

## BKT PHASE TRANSITION IN A 2D SYSTEM WITH LONG-RANGE DIPOLE–DIPOLE INTERACTION

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We consider phase transitions in 2D XY-like systems with long-range dipole–dipole interactions and demonstrate that BKT-type phase transition always occurs separating the ordered (ferroelectric) and the disordered (paraelectric) phases. The low-temperature phase corresponds to a thermal state with bound vortex–antivortex pairs characterized by linear attraction at large distances. Using the Maier–Schwabl topological charge model, we show that bound vortex pairs polarize and screen the vortex–antivortex interaction, leaving only the logarithmic attraction at sufficiently large separations between the vortices. At higher temperatures the pairs dissociate and the phase transition similar to BKT occurs, though at a larger temperature than in a system without the dipole–dipole interaction.

Рассмотрен фазовый переход в двумерной XY-модели с дальнедействующим диполь-дипольным взаимодействием. Показано, что при определенной температуре происходит фазовый переход Березинского–Костерлица–Таулесса (БКТ) из упорядоченного сегнетоэлектрического состояния в неупорядоченное параэлектрическое. В низкотемпературной фазе на фоне однородной поляризации дипольных моментов имеются связанные пары «вихрь–антивихрь», энергия которых линейно возрастает с увеличением расстояния между вихрями в паре. На основе модели топологических зарядов Майера–Швабла показано, что газ связанных пар малых размеров поляризует и экранирует взаимодействие между вихрем и антивихрем в парах большого размера. В результате линейный закон взаимодействия в парах сменяется логарифмическим. Вследствие этого при повышении температуры пары диссоциируют, т. е. происходит фазовый переход, аналогичный переходу БКТ. Температура перехода при этом выше, чем в отсутствие диполь-дипольного взаимодействия. Указанный фазовый переход приводит к одновременному исчезновению как ближнего, так и дальнего порядка в системе диполей.

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Phase transitions and long-range order in 2D systems are intimately related with dynamics of topological excitations (vortices) with logarithmic interactions: the transition occurs when vortex–antivortex pairs dissociate at a certain finite temperature [1–3]. The ground state of the system is not truly ordered (there can be no true long-range order) but superfluid at temperatures below the phase transition point. The transition has been observed in numerous experimental systems, such as weakly interacting ultracold gases [4].

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There are situations though when the microscopic interactions are long-range, as in a case of a system with appreciable dipole–dipole forces. The nature of the phase transition under the circumstances has a long history [5–11]. It should be noted that there is no clear understanding of related phenomena so far [12]. Dipole–dipole interactions are shown to lead to formation of a true long-range order at small temperatures, so that the ground state of a 2D system of magnetic dipoles is a ferromagnet [5]. The spontaneous polarization of the ground state has also been pointed out in [6]. Both works do not clear out the nature of the phase transition, which has been studied first in [13], where the correction to BKT temperature was obtained in the limit when the dipole–dipole interaction is much smaller than the exchange term, and hence the correction to the transition temperature is small. The author made an important observation though: the interaction between the vortices was found to be linear at sufficiently large distances (see [10] for a recent discussion). Ferromagnetic ordering was studied in various model dipole systems theoretically in [7–9] and experimentally in [11]. It was shown in [14, 15] that the ground state of the two-dimensional dipole systems has a striped structure. It should be noted here that the long-range structure of the ground state crucially depends on the short-range form of the dipole lattice [7–9]. The long-range order can be destroyed by the short-range disorder [16, 17] relevant for the liquid state considered in our paper. Arguably, dipole–dipole forces between water molecules on hydrophobic surfaces can lead to macroscopic ordering of the molecular dipoles such as creation of macroscopic hydrogen-bond networks in biological systems [18–20].

In spite of vast volume of the research, the nature of the disordering phase transition appears to be somewhat controversial. There are claims that the long-range dipole–dipole forces change the physics of the phase transition, entirely transforming BKT vortex pairs dissociation transition in a nonpolar system to a deconfinement transition similar to that in quark–gluon plasma [10]. Note that transition temperature is predicted to be four times higher than BKT temperature. Below we perform a systematic study of dipole–dipole interaction influence in 2D systems at finite temperatures. We show that the interaction leads to ferromagnetic ordering at low temperatures without contradiction to the Mermin–Peierls theorem [21] and in a full accordance to the earlier statements. The phase transition itself turns out, as in BKT case, to be associated with dissociation of vortices. We calculate the transition temperature,  $T_C$ , as a function of the interactions parameters. We show that the linear interaction between the vortices does change the transition temperature, though the vortex gas polarization screens the long-range linear potential and transforms it to a logarithmic interaction. Therefore, in a system with dipole–dipole interaction the phase transition is essentially BKT, though the transition temperature itself has a complicated dependence on the model parameters.

