

Bound Dimers in Bilayers of Cold Polar Molecules

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Abstract. - The exploration of cold polar molecules in different geometries is a rapidly developing experimental and theoretical pursuit. Recently, the implementation of optical lattices has enabled confinement in stacks of planes, the number of which is also controllable. Here we consider the bound state structure of two polar molecules confined in two adjacent planes as function of the polarization angle of the dipole moment of the molecules. We present evidence for the existence of bound states for arbitrary dipole moments and polarization angles in this two-dimensional geometry. The spatial structure of the bound states is dominated by two-dimensional *s*- and *p*-waves, where the latter exceeds 40 percent over a large range of polarization angles for intermediate or strong dipole strength. Finally, we consider the influence of the dimer bound states on the potential many-body ground-state of the system.

Introduction. – A strong experimental drive in the field of polar atoms and molecules has realized controllable samples in the rotational and vibrational ground-state that are close to quantum degeneracy [1–6]. These heteronuclear systems have a number of very interesting properties due to the long-range and anisotropic dipole-dipole force which can give rise to highly non-trivial many-body states in both the weak- and strong-coupling regime [7, 8]. The attractive head-to-tail configuration can, however, lead to collapse of the system [9], and confinement in optical lattices has been suggested as a means of avoiding this problem [10]. These confined one- or two-dimensional geometries have led to a number of predictions of novel few- and many-body states [10–18], and very recently the first experimental implementation of a multilayered stack of pancakes containing fermionic polar molecules was reported [19].

Here we consider the case of two adjacent layers. However, even in this seemingly simple case there is a competition of intra- and interlayer interactions which can vary between repulsion and attraction as one changes the polarization angle of the dipole moments with respect to the layers. In the present paper we will be concerned with few-body states with one particle in each layer in order to describe the simplest complex in such a system in detail.

The case of dipoles oriented perpendicular to the layers was considered from the few-body bound state and scattering point of view in previous works [20–23]. At the so-called ‘magic’ angle where the intralayer repulsion vanishes in a one-dimensional trap the few-body bound state structure was also discussed [24]. However, to our knowledge no paper has addressed the full two-body bound-state problem as a function of the polarization angle and the dipole moment. This problem is highly non-trivial due to (i) the anisotropy and (ii) the vanishing integral over space of the potential for arbitrary polarization angle.

We solve for the bound states using a powerful stochastic variational approach from which we can get energies and expectation values of relevant operators. One of our main results is that the bilayer system has a bound state for *any* polarization angle and *any* value of the dipole moment. We also calculate a partial-wave decomposition that characterizes the geometric structure of the wavefunction which indicates the likely symmetries of the corresponding many-body problem. As a first application of our results in a many-body context we consider the limit of strong coupling where the system forms bound bosonic dimers that can potentially form a (quasi)-condensate.

Theoretical Model. – The potential of two dipoles in layers separated by a distance d has the following form in the ideal limit of zero-width layers

$$V(x, y) = D^2 \frac{\rho^2 + d^2 - 3(x \cos \theta + d \sin \theta)^2}{(\rho^2 + d^2)^{5/2}}, \quad (1)$$

where $\rho^2 = x^2 + y^2$, $x = \rho \cos \phi$ and $y = \rho \sin \phi$, D is the dipole moment¹, and θ denotes the polarization angle measured from the layer plane to the z -axis which intersects the two layers at right angles. Thus, for $\theta = \pi/2$, the dipoles are oriented perpendicular to the layers as in [20, 22, 23]. One can take corrections to the zero-width layer limit into account by integrating out a gaussian in the transverse direction. However, the corrections are second-order in the width, w , and we neglect them as we are interested in the $w \ll d$ limit. We solve the 2 dimensional (2D) Schrödinger equation with the potential in Eq. 1, i.e.

$$\left[-\frac{1}{2} \left(\frac{d^2}{d\tilde{x}^2} + \frac{d^2}{d\tilde{y}^2} \right) + \frac{Md^2}{2\hbar^2} V(x, y) \right] \Psi = \tilde{E} \Psi, \quad (2)$$

where \tilde{x} and \tilde{y} are measured in units of d , M is the molecular mass and $\tilde{E} = Md^2E/2\hbar^2$. The factor of 2 on the potential and energy comes from the reduced mass. The bound state energy is now a function of the dimensionless dipolar strength quantity $U = MD^2/(\hbar^2d)$, which is the ratio of potential to kinetic energy. We will also consider the case where $U < 0$ which is also physically realizable as explained below. Since the potential is in general anisotropic the wave equation is not easy to solve by discretization or integration. We therefore turn to the stochastic variational approach using gaussian wavefunctions which has been successfully applied to other interactions [25].

