Orientation states of dipoles on 2D Bravais lattices

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The condition that the absolute values of the 2*D* dipole moments remain constant on arbitrary plane Bravais lattices, with an arbitrary anisotropic interaction, substantially restricts the types of periodic configurations of dipole moments which could occur in the ground state (uniform structures with a doubled or quadrupled lattice constant and polydomain periodic structures). This condition also plays an important role in the analysis of the stability of the ground states with respect to thermodynamic fluctuations. The energies and configurations of the dipoles in the ground state on arbitrary 2*D* Bravais lattices are calculated by separating the dipole-dipole interactions into intrachain and interchain parts. The frequencies of the orientation oscillations which are active in the IR absorption spectra are determined. Approximate expressions are derived for the temperatures of orientation phase transitions. These approximations become asymptotically exact in the limit of weak interactions between Ising chains of dipoles.

1. INTRODUCTION

An anisotropic interaction can lead to complex configurations of dipoles in the ground state. Calculations of these configurations are reported in Refs. 1–10 for plane lattices—square, triangular, rectangular, and rhombic. It was proved in Ref. 11 that orientational long-range order prevails in a system of dipoles on arbitrary 2*D* Bravais lattices, except square lattices (in which case a phase with short-range order occurs). That proof provides substantial motivation for calculations of the ground states. If thermodynamic fluctuations were able to disrupt the long-range order at some arbitrarily low (but nonzero) temperature, such calculations would be of purely methodological interest. The primary results of the analysis below are a determination of the characteristics of the ground state of dipoles on an arbitrary 2*D* Bravais lattice and the derivation of correct estimates of the temperatures of the phase transitions to these states.

Our solution of this problem is based on three subproblems, which are examined in the following three sections of this paper. The first subproblem is analyzing the limitations imposed on the types of periodic configurations of dipoles with an arbitrary interaction in the ground state by the condition that the absolute values of the dipole moments remain constant. It turns out that these limitations are substantial, and they select a narrow class of configurations for further analysis.

The second subproblem is constructing a convenient method for evaluating the dipole interactions. This method would make it possible to work with analytic expressions for an arbitrary 2*D* Bravais lattice with adequate accuracy. This method is based on Van der Hoff and Benson's idea[12] of a chain representation of the interactions. That method was used in Ref. 8 for the first analytic analysis of the ground states of dipoles on triangular and square lattices. That analysis (without the help of numerical simulations) revealed the reason for the ferroelectric and layered antiferroelectric (or, as a result of degeneracy, microvortex) ground states on these lattices.

This separation of strong intrachain interactions and weak interchain interactions is responsible for the formulation of the third subproblem—that of evaluating the phase-transition temperatures. It is this suggestion regarding the interactions which made it possible, in Refs. 13 and 14, to introduce a generalized self-consistent-field approximation which leads to asymptotically exact results for a 2*D* Ising model in which the constants of the interactions between nearest neighbors along the axes of the square lattice are very different. In the present paper we generalize that approximation to systems with a long-range dipole interaction. This approximation makes it possible to explain the low phase-transition temperatures indicated by the calculations in Ref. 11.

In the final section of this paper we discuss the results of this study. We also analyze the results of Refs. 10 and 15, where the existence of a long-range order in certain specific dipole systems and also in arbitrary dipole systems was questioned.

2. STRUCTURE OF THE GROUND STATES OF PERIODIC CONFIGURATIONS OF 2D UNIT VECTORS ON 2D BRAVAIS LATTICES

We consider a system of 2*D* unit vectors $\mathbf{e}_\mathbf{r} = (\cos\theta_\mathbf{r}, \sin\theta_\mathbf{r})$ on an arbitrary 2*D* Bravais lattice ($\mathbf{r} = n_1\mathbf{a}_1 + n_2\mathbf{a}_2$, where $\mathbf{a}_1$, $\mathbf{a}_2$ are the fundamental lattice vectors, and n_1, n_2 are integers). The interaction between these unit vectors is specified by an arbitrary tensor $V^{\alpha\beta}(\mathbf{r}) = V^{\alpha\beta}(-\mathbf{r}) = V^{\beta\alpha}(\mathbf{r})$:

$$H = \frac{1}{2}\sum_{\mathbf{r},\mathbf{r}'} V^{\alpha\beta}(\mathbf{r}-\mathbf{r}')e_\mathbf{r}^{\alpha}e_{\mathbf{r}'}^{\beta} \tag{1}$$

(a repeated Greek index $\alpha,\beta = x,y$ means a summation). We go over to the Fourier representation in the wave vector $\mathbf{k}$:

$$\tilde{V}^{\alpha\beta}(\mathbf{k}) = \sum_\mathbf{r} V^{\alpha\beta}(\mathbf{r})\cos(\mathbf{kr}), \quad \tilde{e}^{\alpha}(\mathbf{k}) = N^{-1}\sum_\mathbf{r} e_\mathbf{r}^{\alpha}\exp(-i\mathbf{kr}). \tag{2}$$

In this representation, the Hamiltonian (1) becomes

$$H = \frac{1}{2}N\sum_\mathbf{k} \tilde{V}^{\alpha\beta}(\mathbf{k})\tilde{e}^{\alpha}(\mathbf{k})\tilde{e}^{\beta}(-\mathbf{k})$$
$$= \frac{1}{2}N\sum_{\mathbf{k}j} \tilde{V}_j(\mathbf{k})\,|\tilde{\mathbf{e}}(\mathbf{k})\tilde{\xi}_j(\mathbf{k})|^2, \tag{3}$$

where $\tilde{V}_j(\mathbf{k})$ and $\tilde{\xi}_j(\mathbf{k})$ are real eigenvalues and vectors of

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the tensor $\tilde{V}^{\alpha\beta}(\mathbf{k})$:

$$\tilde{V}^{\alpha\beta}(\mathbf{k})\tilde{\xi}_j^{\beta}(\mathbf{k})=\tilde{V}_j(\mathbf{k})\tilde{\xi}_j^{\alpha}(\mathbf{k}),$$
$$\tilde{\xi}_j^{\alpha}(\mathbf{k})\tilde{\xi}_{j'}^{\alpha}(-\mathbf{k})=\delta_{jj'}, \quad j=1, 2. \tag{4}$$

Without any loss of generality we can assume

$$\tilde{V}_1(\mathbf{k})\leqslant\tilde{V}_2(\mathbf{k}), \quad \min\tilde{V}_1(\mathbf{k})=\tilde{V}_1(\mathbf{k}_l)\equiv\tilde{V}_1, \quad l=1, 2, \ldots, L. \tag{5}$$

