

Correction of the proof of detailed balance in Wanjura *et al.*, Granular Matter **22**, 91 (2020)

Two aims to this document. One is to show that the theoretical proof by Wanjura *et al.* [1] is limited to very dense systems with highest cell order at most 5. The other is to demonstrate that the steady states of such systems *cannot* support any cycle and therefore must satisfy detailed balance (DB) by default.

To substantiate these claim, consider first the steady states of systems with only 3- and 4-cells. In such systems only one process is possible: $A \equiv 3 + 3 \rightleftharpoons 4$. Using the evolution equations (3) and (6) in [1], the steady state is governed by the equations

$$\begin{aligned}\dot{Q}_3 &= 0 = (Q_3 - 2) \eta_{3,3} \\ \dot{Q}_4 &= 0 = (Q_4 + 1) \eta_{3,3} ,\end{aligned}\tag{1}$$

with $\eta_{3,3} = p_{3,3}Q_3^2 - q_{4,3}Q_4$. These equations are dependent because they satisfy automatically the normalisation condition, $Q_3 + Q_4 = 1$. This leaves one equation with one unknown – $\eta_{3,3}$. Since at least one of these fractions must be finite, the condition for the steady state is $\eta_{3,3} = 0$. Since the entire dynamics consists of only possible process, $3 + 3 \rightleftharpoons 4$, and this process must be balanced by virtue of the state being steady, no cycle is possible and these systems satisfy DB by default.

Next, consider systems with highest order 5. In these systems, the cell orders evolve only via two processes: $A \equiv 3+3 \rightleftharpoons 4$, with $\eta_{3,3}$ as above, and $B \equiv 3+4 \rightleftharpoons 5$, with $\eta_{3,4} = p_{3,4}Q_3Q_4 - q_{4,3}Q_5$. Using again eqs. (3) in reference [1], the evolution equation at steady state reduce to

$$\begin{aligned}\dot{Q}_3 &= 0 = (Q_3 - 2) \eta_{3,3} + (Q_3 - 1) \eta_{3,4} \\ \dot{Q}_4 &= 0 = (Q_4 + 1) \eta_{3,3} + (Q_4 - 1) \eta_{3,4} \\ \dot{Q}_5 &= 0 = Q_5 \eta_{3,3} + (Q_5 + 1) \eta_{3,4} .\end{aligned}\tag{2}$$

Here, too, the equations are dependent because of the normalisation condition, which leaves us with two equations for the two unknowns $\eta_{3,3}$ and $\eta_{3,4}$. The solution gives the ratio of these unknowns:

$$\eta_{3,4} = \frac{2 - Q_3}{1 - Q_3} \eta_{3,3} = -\frac{1 + Q_4}{1 - Q_4} \eta_{3,3} .\tag{3}$$

Since at least one of the fractions must be finite we get $\eta_{3,4} = \eta_{3,3} = 0$. In such systems, there is only one process leading to and from 5-cells, $3 + 4 \rightleftharpoons 5$ then the 5-cells cannot be party to any cycle and this process must itself be balanced. Since what is left is only the one other process, $3 + 3 \rightleftharpoons 4$, then it also must be balanced and these steady states also satisfy DB by default.

Finally, consider systems with highest order 6. Next, I show that In these systems one cannot prove DB, following the same analysis as above. In these systems, the cell orders evolve via four processes: $A \equiv 3 + 3 \rightleftharpoons 4$, with $\eta_{3,3}$ as above, $B \equiv 3 + 4 \rightleftharpoons 5$, with $\eta_{3,4}$ as above, $C \equiv 4 + 4 \rightleftharpoons 6$, with $\eta_{4,4} = p_{4,4}Q_4^2 - q_{6,4}Q_6$, and $D \equiv 3 + 5 \rightleftharpoons 6$, with $\eta_{3,5} = p_{3,5}Q_3Q_5 - q_{6,3}Q_6$. In these systems there are two processes affecting 6-cells, C and D , and therefore at least one cycle is possible. For

example, $A \rightarrow B \rightarrow D \rightarrow C \rightarrow A$. Indeed, in these systems, DB cannot be shown because there are four equations:

$$\begin{aligned}
 \dot{Q}_3 &= 0 = (Q_3 - 2) \eta_{3,3} + (Q_3 - 1) (\eta_{3,4} + \eta_{3,5}) \\
 \dot{Q}_4 &= 0 = (Q_4 - 2) \eta_{4,4} + (Q_4 + 1) \eta_{3,3} + (Q_4 - 1) \eta_{3,4} \\
 \dot{Q}_5 &= 0 = Q_5 (\eta_{3,3} + \eta_{4,4}) + (Q_5 + 1) \eta_{3,4} + (Q_5 - 1) \eta_{3,5} \\
 \dot{Q}_6 &= 0 = Q_6 (\eta_{3,3} + \eta_{3,4}) + (Q_6 + 1) (\eta_{4,4} + \eta_{3,5}) ,
 \end{aligned} \tag{4}$$

of which one is dependent because of the normalisation condition. This leaves three independent equations with four unknowns: $\eta_{3,3}, \eta_{3,4}, \eta_{4,4}$, and $\eta_{3,5}$. This system of equations is underdetermined and the best one can do is express three of the unknowns in terms of the fourth. It follows that there is an infinite family of finite solutions.

This establishes that the proof in [1] holds only for steady states of far-from-equilibrium granular systems that contain no cycle processes.

References

- [1] C.C. Wanjura, P. Gago, T. Matsushima, R.Blumenfeld, *Granular Matter* **22**, 1 (2020)