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Ground state and vibrations of dipoles on a honeycomb lattice

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For long-range dipole-dipole interactions, the energies of various periodic dipole orientations on a complex honeycomb lattice have been calculated based on the lattice-sublattice relations between Fourier components of interaction tensors. In the ground state of the system, vortex configurations of dipoles with their orientations differing in angles by $\pm 2\pi/3$ have been shown to be formed in the lattice plane. Provided any adsorption potentials force polar molecules out of the surface plane, Davydov splitting occurs in the vibration spectrum of the molecular ensemble.

Dipole-dipole interactions in two-dimensional systems were correctly taken into account in the analysis of the vibration spectrum of Wigner-crystal electrons over the liquid-helium surface.[1] Subsequently, interactions of this kind were treated as small (stabilizing long-range order) corrections to exchange interactions in planar magnetics[2,3] and as lateral interactions predominating in the systems of adsorbed polar molecules.[4,5] As predicted previously,[6–8] there are certain types of planar ordering of adsorbed polar molecules; this has recently been proved experimentally for the CO/NaCl (100) system[9] exhibiting low-temperature Davydov splitting of CO vibration lines.

A dipole force anisotropy gives rise to complex ground-state configurations of dipole moments which were calculated by various methods taking account of long-range interactions for square,[8,10] triangular,[8] rhombic,[11] and arbitrary planar Bravais lattices.[12] In the present paper, it is our intention to present the lattice-sublattice relations for Fourier components of a dipole-dipole interaction tensor, which enable the energies of various orientation structures on a complex honeycomb lattice to be expressed in terms of known characteristics of its triangular sublattices. In the framework of such treatment, the ground state of the system concerned, as well as the infrared-active spectral frequencies of radial (valence) dipole vibrations, will be elucidated.

Let us consider an arbitrary planar lattice that consists of n Bravais sublattices having unit cell $\mathbf{A}_1$, $\mathbf{A}_2$. Then the sites of the complex lattice are given by the vectors $\mathbf{R}+\mathbf{r}_j$, where $\mathbf{R}=m_1\mathbf{A}_1+m_2\mathbf{A}_2$ specifies the sites of the basic Bravais sublattice and $\mathbf{r}_j$ characterizes the positions of the jth sublattice's sites within the unit cell defined above. The sought-for energy of interactions between dipole moments $\boldsymbol{\mu}_{\mathbf{R}j}=\mu\mathbf{e}_{\mathbf{R}j}$ with $\mathbf{e}_{\mathbf{R}j}$ denoting unit vectors in the sites $\mathbf{R}+\mathbf{r}_j$ can be expressed as

$$H=\tfrac{1}{2}\sum_{\mathbf{R}j,\mathbf{R}'j'} V^{\alpha\beta}(\mathbf{R}-\mathbf{R}'+\mathbf{r}_{jj'})e^{\alpha}_{\mathbf{R}j}e^{\beta}_{\mathbf{R}'j'}$$
$$=\tfrac{1}{2}N_0\sum_{\mathbf{K},j,j'}\tilde{V}^{\alpha\beta}_{jj'}(\mathbf{K})\tilde{e}^{\alpha}_{j}(-\mathbf{K})\tilde{e}^{\beta}_{j'}(\mathbf{K}) , \qquad (1)$$

where

$$V^{\alpha\beta}(\mathbf{R})=\mu^2\left[\frac{\delta_{\alpha\beta}}{R^3}-\frac{R_\alpha R_\beta}{R^5}\right] \qquad (2)$$

is a dipole-dipole interaction tensor. The second equality in Eq. (1) corresponds to switching to Fourier components within the main area of the basic Bravais sublattice involving N_0 sites; hereinafter twice repeated Greek indices of Cartesian coordinate axes $(\alpha,\beta=x,y,z)$ imply the summation over them. After finding eigenvalues and eigenvectors of the tensor $\tilde{V}^{\alpha\beta}_{jj'}(\mathbf{K})$

$$\sum_{j'}\tilde{V}^{\alpha\beta}_{jj'}(\mathbf{K})C^{\beta\nu}_{j'p}(\mathbf{K})=V^{\nu}_{p}(\mathbf{K})C^{\alpha\nu}_{jp}(\mathbf{K}) ,$$
$$j=1,\ldots,n;\quad \nu=1,2,3 \qquad (3)$$

