

Evidence of dipolar magnetic interactions in the self-assembly of two-dimensional magnetic nanocubes:

Supplementary information

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Fig. S1 presents a typical zero-field-cool and field-cool (ZFC-FC) behavior of the sample with high nanoparticle concentration (in sub- 20 nm range).

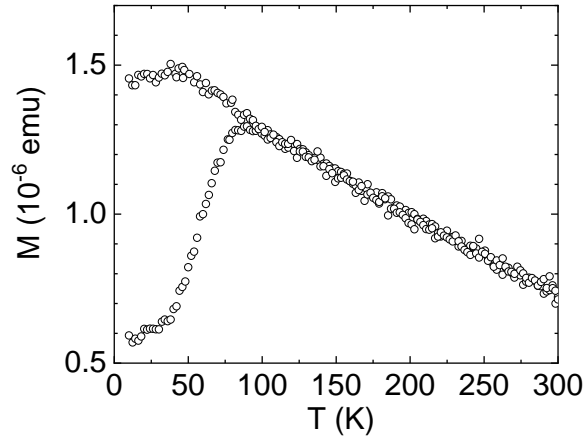


Fig. S1 Typical ZFC-FC behavior for a sample with high nanoparticle concentration.

Fig. S2 presents the cluster size distribution for all sub-threshold concentrations (corresponding to Figs. 2(a)–(d) of the main text).

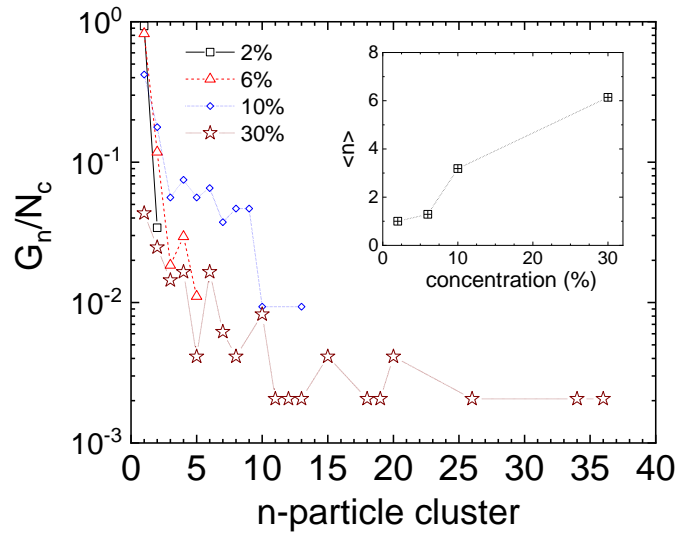


Fig. S2 Number of n-particle clusters with cubic particles G_n per total number of particles N_c , visualized from SEM images in Figs. 2(a)-(d) of the main text. The inset displays the average cluster size $\langle n \rangle$ as a function of concentration of particles c expressed in percent.

As evident from the SEM micrographs in Figs. 2(a)–(d) of the main text, the number of clusters containing n nanoparticles, denoted as G_n , decreases as cluster size increases across all samples. It is noteworthy that with increasing nanoparticle concentration, the average cluster size exhibits a clear upward trend (see corresponding inset) as estimated from: $\langle n \rangle = \sum nG_n / \sum G_n$.¹

In Figs. 6(a1)–(a4) of the main text we consider topography of magnetic nanocubes recorded in contact mode simultaneously during AFM manipulations at gradually increasing normal load and for four manipulation steps from 1 to 4. With a further increase in normal load during AFM manipulation, the chains nearly disappear, as shown in Figs. S3(a)–(c) displaying topographic images recorded during the three additional manipulation steps. The normal load in these three manipulation steps was varied within the following ranges: 6.50–7.80 nN in the fifth manipulation step (Fig. S3(a)), 8–9.40 nN in the sixth step (Fig. S3(b)), and 10.90–15.60 nN in the seventh step (Fig. S3(c)). Instead of chains, the nanoparticles tend to form compact clusters comprising just a few individual particles. Since the AFM measurements were performed in contact mode simultaneously with manipulation, the images in Fig. 7(a)–(c) lack the resolution necessary to clearly resolve individual nanocubes. Nevertheless, the observed clusters closely resemble those observed in the SEM images shown in Fig. 3(d) of the main text.

