Supplementary Information: Directional self-assembly of magnetic nanotubes in quasi two dimensional layers

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I. ENERGY OF A REGULAR SQUARE LATTICE OF DIPOLAR NANOCUBES

We include here the details of the calculation to determine the energy of an arbitrarily sized regular square lattice of dipolar nanocubes. To recap, the lattice structures observed in simulation adopted configurations with four distinct dipole orientations. In order to keep track of the repeating patterns, we associated each of these dipoles with a symbol ($\bullet$, $\times$, $\triangle$, $\circ$); the identifications and the locations of the dipoles within the lattice were as follows,

$$\left.\begin{array}{c}\mathbf{m}_{\bullet}\\ \mathbf{m}_{\times}\\ \mathbf{m}_{\triangle}\\ \mathbf{m}_{\circ}\end{array}\right\} = \frac{|\mathbf{m}|}{\sqrt{3}}\begin{cases}(-1,1,1) & x,\ y \text{ even or } 0,\\ (1,1,-1) & x \text{ even or } 0;\ y \text{ odd},\\ (1,-1,1) & x,\ y \text{ odd},\\ -(1,1,1) & x \text{ odd};\ y \text{ even or } 0.\end{cases} \tag{1}$$

Noting that $\mathbf{m}_{\bullet}$ was placed at the origin purely for convenience. Consider FIG.1, a schematic of the system; this figure should be kept in mind throughout the remainder of the derivation.

To begin, we define the position of two arbitrarily chosen dipoles in the lattice by the following vectors,

$$\mathbf{r}_1(i,j) = hi\hat{\mathbf{x}} + hj\hat{\mathbf{y}}, \qquad \mathbf{r}_2(i',j') = hi'\hat{\mathbf{x}} + hj'\hat{\mathbf{y}}. \tag{2}$$

The corresponding coordinates are $h(i,j)$ and $h(i',j')$ respectively, with $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ denoting unit vectors in the x and y direction. The length of the cube side is denoted h and for a simple square lattice is the value of the lattice spacing. The resulting displacement vector between the dipoles is $\mathbf{r}(i,j,i',j') = \mathbf{r}_2 - \mathbf{r}_1$, with a corresponding magnitude of $|\mathbf{r}(i,j,i',j')| = h\sqrt{(i'-i)^2 + (j'-j)^2}$. The dipole interaction between two arbitrary position in the lattice is

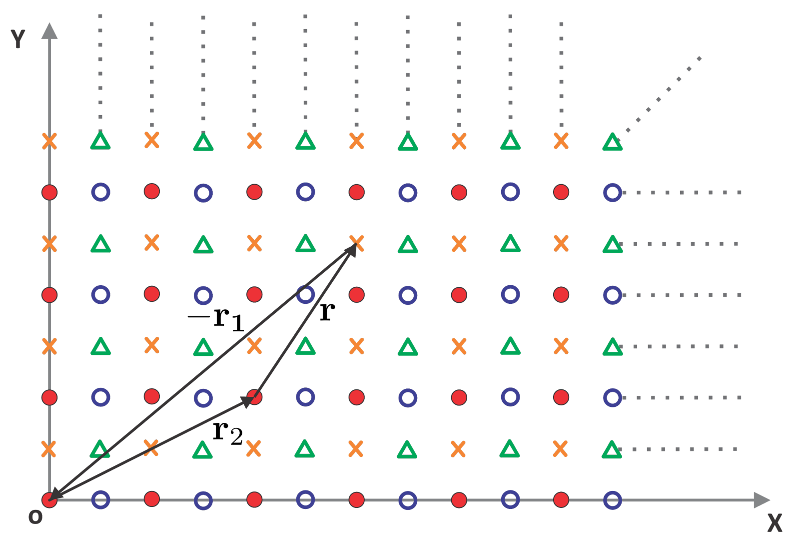

FIG. 1. Symbolic representation of the dipolar lattice structure. The position vectors of two arbitrary particles are given as $\mathbf{r}_1$ and $\mathbf{r}_2$; the displacement between the two is denoted $\mathbf{r}$. The lattice spacing is equivalent to h, the length of the cube side. The grey dashed lines indicate that the size of the lattice is unconstrained.