The potential has the peculiar property that $\int dx dy V(x, y) = 0$ for any θ . In particular, it does not fulfil the Landau criterion for bound states in two dimensions [26]. In the spherically symmetric case, $\theta = \pi/2$, an early existence proof was given in [27]. Here we are interested in the appearance and properties of bound states for arbitrary θ . A partial-wave decomposition of the potential in the basis $\{1, \cos \phi, \cos(2\phi)\}$ (which are the only non-zero terms) leads to

$$V_0(\rho) = U \frac{[3 \sin^2 \theta - 1][\rho^2/2 - 1]}{(\rho^2 + 1)^{5/2}}, \quad (3)$$

$$V_1(\rho) = -3U \frac{\rho \sin 2\theta}{(\rho^2 + 1)^{5/2}}, \quad (4)$$

$$V_2(\rho) = -\frac{3}{2}U \frac{\rho^2 \cos^2 \theta}{(\rho^2 + 1)^{5/2}}, \quad (5)$$

which we will refer to as monopole, dipole, and quadrupole terms, respectively. Here ρ is measured in units of d . Notice that for $\theta = \theta_c$ where $\sin^2 \theta = 1/3$, the monopole term vanishes. The dipole term only vanishes for $\theta = 0$

¹In SI units we have $D^2 = d^2/4\pi\epsilon_0$ when d is dipole moment of the molecules. However, in this letter we use d to denote the interlayer distance.

and $\pi/2$, whereas the quadrupole term is finite except at $\theta = \pi/2$. Thus for $\theta > \theta_c$ and $U > 0$, the monopole term has an inner attractive pocket and a repulsive barrier outside $\rho/d = \sqrt{2}$, and vice versa for $\theta < \theta_c$. For $U < 0$ the story is reversed. We expect the monopole term to be the most important one for determining the system properties, at least when it is non-vanishing away from $\theta = \theta_c$. However, the monopole term is, except for the factor of $3 \sin^2 \theta - 1$, identical to the full potential at $\pi/2$, i.e. we know from previous work that it always supports bound states [22, 23, 27]. We also know that the configuration with an attractive inner pocket and a repulsive outer barrier leads to considerably stronger binding than in the reversed case [22]. We will see this explicitly in the energies presented below.

It is very important to notice that the angle θ_c is different from the magic angle where the potential of two dipoles moving in one dimension vanishes (determined by $\cos^2 \theta = 1/3$) [8]. This demonstrates an important difference between one- and two-dimensional dipolar systems. We will address this fact in more detail when we discuss many-body physics below.

The bound state wavefunction, Ψ , in spherical coordinates can be decomposed into partial waves,

$$\Phi(\rho, \phi) = \sum_m \Psi_m(\rho) \cos(m\phi) \quad (6)$$

$$\Psi_m(\rho) = \frac{1}{(1+\delta_{m0})\pi} \int_0^{2\pi} d\phi \cos(m\phi) \Psi(\rho, \phi), \quad (7)$$

where the corresponding contribution of the $\sin(m\phi)$ terms is zero since the wavefunction has the same $y \rightarrow -y$ invariance as the potential. From $\Psi_m(\rho)$ we define the probability as

$$P_m = \pi(1 + \delta_{m0}) \int_0^\infty d\rho \rho |\Psi_m(\rho)|^2, \quad (8)$$

that gives the total contribution of each partial wave, normalized as $\sum_m P_m = 1$.

When U becomes small we expect universal behavior of energies and radii [28, 29]. Using the stochastic variational approach in the small U limit, our results for $\theta = \pi/2$ approach the universal behavior of the energy which to leading order scales like $\text{Log}(-E) \propto -1/U^2$ as discussed previously [22, 23, 27]. For other values of θ we expect the same scaling for very small U , however, the range of U around zero where this applies will very likely have a strong dependence on θ as indicated by the energies presented below. We also expect differences for general θ between positive and negative U in the limit $U \rightarrow 0$ as for $\theta = \pi/2$ [22]. The questions of critical behavior of the binding energy and approach to universality need to be investigated in more details.