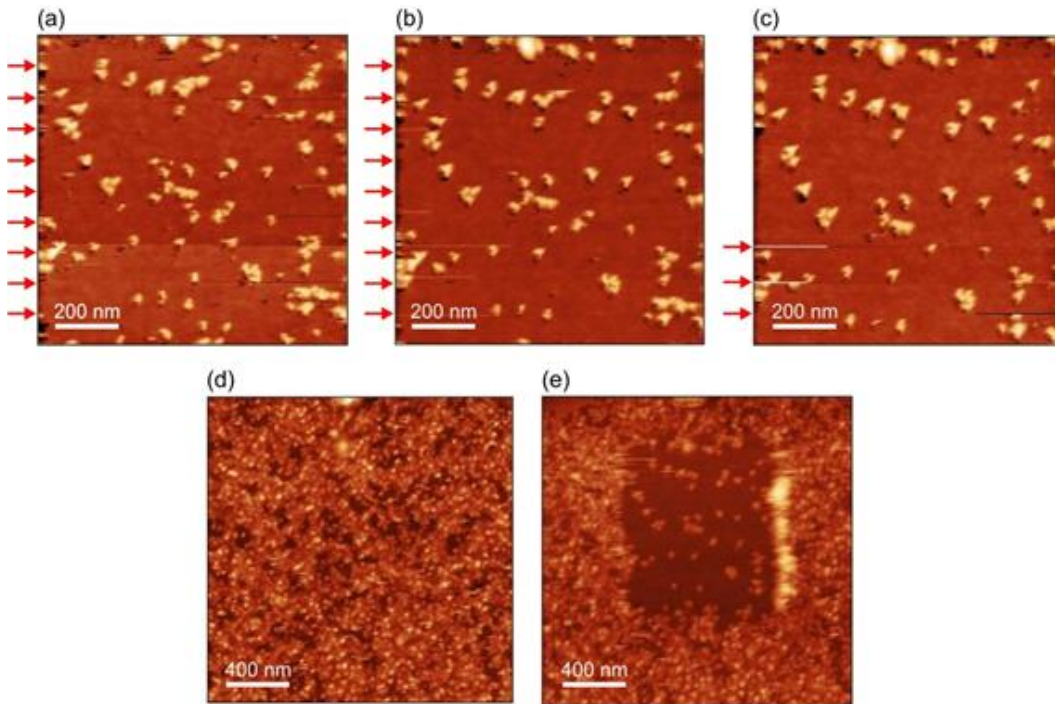


Fig. S3 Continuation of Figs. 6(a1)–6(a4) of the main text, illustrating three additional manipulation steps: (a)–(c) Topography (z-scale is 0–30 nm) of magnetic nanocubes recorded in contact mode simultaneously during AFM manipulations (the manipulation steps from 5 to 7, respectively). In each image, the normal force was incrementally increased from the bottom to the top (with the load held constant within each individual stripe which marked by the arrows). (d) The initial topographic image (z-scale is 0–45 nm) of the layer of magnetic nanocubes and (e) the final image (z-scale is 0–65 nm) recorded after all manipulation steps (1–7).

¹ D. Zablotsky, L.L. Rusevich, G. Zvejnieks, V. Kuzovkov, E. Kotomin, *Nanoscale*, 2019, **11**, 7293.

Figures S3(d) and S3(e) show the initial topography of the layer of magnetic nanocubes and the final image recorded after all seven manipulation steps, respectively. Both images were acquired in contact mode at a low normal force, ensuring stable imaging without undesired particle displacement. As seen, most of the nanocubes were removed from the central square region where the manipulation was performed and they were redeposited along the right edge of this region. Since scanning began at the bottom-left corner, with the forward scan in the direction from the left to the right edge, the nanocubes were dominantly displaced during the forward scan. Notably, the left, bottom, and top edges of the square region appear irregular rather than straight, further indicating reorganization of the nanocubes due to their mutual interactions after the AFM tip passes over them.