It is easy to see that the vector

$$\tilde{\mathbf{e}}(\mathbf{k})=\sum_{l=1}^{L}\delta_{\mathbf{k},\mathbf{k}_l}C_{1l}\tilde{\xi}_1(\mathbf{k}_l), \qquad \sum_{l=1}^{L}C_{1l}^2=1 \tag{6}$$

corresponds to the minimum value of the Hamiltonian (3),

$$H_{min}={}^1/_2N\tilde{V}_1, \tag{7}$$

and satisfies the condition

$$\frac{1}{N}\sum_{\mathbf{r}}\mathbf{e}_{\mathbf{r}}^2=\sum_{\mathbf{k}}\tilde{\mathbf{e}}(\mathbf{k})\tilde{\mathbf{e}}(-\mathbf{k})=1, \tag{8}$$

which underlies the spherical model.[16] Nevertheless, if the vector (6) is to satisfy the stronger condition of the uniqueness of $|\mathbf{e}_{\mathbf{r}}|$ at each lattice site, it is necessary to impose the following equality:

$$\sum_{\mathbf{k}\neq\mathbf{k}'}\tilde{\mathbf{e}}(\mathbf{k})\tilde{\mathbf{e}}(-\mathbf{k}')\exp[i(\mathbf{k}-\mathbf{k}')\mathbf{r}]=0. \tag{9}$$

Identical minimum values $\tilde{V}_1(\mathbf{k})$ could be achieved for realistic interactions only at several points of the first Brillouin zone, $\mathbf{k}=\mathbf{k}_l$, which transform into each other under the lattice-symmetry operations. If there are only two such points for the $2D$ lattices under consideration here, i.e., if there are only $\mathbf{k}=\pm\mathbf{k}_1\neq\mathbf{h}/2$ (i.e., $l=1,2$), where $\mathbf{h}=h_1\mathbf{b}_1+h_2\mathbf{b}_2$ is an arbitrary reciprocal-lattice vector ($\mathbf{a}_i\mathbf{b}_j=2\pi\delta_{ij}$, where h_1 and h_2 are integers), Eq. (9) becomes

$$\tilde{\mathbf{e}}^2(\mathbf{k}_1)\exp(2i\mathbf{k}_1\mathbf{r})+\tilde{\mathbf{e}}^2(-\mathbf{k}_1)\exp(-2i\mathbf{k}_1\mathbf{r})=0. \tag{10}$$

With $\mathbf{k}_1=\mathbf{h}/4$ we find from (10) the condition $\operatorname{Re}\tilde{\mathbf{e}}^2(\pm\mathbf{h}/4)=0$, which can easily be satisfied through the substitution

$$\tilde{\mathbf{e}}(\pm\mathbf{h}/4)=2^{-1/2}\exp(\pm i\pi/4)\tilde{\xi}_1(\mathbf{h}/4),$$

where $\tilde{\xi}_1(\mathbf{h}/4)$ is the single real eigenvector of $\tilde{V}^{\alpha\beta}(\mathbf{h}/4)$ in Eq. (4) [this representation was used in Refs. 3, 4, and 11 for complex vectors $\tilde{\mathbf{e}}(\pm\mathbf{h}/2)$]. In the case $\mathbf{k}_1\neq\mathbf{h}/2, \mathbf{h}/4$, condition (10) leads to $\tilde{\mathbf{e}}^2(\pm\mathbf{k}_1)=\tilde{\xi}_1^2(\pm\mathbf{k}_1)=0$ because of the arbitrary values of the exponential functions at each lattice site $\mathbf{r}$. For $2D$ vectors, this condition holds only if $\tilde{\xi}_1^x(\pm\mathbf{k}_1)=i\tilde{\xi}_1^y(\pm\mathbf{k}_1)$. Since the components of the tensor $\tilde{V}^{\alpha\beta}(\mathbf{k}_1)$ are real, however, the vectors $\tilde{\xi}_1(\pm\mathbf{k}_1)$ can always be chosen to be real, and the condition $\tilde{\xi}_1^x(\pm\mathbf{k}_1)=i\tilde{\xi}_1^y(\pm\mathbf{k}_1)$ is incompatible with (4) except in the special case in which the tensor $\tilde{V}^{\alpha\beta}(\mathbf{k})$ is isotropic at the point $\mathbf{k}=\pm\mathbf{k}_1$ [$\tilde{V}^{\alpha\beta}(\pm\mathbf{k}_1)=\tilde{V}_1\delta_{\alpha\beta}$].[1)] In this manner, it is shown that for realistic anisotropic interactions there cannot exist structures which correspond to only two harmonics, $\mathbf{k}=\pm\mathbf{k}_1$, with $\mathbf{k}_1\neq\mathbf{h}/2, \mathbf{h}/4$ and to the lowest energy of the system, (7). The structure of the ground state in this case will include some additional harmonics, which raise the total energy of the system [since the additional harmonics would in general correspond to $\tilde{V}_1(\mathbf{k})>\tilde{V}_1$]. The ground-state structure will have a periodic polydomain structure with sharp changes in the orientations of $\mathbf{e}_{\mathbf{r}}$ at the boundaries of domains.

The appearance of minima of $\tilde{V}_1(\mathbf{k})$ at the points $\mathbf{k}\neq\mathbf{h}/2$ ("accidental minima") is characteristic of systems with different and competing interactions (as in Ref. 17, for example, with a competition between exchange and dipole interactions). Direct calculations on the ground states of various dipole lattices have shown[1-10,18-20] that the structures of these states do indeed correspond to values $\mathbf{k}=\mathbf{h}/2$ for which the necessary condition of an extremum of $\tilde{V}_1(\mathbf{k})$ holds [for $\mathbf{k}=\mathbf{h}/2$ we find $d\tilde{V}^{\alpha\beta}(\mathbf{k})/d\mathbf{k}=0$ from (2)]. Accordingly, structures with $\mathbf{k}\neq\mathbf{h}/2$ will be ignored in the following sections of this paper, which are concerned exclusively with dipole-dipole interactions. At this point we would like to note that as the temperature rises the harmonics with $\mathbf{k}_1\neq\mathbf{h}/2, \mathbf{h}/4$, which are forbidden in the ground state, may arise as an intermediate phase, since the strong condition of the uniqueness of $|\mathbf{e}_{\mathbf{r}}|$ in the ground state is replaced by the inequality $|\langle\mathbf{e}_{\mathbf{r}}\rangle|<1$ for thermodynamic averages. A phenomenological theory of phase transitions to an incommensurate phase was examined in Refs. 17, 21, and 22. The thermodynamic stability of an incommensurate phase (and, in our case, of a phase with $\mathbf{k}_1\neq\mathbf{h}/2, \pm\mathbf{h}/4$) at specifically intermediate temperatures stems (on the one hand) from the inequality $\tilde{V}_1(\mathbf{k}_1)<\tilde{V}_1(\mathbf{h}/2)$ and (on the other) from the combinatorial increase in the coefficient of the fourth power of the order parameter in the Landau expansion for the phase with $\mathbf{k}_1$, with respect to the phase with $\mathbf{k}=\mathbf{h}/2$.