At last we apply the developed model to water–solute boundaries using the phenomenological vector model of polar liquids [18, 20]. We show that spontaneous polarization of molecular dipoles next to hydrophobic boundaries may occur and a ferroelectric liquid film may form. The vector model naturally describes topological excitations on the solute boundaries. The dissociation of the vortex pairs can be associated with disappearance of hydrogen bond networks in the course of order–disorder phase transition in the hydration water layer [22–24].

Consider a plane layer of a thickness  $\lambda$  composed of interacting dipoles. The unit vector  $\mathbf{S}(\mathbf{r})$  is taken parallel to the dipole moment of a molecule residing at a point  $\mathbf{r}$  and is characterized by the orientation angle  $\theta(\mathbf{r})$ :  $\mathbf{S} = (\cos \theta(\mathbf{r}), \sin \theta(\mathbf{r}))$ . The Hamiltonian of the

interacting system consists of the two parts:

$$G_S = G_H + G_{dd}, \quad (1)$$

where the short-ranged gradient term gives the energy of a nonpolar system

$$G_H = \frac{1}{2}M \int_{\Gamma} df (\nabla\theta)^2, \quad (2)$$

and the long-range dipole–dipole interaction is represented by the following model term:

$$G_{dd} = \frac{1}{2}K \int_{\Gamma} df df' \frac{(\nabla \cdot \mathbf{S})(\nabla' \cdot \mathbf{S}')}{|\mathbf{r} - \mathbf{r}'|}, \quad (3)$$

where  $df$  is the element of the surface  $\Gamma$ , whereas the constants  $M$  and  $K$  characterize the strength of the interactions. The Hamiltonian has been studied in the limit  $M \gg \lambda K$  in [5,13].

Consider the case of an arbitrary relation between parameters  $\lambda K$  and  $M$ . At small temperatures  $T$  the dipole system is ordered; i.e., all the dipoles point at the same direction, for example, along the  $x$  axis ( $\theta(\mathbf{r}) = 0$ ). To prove that, let us consider the correlation function of the dipole orientation fluctuations:  $K_2(\mathbf{r}) = \langle \theta(0)\theta(\mathbf{r}) \rangle$ ,  $|\theta| \ll 1$ . Keeping quadratic terms in  $\theta$  only and linearizing the Hamiltonian, we find that

$$G_S \approx \frac{1}{2}M \int_{\Gamma} df (\nabla\theta)^2 + \frac{1}{2}K \int_{\Gamma} df df' \frac{\theta_y(\mathbf{r})\theta_y(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|},$$

so that the correlation function takes the form

$$K_2(\mathbf{r}) = T \int d^2k \frac{\exp(i\mathbf{k}\mathbf{r})}{Mk^2 + 2\pi Kk \sin^2 \alpha}, \quad (4)$$

where  $\alpha$  is the angle between wave vector  $\mathbf{k}$  and  $x$  axes. The integral in the r.h.s. converges at small values of  $k$ ; therefore,  $K_2(0)$  is finite and hence the ordered state is thermally stable. In fact, the appearance of the last term in the integrand denominator resolves the contradiction with the Mermin–Peierls theorem [21]. It may even seem that the correlation function integral converges solely due to the finite value of the dipole–dipole interaction strength ( $K \neq 0$ ). The conclusion is in fact wrong: as is clear from Eq. (4), at  $M \rightarrow 0$  the integral diverges at  $\alpha \rightarrow 0$ . It means that both terms in Eq. (1) denominator are equally important. Mathematically, this analysis is in full agreement with a well-known feature of classic electrostatics, the Earnshaw theorem: a system of classical charges interacting with electrostatic forces only can have no stable state [25,26].

To elucidate the nature of the ordered state, let us consider the correlation function at large distances  $r$ . The main contribution to the integral in Eq. (4) comes from the two separate regions:  $|\alpha| \ll 1$  and  $|\alpha - \pi| \ll 1$ , so that