expressed in terms of a single fraction as,

$$U(i,j,i',j') = -\frac{\mu_0}{4\pi}\left[\frac{3(\mathbf{m}\cdot\mathbf{r})(\mathbf{m}'\cdot\mathbf{r}) - h^2[(i'-i)^2+(j'-j)^2]\mathbf{m}\cdot\mathbf{m}'}{h^5[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}}\right]. \tag{3}$$

We need to sum this interaction in an appropriate manner over a square lattice of size (n_1, n_2), and in the process account for each particle and the four different dipole orientations.

As stated in the manuscript, the number of possible dipole pair interaction types is limited to ten (including those between like dipoles). As such, the energy of the lattice is comprised of the following ten separate terms,

$$u_{lat}(n_1,n_2) = \frac{u_{\bullet\bullet}+u_{\times\times}+u_{\circ\circ}+u_{\triangle\triangle}+u_{\bullet\times}+u_{\bullet\triangle}+u_{\bullet\circ}+u_{\times\triangle}+u_{\times\circ}+u_{\triangle\circ}}{n_1 n_2}. \tag{4}$$

To determine the form of these components, we can begin by deriving the form of the pair interactions for each dipole combination by substitution of the relevant dipole orientations from eqn(1) into eqn(3). Below are the expressions for each combination, along side the conditions on each set of dipole coordinates according to their classification:

$$v_{\bullet\bullet}(i,j,i',j') = \frac{6\sqrt{3}(i'-i)(j'-j)}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,j,i',j' \text{ even or } 0, \tag{5}$$

$$v_{\times\times}(i,j,i',j') = \frac{-6\sqrt{3}(i'-i)(j'-j)}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,i' \text{ even or } 0 : j,j' \text{ odd}, \tag{6}$$

$$v_{\triangle\triangle}(i,j,i',j') = \frac{6\sqrt{3}(i'-i)(j'-j)}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,j,i',j' \text{ odd}, \tag{7}$$

$$v_{\circ\circ}(i,j,i',j') = \frac{-6\sqrt{3}(i'-i)(j'-j)}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,i'\text{odd} : j,j' \text{ even or } 0, \tag{8}$$

$$v_{\bullet\times}(i,j,i',j') = 2\sqrt{3}\frac{(i'-i)^2-2(j'-j)^2}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,j,i' \text{ even or } 0 : j' \text{ odd}, \tag{9}$$

$$v_{\bullet\triangle}(i,j,i',j') = 2\sqrt{3}\frac{(i'-i)^2+(j'-j)^2-3(i'-i)(j'-j)}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,j \text{ even or } 0 : i'j' \text{ odd}, \tag{10}$$

$$v_{\bullet\circ}(i,j,i',j') = 2\sqrt{3}\frac{(j'-j)^2-2(i'-i)^2}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,j,j' \text{ even or } 0 : i' \text{ odd}, \tag{11}$$

$$v_{\times\triangle}(i,j,i',j') = 2\sqrt{3}\frac{(j'-j)^2-2(i'-i)^2}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i \text{ even or } 0 : j,i',j' \text{ odd}, \tag{12}$$

$$v_{\times\circ}(i,j,i',j') = 2\sqrt{3}\frac{(i'-i)^2+(j'-j)^2+3(i'-i)(j'-j)}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad i,j'\text{even or } 0 : j,i' \text{ odd}, \tag{13}$$

$$v_{\triangle\circ}(i,j,i',j') = 2\sqrt{3}\frac{(i'-i)^2-2(j'-j)^2}{[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}} \quad j' \text{ even or } 0 : i,j,i' \text{ odd}. \tag{14}$$

Using this construction, we can consider the complete lattice as a set of ten sub-lattices, each with the defining pair interaction just described.