With this said, we restrict the analysis to ground-state structures with $\mathbf{k}=\mathbf{h}/2$. By virtue of the identity $f(\mathbf{k}+\mathbf{h})=f(\mathbf{k})$ [$f(\mathbf{k})$ is an arbitrary function], we need carry out the summation over only four wave vectors—$\mathbf{k}=0, \mathbf{b}_1/2, \mathbf{b}_2/2, (\mathbf{b}_1+\mathbf{b}_2)/2$—in order to determine the ground state in Hamiltonian (3). The minimization of H with respect to the eight variables embodied in the four real vectors $\tilde{\mathbf{e}}(\mathbf{k})$, should be carried out under the auxiliary condition (8) and under three conditions which follow from (9):

$$\sum_{\nu_1\neq\nu_2\neq\nu_3\neq\nu_4}[\tilde{\mathbf{e}}(\mathbf{k}_{\nu_1})\tilde{\mathbf{e}}(\mathbf{k}_{\nu_2})+\tilde{\mathbf{e}}(\mathbf{k}_{\nu_3})\tilde{\mathbf{e}}(\mathbf{k}_{\nu_4})]=0 \tag{11}$$

($\mathbf{k}$ takes on the four specified values). We are thus left with four independent variables, which correspond to the orientation angles $\theta_{\mathbf{r}}$ of the unit vectors $\mathbf{e}_{\mathbf{r}}$ in the four sublattices considered in the $2D$ analog[2] of the Luttinger–Tisza method.[18,19] It is thus not surprising that minimization[7] of a Hamiltonian by this method with respect to sixteen sublattices has yielded the same result as is found through a minimization with respect to four sublattices.

For realistic interactions (in particular, dipole-dipole interactions), the four values of $\tilde{V}_1(\mathbf{k})$ with the specified $\mathbf{k}=\mathbf{h}/2$ cannot be identical. Only two cases are possible, in which one of these values is the minimum, or (for lattices with symmetry axes of higher than twofold symmetry) the two values are equal, $\tilde{V}_1(\mathbf{b}_1/2)=\tilde{V}_1(\mathbf{b}_2/2)$. In the first of these cases, only the one vector $\tilde{\mathbf{e}}(\mathbf{k}_l)$ is nonzero, while in a second case, two vectors are nonzero: $\tilde{\mathbf{e}}(\mathbf{b}_1/2)$ and $\tilde{\mathbf{e}}(\mathbf{b}_2/2)$. By virtue of condition (11), these two vectors must be or-

thogonal. These arguments show that the Luttinger–Tisza method is overburdened with independent minimization variables: Analysis of the values of the Fourier components $\tilde{V}_1(\mathbf{k})$ immediately makes it possible to eliminate no less than half of the variables and to derive the result vastly more rapidly (compare this situation with, for example, the derivation of the ground state of a square lattice of dipoles by the Luttinger–Tisza method in Ref. 2 and by Fourier analysis in Refs. 3, 4, and 8). The ground state is degenerate both because the minimum values of $\tilde{V}_1(\mathbf{k})$ at two points on the boundary of the first Brillouin zone, $\mathbf{k}=\mathbf{b}_1/2$ and $\mathbf{k}=\mathbf{b}_2/2$, are equal and also because of the equality $\tilde{V}_1(\mathbf{k})=\tilde{V}_2(\mathbf{k})$ at one point, $\mathbf{k}=\mathbf{h}/2$. A natural consequence of the degeneracy of the ground state is the presence of a Goldstone mode in the spectrum of orientation oscillations.[20]

3. GROUND STATES OF 2D DIPOLE BRAVAIS LATTICES

We turn now to calculations on the ground states of $2D$ systems with a specific dipole–dipole interaction:

$$V^{\alpha\beta}(\mathbf{r})=VD^{\alpha\beta}(\mathbf{r}/a_1),\quad V=\mu^2/a_1^3,\quad a_1\leqslant a_2,$$
$$D^{\alpha\beta}(\mathbf{r})=\delta_{\alpha\beta}/r^3-3r_\alpha r_\beta/r^5 \tag{12}$$

(μ is the dipole moment). This interaction generalizes the results of Refs. 1–10 to arbitrary $2D$ Bravais lattices. For this purpose we need to select the smallest of the four eigenvalues $\tilde{V}_1(\mathbf{k})$, with $\mathbf{k}=0$, $\mathbf{b}_1/2$, $\mathbf{b}_2/2$, $(\mathbf{b}_1+\mathbf{b}_2)/2$, and we need to determine the orientation of the corresponding eigenvector (Sec. 2).

This selection process can be simplified substantially by noting that anisotropic dipole forces tend to orient the dipoles in the direction parallel to the line connecting the dipoles. The ground-state energy is dominated by the interaction between parallel dipole moments of the chain with the smallest distances between sites. We thus choose the lattice constants of the $2D$ Bravais lattice in the following way:

$$1=a_1\leqslant a_2\leqslant|\mathbf{a}_1-\mathbf{a}_2|\leqslant|\mathbf{a}_1+\mathbf{a}_2|. \tag{13}$$

Uniform configurations of the orientations of the dipoles should thus be realized within chains parallel to $\mathbf{a}_1$ (the x axis in Fig. 1), while uniform configurations in neighboring chains may alternate (along the y axis). These structures correspond to wave vectors $\mathbf{k}=0$ and $\mathbf{k}=\mathbf{b}_1/2$, and it is for these wave vectors that we need to calculate the values of $\tilde{V}_1(\mathbf{k})$. The analysis of the values of $\tilde{V}_1(\mathbf{k})$ at two other points, $\mathbf{k}=\mathbf{b}_2/2$ and $(\mathbf{b}_1+\mathbf{b}_2)/2$, carried out below, confirms the validity of this choice of the structures to be analyzed.