the quadratic form (1) can be presented as a sum of squares:

$$H=\tfrac{1}{2}N\sum_{\mathbf{K},p,\nu}V^{\nu}_{p}(\mathbf{K})|\eta^{\nu}_{p}(\mathbf{K})|^2 ,$$
$$N=nN_0,\quad \sum_{\mathbf{K},p,\nu}|\eta^{\nu}_{p}(\mathbf{K})|^2=1 , \qquad (4)$$

$$e^{\alpha}_{\mathbf{R}j}=\sqrt{n}\sum_{\mathbf{K},p,\nu}C^{\alpha\gamma}_{jp}(\mathbf{K})\cdot\eta^{\nu}_{p}(\mathbf{K})\cdot\exp[i\mathbf{K}\cdot(\mathbf{R}+\mathbf{r}_j)] . \qquad (5)$$

As is evident now, the ground-state energy is determined by the deepest minimum among those inherent in a family of functions $V^{\nu}_{p}(\mathbf{K})$, and the corresponding dipole moment configuration $\mathbf{e}_{Rj}$ is specified by the eigenvectors $C^{\alpha\nu}_{jp}(\mathbf{K})$ [see Eq. (5)]. The above procedure of finding the ground state of a dipole system on a complex lattice is a generalization of the technique used previously for simple Bravais lattices.[12,13]

The behavior of the quantities $\tilde{V}^{\alpha\beta}_{jj}(\mathbf{K})$ for an arbitrary Bravais sublattice has been studied rather well.[4,5,12] Thus the challenge is to most efficiently calculate the values of $\tilde{V}^{\alpha\beta}_{jj'}(\mathbf{K})$ at $j'\neq j$. We show such intersublattice interactions to be related to the known tensors $\tilde{V}^{\alpha\beta}_{jj}(\mathbf{K})$ for the basic Bravais sublattice. To do this, let us construct a denser Bravais lattice whose site set $\mathbf{r}=m_1\mathbf{a}_1+m_2\mathbf{a}_2$ contains the site set of the complex lattice under study. Naturally, construction of this kind is not always possible; however, in the most interesting case presented by symmetric lattices, the basic vectors for the basic Bravais lattice are expressible as the integer-coefficient linear combinations of those for the dense lattice: $\mathbf{A}_j=n_{j1}\mathbf{a}_1+n_{j2}\mathbf{a}_2$ $(j=1,2;$ $n_{jj'}$ are integers). Then the unit-cell areas for these lattices are in $n=\det\hat{n}$ relationship to each other,

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and the basic vectors for reciprocal lattices satisfying the equations $\mathbf{A}_j \cdot \mathbf{B}_{j'} = 2\pi\delta_{jj'}$ and $\mathbf{a}_j \cdot \mathbf{b}_{j'} = 2\pi\delta_{jj'}$ are mutually expressible as rational-coefficient linear combinations:

$$\mathbf{B}_j = \sum_{j'} (\hat{n}^{-1})_{j'j} \mathbf{b}_{j'} \ . \tag{6}$$

With introducing the Fourier components $\tilde{V}^{\alpha\beta}(\mathbf{K})$ of dipole-dipole interaction tensor for the dense Bravais lattice, the desired quantities $\tilde{V}_{jj'}^{\alpha\beta}(\mathbf{K})$ for the complex lattice can be determined as follows:

$$\tilde{V}_{jj'}^{\alpha\beta}(\mathbf{K}) = \frac{1}{n} \sum_{\mathbf{B}} \tilde{V}^{\alpha\beta}(\mathbf{K}+\mathbf{B}) e^{i\mathbf{B}\cdot\mathbf{r}_{jj'}} \ , \tag{7}$$

where summation is performed over all the integer-coefficient linear combinations of the vectors $\mathbf{B}_1$ and $\mathbf{B}_2$ falling within the first Brillouin zone of the dense Bravais lattice.

