CONFERENCES AND SYMPOSIA

New effects in and control of exciton systems in quasi-two-dimensional structures

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Abstract. New effects in systems of quantum dipoles are discussed, such as anisotropic superfluidity in external fields, strong correlations, crystallization, and the supersolid phase. Roton instability effects typical of strongly correlated Bose systems but also manifesting themselves in weakly interacting systems of titled dipoles are analyzed. Among the interesting physical realizations of the systems under consideration are dipole excitons in single or coupled quantum wells under a strong transverse electric field and in van der Waals heterostructures of new 2D materials, such as transition metal dichalcogenides (TMDCs). The use at ultralow temperatures of polar molecules or atoms with permanent or external-field-induced dipoles is also interesting, as are Rydberg atoms in an external electric field.

We discuss a number of new effects in quantum dipole systems [1–3] (see also [4]) or, more specifically, anisotropic superfluidity in external fields, strong-correlation effects, and crystallization. We also consider the effects of roton instability that are typical of strongly correlated Bose systems but manifest themselves in weakly interacting systems of tilted dipoles.

Dipole systems are implemented in the following ways. First, they are realized as dipole excitons in coupled quantum walls with spatially separated electrons and holes in coupled quantum wells or in a single quantum well in a strong transverse electric field. A system of excitons with spatially separated electrons and holes was considered for the first time in 1973 in Lozovik's report [5]. Later, the superfluidity of that system and its other unusual physical properties were explored in detail in [6–15] and subsequent studies that predicted and thoroughly investigated superfluidity in systems of spatially separated electrons and holes, Josephson-type effects (in systems without superfluidity!) [16–21], drag of excitons by elec-

trons and the control of excitons using electrons [22–24], behavior in external magnetic fields [25–36], unusual coherent optical linear and nonlinear properties [37–42], and a phase diagram for spatially separated electron–hole systems and dipole excitons [43-46]. As a result, a promising new area emerged in the physics of coherent phenomena in systems of excitons in quantum wells and new 2D systems. There are also new and interesting accomplishments in theory and computer simulations; a number of experimental teams have obtained significant experimental results [47–60].

In a system of spatially distributed electrons and holes, implementation with a high density of electrons and holes is also possible. In the case of high density, 2D electrons and holes create Fermi circles or segments, and, if those Fermi circles are almost congruent, a coherent state similar to the Bardeen–Cooper–Schrieffer (BCS) state with a spectrum gap due to coupling of spatially separated electrons and holes can emerge as the temperature is decreased [6–14]. If those Fermi lines for 2D systems are not fully congruent, a BCS-type coherent state emerges under the condition that the emerging gap (which determines the energy gain in the e – h coupling) is larger than the energy difference between the Fermi lines of electrons and holes. Tunneling between layers results in fixing the order parameter phase and the occurrence of the internal Josephson effect [16–21]. The main difference between the two-layer system and the 3D uniform system with the coupling of electrons and holes considered by Keldysh and Kopaev [61], which describes semimetal–insulator transitions, etc., is that superfluid currents can emerge in the former system. The predicted superfluidity was confirmed in the Eisenstein group experiments for double electronic layers in a strong magnetic field with half-filled Landau levels [62].

The dipole systems considered in this review also include excitons in (now popular) van der Waals heterogeneous structures consisting of two layers of new materials, such as transition metal chalcogenides (see [63, 64] and the references therein) or graphene with a created gap (see a discussion of the e – h coupling in dense spatially separated 2D materials in [65–71] and the references therein). Structures based on transition metal chalcogenides are also promising from the standpoint of attaining high-temperature superfluidity of excitons.

Of significant interest are also realizations of dipole polar molecules and atoms with permanent dipoles or those induced by external fields at superlow temperatures [72–74]. Some interesting features are exhibited by systems of Rydberg atoms in an electric field, where very large dipole moments occur such that strong correlations and possibly crystallization occur even in a system of relatively rarefied Rydberg atoms.

