A SYSTEM EXHIBITING TOROIDAL ORDER

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A two dimensional system of discs upon which a triangle of spins are mounted is shown to undergo a sequence of interesting phase transitions as the temperature is lowered. We are mainly concerned with the ‘solid’ phase in which bond orientational order but not positional order is long ranged. As the temperature is lowered in the ‘solid’ phase, the first phase transition involving the orientation or toroidal charge of the discs is into a ‘gauge toroid’ phase in which the product of a magnetic toroidal parameter and an orientation variable (for the discs) orders but due to a local gauge symmetry these variables themselves do not individually order. Finally, in the lowest temperature phase the gauge symmetry is broken and toroidal order and orientational order both develop. In the ‘gauge toroidal’ phase time reversal invariance is broken and in the lowest temperature phase inversion symmetry is also broken. In none of these phases is there long range order in any Fourier component of the average spin. A definition of the toroidal magnetic moment $T_i$ of the $i$th plaquette is proposed such that the magnetostatic interaction between plaquettes $i$ and $j$ is proportional to $T_i T_j$. Symmetry considerations are used to construct the magnetoelectric free energy and thereby to deduce which coefficients of the linear magnetoelectric tensor are allowed to be nonzero. In none of the phases does symmetry permit a spontaneous polarization.

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I. INTRODUCTION

The theoretical analysis of toroidal ordering in electric\cite{1,2} and magnetic\cite{1,4,5} systems has recently been investigated. Examples of such states in which plaquettes of spins assume a chiral configuration have been known for some time.\cite{6} However, it is widely believed that toroidal magnetic order should always be subservient to the primary order parameter, a Fourier component of the average spin. Here we address the possibility of defining a toroidal order parameter for a system in which it is the primary magnetic order parameter. The major problem is to identify a situation in which there is ferrotoroidicity but there is no nonzero Fourier component of average spin. In this paper we consider a two dimensional system of toroidal spin plaquettes, such as those shown in Fig. 1 which exhibits the desired behavior.

![Fig. 1: Toroidal plaquettes with 2, 3, or 4 moments.](image1)

II. THE MODEL

We consider a system of microscopic circular discs (confined to lie in the $x$-$y$ plane) which contain three spins in a triangular configuration as in the center panel of Fig. 1. Each spin has a large single ion anisotropy so that it is aligned either parallel or antiparallel to its local axis fixed in the plane of the disc, as shown in Fig. 2. The intrplaquette dipolar interactions are strong enough so that at temperatures of interest the spins in each plaquette come to thermal equilibrium in one of the two degenerate ground states as shown in Fig. 1. The magnetic dipole moment of each spin is mimicked by a pair of opposite charges ($\pm Q$), as shown in Fig. 2. If $r$ is the distance of the charges from the center of the plaquette, then the magnitude of the dipole moment is $p = 2Qr \sin \chi$. The orientation of the disc is defined by the angle $\phi$ between the $x$-direction and the dashed line fixed on the plaquette.

![Fig. 2: A plaquette with three spins in one of their dipolar ground states. Each magnetic dipole is represented by a pair of charges $\pm Q$ whose positions are fixed by the angle $\chi$.](image2)
electrostatic energy (which is proportional to the desired magnetic dipole-dipole energy) between the charges on the two plaquettes. This energy is a sum over terms of the form \((1/R)F_i(\phi_A, \phi_B, \Psi)\exp^{\sigma n}\), where \(z \equiv r/R\). The positions are functions of \(r \exp(\pm i\chi_i)\) and the symmetry of the plaquette indicates that the result can only involve factors of \(r^3 \exp(\pm 3i\chi_i)\). Also this term must be odd in both \(\chi_A\) and \(\chi_B\). Accordingly, the lowest order nonzero term is of order \(z^6/R^7\). So at this order

\[
V_{AB} = \Lambda(\phi_A, \phi_B, \Psi)Q_A \sin(3\chi_A)Q_B \sin(3\chi_B)r^6/R^7 ,
\]

(1)

where \(Q_A\) (\(Q_B\)) is the amplitude of the charges on plaquette \(A\) (\(B\)). We only need this for small \(\chi\), so

\[
V_{AB} = 9\Lambda(\phi_A, \phi_B, \Psi)Q_A\chi_AQ_B\chi_Br^6/R^7 ,
\]

\[
= \frac{9}{4R^7}m_A m_B r^4 \Lambda(\phi_A, \phi_B, \Psi) ,
\]

(2)

where \(m_i = 2Q_i r\chi_i\) is the \(i\)th magnetic moment. For the present situation it seems reasonable to define the toroidicity \(T_i\) of the \(i\)th plaquette such that

\[
V_{AB} = \Lambda(\phi_A, \phi_B, \Psi)T_A T_B / R^7 ,
\]

(3)

so that

\[
T_i = (3/2)m_i r^2 = 3Q_i r^3 .
\]

(4)

Note that this definition, unlike the standard one, has the property that the magnetostatic interaction energy between two toroids is proportional to product of their toroidal ‘strengths’ \(T_i\). To get \(\Lambda(\phi_A, \phi_B, \Psi)\) we may consider the lowest order terms in the expansion of \(V_{AB}\) in powers of \(\chi_A\) and \(\chi_B\). Thus

\[
\Lambda(\phi_A, \phi_B, \Psi) = \frac{R^7}{9QAQB} \frac{d^2}{d\chi_A d\chi_B} \frac{d^6}{d\phi_A d\phi_B} V_{\chi_A=\chi_B=r=0} .
\]