Equation (7) proves very convenient both in elucidating the relationships between tensors $\tilde{V}^{\alpha\beta}(\mathbf{K})$ in different points of the first Brillouin zone and in calculating intersublattice interactions within the complex lattice. This is typified by a complex honeycomb lattice, with the hexagon side being equal to a. The basic vectors $\mathbf{A}_1 = \sqrt{3}a(1,0,0)$ and $\mathbf{A}_2 = \sqrt{3}a(1/2, \sqrt{3}/2, 0)$ for the basic triangular sublattice are expressible in terms of the basic vectors $\mathbf{a}_1 = a(0,-1,0)$ and $\mathbf{a}_2 = a(\sqrt{3}/2, -1/2, 0)$ for the dense triangular lattice in the following way: $\mathbf{A}_1 = 2\mathbf{a}_2 - \mathbf{a}_1$; $\mathbf{A}_2 = \mathbf{a}_2 - 2\mathbf{a}_1$ [see Fig. 1(a)]. Further, $\mathbf{r}_{21} = a(\sqrt{3}/2, 1/2, 0) = \mathbf{a}_2 - \mathbf{a}_1$ and all the sites of the complex lattice are contained in the site set of the dense Bravais lattice. The basic vectors for the reciprocal lattices are interrelated: $\mathbf{B}_1 = (\mathbf{b}_1 + 2\mathbf{b}_2)/3$ and $\mathbf{B}_2 = -(2\mathbf{b}_1 + \mathbf{b}_2)/3$, so that the corresponding first Brillouin zones appear as in Fig. 1(b) and the summation in Eq. (7) is confined to the vectors $\mathbf{B} = -\mathbf{B}_2, 0, \mathbf{B}_2$. Thus we come to

$$\begin{aligned} \tilde{V}_{11}^{\alpha\beta}(\mathbf{K}) &= \tilde{V}_{22}^{\alpha\beta}(\mathbf{K}) \\ &= \tfrac{1}{3}[\tilde{V}^{\alpha\beta}(\mathbf{K}) + \tilde{V}^{\alpha\beta}(\mathbf{K}+\mathbf{B}_2) + \tilde{V}^{\alpha\beta}(\mathbf{K}-\mathbf{B}_2)] \ , \end{aligned} \tag{8}$$

$$\begin{aligned} \tilde{V}_{21}^{\alpha\beta}(\mathbf{K}) &= [\tilde{V}_{12}^{\alpha\beta}(\mathbf{K})]^* \\ &= \frac{1}{3}\left\{ \tilde{V}^{\alpha\beta}(\mathbf{K}) - \tfrac{1}{2}[\tilde{V}^{\alpha\beta}(\mathbf{K}+\mathbf{B}_2) + \tilde{V}^{\alpha\beta}(\mathbf{K}-\mathbf{B}_2)] \right. \\ &\qquad \left. + i\frac{\sqrt{3}}{2}[\tilde{V}^{\alpha\beta}(\mathbf{K}+\mathbf{B}_2) - \tilde{V}^{\alpha\beta}(\mathbf{K}-\mathbf{B}_2)] \right\} . \end{aligned} \tag{9}$$

The dense Bravais lattice is none other than the same basic triangular lattice merely rotated around the axis OZ through 90 degrees clockwise. With the appropriate rotation matrix $\hat{S}$ introduced we obtain the identity of importance:

$$\hat{\tilde{V}}_{jj}(\mathbf{K}) = 3^{-3/2} \hat{S} \hat{\tilde{V}}(\sqrt{3}\hat{S}^{-1}\mathbf{K}) \hat{S}^{-1} \ , \tag{10}$$

which, along with Eq. (8), allows the values $\tilde{V}^{\alpha\beta}(\mathbf{K})$ for different points of the first Brillouin zone to be interrelated.

Specifically, for the symmetric points $\mathbf{K}=0$, $\mathbf{k}_A = \mathbf{b}_2/2$, $\mathbf{k}_J = -\mathbf{B}_2$ [see Fig. 1(b)], and for the point $\mathbf{K} = \mathbf{K}_J = \mathbf{b}_2/3$, the following expression is obtained:

$$\begin{aligned} &\tilde{V}^{\alpha\beta}(0) = D_F V \delta_{\alpha\beta}, \quad \tilde{V}^{\alpha\beta}(\mathbf{k}_J) = \tfrac{1}{2}(3^{-1/2} - 1) D_F V \delta_{\alpha\beta} \ , \\ &V = \frac{\mu^2}{a^3}, \quad \alpha,\beta = x,y \ , \\ &\tilde{V}^{xx}(\mathbf{k}_A) = D_A V, \quad \tilde{V}^{yy}(\mathbf{k}_A) = -(D_A + D_F/3)V \ , \\ &\tilde{V}^{xy}(\mathbf{k}_A) = 0 \ , \\ &\tilde{V}^{xx}(\mathbf{K}_J) = -4.453\,809 V \ , \\ &\tilde{V}^{yy}(\mathbf{K}_J) = 3^{-3/2}(3^{-1/2} - 1) D_F V - \tilde{V}^{xx}(\mathbf{K}_J) \ , \\ &\tilde{V}^{xy}(\mathbf{K}_J) = 0 \ , \\ &\tilde{V}^{zz}(\mathbf{K}) = -\tilde{V}^{xx}(K) - \tilde{V}^{yy}(K) \ , \end{aligned} \tag{11}$$

where the parameters $D_F = -5.517\,088$ and $D_A = -4.094\,910$ determine the energies of ferroelectric and antiferroelectric states of dipole systems on a triangular lattice.[8] Now, the values of $\tilde{V}_{21}^{\alpha\beta}(\mathbf{K})$ at $\mathbf{K}=0$, $\mathbf{K}_A = \mathbf{B}_2/2$, and $\mathbf{K}_J = \mathbf{b}_2/3$ [see Fig. 1(b)] are derivable from Eq. (9):

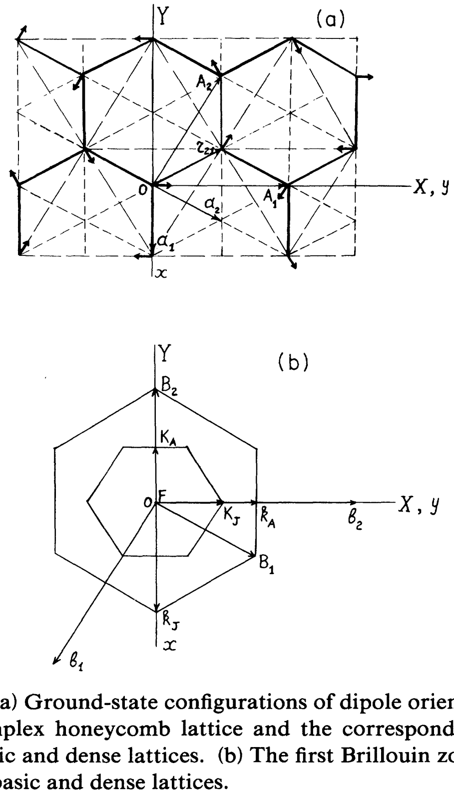

FIG. 1. (a) Ground-state configurations of dipole orientations on the complex honeycomb lattice and the corresponding triangular basic and dense lattices. (b) The first Brillouin zones for reciprocal basic and dense lattices.

$$
\begin{aligned}
&V_{21}^{\alpha\beta}(0)=\tfrac{1}{2}(1-3^{-3/2})D_F V\delta_{\alpha\beta}\ ,\\
&\widetilde{V}_{21}^{XX}(\mathbf{K}_A)=\tfrac{1}{2}[(1+3^{-3/2})D_A+\tfrac{1}{3}D_F]\exp\left[-i\frac{\pi}{3}\right]V\ ,\\
&\widetilde{V}_{21}^{YY}(\mathbf{K}_A)=-\tfrac{1}{2}[(1+3^{-3/2})D_A\\
&\qquad\qquad+3^{-5/2}D_F]\exp\left[-i\frac{\pi}{3}\right]V\ ,\\
&\widetilde{V}_{21}^{XY}(\mathbf{K}_A)=0\ ,\\
&\widetilde{V}_{21}^{XX}(\mathbf{K}_J)=-\widetilde{V}_{21}^{YY}(\mathbf{K}_J)\\
&\qquad=-i\widetilde{V}_{21}^{XY}(\mathbf{K}_J)\\
&\qquad=\tfrac{1}{4}3^{-3/2}(3^{-1/2}-1)D_F V-\tfrac{1}{2}\widetilde{V}^{xx}(\mathbf{K}_J)\ ,\\
&\widetilde{V}_{21}^{ZZ}(\mathbf{K})=-\widetilde{V}_{21}^{XX}(\mathbf{K})-\widetilde{V}_{21}^{YY}(\mathbf{K})\ .
\end{aligned}
\tag{12}
$$

