Electronic Supplementary Material (ESI) for Soft Matter. This journal is © The Royal Society of Chemistry 2023

Extended Data

for

Melting of a Macroscale Binary Coulombic Material

Sarah Battat¹, David A. Weitz^{1,2,3*}, George M. Whitesides^{4*}

¹ John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, United States

² Department of Physics, Harvard University, Cambridge, Massachusetts 02138, United States

³ Wyss Institute for Biologically Inspired Engineering, Harvard University, Boston, Massachusetts 02115, United States

⁴ Department of Chemistry and Chemical Biology, Harvard University, Cambridge, Massachusetts 02138, United States

* To whom correspondence should be addressed: weitz@seas.harvard.edu, gwhitesides@gmwgroup.harvard.edu



Figure S1. (left) Photograph of the humidity-controlled chamber in which the experiment is conducted. Three lights (not pictured) are mounted outside of the chamber to enhance the quality of the video. (**right**) Overhead image of the gold-coated Petri dish, the optically transparent lid, and the wire connection to electrical ground. The scale bar corresponds to two centimeters.



Figure S2. Video stills captured at select times depicting the process of melting a square composed of 200 nylon beads (blue), 200 PTFE beads (white), and one impurity (gold-coated nylon bead). The impurity is outlined in black in each frame. The crystal is mechanically agitated on an orbital shaker at a centripetal acceleration of $21.1 \text{ } m/s^2$. Square structures and hexagonally layered structures are outlined in red. The line in the second to last time panel corresponds to the reflection of the wire that connects the lid of the dish to the electrical ground. All scale bars correspond to one centimeter. The color of the images is altered for improved contrast. (inset) The shear transformation of a square lattice to a hexagonally layered structure is depicted schematically.



Figure S3. Video stills captured at select times depicting the process of melting a square composed of 200 nylon beads (blue), 200 PTFE beads (white), and 50 impurities (gold-coated nylon beads). The impurities are outlined in black in each frame. The crystal is mechanically agitated on an orbital shaker at a centripetal acceleration of $21.1 m/s^2$. Square structures and hexagonally layered structures are outlined in red. All scale bars correspond to one centimeter. The lines that appear in some time panels correspond to the reflection of the wire that connects the lid of the dish to the electrical ground. The color of the images is altered for improved contrast.



Figure S4. The average of the modulus of the *k*-atic order parameter, $\langle |\psi_k| \rangle$, where $k = \{2, 3, 4, 5, 6\}$, for all PTFE beads in each frame. The *k*-atic order parameter for the PTFE beads is measured relative to their PTFE neighbors. Video frames are only included in the analysis if at least 80% of the nylon beads and 90% of the PTFE beads are identified. The results correspond to the melting of a square crystal composed of 200 nylon beads and 200 PTFE beads (**A**) without impurities when agitated orbitally at a centripetal acceleration of $26 m/s^2$, (**B**) without impurities when agitated orbitally at a centripetal acceleration of $21.1 m/s^2$, (**C**) with one impurities when agitated orbitally at a centripetal acceleration of $21.1 m/s^2$. Each bolded trendline corresponds to the rolling average of the raw data, which is depicted in the corresponding lightened hue.



Figure S5. The fraction of nylon and PTFE beads at each time point whose modulus of the *k*-atic order parameter, $|\psi_k|$, is greatest. For instance, for any given bead, if the modulus of its 2-atic order parameter is greatest in comparison to the modulus of its 3-atic, 4-atic, 5-atic, or 6-atic order parameter, then $k_{max} = 2$ for that bead. Video frames are only included in the analysis if at least 80% of the nylon beads and 90% of the PTFE beads are identified. The results correspond to the melting of a square crystal composed of 200 nylon beads and 200 PTFE beads (**A**) without impurities when agitated orbitally at a centripetal acceleration of $21.1 \ m/s^2$, (**B**) without impurities when agitated orbitally at a centripetal acceleration of $21.1 \ m/s^2$, and (**D**) with 50 impurities when agitated orbitally at a centripetal acceleration of $21.1 \ m/s^2$. Each bolded trendline corresponds to the rolling average of the raw data, which is depicted in the corresponding lightened hue.



Figure S6. Plot of the average coordination number of the beads within the aggregate as a function of their distance from the center of mass of the aggregate at specific times. The coordination number of a bead corresponds to the number of beads, agnostic of their kind, within 1.2 bead diameters from the bead's center. The results correspond to the melting of a square crystal composed of 200 nylon beads and 200 PTFE beads (A) without impurities when agitated orbitally at a centripetal acceleration of $26 \ m/s^2$, (B) without impurities when agitated orbitally at a centripetal acceleration of $21.1 \ m/s^2$, (C) with one impurity when agitated orbitally at a centripetal acceleration of $21.1 \ m/s^2$.