(5)

The phase space of this model is specified as follows. Each plaquette is characterized by its center of mass position \(R\) inside a two dimensional box in the \(x-y\) plane with respect to which the plaquette has mirror symmetry. The \(i\)th plaquette can assume an orientation specified by \(\phi_i\) and has a toroidal strength \(T_i\) which is an Ising-like variable: \(T_i = (\pm 3/2)|m_i r^2|\), according to which of the two ground states the spins occupy. We assume an orientationally independent interaction between plaquettes which for concreteness we take to be the Lennard-Jones potential,

\[
W_{AB} = 4\epsilon \left( \frac{\sigma}{R} \right)^{12} - \left( \frac{\sigma}{R} \right)^6 ,
\]

(6)

where \(\sigma\) and \(\epsilon\) are constants. Because the system is two dimensional, the plaquettes can not develop long range order characteristic of a three dimensional solid. Instead the system can develop various behaviors intermediate between a conventional solid and an isotropic liquid.

As shown in Ref. [10] the system exhibits a ‘solid’ phase in which bond-orientational order is truly long range but position correlations exhibit power-law decay. The ‘solid’ melts either directly or indirectly (via a hexatic phase) into an isotropic liquid phase. Here we focus on the transition as the temperature is lowered through the value \(T_1\) at which the ‘solid’ phase appears and assume that \(kT_1\) is much larger than the magnetic interactions between plaquettes. Even in the ‘solid’ phase, there is no true long range positional order. Then the spin correlation function will involve an average over correlations between a spin at the origin and spins in a distant plaquette. Since the average spin in a plaquette is zero, the spin correlation function, like the positional correlation function, can not display long range order. In contrast, because there is long-range bond orientational order, lowering the temperature can lead to phase transitions due to the interplaquette dipolar interactions. It is the purpose of this paper to analyze the symmetry of the resulting ordered phases. In a likely scenario we find that the ‘solid’ undergoes two such phase transitions. At the first (higher temperature) transition we find that time reversal symmetry is broken and at the second spatial inversion symmetry is also broken. Since the underlying system does not have long range positional ordering, the spin correlation function itself is never long ranged. To substantiate this picture it is necessary to analyze the interplaquette interactions and thereby verify that they lead to Ising-like transitions.

A. Interplaquette Interaction

To calculate the interaction between two plaquettes, each of which is confined to the \(x-y\) plane, we assume (see Fig. 2) that plaquette A has charges \(\sigma_A Q_A\) where \(\sigma_A = \pm 1\), at (positions relative to the center of the plaquette)

\[
r(n_A, \sigma_A) = [x(n_A, \sigma_A), y(n_A, \sigma_A)]
\]

(7)

where

\[
x(n_A, \sigma_A) = r \cos(\sigma_A \chi_A + 2n_A r \pi/3 + \phi_A)
\]

\[
= [r \cos(\chi_A) \cos(2n_A r \pi/3 + \phi_A) - \sigma_A r \sin(\chi_A) \sin(2n_A r \pi/3 + \phi_A)] ,
\]

(8)

\[
y(n_A, \sigma_A) = r \sin(\sigma_A \chi_A + 2n_A r \pi/3 + \phi_A)
\]

\[
= \sigma_A r \sin(\chi_A) \cos(2n_A r \pi/3 + \phi_A) + r \cos(\chi_A) \sin(2n_A r \pi/3 + \phi_A) \]

(9)

for \(n_A = 1, 2, 3\) and similarly for plaquette B.

Then the interaction energy between the two plaquettes is

\[
V_{AB} = Q_A Q_B \sum_{n_A, n_B=1}^{3} \sum_{\sigma_A \sigma_B = \pm 1} \sigma_A \sigma_B \left[ R^2 + 2R\xi [x(n_A, \sigma_A) - x(n_B, \sigma_B)] \right]
\]
where $\xi \equiv \cos(\Psi) = X/R$, and $\eta \equiv \sin \Psi = Y/R$. We drop the last term in $r^2$ since it cannot contribute to a term involving $\sin(3\chi_A)\sin(3\chi_B)$. We want to expand this in powers of $z$ and now we work to first order in $\chi_A$ and $\chi_B$ by setting $\sin(\chi_X) = \chi_X$ and $\cos(\chi_X) = 1$, where $X$ is either $A$ or $B$. Thus

$$V_{AB} = \frac{Q_A Q_B}{R} \sum_{n_a, n_B = 1}^{3} \sum_{\sigma_A, \sigma_B = \pm 1} \sigma_A \sigma_B \mathcal{R}^{-1/2},$$

