problem of having non-overlapping arrangements of close-packed images of a given object, and its application to molecular close-packing. Our approach centers on the concept of "Maximal Boundary Distance" (MBD) where we explore how maintaining specific distances be-

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tween adjacent images of an object guarantees contact without overlap. While the concept is intuitive, its systematic application across different dimensions offers novel insights and practical solutions. By identifying critical boundary points and leveraging translations, we decipher the conditions necessary to achieve non-overlapping arrangements in two and three dimensions. Furthermore, our approach paves the way for future explorations into more complex shapes and their arrangements across diverse domains, especially in the field of molecular close-packing. This work is organized as follows: in the second section we discuss the main idea and provide a mathematical proof for it, then in the third section we present a two dimensional example, where we use the MBD to accommodate 2D shapes in contact. In the fourth section we show how to do the same with 3D molecular structures, and finally in the fifth section we extend this approach to form close-packed molecular crystals of Pentacenetetrone, by implementing a random exploration of molecular orientations and corresponding unit cell configurations, and the stability of these crystals is studied with the Density Functional Tight Binding (DFTB) method.

2 Methodology

We will study an arbitrary shape and images of it such that they are arranged along the horizontal direction, touching each other but not overlapping. For the sake of simplicity, we consider single-valued two-dimensional shapes and we investigate the maximal boundary distance in the horizontal (x) dimension as follows. We will prove that the distance between adjacent shapes is equal to the largest horizontal length between opposite points in the original shape, which we call d_1^{max} and that is the MBD. We probe this by showing that when the distance between adjacent shapes is d_1^{max}:

1) contact between adjacent shapes is ensured, and

2) overlap of the shapes does not occur.

So, for any other distance than d_1^{max}, there will be no contact, or conversely, the shapes will overlap. This result can be generalized to any other direction and, also to the three-dimensional case. The implications of this idea can pave the way to new strategies to close-pack objects, and this is specially important for the prediction of molecular crystal structures.

2.1 Proof for Non-Overlap based on Maximal Boundary Distance

First, onsider an arbitrary two-dimensional shape S (See Fig. 1). Let's examine two points x_1 and x_2 on the shape's boundary such that both lie on a horizontal line of constant height. Let:

- x_1 be the leftmost point, and
- x_2 be the rightmost point

.

We define the maximum boundary distance d_1^{max} at height y as

$$d_1^{max} = x_2(y) - x_1(y). \tag{1}$$

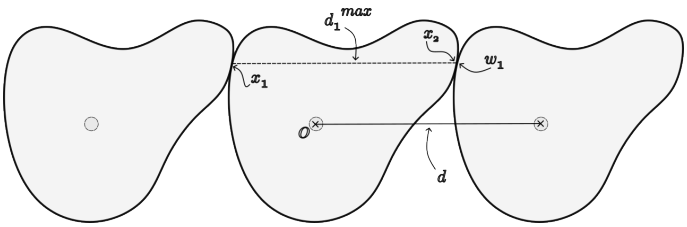

Fig. 1 Representation of an arbitrary shape centered at origin O and its images to the left and right in the horizontal x-direction. It is visually noticed that the center-to-center distance is equal to the lenght d_1^{max} between points x_1 and x_2 which is the largest distance between opposite points of S intersecting a horizontal line. On the right image, w_1 is shown to be equivalent to x_1 and having the same value as x_2.

Notice that at any other height, the distance between x_2 and x_1 will be less than or at most equal to d_1^{max}. Consider another image of S positioned to the right. Let w_1 and w_2 be the leftmost and rightmost points on this image, respectively, at height y. Thus, necessarily we must have:

$$w_1(y) = x_1(y) + d, \tag{2}$$

where d is the center to center separation between both shapes. To ensure contact, we require that

$$w_1(y) = x_2(y). \tag{3}$$

This implies that:

$$d = d_1^{max} \tag{4}$$

So, this leads to the fact that *the distance between images must be equal to d_1^{max} in order to have contact between them*.

Second, we should prove that any other pair of points not satisfying eq. 1 will not make contact, nor will make the shapes overlap.