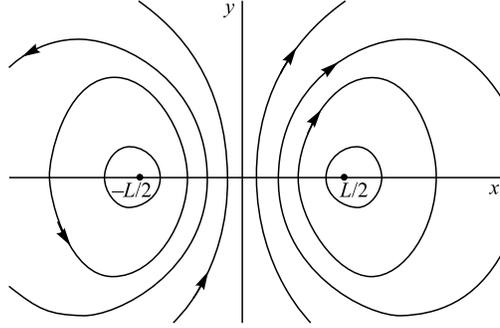
$$K_2(\mathbf{r}) \approx \frac{4T}{M\sqrt{\pi}} \int_0^{\infty} d\tau \cos(x\tau^2) \exp\left(-\frac{|y|\tau^3}{\sqrt{\gamma}}\right) \quad (5)$$

with  $\gamma = 2\pi K/M$ . The calculation in Eq. (5) is simplified in the two following limiting cases:

$$K_2 \approx \frac{\sqrt{2}T}{M\sqrt{|x|}}, \quad |y| \ll \sqrt{\gamma}|x|^{3/2},$$

$$K_2 \approx \frac{4T}{3M\sqrt{\pi}|y|^{1/3}}\Gamma\left(\frac{1}{3}\right), \quad |y| \gg \sqrt{\gamma}|x|^{3/2}.$$

The correlations at large temperatures are addressed in [10] and turn out to decay exponentially. The crossover between the power laws found here for small  $T$  and the exponential decay at large  $T$  suggests a phase transition at some intermediate temperature  $T_C$ .



Polarization configuration  $\mathbf{S}(x, y)$  corresponding to a vortex–antivortex pair (VAP)

To establish the transition temperature, we apply a set of familiar arguments (see, e.g., [2] for further explanations). Thermodynamics of a 2D system can be mapped to thermodynamics of a gas of interacting vortex–antivortex pairs (VAPs) [27]. Consider first the small temperatures limit:  $T < T_C$ ,  $T_C - T \ll T_C$ . The thermal state in this case can be viewed as a gas of bound VAPs. Since the considered temperature is still below the transition point, VAPs approach the dissociation limit and the pairs with large distances  $r$  between the vortices dominate. At such separations the vortex–antivortex interaction is described by the linear term and the energy of the system can be approximated as [10]

$$F \approx 2NE_0 + \frac{1}{2}K \sum_{i,j} q_i q_j (-r_{ij}) = \frac{1}{2} \sum_i q_i \Phi(\mathbf{r}_i), \quad (6)$$

where  $N$  is the number of the VAPs and  $q_j = \pm 1$  are the topological charges associated with the vortices [28]. The «rest energy»,  $E_0$ , is the energy associated with a pair of a minimum possible separation  $r \sim \lambda$ , where  $\lambda$  is the size of the vortex core. The latter quantity is small, model-dependent and depends on microscopic details of the underlying physical system (such as thickness of the layer). The (quasi-electric) potential  $\Phi(\mathbf{r})$  is introduced in analogy with electrostatics and amounts to the energy associated with the interaction of a given vortex (charge) with all other vortices in the system:

$$\Phi(\mathbf{r}) = -K \sum_j q_j |\mathbf{r} - \mathbf{r}_j| = -K \int |\mathbf{r} - \mathbf{r}'| \rho(\mathbf{r}') d^2\mathbf{r}', \quad (7)$$

where  $\rho(\mathbf{r}) = \sum_j q_j \delta^{(2)}(\mathbf{r} - \mathbf{r}_j)$  is the vortex charge density.

The topological charge model (6) seems to be questionable if one takes into account the paper [29] in which the existence of the strings in the vortex–antivortex pair was reported in the case when the pair axis is parallel to the overall polarization vector,  $\varphi = 0$ , where  $\varphi$  is the angle formed by the polarization vector and the axis. At the first sight, it means that the superposition principle behind the expression (6) fails. To study this question, we found numerically the ground state of Hamiltonian (1) on numerous lattices. We found that the conclusion of [29] is correct only in case  $\varphi = 0$ . We turned the vortex–antivortex pair axes and found that the string disintegrates at  $\varphi \neq 0$ . The energy of pair is a function of  $\varphi$  and the distance  $L$  between vortex cores behaves approximately as follows from Eq.(6). It means that the expression (6) is capable of giving, at least, a qualitative answer on the nature of the phase transition in 2D dipole system in accordance with [10]. It is shown below that given (6) it is possible to achieve more definite analytical results than those obtained in [10]. It seems to be important if we also take into account that it is difficult to obtain a clear and robust picture knowing only the results of numerical experiments [12]. We hope that the topological charge model (6) can shed a light on physics of the phase transition in 2D dipole systems.

According to its definition (7), the potential  $\Phi(\mathbf{r})$  satisfies the analogue of the Poisson equation  $\hat{L}_r \Phi(\mathbf{r}) = K\rho(\mathbf{r})$ , where the linear operator  $\hat{L}_r$  is defined so that

$$\hat{L}_r(-|\mathbf{r} - \mathbf{r}'|) = \delta^{(2)}(\mathbf{r} - \mathbf{r}'). \quad (8)$$

Let us follow the electrostatic analogy even further: since the energy of a charge (vortex)  $q$  placed in the external potential  $\Phi$  is  $U = q\Phi$ , the force acting on the charge (vortex) is  $\mathbf{F} = q\mathbf{E}$ , where the quasi-electric field vector  $\mathbf{E} = -\nabla\Phi$  is associated with the potential  $\Phi$  in a normal way.