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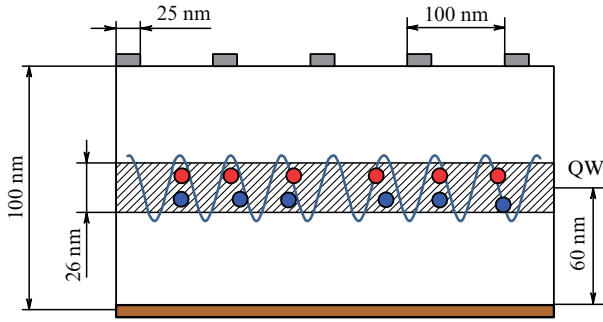E-mail: lozovik@isan.troitsk.ru

Figure 1. Possible experimental realization of a system with anisotropic supefluidity: a quantum well (QW) (or coupled quantum wells or a van der Waals structure consisting of two 2D transition metal dichalcogenides) with dipole excitons controlled by the potential of external electrodes.

Figure 2. An external force pushes the system in one direction (momentum **P**) while the current $\mathbf{J}_{sys}$ in the anisotropic system under consideration flows in another direction.

Next, for the 2D dipole systems described above, we consider how their properties can be controlled and the anisotropic superfluidity in external periodic fields created by specially shaped electrodes, as we predicted in [1] (Fig. 1). We note that the anisotropic superfluidity was considered previously for He^3 [75–77] and cooled atoms in optical superlattices [78, 79].

It is known that a persistent quasi-equilibrium current **J** can be created in a superfluid system, the value of that current being proportional to the total momentum of the system **P** with the proportionality coefficient equal to the absolute value of the helicity Y_s. The last quantity is, in turn, proportional to the density n_s of the superfluid component:

$$\frac{\mathbf{J}(T)}{S} = Y_s(T)\mathbf{P} = \frac{n_s(T)}{m}\mathbf{P}, \tag{1}$$

where S is the area of the quasi-2D systems under consideration. Similarly, the total momentum **P** is also proportional to the superfluid density and the system velocity **v**:

$$\frac{\mathbf{P}(T)}{S} = \rho_s(T)\mathbf{v} = n_s(T)\,m\mathbf{v}. \tag{2}$$

In an anisotropic system, the density of the superfluid component does not have its usual meaning, because it is no longer a scalar but rather a tensor, such that

$$\frac{\mathbf{P}_i(T)}{S} = \sum_j \rho_s^{ij}(T)v_j, \tag{3}$$

which means that as a result of an external effect, the system gains momentum in one direction but moves in another direction (Fig. 2).

To make the task simpler, we consider an exciton system with weak interaction at a zero temperature. The condensate depletion is in this case small, and the number of particles above the condensate is also small compared to the condensate density. The Gross–Pitaevskii equation for a periodic external field can therefore be used for the condensate part. To solve the equation, we used two approaches: (1) the Gross–Pitaevskii functional in a periodic external field was numerically minimized and (2) an analytic solution was found in the second order of perturbation by the periodic external field.

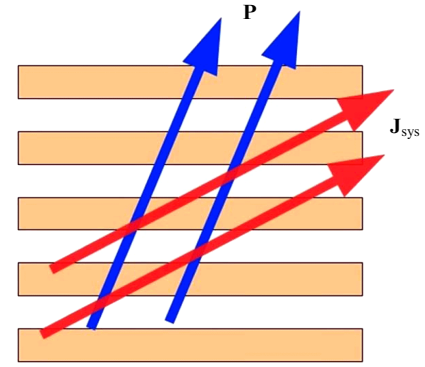If the field amplitude is not too large, the analytic results obtained in the second order of the perturbation theory by a periodic field agree well with numerical results. To calculate the anisotropic properties of the excited system (in particular, the anisotropic speed of sound), we diagonalized the above-condensate Hamiltonian in the Bogoliubov approximation. The excitation spectrum exhibits Bloch discontinuities at momenta equal to the inverse period of the periodic field. The anisotropy of all physical quantities rapidly increases in the vicinity of those discontinuities.

The angular distribution of the recombination radiation from the 2D system of dipole excitons in a condensed state, which is considered here, differs from that for a homogeneous system: in addition to the beam perpendicular to the 2D system, due to periodicity, additional lateral beams appear whose intensity is proportional to the depth of modulation of excitons obtained using controlling electrodes. Moreover, the transverse section of the luminescent beams emitted by the exciton system in a direction other than the normal to the system plane acquires an elliptic shape. The vortices in the system also feature anisotropic properties.

We note that if the periodic external field is strong enough in the system under consideration (see Fig. 1), narrow superbands for excitons can emerge, and a phase transition from the superfluid state to the glassy state can occur, which was theoretically studied in [4].