Consider another height y' for which the distance between x_1 and x_2 is not maximum. Thus,

$$d_1^{max} > x_2(y') - x_1(y'). \tag{5}$$

Given the established relationship from Equation (2), it can be inferred that:

$$w_1(y') = x_1(y') + d_1^{max}. \tag{6}$$

Combining the last two equations, we conclude:

$$w_1(y') - x_1(y') > x_2(y') - x_1(y'), \tag{7}$$

or

$$w_1(y') > x_2(y'), \tag{8}$$

which confirms no overlap or contact at height y'. This relationship guarantees that, by selecting the maximum boundary distance as the spacing, adjacent images of S will touch without overlapping.

These findings are valid even when the shape suffers arbitrary rotations, which can be used to define a systematic approach for optimizing close-packing by minimizing the value of d_1^{max} by rotating the shape.

Generalization for three dimensions is possible just by replac-

ing y and y' in the previous equations by y,z and y',z' respectively, where the first pair is the point in the yz plane where the maximum distance between boundary points will occur.

3 Example in two dimensions

In this section, we will review a simple case of a two dimensional shape formed by the superposition of two circles, forming a snowman like shape using simple geometric relations. For this, we can define two circles of radii r_1 and r_2, centered in (a_1,b_1) and (a_2,b_2) respectively. Then, we can find that the height y where they will touch is given by solving:

$$(r_2^2-r_1^2)y^2-2(b_1r_2^2-b_2r_1^2)y+(b_1^2r_2^2-b_2^2r_1^2)=0 \tag{9}$$

Knowing the solutions, it is possible to find the extreme values for the x coordinates in the composed shape. In Fig.2 we show the "snowman" and its images in different orientations, and it can be seen that they always touch each other, at least in one point. The code for this implementation is found in `https://github.com/napoles-uach/max_distance/blob/main/Snowman.ipynb`. We can observe that there are angles for which the molecules are more closely accommodated. More general shapes could be difficult to study analytically, but it could be possible to approximate them by taking many circles to fit the contour and follow a similar approach as in this example.

4 Application to Molecules

A common representation for a molecule is to consider it as a discrete object formed by a collection of spheres of van der Waals radii. For example, Carbon has a radius of 1.7 Å. In this way, we can find the displacement distance to have contact without overlap by making geometric considerations as follows. Let us consider a given pair of atoms A and B in a molecule, such that atom B touches atom A' in the neighbor molecule displaced along a given direction vector $\mathbf{v}$. In this case, A' is equivalent to A in the image molecule. Our purpose is to find the distance between A and A'. In Fig. 3, we are seeing the plane formed by the centers of atoms A, B, and A', and the direction vector $\mathbf{v}$ lies in the horizontal direction.

According to Fig. 3:

- $\mathbf{r}_i$ is the position of atom A
- $\mathbf{r}_j$ is the position of atom B
- a is the radius of atom A, and A'
- b is the radius of atom B
- d is the distance between A and B
- P_1 is the projection of ($\mathbf{r}_j$-$\mathbf{r}_i$) on $\mathbf{v}$
- S is $\sqrt{d^2-P_1^2}$
- P_2 is $\sqrt{(a+b)^2-S^2}$

Thus, by trigonometry we can find that the distance between A and A' by evaluating P_1+P_2. The next step is to iterate over all pairs of atoms satisfying:

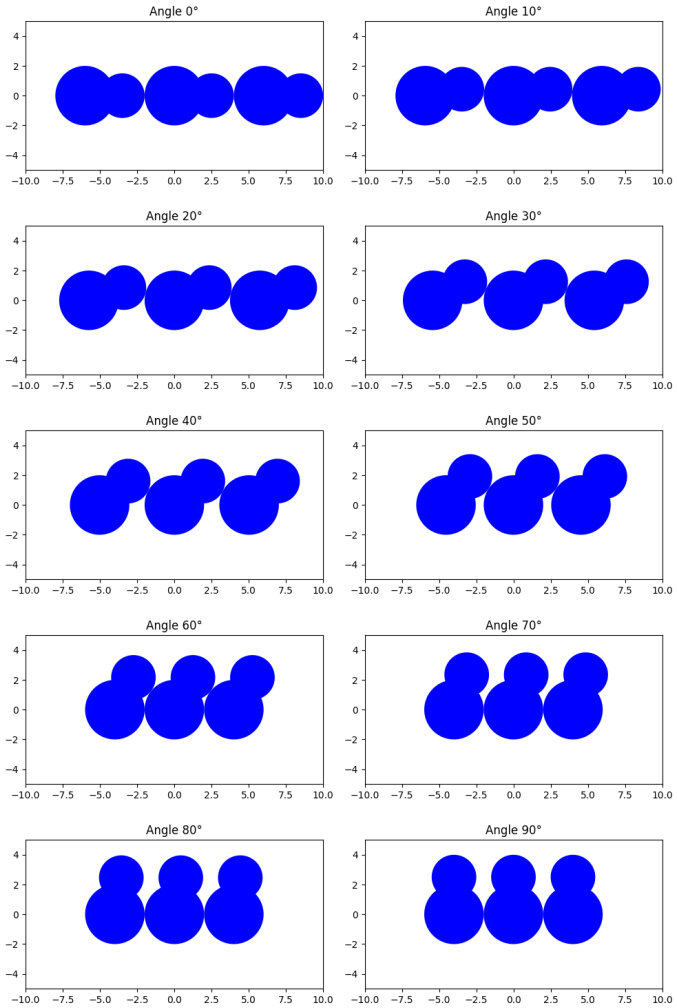

Fig. 2 "Snowman" shape and its left and right images for different orientations between 0 and 90 degrees. It can be noticed that for all cases, there is contact in at least one point, and there is not overlap in any case.

$$a+b\geq S \tag{10}$$

otherwise, they would not touch each other. In each iteration, we should store the resulting distance between A and A' in order to get the largest, which is the distance the image molecule should be displaced to ensure contact without overlap.

This procedure has been implemented in a Streamlit web app, (`https://molecular-contact.streamlit.app/`). By using the Stmol component,[12] it is possible to select some molecules, or upload any SDF file with a molecule. In Fig. 4 we can see some outputs for fullerene PCBM, and Cholesterol where it can be observed that the molecules touch each other without overlapping. The code is open sourced in this github repository: `https://github.com/napoles-uach/max_distance` As can be seen, different molecule orientations give place to different displacements, which means that this length can be optimized by performing rotations on the original molecule. This procedure can be extended, for example, to find close-packed molecules in

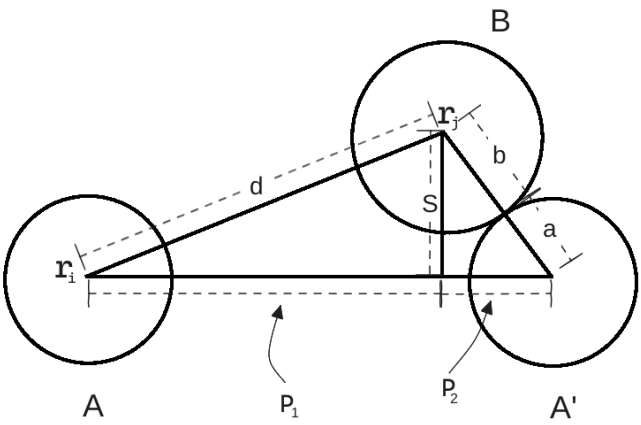

Fig. 3 Schematic representation of atoms A and B in a molecule, and antom A' (image of A) touching atom B. The sum of distances P_1 and P_2 are used to define the maximum boundary distance.

a unit cell to form molecular crystals, as we will show in the following section.

5 Molecular Crystal of Close-Packed Pentacenetetrone

As an example of how to build a crystal using the ideas presented in the previous section, let us build a close-packed crystal using the Pentacenetetrone molecule, which is known to build close-packed crystals with the molecules stacked[13]. To keep simplicity, the approach we will follow is like this: let us take a molecule with a random orientation to find how large should be a Simple Cubic unit cell having this molecule as repetition motif by ensuring that the molecule can be in contact with its nearest neighbors, without overlap. As before, the atoms are taken as rigid spheres with Van der Waals radii. If we try several random orientations, the more stable structures may likely be the ones with the lowest lattice parameter, provided that close-packing is the driving force to form the molecular crystals. We implemented this in a simple code where we explored 1500 random orientations. Algorithm 1 shows in detail how is the procedure using a random exploration to try different molecular orientations. For each iteration, the distances for contact were evaluated along 13 directions, defined by the unit vectors a=[1,0,0], b=[0,1,0],c=[0,0,1], two vector combinations: [b-c],[b-a],[c-a],[a+b],[a+c],[b+c], and three vector combinations: [a+b+c],[a+c-b],[a+b-c],[b+c-a] representing all the diagonals formed between the corners of the unit cell. These distances are used to adjust the lattice parameter to fit the molecule in close-packing arrangement. The code for this exploration can be found in `https://github.com/napoles-uach/max_distance/blob/main/Random_ClosePack.ipynb`