To find out the energy of a VAP in a self-consistent «electric» field of all other pairs, let us define the «dipole moment» of a pair according to

$$\mathbf{d} = \sum_j q_j \mathbf{r}_j = q_+ \mathbf{r}_+ + q_- \mathbf{r}_- \equiv \mathbf{r}, \quad (9)$$

where  $\mathbf{r} = \mathbf{r}_+ - \mathbf{r}_-$ . Then, the energy of the pair is  $U = q_+ \Phi(\mathbf{r}_+) + q_- \Phi(\mathbf{r}_-) \approx -\mathbf{d} \cdot \mathbf{E}$ . Let us pursue the analogy and calculate first the polarizability  $\alpha_P$  of a single VAP. The dipole moment of a pair in a weak external field  $\mathbf{E}$  is given by a standard relation

$$\langle \mathbf{d} \rangle = \frac{\int d\mathbf{f} \mathbf{r} \exp\left(\frac{\mathbf{r} \cdot \mathbf{E}}{T} - \frac{K}{T} r\right)}{\int d\mathbf{f} \exp\left(\frac{\mathbf{r} \cdot \mathbf{E}}{T} - \frac{K}{T} r\right)} \approx \alpha_P \mathbf{E}, \quad (10)$$

where  $\alpha_P = T/K^2$  is nothing else but the pair polarizability.

At the transition temperature,  $T = T_C$ , VAPs begin to dissociate. It means that at  $T_C - T \ll T_C$  only a small fraction of the pairs are very large and close to dissociation. For this reason, it is possible to neglect the interactions between the largest VAPs and calculate the energy of single large pair approaching its dissociation limit in a cloud of comparatively small bound VAPs. As we have demonstrated above, the bound pairs are polarizable and therefore the field of a charge is screened by the polarization of VAPs gas, thus influencing

the potential energy of a large VAP. To find out how exactly, let us consider the effect of shielding of a probe point charge  $Q$  or the charge density  $\rho_Q = Q\delta^{(2)}(\mathbf{r})$  placed at the origin. The complete «electrostatic» potential  $\Phi(\mathbf{r})$  is produced both by the probe charge and the polarization charges of VAPs. The density  $\rho_P$  of the polarization charges is given by usual expressions following from Eq.(9):  $\rho_P = -\nabla \cdot \mathbf{P}$ , where  $\mathbf{P}$  is polarization of the vortex gas:

$$\mathbf{P} = n_P \langle \mathbf{d} \rangle = \chi \mathbf{E} = -\chi \nabla \Phi, \quad (11)$$

with  $\chi = \alpha_P n_P$  and  $n_P$  being the «dielectric susceptibility» of the gas of VAPs and the concentration of the pairs, correspondingly.

Combining Eqs.(8), (11) and (7), we obtain the complete equation for  $\Phi(\mathbf{r})$ :

$$\hat{L}_{\mathbf{r}} \Phi(\mathbf{r}) = K(\rho_Q + \rho_P) = KQ\delta^{(2)}(\mathbf{r}) + K\chi \Delta \Phi(\mathbf{r}). \quad (12)$$

Since the Fourier component of the operator  $\bar{L}_{\mathbf{r}}$  is  $L_{\mathbf{k}} = k^3/2\pi$ , we find that

$$\Phi(\mathbf{r}) = QK \int \frac{d^2k}{(2\pi)^2} \frac{\exp(i\mathbf{k}\mathbf{r})}{\frac{k^3}{2\pi} + K\chi k^2}. \quad (13)$$

At large distances,  $r \gg r_0$ , where  $r_0 = 1/2\pi K\chi$ , most contribution to the integral comes from small values of  $k$ , where  $k^3$  term in the denominator is negligible. Therefore, the potential of the «charge» at large distances is logarithmic:  $\Phi(\mathbf{r}) = -(Q/2\pi\chi) \log(r/C_1)$ ,  $r \gg r_0$ , where  $C_1 \sim r_0$ . In the opposite limit,  $r \ll r_0$ , the potential is linear:  $\Phi(\mathbf{r}) \approx -Kr$ ,  $r \ll r_0$ . Below we propose a simple expression interpolating between the two results:  $\Phi(\mathbf{r}) \approx -(Q/2\pi\chi) \log(1 + r/r_0)$ , so that the energy of a large pair of a size  $R$  is given by

$$E_{\text{VAP}}(R) \approx 2E_0 + \frac{1}{2\pi\chi} \log\left(1 + \frac{R}{r_0}\right). \quad (14)$$

We note that although the dipole–dipole interactions do change the interactions between the vortices at small distances, the polarization of the VAPs destroys the linear attraction at large separations between the vortices and hence the phase transition associated with the dissociation of the pairs is qualitatively very similar to BKT transition in a nonpolar system.