(11)

where

$$\mathcal{R} = 1 + 2\xi z \left[ \cos(2n_A \pi/3 + \phi_A) - \cos(2n_B \pi/3 + \phi_B) \right]
- \sigma_A \chi_A \sin(2n_A \pi/3 + \phi_A) + \sigma_B \chi_B \sin(2n_B \pi/3 + \phi_B)
+ 2\eta a \left[ \sigma_A \chi_A \cos(2n_A \pi/3 + \phi_A) + \sin(2n_A \pi/3 + \phi_A) \right]
- \sigma_B \chi_B \cos(2n_B \pi/3 + \phi_B)
+ \sin(2n_A \pi/3 + \phi_A) - \sin(2n_B \pi/3 + \phi_B)
- 2\xi a \left[ \cos(2n_A \pi/3 + \phi_A) - \sigma_A \chi_A \sin(2n_A \pi/3 + \phi_A) \right]
\times \left[ \cos(2n_B \pi/3 + \phi_B) - \sigma_B \chi_B \sin(2n_B \pi/3 + \phi_B) \right]
- 2\xi a \left[ \sigma_A \chi_A \cos(2n_A \pi/3 + \phi_A) + \sin(2n_A \pi/3 + \phi_A) \right]
\times \left( \sigma_B \chi_B \cos(2n_B \pi/3 + \phi_B) + \sin(2n_B \pi/3 + \phi_B) \right).

(12)

We now analyze how $V_{AB}$ depends on $\Psi$, i.e., how it depends on $\xi$ and $\eta$. Note that $\xi$ and $\eta$ each carry a factor of $z$. However, factors that do not depend on $\eta$ and $\xi$ carry factors of 1 or $z^2$, so that can be no terms at order $z^6$ which are cubic in $\eta$ or $\xi$. Therefore $V_{AB}$ can only depend on $\Psi$ through the argument $6\Psi$. In addition, for $V_{AB}$ to be invariant under a global rotation, it can only depend on differences in angle. Also the result can only be a function of $3\phi_A$ and $3\phi_B$ because it must be invariant under $\phi_i \to \phi_i + 2\pi/3$. Thus $\Lambda$ must be of the form

$$\Lambda(\phi_A, \phi_B; \Psi) = a + b \cos(6\Psi - 3\phi_A - 3\phi_B)
+ c \cos(3\phi_A - 3\phi_B)
+ d \sin(6\Psi - 3\phi_A - 3\phi_B)
+ e \sin(3\phi_A - 3\phi_B).$$

(13)

One can view the system looking either along the positive $z$ axis or along the negative $z$ axis. Comparing these two views, one sees that a positive rotation is equivalent to changing the sign of the charges followed by a negative rotation. But the interaction energy is invariant under charge conjugation. So

$$\Lambda(\phi_A, \phi_B, \Psi) = a + b \cos(6\Psi - 3\phi_A - 3\phi_B)
+ c \cos(3\phi_A - 3\phi_B).$$

(14)

Furthermore, consider what happens if we average the interaction over the orientation $\phi_i$ of one of the plaquettes. This superposition of charges leads to a uniformly charge-neutral plaquette. This argument tells us that $a$ in Eq. (14) must be zero. To identify the coefficients $b$ and $c$ it suffices to evaluate

$$\Lambda(0, 0, \Psi) = b \cos(6\Psi) + c.$$ 

(15)

The explicit evaluation of $\Lambda(0, 0, \Psi)$ is carried out in the Appendix. Then use of Eq. (14) implies the result

$$\Lambda = \frac{10, 395}{32} \cos(6\Psi - 3\phi_A - 3\phi_B)
- \frac{225}{32} \cos(3\phi_A - 3\phi_B).$$

(16)

It is important to note a local symmetry. With the interactions so far postulated, the Hamiltonian is invariant under the local transformation

$$Q_i \to -Q_i, \quad \phi_i \to \phi_i + \pi.$$ 

(17)

This is a nontrivial symmetry that indicates that the two configurations shown in Fig. 3 have the same energy at leading order in the multipole expansion. Note that changing the sign of $Q_i$ is equivalent to changing the sign of the toroidal moment $T_i$. As a result of this local gauge symmetry it follows from Elitzur’s theorem that even though there is long range order in the variable $Q \sin(3\phi)$, there is no long range order in either $Q$ or $\sin(3\phi)$.

### III. PHASE TRANSITIONS OF THIS MODEL

Here we give a more detailed analysis of the phase transitions within this model. We assume that the magnetic anisotropy energy that aligns the spins along a fixed direction in each plaquette and the isotropic interactions of Eq. (3) between plaquettes are dominant. These assumed interactions do not depend on the orientation of either of the interacting plaquettes. (With this assumption Elitzur’s theorem applies.) Accordingly, as the temperature is lowered, this two-dimensional system will undergo a phase transition at a temperature $T_f$ into a ‘solid’ phase with long-range bond-orientational order, but no long range positional order. It is obvious that in this phase both spatial inversion and time reversal symmetries are maintained.

As one further reduces the temperature, the interplaquette dipolar interactions come into play and can cause
order to develop consistent with the local gauge symmetry. To see what sort of order develops we introduce the appropriate gauge invariant variables

\[ X_i = T_i \cos(3\phi_i), \quad Y_i = T_i \sin(3\phi_i). \]  

Because bond orientational order is maintained, we can treat each molecule as being surrounded by a hexagon of neighboring plaquettes and the orientation of this hexagon of neighbors is maintained over the entire system. Accordingly, we can define \( \Psi \) as being measured relative to the direction between the central plaquette and one of its neighbors. So we may take \( 6\Psi/(2\pi) \) to be an integer for all nearest neighbor interactions. We do not consider further neighbor interactions in view of how rapidly the interplaquette interaction falls off with separation. For simplicity we work as if we have a two dimensional triangular lattice. Thus we analyze the model with orientationally-dependent interactions

\[ V_{AB} = \frac{10,170 T_A T_B}{32 R^7} \left[ \cos(3\phi_A) \cos(3\phi_B) - \frac{10,620 T_A T_B}{32 R^7} \sin(3\phi_A) \sin(3\phi_B) \right]. \]  