Energies and Wavefunctions. – The energies have been calculated using the correlated gaussian approach. In Fig. (1) we exhibit the results as a function of $U > 0$ for a selection of polarization angles. At small U the energy decreases very fast with decreasing U as noted already in

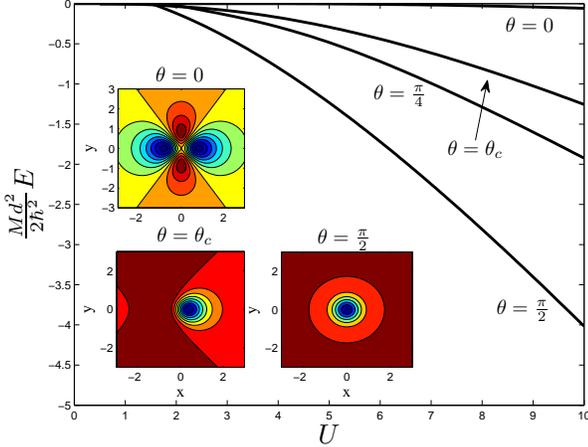


Fig. 1: Bound state energies as a function of U for different angles. Insets show contour plots of the potentials with valleys in bright (blue) and hills in dark (red) colors.

[27], whereas at larger U we find a linear dependence on U as argued in [22] for $\theta = \pi/2$. The binding energies decrease dramatically as θ approaches zero. However, we stress that we find a bound state for any value of U also in this particularly unfavorable case of $\theta = 0$. Notice that there are more bound states for larger U but we restricted our discussion to the single bound state regime.

The $U < 0$ case is also of great interest as that potential can be generated by using microwave-dressed molecules. In [12, 13] an AC light field directed perpendicular to the layers was used to create the $\theta = \pi/2$ potential with $U < 0$. A straightforward calculation shows that if the laser hits the layers at an angle, θ , the potential is the same as for a homogeneous electric field at angle θ but with negative U . For $U < 0$ we again find that for all values of the strength the two-body system has bound states. The results for the binding energy at different angles are shown in Fig. (2) as function of $|U|$. The first thing one notices is that the overall magnitude of the bound state energy is smaller than that for $U > 0$. At $\theta = \pi/2$ this can be understood as the potential has a repulsive core at $\rho = 0$, forcing the state to reside in the shallow attractive pocket at intermediate distance. In turn, this gives a much smaller binding energy. This qualitative behavior of the potential persists until θ decreases below θ_c where the monopole changes sign. Then the potential changes overall character to become more attractive with inner attractive pocket and outer repulsive tail. The $U < 0$ results thus show maximum binding at $\theta = 0$ which is, however, still about a factor of three smaller than the $U > 0$ case at its most favorable angle of $\theta = \pi/2$.

The structure of the bound state wavefunctions can be seen from the partial wave decomposition. The results are shown in Fig. (3) for a strong coupling of $U = 10$ and a weaker one of $U = 4$. The probabilities are normalized so that they sum to one. We note that the contribution of

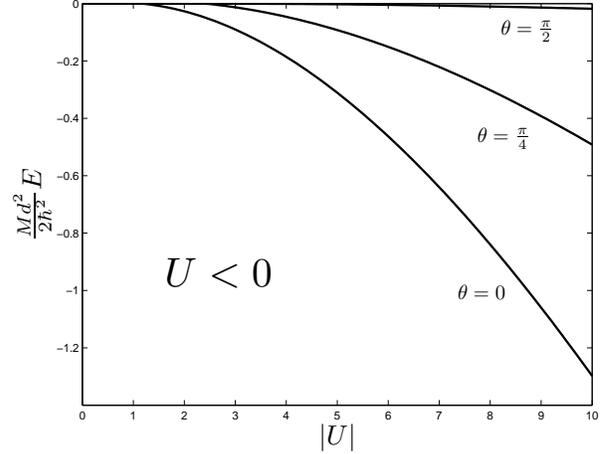


Fig. 2: Same as Fig. (1) but for $U < 0$. Note that the vertical scale is different from that of Fig. (1).

