Brownian motion and Edagawa phasons

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Abstract. The so far most direct observation of phasons was reported in the year 2000 by Edagawa et al. In HRTEM images of decagonal Al-Cu-Co they detected white spots which formed vertices of a quasiperiodic tiling. Some spots vanished and reappeared in phason flipped positions within seconds to minutes. Already at ICQ13 we had applied a structure model of Zeger and Trebin of the year 1996 to explain the physical origin of the spots and the time scale of the flips. We found that the spots are columns of ten-rings of atoms which change positions by fluctuations of atomistic phason flips. Thus they resemble Brownian motion of mesoscopic particles, which are pushed by fluctuating knocks of fluid molecules. Whereas Brownian motion is described by a random walk in continuous space, Edagawa phasons can be conceived as a random walk on a quasilattice.

1. Introduction
It is almost two decades ago that in a seminal paper Edagawa et al. [1] published High Resolution Transmission Electron images of decagonal Al-Cu-Co along the decagonal axis. They observed white spots, which, when connected, form the vertices of a quasiperiodic decagonal tiling. Within seconds to minutes some of the spots disappeared and reappeared at phason flipped positions. To explain the physical origin of the white spots and the time scales already at ICQ13 we had presented a model for structure and dynamics of the system [2]. It turned out that the spots and their flips resemble atomistic phason flips in a self-similar fashion, both for structure and dynamics. They are an inflated image of atomistic motions. In this respect they are comparable to Brownian motion. In the following we will work out this analogy after deepening the insight into our model and its dynamics.

In Brownian motion mesoscopic particles – originally pollen – are pushed by fluid molecules. The pollen motion resembles the motion of the molecules but with differences in size – micrometers instead of Ångströms – and time scales. The velocities of the particles are micrometers per second, those of the molecules kilometers per second, nine orders of magnitude faster. Brownian motion is described as a symmetric random walk, where in a simplified one-dimensional form the probability for a step to the right equals that of a step to the left, independent of history: \( p_\uparrow = p_\downarrow = \frac{1}{2} \). The global motion is diffusive, the averaged squared distance to the origin is proportional to time: \( \langle r^2 \rangle = 2Dt \).
2. Model for d-Al-Cu-Co

2.1. Structure

We use a structure model of Burkov [3] modified by Zeger and Trebin [4]. It consists of two layers stacked periodically by 4.18 Å. The projection of the two layers is a Tübingen triangle tiling with a complex decoration of the two golden triangles (Fig. 1). The vertices are centers of rings of ten atoms which form a column along the stacking axis. These columns are supposed to form the white spots under certain conditions.

![Symbols: ▲ Cu ■ Co • Al](image1)

**Figure 1.** (From Ref. [2]) Double layer in the model structure for d-Al-Cu-Co. Empty and filled symbols denote atoms in the lower and upper layer, respectively. Each vertex is surrounded by a ring of ten atoms. The aluminum atoms of a ring belong to rhombs (dashed) of the deflated tilings (left figure). A rhomb can change from its original state \(x = 0\) to a reflected one \(x = 1\) by a simpleton flip. While doing so the aluminum atom inside the large triangle moves by 0.87 Å to the new vertex position; the previous vertex atom leaves by the same distance for the interior of the freshly formed large triangle. If both rhombs perform this flip (from state 00 to 11) an entire ten-ring is jumping in the original, undeflated tiling (right figure) [4].

Let us concentrate on the oriented rhomb in the lower left part of Fig. 1 and on the ring around the central vertex. Several atoms of the ring belong to equally oriented smaller rhombs of a three times deflated triangle tiling, one rhomb in the lower layer and one slightly displaced in the upper layer. These rhombs are stacked zig-zag in a column perpendicular to the paper plane. They can change their orientation from state 0 to 1 and vice versa in a correlated flip of two aluminum atoms by 0.87 Å, which we denote “simpleton flip”. If this flip occurs simultaneous in both layers (from state 00 to 11), the entire ten ring performs an identical flip in the inflated structure. We observe a self-similarity in both structure and kinetics. If the small rhombs are in different states (01 or 10), there is no ten ring. Be aware that along the stacking axis successive single layers can alternate in states 0 and 1. Therefore we have to study in detail an entire column as in Fig. 2.

2.2. Visibility of white spots

In Fig. 2 a column of small rhombs in \(N = 30\) single layers is depicted symbolically, each in state 0 or simpleton flipped state 1. Note that the small rhombs are not exactly on top of each other, but those in the even numbered layers are slightly displaced with respect to those in the
Figure 2. (Cp. Ref. [2])

First column: numbering of the single layers. Second column: relative flipping probability of a single small rhomb. Third column: state of the small rhomb and cluster structure. Forth column and adjacent rings: state of the double layer, forming a ten-ring to the left if of type 00, to the right if of type 11.

odd numbered. If two consecutive layers are in state 00, we get a ring to the left, if in state 11 to the right. In this example there are $n_{00} = 10$ rings to the left and $n_{11} = 7$ rings to the right. No ring is visible, if the states are different. Now we introduce a visibility parameter $\alpha$ between 0 and 1 and postulate, that a white ring appears left or right if the numbers $n_{00}$ or $n_{11}$, respectively, exceed the critical fraction $\alpha N$ of total layers.