So we have a two dimensional anisotropic rotor model which is in the same universality class as the two dimensional Ising model. Such models have been widely studied.\(^\text{12,13}\) To analyze the phase transitions within this model we invoke mean-field theory, within which the Landau free energy, \( \mathcal{F} \), in terms of the Fourier transforms of \( X_i \) and \( Y_i \) (temporarily assuming a triangular lattice of lattice constant \( a \)) assumes the form

\[ \mathcal{F} = \frac{1}{2} \sum_q \left( [ckT + \mu(q)]X(q)X((q)^* \right. \]

\[ +\left. [ckT + \nu(q)]Y(q)Y((q)^*) \right), \]  

at quadratic order, where \( c \) is a constant of order unity, and \( \mu(q) \) and \( \nu(q) \) are the Fourier transforms of the potential:

\[ \mu(q) = 2A[\cos(aq_x) + 2 \cos(aq_x/2) \cos(\sqrt{3}aq_y/2)] \]

\[ \nu(q) = 2B[\cos(aq_x) + 2 \cos(aq_x/2) \cos(\sqrt{3}aq_y/2)], \]  

where

\[ A = \frac{10,170 T^2}{32 R^7}, \quad B = -\frac{10,620 T^2}{32 R^7}, \]  

where \( T = (3/2)mr^2 \). Thus as the temperature is lowered the system will develop long range “ferro” order (order at zero wave vector) in the variable \( Y = T \sin(3\phi) \), but not, as was said, in either \( T \) or \( \sin(3\phi) \) separately. This type of order is illustrated in Fig. [4]. This transition occurs at a temperature of order \( kT_{II} = 6|B| \approx 2000T^2/R^7 \).

We now discuss whether time reversal (T) symmetry or spatial inversion (P) symmetry is broken in this phase. As a preliminary, note that the single particle density matrix assigns the probabilities \( p/2, p/2, \) and \( 1 - p \) to the states \( S, S', \) and \( S'' \), respectively, where \( S \) is the state of the top-most plaquette in Fig. [4], \( S' \) is the state of a plaquette in the bottom row of Fig. [4] and \( S'' \) is the
completely disordered state. The interpretation of this density matrix is shown in Fig. 3, where we see that time reversal symmetry is broken but inversion symmetry is maintained. (If the dipole were electric dipoles, then inversion symmetry would be broken.)

\[ V' = v \cos(6\Psi - 3\phi_A - 3\phi_B) \]  
\[ V' = -v \sin(3\phi_A) \sin(3\phi_B) \]

Now we again set \( 6\Psi/2\pi \) to be an integer and we only need to consider interactions involving \( \sin(3\phi) \), so effectively

\[ V' = v \cos(6\Psi - 3\phi_A - 3\phi_B) \]  
\[ V' = -v \sin(3\phi_A) \sin(3\phi_B) \]

Now what happens depends on the sign of \( v \). If \( v \) is positive, then we have a ferro arrangement of plaquettes, so that all plaquettes are in the same state (either as those in the bottom row of Fig. 3 or as that in the top row of Fig. 4). Because toroidicity and orientation are strongly coupled, this state is ferrotoroidal. If \( v \) is negative, then we have an antiferro arrangement of plaquettes into the so-called ‘root-3’ structure discussed recently in connection with charge ordering in lutetium ferrite. In this state we have antiferrotoroidicity. Here we are mainly interested in displaying a ferro state, so we take \( v \) to be positive. This final ordering transition which breaks local gauge symmetry will occur at a temperature of order \( kT_{II} = 6v \), which we assume to be much smaller than \( kT_I \). This transition is also in the same universality class as the two dimensional Ising model. At this transition spatial inversion symmetry is broken. The symmetry of the various phases is summarized in Fig. 6.

\[ \begin{align*}
&\text{new long-range order} \quad \text{bond orientation} \\
&\begin{array}{c|c|c|c}
\text{symmetry} & \text{PT} & \text{P} & \Theta P T \\
\hline
\text{phase} & \text{ferro-toroid} & \text{gauge toroid} & \text{nonmagnetic} & \text{hexatic or isotropic} \\
\hline
\text{I} & \text{I} & \text{I} & \text{I} & \text{I} \\
\text{II} & \text{II} & \text{II} & \text{II} & \text{II} \\
\text{III} & \text{III} & \text{III} & \text{III} & \text{III} \\
\end{array}
\end{align*} \]

FIG. 6: Symmetry of the various phases. Here the symmetries are \( T = \) time reversal symmetry, \( P = \) spatial inversion symmetry, and \( \Theta = \) continuous rotational symmetry.