$m > 2$ is only a few percent with a maximum at $\theta = 0$ of 5% in $m > 2$ terms. As expected we find that $m = 0$ becomes dominant for $\theta \rightarrow \pi/2$ as we approach spherical symmetry. Interestingly, close to $\theta = 0$ we also find a very large $m = 0$ component, no $m = 1$ content, and a significant $m = 2$ contribution. The remaining content of the wavefunction is found in the higher m contributions. The fact that $m = 1$ has no weight for $\theta = 0$ can be understood from the symmetry of the potential. For $x \rightarrow -x$ the $m = 1$ term changes sign, whereas the potential is invariant. Interestingly, as we go away from $\theta = 0$, the $m = 1$ component raises rapidly and stays on the order of 40% until we reach $\theta = \theta_c$ at which it starts to decline as for $m = 2$, in line with the restoration of spherical symmetry at $\theta = \pi/2$. For $U > 10$ the $m = 0$ component can be even more suppressed in comparison to $m > 0$ for intermediate θ , whereas for positive $U < 4$ the $m = 0$ component will eventually dominate as one approaches the universal limit discussed above.

We have found similar results for the $U < 0$ when taking into account that the angle θ for $U > 0$ correspond to angle $\pi/2 - \theta$ for $U < 0$ and vice versa. This is in fact an exact symmetry of the dipole part of the potential and an approximate one for the monopole term since θ_c is close to $\pi/4$. For $U = -10$ we find that there is a window $\theta_c < \theta < 1.1$ in which the $m = 1$ term is around 40%. Interestingly, we find that the partial-wave content for $U < 0$ is almost exclusively $m = 0$ and $m = 1$. This is perhaps surprising as the potential in the $m = 2$ channel is non-vanishing except at $\theta = \pi/2$. For small θ , the potential looks like a harmonic oscillator along the y -axis for small x and y . The depth of this harmonic well around zero is about twice as large for $\theta = 0$ and $U < 0$ as opposed to the depth of the two wells in the x -direction for $U > 0$ shown in Fig. (1). A simple gaussian wavefunction should therefore be a fair approximation to the full problem and in turn the lowest

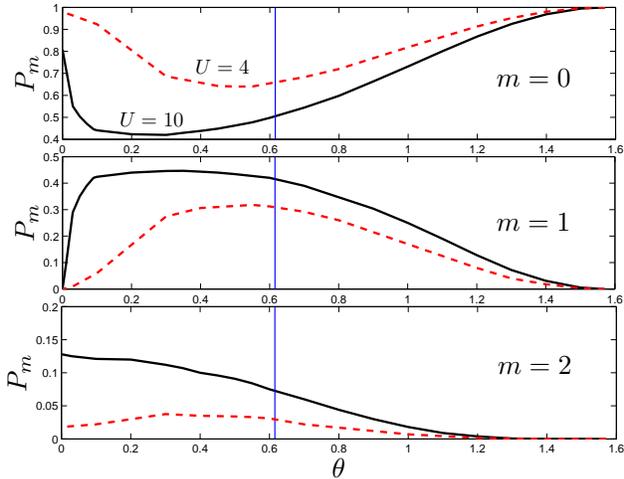


Fig. 3: Partial waves probabilities, P_m , for the bound state wavefunction at $U = 10$ (solid black) and $U = 4$ (dashed red) as function of polarization angle θ for $m = 0, 1$, and 2 . The vertical line indicates $\theta = \theta_c$. Note the different vertical scales.

partial-waves should dominate.

The Many-Body Bilayer System. – The bilayer system has an interesting many-body structure with combination of attractive interactions that can induce pairing and repulsive interaction that tend to suppress such effects. This was discussed recently for the $\theta = \pi/2$ case in [14, 15]. Here we consider the strongly-coupled limit (large U) where the bound two-body dimers are expected to be the relevant degrees of freedom. As the dimers are effectively bosons, they are capable of forming a (quasi)-condensate under the right conditions [14]. However, as is well-known from BCS-BEC crossover studies [30], this is only expected to occur when the density is low. Unfortunately, the Berezinskii-Kosterlitz-Thouless (BKT) transition [31, 32] that governs this two-dimensional system has a critical temperature that is proportional to density [14, 15]. Therefore, a compromise where the dimer condensate occurs at not too low densities would be optimal to allow experimental access to this unusual many-body state.