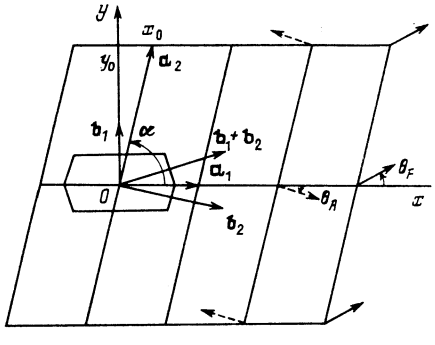

FIG. 1. Two-dimensional Bravais lattice with fundamental vectors $\mathbf{a}_1$, $\mathbf{a}_2$ and reciprocal-lattice vectors $\mathbf{b}_1$, $\mathbf{b}_2$. The solid and dashed arrows at the angles θ_F and θ_A specify the ferroelectric and antiferroelectric configurations of dipoles in the ground state.

Van der Hoff and Benson's method[12] for evaluating lattice sums in the chain representation was developed further in Refs. 23 and 24 for calculations of dipole sums and Lorentz factors of orthorhombic $3D$ lattices. The advantage of that method is that it singles out the intrachain interactions, which are expressed in terms of a zeta function $\zeta(p)$, and the interchain interactions are written as rapidly converging sums of modified Bessel functions $K_p(z)$. Explicit expressions for the interchain interactions for a uniform structure and for a periodic structure (with arbitrary $\mathbf{k}$) of orientations of dipoles are given in Refs. 8 and 25, respectively. Making use of those results, we can immediately write the Fourier components of the dimensionless dipole interaction tensor $\tilde{D}^{\alpha\beta}(\nu\mathbf{b}_1/2)$ $(\nu=0,1)$ for an arbitrary $2D$ Bravais lattice:

$$\tilde{D}^{xx}(\nu\mathbf{b}_1/2)=\tilde{D}_{ch}+(-1)^\nu\tilde{D}_{ch}^{int},\qquad \tilde{D}_{ch}=-4\zeta(3), \tag{14}$$

$$\tilde{D}_{ch}^{int}=32\pi^2\sum_{n,h=1}^{\infty}(-1)^{(n-1)\nu}h^2K_0(2\pi nhy_0)\cos(2\pi nhx_0)$$
$$\approx 16\pi^2y_0^{-1/2}\exp(-2\pi y_0)\cos(2\pi x_0),$$
$$\tilde{D}^{yy}(\nu\mathbf{b}_1/2)=2\zeta(3)-[1+3(-1)^\nu]y_0^{-2}\zeta(2)-(-1)^\nu\tilde{D}_{ch}^{int}$$
$$-16\pi y_0^{-1}\sum_{n,h=1}^{\infty}(-1)^{n\nu}n^{-1}hK_1(2\pi nhy_0)\cos(2\pi nhx_0)$$
$$\approx 2\zeta(3)-[1+3(-1)^\nu]y_0^{-2}\zeta(2)-(-1)^\nu[1+(2\pi y_0)^{-1}]\tilde{D}_{ch}^{int}, \tag{15}$$

$$\tilde{D}^{xy}(\nu\mathbf{b}_1/2)=-32\pi^2\sum_{n,h=1}^{\infty}(-1)^{n\nu}h^2K_1(2\pi nhy_0)\sin(2\pi nhx_0)$$
$$\approx-(-1)^\nu 16\pi^2y_0^{-1/2}\exp(-2\pi y_0)\sin(2\pi x_0). \tag{16}$$

Here we have written $x_0=a_2\cos\alpha$, $y_0=a_2\sin\alpha$ (Fig. 1). By virtue of inequalities (13) we have

$$0\leqslant x_0\leqslant 1/2,\quad y_0\geqslant(1-x_0^2)^{1/2}\geqslant\sqrt{3}/2,\quad 60^\circ\leqslant\alpha\leqslant 90^\circ. \tag{17}$$

The approximate equalities in (14)–(16) are asymptotically exact in the limit $2\pi y_0\gg 1$. For the smallest value, $y_0=\sqrt{3}/2$ ($x_0=1/2$, $a_2=a_1=1$, $\alpha=60^\circ$—a triangular lattice), the approximate equalities in (14) and (15) lead to $\tilde{D}^{xx}(0)\approx-5.544$, $\tilde{D}^{yy}(0)\approx-5.498$. An exact calculation, on the other hand, yields $\tilde{D}^{xx}(0)=\tilde{D}^{yy}(0)=-5.517$. Consequently, the relative error of the approximate equalities is less than 0.5%, so these equalities can be used in the calculations below.

The eigenvalues of the tensor $\tilde{D}^{\alpha\beta}(\nu\mathbf{b}_1/2)$ and the orientation angles θ_ν of the unit vectors $\tilde{\xi}_1(\nu\mathbf{b}_1/2)$ with respect to the x axis [see (4) and (5)], which we are seeking, are given by

$$\tilde{D}_j=1/2\{\tilde{D}^{xx}+\tilde{D}^{yy}+(-1)^j[(\tilde{D}^{xx}-\tilde{D}^{yy})^2+4(\tilde{D}^{xy})^2]^{1/2}\},$$
$$\theta=\operatorname{arctg}[\tilde{D}^{xy}/(\tilde{D}_1-\tilde{D}^{yy})]. \tag{18}$$

For brevity we have omitted the arguments of the functions $\nu\mathbf{b}_1/2$.

We first note that at $x_0=0$ and $1/2$ we have

$$\tilde{D}^{xy}(\nu\mathbf{b}_1/2)=0,\quad \theta=0,\quad \tilde{D}_1(\nu\mathbf{b}_1/2)=\tilde{D}_{ch}+(-1)^\nu\tilde{D}_{ch}^{int}.$$

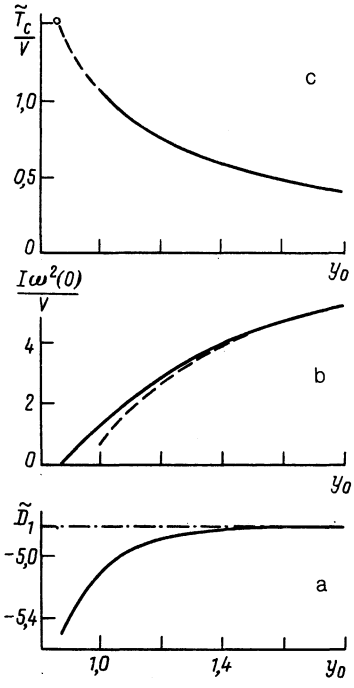

FIG. 2. Characteristics of the ground state of $2D$ Bravais lattices with $x_0 = 0, 1/2$ versus the interchain distance y_0. a: Twice the dimensional energy of the ground state, $\tilde{D}_1$ (the region with $\sqrt{3}/2 \leqslant y_0 < 1$ corresponds to only rhombic lattices with $x_0 = 1/2$). b: Squared frequencies of orientation oscillations which are active in the *ir* absorption spectra (the solid and dashed lines correspond to rhombic and rectangular lattices). c: Temperature of the phase transition in a generalized approximation of an interchain self-consistent field (the dashed line is an interpolation between the points $y_0 = \sqrt{3}/2$ and 1).