In Ref. 5 toroidicity has been discussed in connection with the magnetoelectric effect. However, instead of using the symmetry of the crystal (see Ref. 16), they used the symmetry of free space to obtain a simplified relation between the toroidicity and the linear magnetoelectric tensor. Here we invoke the symmetry of the two dimensional system to write the magnetoelectric free energy as a function of the electric field \( E \) and the magnetic field \( H \) as

\[ F_{ME} = T_z \left( A[H_x E_y - H_y E_x] + BH_x E_z \\
+ C[H_x E_x + H_y E_y] \right), \]

where \( T_z \) is the \( z \)-component of the toroidicity, \( T \). We may check that this form is consistent with the symmetry of the system, keeping in mind that spin and the magnetic field are both pseudo vectors but \( T \) is a real vector. Accordingly, both \( T_z \) and the factor in the large brackets are odd under the mirror \( z \rightarrow -z \). Also all the terms are invariant under a rotation about the \( z \) axis. Thus

\[ \begin{align*}
- \frac{\partial^2 F}{\partial H_x \partial E_y} &= \frac{\partial M_x}{\partial E_y} = \frac{\partial P_y}{\partial H_x} = -A \\
- \frac{\partial^2 F}{\partial H_y \partial E_x} &= \frac{\partial M_y}{\partial E_x} = \frac{\partial P_x}{\partial H_y} = A \\
- \frac{\partial^2 F}{\partial H_z \partial E_z} &= \frac{\partial M_z}{\partial E_z} = \frac{\partial P_z}{\partial H_z} = -B \\
- \frac{\partial^2 F}{\partial H_x \partial E_x} &= \frac{\partial M_x}{\partial E_x} = \frac{\partial P_x}{\partial H_x} = -C \\
- \frac{\partial^2 F}{\partial H_y \partial E_y} &= \frac{\partial M_y}{\partial E_y} = \frac{\partial P_y}{\partial H_y} = -C.
\end{align*} \]
These elements of the linear magnetoelectric tensor are only nonzero in the ferrotoroidal phase. The combination of the three-fold axis and the \(x-y\) reflection plane guarantee that the spontaneous polarization is zero in all these phases. (If the discs were asymmetric with respect to this mirror, then a spontaneous polarization along \(z\) would be allowed in the ferrotoroidal phase\(^{12}\).)

IV. DISCUSSION AND CONCLUSION

The nonmagnetic ‘solid’ to gauge toroid transition at \(T_{1A}\) was analyzed assuming no gauge breaking interactions. We argue that the introduction of the small gauge breaking interaction will not qualitatively modify the phase diagram of Fig. 6 because it will take a finite interaction to orientationally order the plaquettes. Similarly, including higher order terms in the multipole expansion will not alter our conclusions as long as \(z = r/R \ll 1\). One possible difficulty in constructing the system of discs analyzed in this paper is that it may be difficult to achieve equilibrium statistics within the manifold of the dipolar spin ground states. But perhaps it is not crucial that the gap between the two spin ground states and the qualitatively different excited spin states is very large. It would, of course, be extremely interesting to observe the linear magnetoelectric effect in a system such as this.

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Appendix A: Evaluation of \(\Lambda(\phi, \phi_B, \Psi)\)

Write

\[
V_{AB} = \frac{4QA_Q R}{3 \sum_{A,B} \sum_{A,B} \sigma A \sigma B} \left[ 1 + Az + B_z \sigma A \chi A + B_z \sigma B \chi B + C_1 z^2 \sigma A \chi A + C_2 z^2 \sigma B \chi B + Dz^2 \sigma A \sigma B \chi A \chi B + Ez^2 \right]^{-1/2}.
\]

Here

\[
A = 2 \xi [\cos(2n_A \pi /3 + \phi_A) \cos(2n_B \pi /3 + \phi_B)]
+ 2 \eta [\sin(2n_A \pi /3 + \phi_A) - \sin(2n_B \pi /3 + \phi_B)]
B_1 = -2 \xi sin(2n_A \pi /3 + \phi_A) + 2 \eta \cos(2n_A \pi /3 + \phi_A)
B_2 = 2 \xi \sin(2n_B \pi /3 + \phi_B) - 2 \eta \cos(2n_B \pi /3 + \phi_B)
C_1 = 2 \sin(2n_A \pi /3 + \phi_A) \cos(2n_B \pi /3 + \phi_B)
- 2 \cos(2n_A \pi /3 + \phi_A) \sin(2n_B \pi /3 + \phi_B)
C_2 = 2 \cos(2n_A \pi /3 + \phi_A) \sin(2n_B \pi /3 + \phi_B)
- 2 \sin(2n_A \pi /3 + \phi_A) \cos(2n_B \pi /3 + \phi_B)
D = -2 \sin(2n_A \pi /3 + \phi_A) \sin(2n_B \pi /3 + \phi_B)
- 2 \cos(2n_A \pi /3 + \phi_A) \cos(2n_B \pi /3 + \phi_B)
E = -2 \cos(2n_A \pi /3 + \phi_A) \cos(2n_B \pi /3 + \phi_B)
- 2 \sin(2n_A \pi /3 + \phi_A) \sin(2n_B \pi /3 + \phi_B) = D.
\]

Thus, when the sums over \(\sigma_A\) and \(\sigma_B\) are performed, we have

\[
V_{AB} = \frac{4QA_Q R}{3 \sum_{A,B} \sum_{A,B} \sigma A \sigma B} \left[ 1 + Az + B_z \sigma A \chi A + B_z \sigma B \chi B + C_1 z^2 \sigma A \chi A + C_2 z^2 \sigma B \chi B + Dz^2 \sigma A \sigma B \chi A \chi B + Ez^2 \right]^{-1/2}
\]

\[
\times \left[ 1 + Az + Dz^2 + \frac{3[B_1 B_2 z^2 + (B_2 - B_1) C_1 z^3 - C_2 z^4]}{4(1 + Az + Dz^2)^5/2} \right].
\]