As the criterion for the onset of condensation of dimers, we consider the point at which the chemical potential becomes negative [14], i.e.

$$\mu(U, \theta) = \frac{1}{2}nV_{\text{eff}}(U, \theta) + E_F - \frac{1}{2}E_B(U, \theta), \quad (9)$$

where E_B is the dimer binding energy and V_{eff} is the long-wavelength (zero momentum) effective momentum-space interaction between two dimers. Here we include both the binding energy and the dimer-dimer interaction, and we also include a term for the Fermi energy, E_F , that the constituents of the dimer inherit from their layer. The density of dimers (equal to the single-layer density when the layers have an equal number of molecules) is denoted by n . To

obtain the effective interaction, one must in principle integrate out the wavefunction of the dimer and include all inter- and intralayer two-body terms [14]. However, here we are only interested in the long-wavelength limit (momentum zero) in which the interlayer term vanishes [17]. This gives

$$V_{\text{eff}}(U, \theta) = \frac{\hbar^2}{M} \frac{4U}{3\sqrt{2\pi}} \left(\frac{d}{w}\right) 4\pi P_2(\sin \theta), \quad (10)$$

where $P_2(x) = (3x^2 - 1)/2$. For the layer width, we take $w/d = 0.2$ in the following. Notice that V_{eff} is attractive for $\theta < \theta_c$, vanishes at θ_c , and repulsive for $\theta > \theta_c$. The attraction for $\theta < \theta_c$ results in a negative compressibility in a single layer [16]. We stress again that θ_c is much smaller than the angle at which the intralayer repulsion vanishes in a one-dimensional system. In this sense θ_c is a special angle for the intralayer repulsion, whereas it has no dramatic effect on the binding energies which vary smoothly around $\theta = \theta_c$. Combining the formula above, the final expression for μ becomes

$$\frac{Md^2}{\hbar^2}\mu = \frac{(k_F d)^2}{2} \left(\frac{4U}{3\sqrt{2\pi}} \left(\frac{d}{w}\right) P_2(\sin \theta) + 1\right) - \frac{Md^2}{2\hbar^2}E_B, \quad (11)$$

where we use the Fermi momentum $k_F^2 = 4\pi n$ for fermions in a single layer in place of n .

The lines of $\mu = 0$ for selected angles are shown in Fig. (4) in the $(U, k_F d)$ plane for $U > 2.5$ where the dimers are strongly bound and can be treated as localized bosonic objects. For $\theta = \pi/2$ we present results both with and without the intralayer term which is clearly seen to shrink the region of potential dimer condensation. For $\theta = \theta_c$, the intralayer term vanishes and we find a larger region of $\mu < 0$. For $\theta < \theta_c$ the region would in principle become even larger, however, the intralayer term is then attractive and can lead to instability and collapse [16]. We therefore expect the line for $\theta = \theta_c$ to provide a boundary for how large the BEC region can become when tuning the angle within our approximations. In the large U limit, the BKT transition temperature is maximal at $k_B T_{BKT} = E_F/8$ [12, 14, 15], or

$$T_{BKT} = 765 \frac{n}{10^8 \text{ cm}^{-2}} \frac{\text{amu}}{M} \text{ nK}. \quad (12)$$

If we consider LiCs molecules (which can have a dipole moment of up to 5.5 Debye) with $d = 0.5 \mu\text{m}$, then the $\mu = 0$ phase for $\theta = \theta_c$ at $U = 10$ is at $n = 8.1 \cdot 10^7 \text{ cm}^{-2}$, and thus $T_{BKT} \sim 4.9 \text{ nK}$. While still very low, this is a significant increase over the sub-nano Kelvin temperatures for $\theta = \pi/2$.