We now consider strong-correlation effects and new exciton phases. In our studies [43–46], we predicted and used the quantum Monte Carlo method to explore new phases of the exciton system: a crystal phase [80–84] (see also [85]) and a supersolid phase [86] (which simultaneously exhibits transverse rigidity, superfluidity, and diagonal and nondiagonal order); both the structure and optical properties have been studied. The quantum Monte Carlo method was used to confirm the existence of supersolids in mesoscopic exciton systems [87, 88].

In a 2D dipole system, as the dimensionless concentration of particles nr_0^2 increases (where $r_0 = me^2 D^2/\varepsilon\,(2\pi\hbar)^2$ is the distance at which the quantum kinetic energy is equal to the dipole–dipole interaction energy, D is the characteristic distance between the electron and hole that form a dipole, and ε is the dielectric permittivity), strong correlation effects occur: depletion of the condensate, the onset of short-range order, significant modification of the excitation spectrum, and emergence of the roton minimum at $nr_0^2 = 16$ (Fig. 3).

At $nr_0^2 = 290$ (the Lindeman parameter equal to 0.23), crystallization occurs in the dipole system [80–82] (Fig. 4).
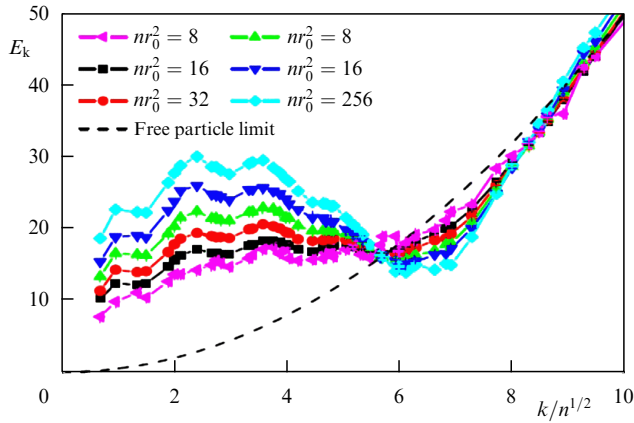
Figure 3. Modification of the excitation spectrum and emergence of the roton minimum in a 2D dipole system as the dimensionless concentration nr_0^2 increases (see [80–82]).
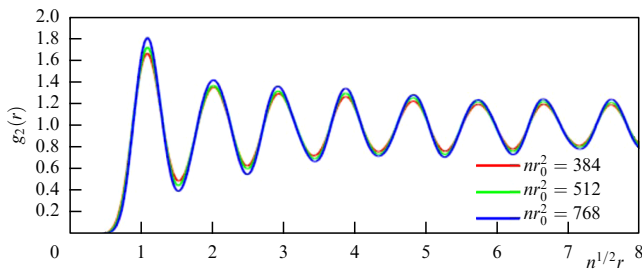

Figure 4. Pair distribution function in a 2D dipole system in the crystalline phase (see [80–82]).

Figure 5. Qualitative view of the excitation spectra squared of dipole excitons in 3D (solid curve), 2D (dashed-dotted curve), and quasi-2D (Q2D) (dotted curve) cases. The widths of the wells in Fig. c: $L_1 < L_2 < \ldots < L_6$. The arrow in Fig. d shows the momentum $\mathbf{p}_i$ of the unstable mode.

Interesting structural features of the dipole system are clearly exhibited near the instability threshold, where the roton gap in the excitation spectrum is close to zero. However, the roton minimum for perpendicular dipoles may not touch zero because the condensate depletion diverges when the spectrum touches the zero energy. Stated differently, the condensate vanishes prior to the spectrum touching zero energy, and the system switches to a strong correlation regime. Due to this, all the structural properties that can be predicted for perpendicular dipoles at the roton instability threshold are not attainable.

However, the condensate depletion divergence at the threshold where the spectrum touches zero energy can be removed if the dipoles are tilted, and the rotational symmetry in the layer plane is broken as a result. Namely, the spectrum touches zero in this case not on a circle but only at two points. Therefore, in the case of tilted dipoles, the condensate depletion divergence at the point where the spectrum touches zero energy vanishes, thus making the roton instability threshold attainable. Owing to this, broad perspectives open for studying the structural properties of the dipole condensate near the roton instability threshold.