Algorithm 1 Random Exploration of Molecule Rotations

Input: molecule, number of iterations N, 13 Linear combinations of unit vectors V
Initialize:
Initialize lists for storing rotation results and displacement data
for $i = 1$ to N **do**
  Generate random rotation angles $(\theta_x, \theta_y, \theta_z)$
  $molecule_{rot} \leftarrow$ ApplyRotation($molecule$, $(\theta_x, \theta_y, \theta_z)$)
  Initialize list for adjusted lattice parameters
  for each vector v in V **do**
    $max\_displacement \leftarrow$ CalculateDisplacement($molecule_{rot}$, v)
    if $index(v) < 3$ **then**
      $adjusted\_displacement \leftarrow max\_displacement$ ▷ No adjustment for lattice vectors
    else if $index(v) < 9$ **then**
      $adjusted\_displacement \leftarrow \frac{max\_displacement}{\sqrt{2}}$ ▷ Adjustment for two-vector combinations
    else
      $adjusted\_displacement \leftarrow \frac{max\_displacement}{\sqrt{3}}$ ▷ Adjustment for three-vector combinations
    end if
    Append $adjusted\_displacement$ to list of lattice parameters
  end for
  $max\_param \leftarrow$ Max value from list of adjusted lattice parameters
  Store $(\theta_x, \theta_y, \theta_z, max\_param)$ in results list
end for
Output: Return list of rotation angles and corresponding maximum parameters

The total energies of all the crystals were calculated using Density Functional Tight Binding (DFTB) method, with the 3ob parametrization [DFTN,3OB] as implemented in the DFTB+ software.[14–16] We can see in Fig. 5 that the lowest values for the Energy are for the lowest lattice parameters, as we expected. Although the experimental crystalline structure is not Simple Cubic, this support the idea that Pentacenetetrone tends to form close-packed structures. A more elaborate approach should be followed to find the true lattice, but this is beyond the scope of this work.

6 Conclusions

We discovered that the optimal relative positions of identical, translated images of the same object—necessary for them to touch but not overlap—can indeed be derived from the geometry of the original object. Our finding is that the largest distance between opposite boundary points or Maximum Boundary Distance (MBD) in a given direction (the direction in which we want to perform the translation) is the length that images must be separated to fulfill both conditions. This fact establishes a direct and reliable approach to determining the optimal spacing between adjacent images of any arbitrary object, ensuring contact without overlap. By focusing on the MBD, the proof is both intuitive and robust. The methodology and proof presented can be a foundation for further explorations into the arrangement of more complex shapes and can find applications in various domains, for example,