Thus when the sums over \(\sigma_A\) and \(\sigma_B\) are performed, we have

\[
\Lambda(\phi_A, \phi_B, \Psi) = \frac{4}{9(6!)} \frac{\partial^6}{\partial z^6} \left[ E \right]_{z=0} \sum_{n_A, n_B} \sum_{n_A, n_B} \left[ -\frac{3(-3/2!)}{2(-7/2!)(2!)} - \frac{3(-3/2!)}{2(-9/2!)(1!)(2!)} \right]
\]

\[
\times \left[ -\frac{1}{2(1 + Az + Dz^2)^5/2} + \frac{3[B_1 B_2 z^2 + (B_2 - B_1) C_1 z^3 - C_2 z^4]}{4(1 + Az + Dz^2)^5/2} \right].
\]

Thus

\[
\Lambda = \frac{4}{9} \sum_{n_A, n_B} \sum_{n_A, n_B} \left[ -\frac{3(-3/2!)}{2(-7/2!)(2!)} - \frac{3(-3/2!)}{2(-9/2!)(1!)(2!)} \right]
\]

\[
- \frac{1}{2(1 + Az + Dz^2)^5/2} + \frac{3[B_1 B_2 z^2 + (B_2 - B_1) C_1 z^3 - C_2 z^4]}{4(1 + Az + Dz^2)^5/2} \right]
\]

\[
\times \left[ -\frac{1}{2(1 + Az + Dz^2)^5/2} + \frac{3[B_1 B_2 z^2 + (B_2 - B_1) C_1 z^3 - C_2 z^4]}{4(1 + Az + Dz^2)^5/2} \right].
\]

\[
\sum_{n=1}^{12} \Lambda_n,
\]

where we number the terms with \((B_2 - B_1)\) separately.

To implement Eq. 15 we write

\[
\Lambda(0, 0, \Psi) = (4/9) [\Lambda_6(0, 0, \Psi) + \Delta \Lambda(0, 0, \Psi)]
\]