A reassignment of variables can be use to explicitly enforce the conditions on $(i,\ j,\ i',\ j')$. It follows that the condition for coordinates being even or zero can be attained by substitution of, for example, $i \equiv 2i$. Similarly for coordinates that are odd, the equivalent substitution is, for example, $i \equiv 2i+1$. If we consider the first interaction term $v_{\bullet\bullet}$, the appropriate change of coordinates would be $(i,\ j,\ i',\ j') \equiv (2i,\ 2j,\ 2i',\ 2j')$, and so on for each. Applying this change of variables, we arrive at the following for each interaction:

$$v_{\bullet\bullet}(2i,2j,2i',2j') = \frac{3\sqrt{3}(i'-i)(j'-j)}{4[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}}, \tag{15}$$

$$v_{\times\times}(2i,2j+1,2i',2j'+1) = \frac{-3\sqrt{3}(i'-i)(j'-j)}{4[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}}, \tag{16}$$

$$v_{\triangle\triangle}(2i+1,2j+1,2i'+1,2j'+1) = \frac{3\sqrt{3}(i'-i)(j'-j)}{4[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}}, \tag{17}$$

$$v_{\circ\circ}(2i+1,2j,2i'+1,2j') = \frac{-3\sqrt{3}(i'-i)(j'-j)}{4[(i'-i)^2+(j'-j)^2]^{\frac{5}{2}}}, \tag{18}$$

$$v_{\bullet\times}(2i,2j,2i',2j'+1) = 4\sqrt{3}\frac{2(i'-i)^2-(2(j'-j)+1)^2}{[(i'-i)^2+(2(j'-j)+1)^2]^{\frac{5}{2}}}, \tag{19}$$

$$v_{\bullet\triangle}(2i,2j,2i'+1,2j'+1) = 2\sqrt{3}\frac{(2(i'-i)+1)^2+(2(j'-j)+1)^2-3(2(i'-i)+1)(2(j'-j)+1)}{[(2(i'-i)+1)^2+(2(j'-j)+1)^2]^{\frac{5}{2}}}, \tag{20}$$

$$v_{\bullet\circ}(2i,2j,2i'+1,2j') = 4\sqrt{3}\frac{2(j'-j)^2-(2(i'-i)+1)^2}{[(2(i'-i)+1)^2+4(j'-j)^2]^{\frac{5}{2}}}, \tag{21}$$

$$v_{\times\triangle}(2i,2j+1,2i'+1,2j'+1) = 4\sqrt{3}\frac{2(j'-j)^2-(2(i'-i)+1)^2}{[(2(i'-i)+1)^2+4(j'-j)^2]^{\frac{5}{2}}}, \tag{22}$$

$$v_{\times\circ}(2i,2j+1,2i'+1,2j') = 2\sqrt{3}\frac{(2(i'-i)+1)^2+(2(j'-j)-1)^2-3(2(i'-i)+1)(2(j'-j)-1)}{[(2(i'-i)+1)^2+(2(j'-j)-1)^2]^{\frac{5}{2}}}, \tag{23}$$

$$v_{\triangle\circ}(2i+1,2j+1,2i'+1,2j') = 4\sqrt{3}\frac{2(i'-i)^2-(2(j'-j)-1)^2}{[4(i'-i)^2+(2(j'-j)-1)^2]^{\frac{5}{2}}}. \tag{24}$$

To determine the energy of each interaction lattice, an appropriate summation scheme is required. The size of the composite lattices vary for a given total lattice size of (n_1, n_2). This is accounted for in the upper summation limits, which are determined by the odd or even conditions on $(i,\ j,\ i',\ j')$. For example, an even condition on i has the corresponding limit of $\left[\frac{n_1-1}{2}\right]$, where the $[\ldots]$ denote that the integer part of the expression is used. In a similar manner, an even condition on j yields $\left[\frac{n_2-1}{2}\right]$. For coordinates with an odd condition the limits for x and y are $\left[\frac{n_1}{2}-1\right]$ and $\left[\frac{n_2}{2}-1\right]$ respectively. The sub-lattices consisting of like dipoles provide a convenient simplification to the calculation: each term individually has a zero net energy contribution to the lattice. We find that, regardless of cluster size, the individual sub-lattices of like dipoles combine in a manner where any contribution from a single pair interaction is cancelled out by another. The remaining six interactions between differing dipoles have the form,