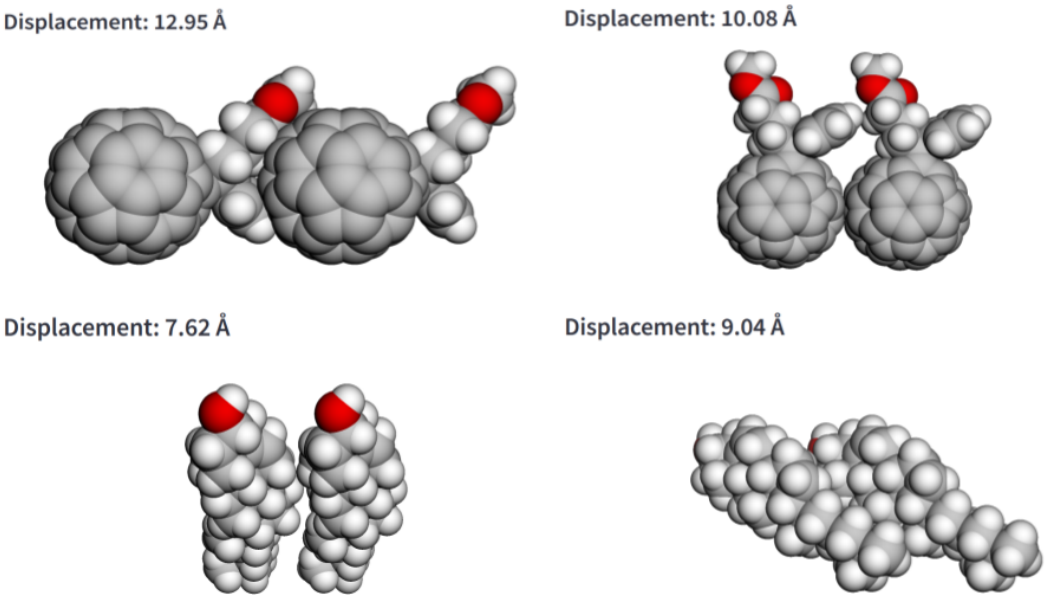

Fig. 4 Screenshots of the Streamlit web app showing PCBM fullerene (Up), and Cholesterol (down) for two different molecular orientations showing the contact without overlap condition satisfied.

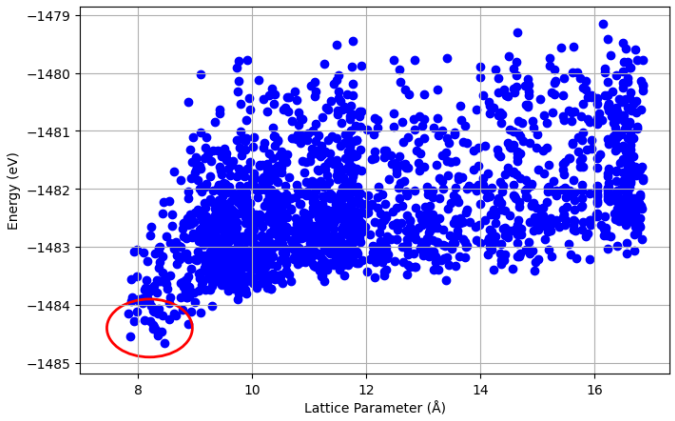

Fig. 5 Total energy of 1500 crystals formed by randomly rotating the motif molecule in simple cubic unit cells. It can be noticed that as the lattice parameter goes to a minimum, the crystal energy tends to be lower.

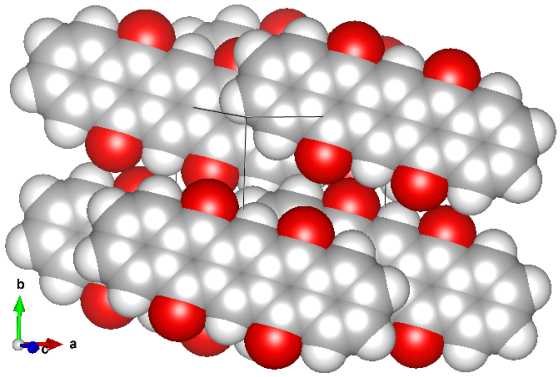

Fig. 6 Unit cell of simple cubic Close-packed Pentacenetetrone for the lowest lattice parameter, showing some of the repetition molecules of neighbor cells.

molecular crystals, protein docking, videogame development, and any field where it is important to optimize the organization of objects. Looking ahead, we plan to extend the application of MBD by incorporating group theory, which will enhance our understanding of symmetry in molecular crystal packing. Additionally, we envision adapting the MBD concept for configurations involving different objects, broadening the scope of our study to include a more diverse range of applications. These future directions not only promise to expand our theoretical framework but also aim to provide practical solutions to complex spatial arrangement challenges in various scientific and industrial fields.

Data and code availability

All data and code is shared in the following github repository `https://github.com/napoles-uach/max_distance`

Author Contributions

Conceptualization: J. M. N.-D. Formal Analysis: J. M. N.-D., J. P. P. B, Writing – review & editing: J. M. N.-D., R. P.-C, and M. A. C.-R.

Conflicts of interest

There are no conflicts to declare.

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