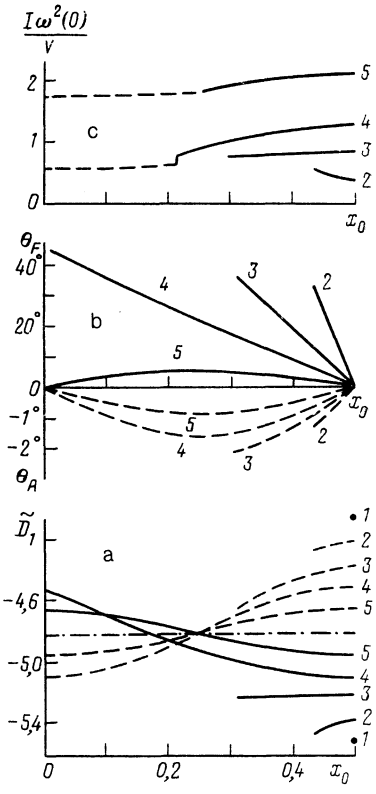

FIG. 3. Characteristics of the ground state of $2D$ Bravais lattices versus the displacements of the sites, x_0, in neighboring chains. 1—$y_0 = \sqrt{3}/2$; 2—0.9; 3—0.95; 4—1; 5—1.1. a: The energy parameter $\tilde{D}_1$. b: The angles θ_F and θ_A, specifying the orientations of the dipoles. c: The frequency parameter $I\omega^2(0)/V$ (the solid and dashed lines correspond to ferroelectric and antiferroelectric ground states).

The case $x_0 = 0$ ($y_0 > 1$) corresponds to a class of rectangular lattices with $\tilde{D}_{ch}^{int} > 0$, $\tilde{D}_1(\mathbf{b}_1/2) < \tilde{D}_1(0)$, for which there is an antiferroelectric ground state with dipole orientations parallel to the x axis, alternating in neighboring chains. For a square lattice ($y_0 = 1$) we have $\tilde{D}_1(\mathbf{b}_2/2) = \tilde{D}_1(\mathbf{b}_1/2)$, and this structure is one of a set of possible microvortex structures of a degenerate ground state.[2-4] The case $x_0 = 1/2$ ($y_0 > \sqrt{3}/2$) corresponds to a class of rhombic lattices with a rhombic angle (the angle between the equal sides, a_2) $\gamma = 180° - 2\alpha < 60°$, for which we have $\tilde{D}_{ch}^{int} < 0$, $\tilde{D}_1(0) < \tilde{D}_1(\mathbf{b}_1/2)$, and there is a ferroelectric ground state with dipoles orientated along the x axis. For a triangular lattice ($y_0 = \sqrt{3}/2$) we have $\tilde{D}_1(0) = \tilde{D}_2(0)$ and a degenerate ferroelectric ground state with arbitrary dipole orientations.[3,4] Figure 2a shows the (doubled) dimensionless energy of the ground states, $\tilde{D}_{ch} - |\tilde{D}_{ch}^{int}|$, versus the interchain distance y_0 for these classes of lattices. As y_0 increases, we find $\tilde{D}_1 \to \tilde{D}_{ch}$.

In the general case $0 \leqslant x_0 \leqslant 1/2$, the dependence of $\tilde{D}_1$ and θ on x_0 is as shown in Fig. 3, a and b, for various values of y_0. We see that at $x_0 \gtrsim 0.22$–0.25 there are ferroelectric ground states with positive values of θ_F, the orientation angles of the dipoles with respect to the x axis (Fig. 1). In the interval $\sqrt{3}/2 < y_0 < 1$ there are solutions which correspond, with $x_0 = (1 - y_0^2)^{1/2}$, to rhombic lattices with unit sides ($\mathbf{a}_1$) and rhombic angles $60° < \alpha < 90°$ (the cutoff points of curves 2 and 3 in Fig. 3, a and b), for which a ferroelectric state with dipoles oriented along the large diagonals of the rhombuses, $\mathbf{a}_1 + \mathbf{a}_2$ ($\theta_F = \alpha/2$), is realized at $60° < \alpha \lesssim 80°$, while at $80° \lesssim \alpha < 90°$ there is an antiferroelectric state with $\theta_A \approx -2°$. The results found for rhombic lattices in the chain representation, (14)–(16), agree with the results of numerical calculations by the Luttinger–Tisza method in Ref. 6 (see also Ref. 25). Rough estimates of the values of the lattice constants corresponding to a transition from a ferroelectric ground state to an antiferroelectric ground state can be found from the condition $\tilde{D}_{ch}^{int} = 0$. This condition yields $x_0 = 1/4$, which in turn leads to a value $\alpha = \arctan[(x_0^{-2} - 1)^{1/2}] = \arctan\sqrt{15} \approx 76°$.

The values of θ_F for arbitrary Bravais lattices with $\sqrt{3}/2 < y_0 < 1$, $(1 - y_0^2)^{1/2} < x_0 < 1/2$, vary from $\alpha/2$ to 0, and the angles θ_A are negative and small in magnitude. For $y_0 > 1$, the dipoles in the ground state are oriented at small angles from the x axis, and the energy of the ground state is dominated by the intrachain interactions (Fig. 2a).

4. FREQUENCIES OF ORIENTATION OSCILLATIONS; ESTIMATES OF THE PHASE-TRANSITION TEMPERATURES

It was proved in Ref. 11 that orientational long-range order exists on arbitrary $2D$ Bravais lattices (except for a square lattice). Consequently, the ground states calculated in the preceding section of this paper specify the structure of an orientational-order phase in a certain region of low temperatures. There are accordingly the problems of determining experimentally observable characteristics of the orientational-order phase and of estimating the temperatures of the phase transitions.