Standard calculation of BKT temperature for a vortex gas with interaction (14) gives the following implicit equation for the transition temperature  $T_C$ :

$$T_C^2 = \frac{K^2}{12\pi n_P(T_C)}. \quad (15)$$

The density  $n_P$  at the transition temperature can be calculated from the following argument. At  $T \approx T_c$  the VAPs just start to dissociate and therefore the size of a typical pair is small and the interaction between the vortices is still linear and unscreened and  $n_P(T) = 2\pi \exp(-2E_0/T) (T^2/\lambda^4 K^2)$ . Therefore, Eq.(15) for the transition temperature takes the form

$$2\pi\sqrt{2} \left(\frac{T_C}{\lambda K}\right)^2 = \exp\left(\frac{E_0}{T_C}\right). \quad (16)$$

From this equation it is clear that its solution always exists and is unique.

Let us assume first that the dipole–dipole interactions are small, i.e.,  $\lambda K \ll M$ . Since the energy of a minimal vortex–antivortex pair  $E_0 \sim M$ , the transition temperature  $T_c \sim M$ , which coincides with the standard BKT result ( $T_{\text{BKT}} = \pi M/2$ ). This is precisely the limit considered in [13]. In the opposite limit,  $M \ll \lambda K$ , the exponent in the r.h.s. of Eq. (16) is approximately 1 and hence  $T_C \sim \lambda K \gg T_{\text{BKT}}$ . The phase transition temperature is indeed larger than BKT temperature, in agreement with [10], although our analysis suggests that  $T_C$  can be arbitrarily larger than  $T_{\text{BKT}}$ .

Layers of water molecules on hydrophobic surfaces such as biological membranes or large biomolecules are another interesting example of 2D systems with dipole–dipole interactions. The orientations of molecules next to a macromolecular surface depend on interplay of long-range interactions of the molecular dipoles and the short-range hydrogen bonds between the adjacent molecules. Since an energy of uncompensated hydrogen bond is large compared to the temperature, no water molecules can point their dipole moments in the direction of a hydrophobic surface, and thus all the water molecules next to the surface have their dipole moments parallel to the surface. Macroscopic polarization of the molecules  $s(\mathbf{r}) = \langle \mathbf{S}(\mathbf{r}) \rangle$  vanishes quickly in the direction of the liquid bulk. Therefore, a water layer next to a hydrophobic boundary can be studied with the help of a model Hamiltonian (1) corresponding to a 2D system of interacting dipoles (see [18,20] and references therein). According to the findings of the current work, the layer of water molecules is completely polarized,  $s \approx s_{\parallel}$ , at very low temperatures. Microscopic parameters of water are such that  $M \sim \lambda K$  and therefore the macroscopic polarization and molecules orientation ordering disappears at temperature  $T_C \sim M \sim E_H s_{\parallel}^2$ , where  $E_H \approx 2500$  K is the characteristic energy of a hydrogen bond. The expression suggests that the transition temperature corresponding to a completely hydrophobic surface ( $s_{\parallel} \approx 1$ ) is always very large, the water molecules are ordered at all realistic temperatures and thus there always exists a macroscopic hydrogen bonds network on the surface of the body. If a surface is partly hydrophilic, i.e., hydrogen bonds donors or acceptors, such as charges,  $s_{\parallel} < 1$  and at  $s_{\parallel} = 0.3$  the transition may occur even at room temperatures. The evidence of such transitions is observed in molecular dynamics calculations [22–24]. So, the presented here simplified model seems to succeed in description of hydration water properties.

It seems to be useful to elucidate the conclusion about the 2D character of phase transition considered here. It immediately follows from expressions (1)–(3) that contain only the surface integrations. As was stated above, at temperatures close to the phase transition point  $T_C$  the free energy part (1) that originates from polarization of polar liquid is dominated by the contribution from VAPs with large distances between their vortices. In any case these distances are large compared with the layer thickness  $\lambda$ . It is the physical reason for the 2D behavior. In this case the starting expression [18,20] for the free energy that is composed of integrals over liquid bulk comes to the surface integrals (1)–(3).

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