For the following a central notion is that of “cluster”, which is a consecutive sequence of layers in identical state, here altogether $S_c = 13$. The total number $S_c$ is important for the visibility of spots. Each cluster contributes one boundary with incomplete ten ring, hence reduces the total number $n_v$ of complete ten-rings by one and leads to the relation $n_v := n_{00} + n_{11} = N - S_c$.

2.3. Dynamics

We investigate the dynamics of the white spots by combinatorial and Kinetic Monte Carlo methods and introduce a relative rate parameter $\gamma > 1$ for the simpleton flips. Each rhomb can flip between 0 and 1 by a rate which is dependent on the state of the rhombs in the neighboring layers. It shall be lowest, namely 1, if the neighbors are in the same state, as for example in layer 8, to stabilize clusters. It shall be highest, namely $\gamma$, if the neighbors are in equal state, but differing from the considered rhomb, as for example in layer 10. And it shall be intermediate, namely $\sqrt{\gamma}$, if the neighbors are in different states, corresponding to half a barrier height in a double well potential. With these rates several statistical data are calculated, for example the average number of clusters $S_c$. Denote the number of clusters of length $n$ by $c_n$, the change of $c_n$ in one simpleton flip by $\Delta c_n$. If the numbers $c_n$ do not change on average: $\langle \Delta c_n \rangle = 0$, we call this state “dynamic equilibrium” and endow the corresponding average data by superscript zero: $c_n^{(0)}$, $S_c^{(0)}$. It turns out that the probability for obtaining $S_c$ clusters is a binomial distribution centered at $S_c^{(0)}$. For counting clusters in the following we use the deviation $2F$ from $S_c^{(0)}$: $S_c = S_c^{(0)} + 2F$. Why the factor 2? Because in a simpleton flip $S_c$ changes by 0 or $\pm 2$ and hence $F$ by 0 or $\pm 1$.

In Fig. 3 the binomial distribution for the probability of observing $S_c$ clusters or the deviation $F$ from dynamic equilibrium is shown for two different rates $\gamma$. It becomes steeper for increasing $\gamma$. Since the number $n_v$ of complete ten rings is related linearly to $S_c$, also this quantity is binomially distributed, and finally, since positions 00 and 11 are statistically equivalent, also the individual numbers of rings $n_{00}$ and $n_{11}$.
2.4. Random walks and time scales

Combinatorics tells us that the binomial distribution of the probability for observing $F$ results from the fact that the probability for increasing or decreasing $F$ by 1 in one simpleton flip close to $S^{(0)}_c$ is $p_F \uparrow = p_F \downarrow = \frac{1}{4}$. The sum of these probabilities is $\frac{1}{2}$. Where is the difference to 1? It is the persistence probability $\frac{1}{2}$, where no change of $F$ occurs due to mere shift of cluster boundaries. We arrive at a microscopic random walk with doubled time scale on the order of picoseconds. It carries over to a macroscopic time scale for the appearance of a white spot due to the following observation: A white spot appears on the left side (and due to the coupling disappears on the right side), when the number of ten rings surmounts the visibility range $\alpha N$. The biggest contribution for such a step is if three clusters combine to a large one, decreasing $F$ by 1. The probability $\frac{1}{4}$ for this event has to be multiplied by the probability that the number of ten rings left is close to $\alpha N$ and that it increases at least by 2:

$$p_{\text{whitespot}}(\alpha N) =: \tilde{p} \uparrow = \frac{1}{4} p_F \downarrow p_{\text{R00}}(n_v, \alpha N) p_{\Delta \text{R00}=2}(n_v, \alpha N) \times 10^{-12}$$

These last probabilities are on the order of $10^{-12}$ and change the time scale from picoseconds to seconds. We obtain a random walk, but only on a limited space of two points. However, vertices on the triangle tiling can proceed by phason flips, as first stated by Kalugin and Katz [5]. Thus we may conjecture that the two-point space can be extended and that Edagawa phasons perform a random walk on a quasilattice.

3. Summary

Brownian motion of a pollen is induced by fluctuations in collective molecular motions. It is a random walk in continuous threedimensional space. The time scales differ by nine orders of magnitude. Edagawa phasons are induced by fluctuations in atomic simpleton flips. The analysis of the cluster dynamics shows that they behave as a random walk on a quasilattice. The time scales of white spot flips and atomistic simpleton flips differ by 12 orders in magnitude.

References

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