The simplest and most convenient way to analyze orientational order is to record the spectrum of IR absorption by

the orientation oscillations which accompany changes in the vector dipole moments in directions perpendicular to their orientations in the ground state.[25] We denote by $\varphi_{\mathbf{r}}$ the angle between the unit vector $\mathbf{e}_{\mathbf{r}}$ and the orientation $\xi_1(\mathbf{r})$ in the ground state. We have the expansion

$$\mathbf{e}_{\mathbf{r}}=\xi_1(\mathbf{r})\cos\varphi_{\mathbf{r}}+\xi_2(\mathbf{r})\sin\varphi_{\mathbf{r}}, \tag{19}$$

where the unit vector $\xi_2(\mathbf{r})$ is perpendicular to $\xi_1(\mathbf{r})$ and is determined by the orientation of $\tilde{\xi}_2(\mathbf{k}_l)$ (Ref. 11). In the limit of extremely low temperatures, the angular oscillations of dipoles with a moment of inertia I can be assumed harmonic. As a result we find the following Hamiltonian for the excitations:[20]

$$H_{ex}=\frac{1}{2}\sum_{\mathbf{r}}[I\dot{\varphi}_{\mathbf{r}}^2-\bar{V}_1\varphi_{\mathbf{r}}^2]+\frac{1}{2}\sum_{\mathbf{r},\mathbf{r}'}\xi_2^{\alpha}(\mathbf{r})V^{\alpha\beta}(\mathbf{r}-\mathbf{r}')\xi_2^{\beta}(\mathbf{r}')\varphi_{\mathbf{r}}\varphi_{\mathbf{r}'}. \tag{20}$$

The corresponding dispersion relation for orientation oscillations is determined by the quantity $\mathbf{J}(\mathbf{k}+\mathbf{h}/2)$ [Eq. (38) from Ref. 11] and can be written in the form

$$\begin{aligned}\omega^2(\mathbf{k})&=I^{-1}[\bar{V}^{\alpha\beta}(\mathbf{k})\tilde{\xi}_2^{\alpha}(\mathbf{h}/2)\xi_2^{\beta}(\mathbf{h}/2)-\bar{V}_1(\mathbf{h}/2)]\\&=I^{-1}[\bar{V}_1(\mathbf{k})\sin^2\vartheta_{\mathbf{k}}+\bar{V}_2(\mathbf{k})\cos^2\vartheta_{\mathbf{k}}-\bar{V}_1],\end{aligned} \tag{21}$$

where $\mathbf{h}/2$ is the wave vector of the orientation structure of the ground state, and $\vartheta_{\mathbf{k}}$ is the angle between $\tilde{\xi}_2(\mathbf{k})$ and $\tilde{\xi}_2(\mathbf{h}/2)$.

The frequencies of orientation oscillations of dipoles on $2D$ Bravais lattices which are observable in the IR absorption spectra correspond to the condition $ka_1\ll 1$ and are given by

$$\omega^2(0)=\frac{V}{I}[\bar{D}_2(0)-\bar{D}_1(0)] \tag{22}$$

for ferroelectric ground states or by

$$\omega^2(0)=\frac{V}{I}\left\{\bar{D}_1(0)-\bar{D}_1\left(\frac{\mathbf{b}_1}{2}\right)+[\bar{D}_2(0)-\bar{D}_1(0)]\cos^2(\theta_F-\theta_A)\right\} \tag{23}$$

for antiferroelectric ground states. Figures 2b and 3c show the squared frequencies in (22) and (23) versus the lattice constants of $2D$ Bravais lattices. At a fixed value of y_0, the oscillation frequencies in the antiferroelectric phase are lower than those in the ferroelectric phase, because of a change in the sign of $\tilde{D}_{\text{ch}}^{\text{int}}$ in (14). For rectangular and rhombic lattices with the same value of y_0, for example, the difference between the values of $I\omega^2(0)/V$ in these phases is $2[1+(2\pi y_0)^{-1}]|\tilde{D}_{\text{ch}}^{\text{int}}|$ according to (15).

Only in the case of a triangular lattice of dipoles does the value of $\omega^2(0)$ vanish. The corresponding long-wavelength asymptotic expression for a Goldstone mode is

$$\omega^2(\mathbf{k})=\frac{V}{I}\left[0{,}2633(ka)^2\sin^2\vartheta_{\mathbf{k}}+\frac{4\pi}{\sqrt{3}}ka\cos^2\vartheta_{\mathbf{k}}\right] \tag{24}$$

[$\vartheta_{\mathbf{k}}$ is the angle between the vector $\mathbf{k}$ and the vector $\tilde{\xi}_2(0)$], while the low-frequency asymptotic behavior of the frequency distribution is

$$g(\omega)\approx 0{,}1062(I/V)^{5/4}\omega^{3/2}. \tag{25}$$

The mean square fluctuations in the angles,

$$\langle\varphi_{\mathbf{r}}^2\rangle=\frac{T}{I}\int_0^{\infty}\frac{g(\omega)\,d\omega}{\omega^2}, \tag{26}$$

are therefore finite, and the ferroelectric ground state of the dipoles on a triangular lattice is stable, in agreement with the results of Ref. 11 but in contradiction of the results of Ref. 10 (this point is discussed in Sec. 5). The low-temperature asymptotic behavior of the specific heat of orientation oscillations of dipoles on a triangular lattice is given by

$$c(T)\approx 0{,}6176(I/\hbar^2V)^{5/4}T^{5/2}. \tag{27}$$

A square lattice of dipoles also has a Goldstone mode of orientation oscillations, but in this case $\omega^2(\mathbf{k})$ vanishes not at $\mathbf{k}=0$ [$\omega^2(0)=0.586V/I$] but at the boundary of the first Brillouin zone. Here we have $g(\omega)\propto\omega$ and $c(T)\propto T^2$, and the integral in (26) diverges logarithmically at its lower limit, indicating the existence of only a short-range order, as in a (Berezinskiĭ-)Kosterlitz-Thouless phase.[11] For all other lattices we have $g(\omega)=0$ at $\omega<\omega(0)$, the mean square fluctuations of the angles in (26) are finite, and the low-temperature asymptotic specific heat is exponentially small. For $y_0\gg 1$, the quantity $I\omega^2(0)/V$ approaches the constant value $6\zeta(3)$, which corresponds to orientation oscillations of dipoles in an isolated chain (these oscillations were studied in Refs. 26 and 27 in the nearest-neighbor approximation).