The eigenvalues of tensor $\widetilde{V}_{jj'}^{\alpha\beta}(\mathbf{K})$ in the symmetric points of the first Brillouin zone that are determined by the formula (3) take the form

$$
\begin{aligned}
&V_1^1(0)=V_1^2(0)=-\tfrac{1}{2}V_1^3(0)=\tfrac{1}{2}(1+3^{-3/2})D_F V\\
&\qquad=-3.289\,426V\ ,\\
&V_2^1(0)=V_2^2(0)=-\tfrac{1}{2}V_2^3(0)=\tfrac{1}{2}(3^{-1/2}-1)D_F V\\
&\qquad=1.165\,898V\ ,\\
&V_1^1(\mathbf{K}_A)=\tfrac{1}{2}[\tfrac{1}{3}D_F+(1+3^{-1/2})D_A]V\\
&\qquad=-4.149\,068V\ ,\\
&V_1^2(\mathbf{K}_A)=-\tfrac{1}{2}[3^{-5/2}D_F-(1-3^{-3/2})D_A]V\\
&\qquad=-1.476\,461V\ ,\\
&V_2^1(\mathbf{K}_A)=-\tfrac{1}{2}[\tfrac{1}{3}D_F+(1-3^{-3/2})D_A]V\\
&\qquad=2.572\,937V\ ,\\
&V_2^2(\mathbf{K}_A)=-\tfrac{1}{2}[3^{-3/2}D_F+(1+3^{-1/2})D_A]V\\
&\qquad=3.760\,436V\ ,
\end{aligned}
\tag{13}
$$

$$
\begin{aligned}
&V_1^3(\mathbf{K}_A)=-V_1^1(\mathbf{K}_A)-V_2^2(\mathbf{K}_A)\ ;\\
&V_2^3(\mathbf{K}_A)=-V_1^2(\mathbf{K}_A)-V_2^1(\mathbf{K}_A)\ ;\\
&V_1^1(\mathbf{K}_J)=\widetilde{V}^{xx}(\mathbf{K}_J)=-4.453\,809V\ ;\\
&V_2^2(\mathbf{K}_J)=\widetilde{V}^{yy}(\mathbf{K}_J)=4.902\,564V\ ,\\
&V_1^2(\mathbf{K}_J)=V_2^1(\mathbf{K}_J)=-\tfrac{1}{2}V_1^3(\mathbf{K}_J)\\
&\qquad=-\tfrac{1}{2}V_2^3(\mathbf{K}_J)\\
&\qquad=\tfrac{1}{2}3^{-3/2}(3^{-1/2}-1)D_F V=0.224\,377V\ .
\end{aligned}
$$

The dependences $V_p^\nu(\mathbf{K})$ calculated along symmetric directions within the first Brillouin zone are given in Fig. 2. The minimum value, $V_1^1(\mathbf{K}_J)$, determines the energy $H=\frac{1}{2}NV_1^1(\mathbf{K}_J)$ of the one-parametric degenerate (in angle φ) ground state with the following configuration of dipole moments:

$$\mathbf{e}_{\mathbf{R}1}=[\cos(\mathbf{K}_J\cdot\mathbf{R}+\varphi),\sin(\mathbf{K}_J\cdot\mathbf{R}+\varphi),0]\ ;$$

$$\mathbf{e}_{\mathbf{R}2}=[-\cos(\mathbf{K}_J\cdot\mathbf{R}+2\pi/3+\varphi),\sin(\mathbf{K}_J\cdot\mathbf{R}+2\pi/3+\varphi),0]$$

[see Fig. 1(a)]. Interestingly, the same ground-state configuration can be arrived at with the consideration of dipole-dipole interactions in terms of the simplest nearest-neighbor approximation [at $V_1^1(\mathbf{K}_J)=-(9/2)V$],[14] whereas for a triangular lattice of dipoles, short- and long-range interaction models lead to the ground-state structures which are qualitatively different.[8]