$$u_{\bullet\times}(n_1,n_2) = \sum_{i=0}^{[\frac{n_1-1}{2}]}\sum_{j=0}^{[\frac{n_2-1}{2}]}\sum_{i'=0}^{[\frac{n_1-1}{2}]}\sum_{j'=0}^{[\frac{n_2}{2}-1]} v_{\bullet\times}(2i,2j,2i',2j'+1), \tag{25}$$

$$u_{\bullet\triangle}(n_1,n_2) = \sum_{i=0}^{[\frac{n_1-1}{2}]}\sum_{j=0}^{[\frac{n_2-1}{2}]}\sum_{i'=0}^{[\frac{n_1}{2}-1]}\sum_{j'=0}^{[\frac{n_2}{2}-1]} v_{\bullet\triangle}(2i,2j,2i'+1,2j'+1), \tag{26}$$

$$\tag{27}$$

$$u_{\bullet\circ}(n_1,n_2) = \sum_{i=0}^{[\frac{n_1-1}{2}]} \sum_{j=0}^{[\frac{n_2-1}{2}]} \sum_{i'=0}^{[\frac{n_1}{2}-1]} \sum_{j'=0}^{[\frac{n_2-1}{2}]} v_{\bullet\circ}(2i,2j,2i'+1,2j'), \tag{28}$$

$$u_{\times\triangle}(n_1,n_2) = \sum_{i=0}^{[\frac{n_1-1}{2}]} \sum_{j=0}^{[\frac{n_2}{2}-1]} \sum_{i'=0}^{[\frac{n_1}{2}-1]} \sum_{j'=0}^{[\frac{n_2}{2}-1]} v_{\times\triangle}(2i,2j+1,2i'+1,2j'+1), \tag{29}$$

$$u_{\times\circ}(n_1,n_2) = \sum_{i=0}^{[\frac{n_1-1}{2}]} \sum_{j=0}^{[\frac{n_2}{2}-1]} \sum_{i'=0}^{[\frac{n_1}{2}-1]} \sum_{j'=0}^{[\frac{n_2-1}{2}]} v_{\times\circ}(2i,2j+1,2i'+1,2j'), \tag{30}$$

$$u_{\triangle\circ}(n_1,n_2) = \sum_{i=0}^{[\frac{n_1}{2}-1]} \sum_{j=0}^{[\frac{n_2}{2}-1]} \sum_{i'=0}^{[\frac{n_1}{2}-1]} \sum_{j'=0}^{[\frac{n_2-1}{2}]} v_{\triangle\circ}(2i+1,2j+1,2i'+1,2j'). \tag{31}$$

For these interaction, sub-lattices each have a non-zero contribution to the overall lattice energy. As such, we can state that the energy of a regular $(n_1 \times n_2)$ square dipole lattice, with this particular dipole configuration, can be expressed as the sum of the terms described by eqns(25-31). Finally we identify the total number of cubes in the lattice as $N = n_1 n_2$; therefore the energy of the lattice per particle can be denoted as,

$$u_{lat}(N) = \frac{u_{\bullet\times} + u_{\bullet\triangle} + u_{\bullet\circ} + u_{\times\triangle} + u_{\times\circ} + u_{\triangle\circ}}{N}. \tag{32}$$

To summarise, interactions between like dipoles contribute zero net influence on the dipole lattice energy. The cluster energy is determined only by interactions between dipoles of different orientations. It is important to note that this calculation is valid for lattices in a regular $n_1 \times n_2$ array. The contribution of additional dipoles within a cluster not in a regular lattice had to be incorporated separately.