We have predicted the roton instability effect [2, 3] for *weakly interacting* tilted dipoles in a 2D homogeneous quantum layer. In the case under consideration, the roton effects typical of quantum systems in the strong-correlation regime are observed in a *weakly interacting* gas. It is important that in contrast to the rotation symmetry of a system of dipoles perpendicular to the dipole plane, the rotation symmetry for a system of *tilted* dipoles is broken,
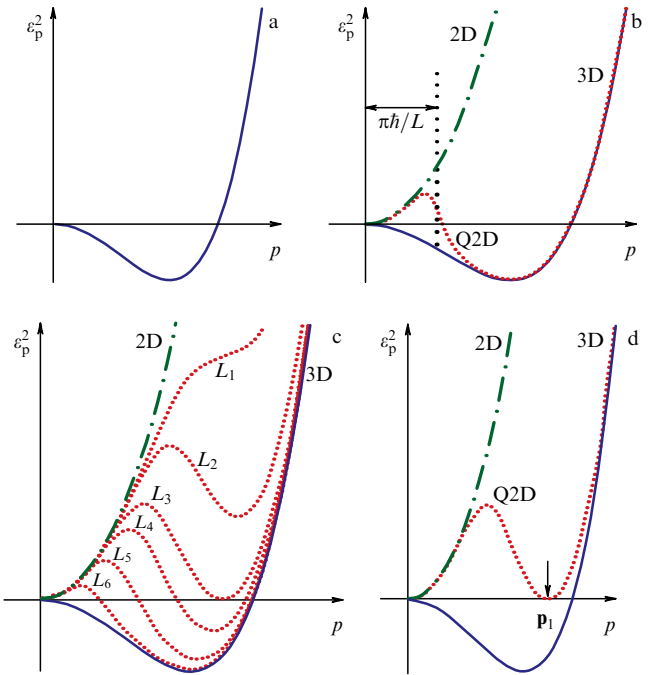
and hence the condensate depletion is finite up to the roton instability threshold, and the mean-field approach is applicable. In [2, 3], we proposed methods to detect those phenomena in the system of dipole excitons and ultracold atoms and polar molecules in optical lattices and estimated optimal experimental parameters.

We have considered two regimes in which the roton instability can be realized in a system of tilted dipoles: a 2D system in a quantum well and a quasi-2D system in a broad quantum well.

To better understand the nature of the roton instability of tilted dipoles in qualitative terms, we put forward two arguments. The first is related to the system of parallel dipoles located in a plane. This system can form a 2D crystal consisting of parallel chains [89], because the dipoles in a 'head-to-tail' arrangement attract (a stable state emerges owing to the repulsion of dipole cores.)

The second argument refers to a broad well. The potential energy of interaction between dipoles is

$$V_{\mathrm{d}}(\mathbf{r}) = \frac{d^2}{\varepsilon} \frac{r^2 - 2z^2}{r^5} \,, \qquad (4)$$

and its Fourier transform is

$$V_{\mathrm{d}}(\mathbf{p}) = \frac{4\pi}{3} \frac{d^2}{\varepsilon} \frac{2p_z^2 - p^2}{p^2} \,, \qquad (5)$$

where $d = eD$ is the dipole moment, ε is the dielectric permittivity of the semiconductor, $\mathbf{p} = \{\mathbf{P}, p_z\}$ and $\mathbf{r} = \{\mathbf{R}, z\}$ are 3D vectors, $\mathbf{P}$ and $\mathbf{R}$ are 2D vectors (in the plane of the well), and $p = |\mathbf{p}| = (\mathbf{P}^2 + p_z^2)^{1/2}$.