Let us analyze the effect caused by the thermodynamic fluctuations of the order parameter on the ground state of a dipole honeycomb lattice. Two factors determine the situation just as they do in the case of a square lattice.[8,10,13] These are the quadratic asymptotics of the minimum-energy branch $V_1^1(\mathbf{K})$ of eigenvalues in the vicinity of the ground state with $\mathbf{K}=\mathbf{K}_J$ and one-parametric ground-state degeneracy in the angle variable. As was shown previously[13] and confirmed by Monte Carlo simulations[15] for a square lattice, these factors lead the system concerned to exist in the Berezinskii-Kosterlitz-Thouless phase at low temperatures. On breaking the degeneracy by any local potentials or multipole corrections, long-range order should be stabilized as is the case for dipoles on the square lattice.[16]

The curves $V_p^\nu(\mathbf{K})$ shown in Fig. 2 correspond to six branches of charge vibrations which can shift relative to the honeycomb lattice sites. As a lattice of this sort is realized on the basic graphite face for which a great body of experimental adsorption evidence is accumulated,[17] here it is appropriate to present the calculated radial vibration frequencies for adsorbed polar molecules that should be observable in infrared spectra.

Adsorption potentials can force the dipole moments of adsorbed molecules out of the lattice plane.[9] Based on the calculations performed previously for square lattice of adsorption centers[18] and with Θ_0 denoting the angle of dipole moments' inclination to the surface-normal direction, we are led to

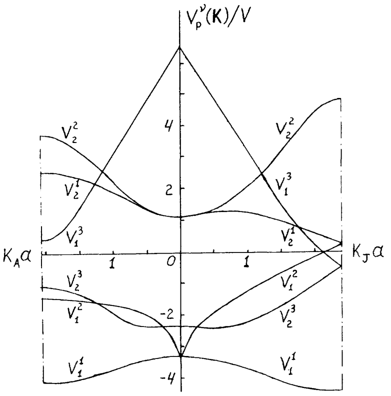

FIG. 2. Fourier component eigenvalues $V_p^\nu(\mathbf{K})$ for the dipole-dipole interaction tensor on a honeycomb lattice versus wave vector $\mathbf{K}$ determined in the first Brillouin zone of the basic sublattice.

$$\omega_Z^2=\omega_0^2+\frac{1+\kappa}{m^*l^2}[V_1^3(0)\cos^2\Theta_0+V_1^1(\mathbf{K}_J)\sin^2\Theta_0]\ ,$$

$$\begin{aligned}\omega_X^2=\omega_Y^2=\omega_0^2+\frac{1}{m^*l^2}\{&[V_1^3(0)\cos^2\Theta_0+V_1^1(\mathbf{K}_J)\sin^2\Theta_0]\kappa\\&+V_1^3(\mathbf{K}_J)\cos^2\Theta_0\\&+\tfrac{1}{2}[V_1^1(0)+V_1^2(\mathbf{K}_J)]\sin^2\Theta_0\}\ .\end{aligned}\tag{14}$$

Here the indices X,Y,Z indicate vibration polarizations, ω_0 is a vibration frequency for an isolated adsorbed molecule, m^* is a reduced mass of an oscillator, $l=\mu/q$ is an equilibrium distance depending on the effective charge q, and κ is a dimensionless characteristic of the anharmonic frequency shift caused by the static electric fields of neighboring dipoles. Finally, Davydov splitting obtained as

$$\begin{aligned}\Delta\omega^2(\Theta_0)&=\omega_Z^2-\omega_X^2\\&=\frac{1}{m^*l^2}([V_1^3(0)-V_1^3(\mathbf{K}_J)]\cos^2\Theta_0\\&\qquad-\{\tfrac{1}{2}[V_1^1(0)+V_1^2(\mathbf{K}_J)]\\&\qquad-V_1^1(\mathbf{K}_J)\}\sin^2\Theta_0)\end{aligned}\tag{15}$$

is independent of anharmonic correction being governed by the values of $V_p^\nu(\mathbf{K})$ [see Eq. (13)] in the symmetric points of the first Brillouin zone. For the planar disposition of dipole moments ($\Theta_0=90°$), no Davydov splitting is observed in the spectrum due to the high symmetry of the ground state.

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