(A6)
where $\Lambda_6$ is the term in $\Lambda$ proportional to $B_1 B_2 A^4$ and
\[ \Delta \Lambda \text{ contains the remaining terms written in Eq. (A5).} \]
Note that $\Lambda_6$ is the only term which is sixth order in $\xi$ and $\eta$ and is therefore the only term which can contribute to the term in Eq. (14) involving $6 \Psi$. Thus we write
\[ \Lambda_6 = \alpha + \beta \cos(6 \Psi - 3 \phi_A - 3 \phi_B) + \gamma \cos(3 \phi_A - 3 \phi_B) \quad (A7) \]
and $\Delta \Lambda$ is a constant which we can evaluate by setting $\Psi = 0$.
\[ \Delta \Lambda = \alpha' + c' \cos(3 \phi_A - 3 \phi_B) . \quad (A8) \]
We first evaluate $\Lambda_6$ for $\phi_A = \phi_B = 0$ with $\Psi$ arbitrary. It is convenient to introduce the notation $(x, y)$, where $x$ and $y$ assume the values 1, $c$, or $s$, for unity, the cosine function and the sine function. The first argument is that for plaquette A and the second is that for plaquette B. Thus $(c, cs) = \cos(2n_A \pi/3) \cos(2n_B \pi/3) \sin(2n_B \pi/3)$ and $(1, c^2) \equiv \cos^2(2n_B \pi/3)$. We have the sums over $n$:
\[
\begin{align*}
\sum_n \cos(2n \pi/3) &= \sum_n \sin(2n \pi/3) \cos^k(2n \pi/3) = 0 \\
\sum_n \sin^2(2n \pi/3) &= \sum_n \cos^2(2n \pi/3) = 3/2 \\
\sum_n \cos^3(2n \pi/3) &= -\sum_n \sin^2(2n \pi/3) \cos(2n \pi/3) = 3/4 \\
\sum_n \cos^2(2n \pi/3) \sin^2(2n \pi/3) &= 3/8 \\
\sum_n \cos^4(2n \pi/3) &= \sum_n \sin^4(2n \pi/3) = 9/8 . \quad (A9)
\end{align*}
\]
With these understandings (and with the sums over $n_A$ and $n_B$ implied) we write
\[ S_6 = \sum_{n_A,n_B=1}^3 B_1 B_2 A^4 , \quad (A10) \]
so that
\[
\begin{align*}
S_6 &= -64[\xi(s, 1) - \eta(c, 1)][\xi(1, s) - \eta(1, c)] \\
&\quad \times \left( \xi[(c, 1) - (1, c)] + \eta[(s, 1) - (1, s)] \right)^4 \\
&= -64[\xi^2(s, s) + \eta^2(c, c) - \xi \eta(s, c) - \xi \eta(c, s)] \\
&\quad \times \left( \xi[(c, 1) - (1, c)] + \eta[(s, 1) - (1, s)] \right)^4 \\
&= -64[\xi^2(s, s)] \left( \eta^4[-4(s^3, s) - 4(s, s^3)] \\
&\quad - 12 \eta^2 \xi^2(s, s)[(c, 1) - (1, c)]^2 \\
&\quad - 64 \eta^2(c, c) \left( \xi^4[(c, 1) - (1, c)]^4 + 6 \xi^2 \eta^2[(c, 1) - (1, c)]^2[(s^2, 1) + (1, s^2)] + \eta^4[(1, s) - (1, s)]^4 \right) \\
&\quad + 64 \xi \eta(s, c) \left( 4 \xi^3 \eta[(c, 1) - (1, c)]^3(s, 1) \\
&\quad + 4 \eta^3 \xi[(c, 1) - (1, c)][(s^3, 1) + 3(s, s^2)] \right) \\
&\quad + 64 \xi \eta(c, s) \left( -4 \xi^3 \eta[(c, 1) - (1, c)]^3(1, s) \right) \\
&\quad - 4 \eta^3 \xi[(c, 1) - (1, c)][(1, s^3) + 3(s, s^2)] \right)
\end{align*}
\]
When simplified this is
\[ S_6 = \eta^6[-64(c, s)[(c, s^2), (s, s^2)] + 512 \eta^4 \xi^2(s^4, s^2) \\
- 768 \eta^4 \xi^2[-2(c^2 s^2, c^2) + (cs, c^3)] \\
+ 512 \eta^4 \xi^2[-(s^4, c^2) + 3(s^2 c^2, c^2) - 3(s^2, s^2 c^2)] \\
+ 768 \eta^2 \xi^4[(s^2 c^2, s^2) - 2(s^2 c, s^2 c) + (s^4, s^2 c^2)] \\
- 64 \eta^2 \xi^4[-4(c, c^3) + 6(c^3, c^3) - 4(c^2, c^4)] \\
+ 512 \eta^2 \xi^4[-3(s^2 c^2, c^2) + 3(s^2 c, c^3) - (s^2, c^4)] \\
= \eta^6[-384 \left[ \frac{3}{4} \right]^2 + 512 \eta^4 \xi^2 \left[ \frac{9}{8} \frac{3}{2} \right] \\
- 768 \eta^4 \xi^2 \left[ -2 \left[ \frac{3}{8} \frac{3}{2} \right] + \left[ \frac{3}{4} \frac{3}{4} \right] \right) \\
+ 512 \eta^4 \xi^2 \left[ - \left[ \frac{9}{8} \frac{3}{2} \right] + 3 \left[ \frac{3}{4} \frac{3}{4} \right] - 3 \left[ \frac{3}{2} \frac{3}{2} \right] \right) \\
+ 768 \eta^2 \xi^4 \left[ \left[ \frac{3}{8} \frac{3}{2} \right] - 2 \left[ \frac{3}{4} \frac{3}{4} \right] + \left[ \frac{3}{2} \frac{3}{2} \right] \right) \\
- 64 \eta^2 \xi^4 \left[ -4 \cdot 2 \left[ \frac{9}{8} \frac{3}{2} \right] + 6 \left[ \frac{3}{4} \frac{3}{4} \right] \right) \
\]
\[ + 512\eta^2\xi^4 \left( -3 \left[ \frac{3}{8} \right] \left[ \frac{3}{2} \right] + 3 \left[ \frac{3}{4} \right] \left[ \frac{3}{2} \right] - \left[ \frac{3}{2} \right] \left[ \frac{3}{8} \right] \right) \]
\[ = -216\eta^6 + \eta^4\xi^2[1296] - \eta^2\xi^4[1944], \quad (A12) \]

This is
\[ S_6 = \left( -108(\xi^2 + \eta^2)^3 \right. \]
\[ -108(\eta^6 - 15\eta^4\xi^2 + 15\eta^2\xi^4 - \xi^6) \right) \]
\[ = 108[\cos(6\Psi) - 1]. \quad (A13) \]

This leads to the result
\[ \Lambda_6(\phi_A, \phi_B, \Psi) = \frac{93,555}{128} \left[ -\cos(3\phi_A - 3\phi_B) \right. \]
\[ + \cos(6\Psi - 3\phi_A - 3\phi_B) \right]. \quad (A14) \]

We now evaluate the other sums. But since \( \Delta A \) does not depend on \( \Psi \), we set \( \xi = 1 \) and \( \eta = 0 \). Then we have (again the sums over \( n_A \) and \( n_B \) are left implicit)
\[ \sum D^3 \equiv S_1 = -8[(s, s) + (c, c)]^3 \]
\[ = -8[3(s^3c^2, s^2c) + (c^3, c^3)] \]
\[ = -8 \left( 3 \left[ \frac{3}{4} \right] \left[ \frac{3}{2} \right] + \left[ \frac{3}{2} \right] \right) = -18. \quad (A15) \]

If \( S_2 \equiv \sum D^2A^2 \), then, with the sums implicit, we have
\[ S_2 = 16[(s^2, s^2) + 2(sc, sc) + (c^2, c^2)] \]
\[ \times [(c, 1) - (1, c)]^2 \]
\[ = 16[(s^2c^2, s^2) + (c^2, c^2) - 2(s^2c, s^2c) - 2(c^3, c^3)] \]
\[ + (s^2, s^2c^2) + (c^2, c^2)] \]
\[ = 32 \left( \left[ \frac{3}{8} \right] \left[ \frac{3}{2} \right] + \left[ \frac{9}{16} \right] \left[ \frac{3}{2} \right] - \left[ \frac{3}{4} \right]^2 \right) \]
\[ = 36. \quad (A16) \]