The roton instability occurs as a result of negative values of the dipole–dipole potential $V_{\mathrm{d}}(\mathbf{p})$ at $|p_z| \ll p$ (Fig. 5), which
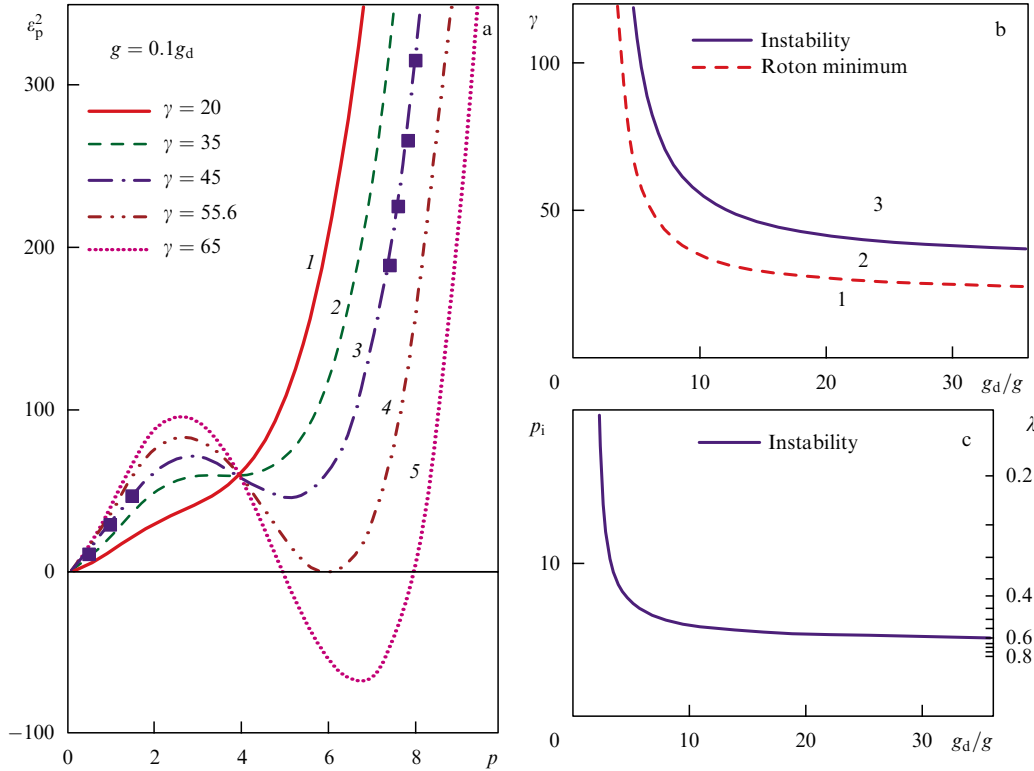
Figure 6. (a) Variational calculation of the lowest dispersion curves ε_p^2 for $g = 0.1 g_d$ at $\gamma = 20$ (curve 1), $\gamma = 35$ (curve 2), $\gamma = 45$ (curve 3), $\gamma = 55.6$ (curve 4), and $\gamma = 65$ (curve 5). (b) Phase diagram of the roton instability and roton minimum threshold in terms of variables $(g_d/g, \gamma)$. The stable phases without (1) and with (2) the roton minimum and the unstable phase (3) are shown. (c) The critical momentum for the instability threshold $\mathbf{p}_i$ and the corresponding period of matter density waves λ as a function of g_d/g. The following system of units is used: $\hbar = m = L = 1$; $g_d = 8\pi d^2/(3\varepsilon)$ and g are the dipole and van der Waals coupling constant, and γ is a dimensionless parameter proportional to the Bose–Einstein condensate density times the dipole coupling constant.

is a consequence of attraction between the dipoles that are transverse to the quantum well plane. Indeed, the momenta $|p_z| \ll p$ correspond to spatial scales $|z| \gg r$, and the dipoles attract at $|z| \gg r$ [see (4)]. As a result, the square of the Bogoliubov spectrum $\varepsilon_\mathbf{p}^2 = p^4/(4m^2) + V_d(\mathbf{p})(n_0/m)\mathbf{p}^2$ at $|p_z| \ll p$ in an infinite homogeneous 3D system is negative at small momenta,

$$\varepsilon_\mathbf{p}^2 = \frac{p^4}{4m^2} - \frac{4\pi}{3}\frac{d^2}{\varepsilon}\frac{n_0}{m}\mathbf{p}^2, \qquad (6)$$

where n_0 is the 3D density of the Bose condensate and m is the particle mass (see Fig. 5). Therefore, at small momenta (in the phonon region) spectrum (6) contains a segment of imaginary energies, which means that the phonon modes in the 3D system are unstable as a result of the formation of a 3D crystal consisting of chains (as the results of our computer simulation show) (see also [90]).