Real dipole systems may have additional n-well local potentials, which form torsional vibrations with a frequency ω_{φ} in the absence of dipole forces.[25] When these local potentials are taken into account, a sum of potential energies of the torsional vibrations, $\frac{1}{2}I\omega_{\varphi}^2\varphi_{\mathbf{r}}^2$, appears in the Hamiltonian (20), and Eqs. (21)–(23) acquire an increment of ω_{φ}^2.

We turn now to estimates of the phase-transition temperatures $\tilde{T}_c$. Rough estimates $\tilde{T}_c\sim|\tilde{V}_1|/2$ are found from the self-consistent field approximation,[3,4] but the calculations of lower limits on $\tilde{T}_c$ carried out in Ref. 11 describe vastly lower values. The low phase-transition temperatures in dipole systems can be explained by the small values of the interchain interactions, since the strong intrachain interaction cannot lead to long-range order in an isolated chain. These considerations show that the most competent description of phase transitions in these systems can be found with the help of a generalized approximation of the interchain self-consistent field, in which the exact solution of the one-dimensional problem would be used, and the weak interchain interactions would be treated as a perturbation. This approximation for quasi-$1D$ systems (which are not dipole systems) was introduced in Ref. 13 and was justified with the help of the Feynman inequality in Ref. 14. Unfortunately, no exact solution is known for a $1D$ system of $2D$ dipoles. We thus consider a $2D$ Ising model with dipole-dipole interactions on an arbitrary Bravais lattice with dipole orientations parallel to the x axis (Fig. 1), $e_{\mathbf{r}}^x=\sigma_n(p)=\pm 1$ (p is the index of the site in the chain, and n is the number of the chain):

$$H=-\frac{1}{2}\sum_{\substack{p,p'\\n,n'}}V_{n-n'}(p-p')\sigma_n(p)\sigma_{n'}(p'),\quad V_n(p)=-V^{xx}(\mathbf{r}). \tag{28}$$

A model of this sort is useful for describing phase transitions in real $2D$ systems with two-well local potentials. In the present approach, this model would correspond to an exactly solvable $1D$ Ising model,

$$H_0=-J\sum_{p,n}\sigma_n(p)\sigma_n(p+1)-h\sum_{p,n}\sigma_n(p), \tag{29}$$

with variable parameters J and h. Following Ref. 14, we write the Feynman inequality:

$$F\leqslant F_0+\langle H-H_0\rangle_0={}^1/_2V[\tilde{D}_{ch}-|\tilde{D}_{ch}^{int}|]m^2-2V(1-m^2)\mathrm{Li}_3(\alpha)$$
$$-T\ln 2+{}^1/_2T\ln[(1+\alpha)^2-m^2(1-\alpha)^2]$$
$$-{}^1/_4T(1-m^2)(1-\alpha)\ln\frac{1+\alpha^2+2\alpha(1+m^2)/(1-m^2)}{(1-\alpha)^2}$$
$$+Tm\,\mathrm{arcth}\left(\frac{1-\alpha}{1+\alpha}m\right), \tag{30}$$

where the intrachain interaction $\tilde{D}_{ch}$ and interchain interaction $\tilde{D}_{ch}^{int}$ are given in (14), while the polarizability m and the correlation parameter α of the $1D$ Ising model are given by

$$m=\langle\sigma_n(p)\rangle_0=\mathrm{sh}\,(h/T)/\Delta,\quad \Delta=[\mathrm{sh}^2\,(h/T)+\exp(-4J/T)]^{1/2},$$
$$\langle\sigma_n(p)\sigma_n(p+1)\rangle_0=m^2+(1-m^2)\alpha, \tag{31}$$
$$\alpha=[\mathrm{ch}\,(h/T)-\Delta]/[\mathrm{ch}\,(h/T)+\Delta],$$

Here $Li_n(\alpha)$ is a polylogarithm of order n:

$$\mathrm{Li}_n(\alpha)=\sum_{p=1}^{\infty}p^{-n}\alpha^p,\quad \mathrm{Li}_n(1)=\zeta(n)$$

Minimizing the right side of inequality (30) with respect to the parameters m and α (this approach is equivalent to minimizing with respect to J and h), we find the following system of equations:

$$T\,\mathrm{arcth}\left(\frac{1-\alpha}{1+\alpha}m\right)$$
$$=mV\left\{4\left[\zeta(3)-\mathrm{Li}_3(\alpha)-\frac{1-\alpha}{\alpha}\mathrm{Li}_2(\alpha)\right]+|\tilde{D}_{ch}^{int}|\right\}, \tag{32}$$

$$T\ln\frac{1+\alpha^2+2\alpha(1+m^2)/(1-m^2)}{(1-\alpha)^2}=\frac{8V}{\alpha}\mathrm{Li}_2(\alpha). \tag{33}$$

In addition to the trivial solution $m=0$, Eqs. (32), (33) have solutions with $m\neq 0$ in a certain temperature region ($T\neq 0$) with $\tilde{D}_{ch}^{int}\neq 0$. At the point of the phase transition ($m\to 0$), the temperature T_c and the parameter α are given by

$$T_c=\frac{4V\,\mathrm{Li}_2(\alpha)}{\alpha\ln[(1+\alpha)/(1-\alpha)]}=\frac{1+\alpha}{1-\alpha}V\left\{4\left[\zeta(3)-\mathrm{Li}_3(\alpha)\right.\right.$$
$$\left.\left.-\frac{1-\alpha}{\alpha}\mathrm{Li}_2(\alpha)\right]+|\tilde{D}_{ch}^{int}|\right\}. \tag{34}$$

In the limit $\tilde{D}_{ch}^{int}\to 0$ we have $\alpha\to 1$, $T_c\to 0$. In the case of weak interchain interactions ($|\tilde{D}_{ch}^{int}/\tilde{D}_{ch}|\ll 1$), the approximate solution of Eqs. (34) for T_c becomes

$$T_c\approx 4V\zeta(2)/\ln|\tilde{D}_{ch}/\tilde{D}_{ch}^{int}|. \tag{35}$$

Expression (35) differs from the known relations of the generalized self-consistent-field approximation for short-range potentials[13,14] (in our notation, $T_c\approx|\tilde{D}_{ch}|/\ln|\tilde{D}_{ch}/\tilde{D}_{ch}^{int}|$) by a factor $\zeta(2)\approx 1.645$, which reflects the long-range intrachain interactions, which raise the value of T_c but which are incapable of leading to a phase transition with $T_c\neq 0$ in the absence of interchain interactions.