If \( S_3 \equiv \sum D^4A^4 \), then, with the sums implicit, we have
\[ S_3 = 32[\xi(s, s) - (c)(c)][(c, 1) - (1, c)]^4 \]
\[ = 64[2(c^4, c^2) - 3(c^3, c^3) + 2(c^2, c^4)] \]
\[ = 64 \left( \left[ \frac{9}{8} \right] \left[ \frac{3}{2} \right] - 3 \left[ \frac{3}{4} \right]^2 \right) \]
\[ = 324. \quad (A17) \]

If \( S_4 \equiv \sum B_1B_2D^2 \), then, with the sums implicit, we have
\[ S_4 = -16(s, s)[(s, s) + (c, c)]^2 \]
\[ = -32(s^2c, s^2c) = -32 \left[ \frac{3}{4} \right] \left[ \frac{3}{2} \right] = -18. \quad (A18) \]

If \( S_5 \equiv \sum B_1B_2D^2A^2 \), then, with the sums implicit, we have
\[ S_5 = 32(s, s)[(s, s) + (c, c)][(c, 1) - (1, c)]^2 \]
\[ = 32(s^2, s^2)[(c^4, 1) - 2(c, c) + (1, c^3)] \]
\[ = 32[(s^2c^2, s^2) - 2(c^2, c^2) + (s^2, s^2c^2)] \]
\[ = 64 \left( \left[ \frac{3}{8} \right] \left[ \frac{3}{2} \right] - \left[ \frac{3}{4} \right]^2 \right) \quad (A19) \]

If \( S_7 \equiv \sum B_2C_1DA \), then, with the sums implicit, we have
\[ S_7 = -16[(s, s) - (c, c)][(c, c) + (s, s)][(c, 1) - (1, c)] \]
\[ = 16[-(s, sc) + (c, s^2)][(c, c) + (s, s)][(c, 1) - (1, c)] \]
\[ = 16[(s^2, s^2c) + (c^2, cs^2)][(c, 1) - (1, c)] \]
\[ = 16[(s^2c, s^2c) + (c^3, cs^2) + (s^2, s^2c^2) - (c^2, c^2s^2)] \]
\[ = 16 \left( \left[ \frac{3}{8} \right] + \left[ \frac{9}{16} \right] \left[ \frac{3}{4} \right] - \left[ \frac{3}{4} \right]^2 \right) = -18. \quad (A20) \]

\[ \sum B_1C_1DA \equiv S_8 = -16[(s, c) - (c, s)][(c, s) + (s, c)][(c, 1) - (1, c)] \]
\[ = -\sum B_2C_1DA \quad (A21) \]

If \( S_9 \equiv \sum B_2C_1A^3 \), then, with the sums implicit, we have
\[ S_9 = 32[(s, c) - (c, s)][(c, 1) - (1, c)] \]
\[ = -32(c^3, c^2) + 3(c^2, cs^2) + 3(c^2, c^2s^2)] \]
\[ = -32 \left( \left[ \frac{9}{8} \right] \left[ \frac{3}{2} \right] - 3 \left[ \frac{3}{4} \right] \left[ \frac{3}{2} \right] + 3 \left[ \frac{3}{2} \right] \left[ \frac{3}{8} \right] \right) \]
\[ = -162. \quad (A22) \]

\[ \sum B_1C_1A^3 \equiv S_{10} = 32[\xi(s, 1) - \eta(c, 1)][(s, c) - (c, s)] \]
\[ \times \left( \xi[(c, 1) + (1, c)] + \eta[(s, 1) + (1, s)] \right)^3 \]
\[ = -\sum B_2C_1A^3 \quad (A23) \]

If \( S_{11} \equiv \sum C_1^2D \), then, with the sums implicit, we have
\[ S_{11} = -8[(s, c) - (c, s)][(c, c) + (s, s)] \]
\[ = -8(s^2, s^2c) + (c^2, cs)(c, c) + 16(cs, cs)(s, s) \]
\[ = -8 \left( \left[ \frac{3}{2} \right] \left[ \frac{3}{4} \right] - 2 \left[ \frac{3}{4} \right]^2 \right) = 18. \quad (A24) \]

If \( S_{12} \equiv \sum C_1^2A^2 \), then, with the sums implicit, we have
\[ S_{12} = 16[(s, c) - (c, s)][(c, 1) - (1, c)]^2 \]
\[ = 16[(s^2, c^2) + (c^2, s^2)][(s^2, 1) - 2(c, c) + (1, c^3)] \]
\[
\begin{align*}
32\left[ (s^2c^2, c^2) + (c^4, c^2) - (s^2c, c^3) - (c^3, s^2c) \right] \\
= 32 \left( \frac{3}{8} + \frac{3}{2} + \frac{3}{8} - 2 \left[ \frac{3}{4} \right] \right) \\
= 108.
\end{align*}
\]

Thus

\[
\sum_n \Lambda_n(0, 0, \Psi) = \frac{135}{8} + \frac{945}{8} - \frac{25,515}{64} \\
\frac{945}{16} + \Lambda_6(0, 0, \Psi) - \frac{945}{8}.
\]

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