However, the dipoles in the quantum well are restricted in the direction of the z axis:

$$0 \leqslant |x|, |y| < \infty, \qquad 0 < z < L, \qquad (7)$$

where L is the width of the quantum well. Therefore, at small scales

$$r \ll L, \qquad p \gg \frac{\pi\hbar}{L}, \qquad (8)$$

i.e., deep inside the well, the excitons treated as pointlike dipoles move as if in three dimensions. At the same time, at

large longitudinal scales,

$$r \gg L, \qquad p \ll \frac{\pi\hbar}{L}, \qquad (9)$$

or at $r \gg z$ and $p \ll |p_z|$, the quantum well is similar to a thin layer, and motion occurs as in two dimensions. The 2D regime, $r \gg L$, apparently exists for any L [see (7)]. However, the 3D regime ($p \gg \pi\hbar/L$ or $L \gg \pi\hbar/p$) is only possible in a well whose width is larger than a certain value

$$L \gg \frac{\pi\hbar}{\sqrt{2m\mu}}, \qquad (10)$$

where μ is the chemical potential of excitons and $\sqrt{2m\mu}$ is the momentum p characteristic of the problem. It is the case of a broad well, which encompasses both the 2D and 3D regimes, that corresponds to the quasi-2D system.

We consider quasi-2D dipoles in more detail. At large momenta $p \gg \pi\hbar/L$, when the 3D regime is implemented, the excitation spectrum is close to the 3D branch (Fig. 5b). At $|p_z| \ll p$, the 3D branch is unstable. At small momenta $p = \pi\hbar/L$, for which the 2D regime is implemented, the excitation spectrum is close to its 2D branch (Fig. 5b). The 2D branch is stable because the dipoles separated by long distances repel at $r \gg z$. At intermediate momenta, $p \sim \pi\hbar/L$, a crossover occurs from the 3D (unstable) branch to the 2D (stable) branch (Fig. 5b). This means that the instability shifts to the region of intermediate momenta in the 2D case. The broader the well (larger L) is, the larger the instability region (Fig. 5c). The narrower the

well (smaller L), the smaller the instability region. At some value of L ($L = L_3$ in Fig. 5c), the instability region collapses into a point, such that at smaller L ($L = L_{1,2}$ in Fig. 5c) the quasi-2D dipole spectrum can only have a roton minimum. The critical value of L ($L = L_3$) at which the roton minimum touches zero energy (Fig. 5c) corresponds to the roton instability threshold of quasi-2D dipoles. Immediately after the threshold, where the lowering roton minimum just crosses zero, the spectrum squared $\varepsilon_{\mathbf{p}}^2$ for a certain mode $\mathbf{p} = \mathbf{p}_i$ becomes negative (Fig. 5d). Due to this, the energy of the $\mathbf{p}_i$ mode becomes negative: $\varepsilon_{\mathbf{p}_i} = \pm i\hbar\kappa$ ($\kappa \neq 0$, $\kappa \approx 0$). As a result, the plane wave corresponding to that unstable mode $\exp\left[i\mathbf{p}_i\mathbf{R}/\hbar \pm i\varepsilon_{\mathbf{p}_i}t/\hbar\right] \propto \exp\left(\kappa t\right)$ ($\kappa > 0$) starts exponentially increasing with time. After some time elapses, the wave becomes microscopic, and spontaneous self-organization occurs: macroscopic population with nonzero momentum — a matter density wave — appears. The system rearranges in the process of self-organizing: the homogeneous, weakly correlated Bose-condensed gas transforms into a crystal-like periodic structure. A more detailed analysis shows that the supersolid phase can emerge in the system where crystalline order and superfluidity coexist. A new phase, the quantum liquid crystal coexisting with superfluidity, is also possible.

Thus, the emergence of the roton minimum, roton instability, and periodic density profile is a consequence of (a) anisotropy and the existence of a positive-valued region in the dipole–dipole potential and (b) the property of quasi-two-dimensionality, i.e., a regime intermediate between unstable three-dimensionality and stable two-dimensionality. Variational calculations of the corresponding dispersion curves and a phase diagram of the system are shown in Fig. 6.

Effects similar to those considered for 2D systems of quantum dipoles are also typical of 2D systems of higher multipoles. In particular, with the increase in density, quantum crystallization occurs in a 2D system of parallel quadrupoles [91]. In a 2D system of tilted quadrupoles, the roton instability and emergence of superfluidity are also possible. The list of interesting physical realizations of quadrupoles includes 3D excitons or Rydberg atoms in a strong magnetic field compressing the particle transversely to the field and ions in superstrong magnetic fields in the core or vicinity of neutron stars.

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