In the model of short-range dipole potentials considered in Refs. 3 and 28, the exact solutions of the $2D$ Ising model have the same asymptotic behavior in the case $|\tilde{D}_{ch}^{int}/\tilde{D}_{ch}|\ll 1$ as in the approximation under consideration here. For the four symmetric orientations of the dipoles in the plane of the lattice, the exact values of T_c are smaller by a factor of 2 than for the two orientations of the Ising model. A corresponding decrease in the values of T_c by a factor of 2 when the $1D$ orientation space is replaced by $2D$ also follows from the equations of the ordinary self-consistent-field approximation. Accordingly, values of T_c smaller by a factor of 2 than in Ref. 35 can be used as estimates of the transition temperatures in dipole systems with $2D$ degenerate orientations of dipoles, whose ground states correspond to orientations along the axes of the lattices (Bravais lattices with $x=0$ and $1/2$). Figure 2c shows $\tilde{T}_c=T_c/2$ as a function of the interchain distance y_0. For triangular and square lattices we have $\tilde{T}_c/V\approx 1.501$ and 1.059, respectively, while the ordinary self-consistent-field approximation yielded values of $\tilde{T}_c/V=-\tilde{D}_1/2\approx 2.759$ and 2.550 in Refs. 3 and 4 for the same lattices. Interestingly, the exact value for $\tilde{T}_c$ for a square lattice of short-range dipoles with four orientations ($\tilde{T}_c/V=1.641$; Ref. 28) is higher than the estimate found for long-range dipoles from (35), since the interaction with the nearest dipoles of the neighboring chains is much stronger than that with all the dipoles of the neighboring chains ($|\tilde{D}_{ch}^{int}|\ll 2<|\tilde{D}_{ch}|$).

For $2D$ Bravais lattices with $y_0>3$, expression (35) can be approximated by the following simple expression, when we use (14):

$$T_c\approx 2V\zeta(2)/\pi y_0,\qquad y_0>3. \tag{36}$$

The temperatures of orientation phase transitions in quasi-$1D$ dipole systems thus fall off in inverse proportion to the distance between chains.

5. DISCUSSION

A point of fundamental importance in describing the ground states of $2D$ dipole systems, in proving the existence of a long-range order, and in reaching an understanding of the reasons for the appearance of intermediate phases (intermediate along the temperature scale) with a wave vector $\mathbf{k}\neq\mathbf{h}/2$ is the condition that the absolute values of the $2D$ dipole moments remain constant. Ferromagnetic ordering of $2D$ systems with dipole-dipole and exchange interactions was examined in the spherical-model approximation in Ref. 10. The basic simplifying assumption of the spherical model is the replacement of the condition $|\mathbf{e}_r|=1$ by the weaker condition (8). If the identity (9) is ignored, the contributions of the eigenvalues $\tilde{V}_1(\mathbf{k})$ and $\tilde{V}_2(\mathbf{k})$ become independent in (for example) the dispersion relation for orientation oscillations [cf. the correct expression, (21)]. It is as if there were two oscillation branches, with frequency distributions $g_1(\omega)\propto\omega$ and $g_2(\omega)\propto\omega^3$, for a triangular lattice of dipoles. Oscillation branches of this sort do indeed occur in the system of electrons of a Wigner crystal above the surface of

liquid helium,[1] since displacements of the electrons from their equilibrium positions are independent along the x and y axes and not of fixed length. The logarithmic divergence of the mean square displacements for the branch with $g_1(\omega)$ [which arises in integrals of the form (26)] indicates that there is no long-range order.[29] A corresponding divergence of course arises in the spherical model also, as a result of the violation of the condition $|\mathbf{e}_r| = 1$. In Ref. 10, this led to the incorrect conclusion that there is no long-range order in $2D$ dipole systems with a continuously degenerate ferromagnetic ground state.

Again in Ref. 15 we find the assertion that there is no long-range order in $2D$ or $3D$ systems. In that paper, the partition function of functionals of Gaussian electric fields was examined. That partition function is equivalent to the partition function from a Hamiltonian which incorporates only the long-wavelength asymptotic behavior of the dipole-interaction tensor, proportional to $k_\alpha k_\beta / k^2$. Consequently, the 2^N-fold degeneracy found for the ground state in Ref. 15 appears to implicitly reflect the indefiniteness in a ferroelectric ground state which stems from the dependence of the depolarizing electric field on the shape of the sample. In fact, as we have also shown here, the ground state in dipole systems may correspond to a wave vector $\mathbf{k}$ at the boundary of the first Brillouin zone, so it would be incorrect to consider only the long-wavelength asymptotic behavior. Furthermore, for $2D$ systems the long-wavelength asymptotic behavior $\tilde{V}^{\alpha\beta}(\mathbf{k}) \propto k_\alpha k_\beta / k$ differs from that of the $3D$ analog in Ref. 15, there is no depolarizing field, and one cannot speak in terms of 2^N-fold degeneracy of ground states of a ferroelectric type. Consequently, despite the elegance of the field-functional formalism for dipole systems, we cannot avoid the discrete nature of the lattice. For this reason, the results of Ref. 15, which were actually derived in the continuum approximation, contradict the conclusions of the present paper and those of Ref. 11.

In research on the properties of dipole systems, a chain representation of the interactions has proved to be the most successful. In the first place, it reflects the tendency for the dipole moments to become ordered along the axes of the chains (the cases $x_0 = 0$ and $1/2$ with $\sqrt{3}/2 \leqslant y_0 \lesssim 1$ and arbitrary x_0 at $y_0 > 1$ on Bravais lattices) with a small ratio of the interchain interaction to the intrachain interaction. Second, that representation makes it possible to work very accurately with analytic expressions which sum the interactions with all the dipoles of the lattice. Third, it is a justification for using the generalized approximation of an interchain self-consistent field to describe orientational phase transitions.

The results of this paper generalize the research on the ground states of individual lattices[1-10] to arbitrary $2D$ Bravais lattices. They prove the stability of the ground states which have been found with respect to thermodynamic fluctuations in the sense that the mean square angular displacements of the orientational excitations are bounded. They provide information on the frequencies of orientational oscillations which are active in the IR absorption spectra and on the phase-transition temperatures. In particular, it has been established here that the temperatures of orientation phase transitions in quasi-$1D$ dipole systems fall off in inverse proportion to the distance between chains.

[1)] This special case is realized on a triangular lattice with a strong antiferromagnetic interaction and a weak dipole–dipole interaction.[17] The ("120°") ground state in this system corresponds to wave vectors of the vertices of the hexagonal boundary of the first Brillouin zone, at which the tensors $\tilde{V}_1^{\alpha\beta}(\mathbf{k}_l)$ of both interactions are isotropic.

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