We expect that the partial-wave analysis presented above can help indicate what symmetries are possible and relevant for the corresponding many-body problem in the large U limit. The problem is of course still that the intralayer term is attractive in the long-wavelength limit for $\theta < \theta_c$, and we thus expect that the most stable system

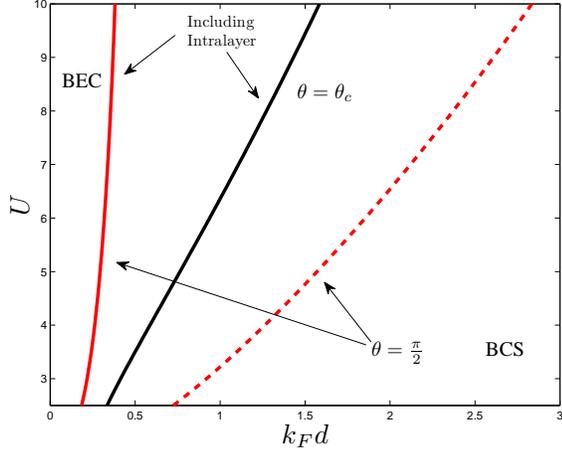


Fig. 4: Lines of vanishing chemical potential for $\sin^2 \theta_c = 1/3$ (solid black) and $\theta = \pi/2$ with (solid red) and without (dashed red) intralayer repulsion. Above the solid lines we expect condensation of dimers to occur, whereas in the lower right part a many-body paired BCS-like state should be the ground-state of the system.

require $\theta > \theta_c$ where the decomposition of the wavefunction is entirely dominated by the $m = 0$ term. However, close to θ_c we still have a substantial $m = 1$ contribution (immediately to the right of the vertical line in Fig. (3)). We therefore expect a region of interest in which an exotic many-body state with non-trivial symmetry like a p -wave dominated or mixed symmetry superfluid would emerge in the bilayer. These indications are consistent with a partial-wave decomposition of the potential itself [33]. Combining this information strongly suggests that there is a very interesting crossover from weak- to strong-coupling in the corresponding many-body system as recently discussed for the $\theta = \pi/2$ case [14, 15]. Similar considerations hold for the $U < 0$ case.

Summary and Outlook. – We have studied a bilayer system of dipolar molecules for arbitrary orientation of the dipoles with respect to the planes. The two-body bound state structure was calculated, including energies and partial-wave decomposition of the wavefunction as function of dipolar strength and polarization angle. We always find a bound two-body state in the system, irrespective of strength and polarization angle of the molecules. We argued that this follows from the fact that for small strength, the wavefunction must reside outside the region where the potential is non-zero, and as such the known proof of bound states applies in the limit. The results apply irrespective of the sign of the interaction strength. Negative strengths invert the dependence of energy on the dipole angle such that perpendicular polarization angle has the smallest binding energy. The structure of the wavefunction is dominated by the monopole component which decreases with the strength of the interaction. Up

to moderate strengths, the monopole component is always larger than 50 percent while the dipole component accounts for most of the remaining probability.

Implications for the many-body physics of a bilayer were discussed in the limit of strong-coupling where the two-body bound states are expected to be the important degrees of freedom. We conclude that the region where (quasi)-condensation of two-molecule dimers is likely to occur can be enhanced by tuning the angle of the dipoles. In particular, at the critical angle, $\sin^2 \theta_c = 1/3$, where the long-wavelength part of the intralayer interaction vanishes, we expect the conditions are most favorable for accessing this phase where dimers condense. This critical angle is different from the ‘magic’ angle at which dipoles moving on a line become non-interacting which has been discussed in a number of previous works [8, 24, 34]. The possible roton instability in the bilayer system, as discussed for the perpendicular case in [14], is currently under active investigation.

The results presented in this letter indicate that bound complexes of more than two particles must exist in the bilayer. In the case of one-dimensional tubes this was studied in some special cases in [24] and [34], and a full investigation of one-dimensional complexes as function of angles and dipole moment is forthcoming [35]. For the two-dimensional case, the method employed here can be extended to complexes of more particles and we plan such investigations in the near future. We also note that the external trapping potential that is present in each layer in experiments [19] can be easily accommodated in the current approach by introducing one-body harmonic oscillator terms.

In conclusion, we find that bound states of dimers in a bilayer consisting of one particle in each layer are generic for particles interacting through the dipole-dipole force, irrespective of the dipole strength or polarization angle of the dipoles with respect to the layers. In general, the wavefunction contains several partial-wave components and therefore has interesting spatial structure. This suggests that few-body states with more than two particles will also have rich structure and it also implies that the many-body physics of the system is highly non-trivial. We sketched a phase diagram for the appearance of a dimer condensate as a function of polarization angle and showed that it is enhanced around the so-called magic angle. At this point the dimer contains a large admixture of higher partial waves and we expect the collective behavior of the system to reflect this fact. The many-body problem of a bilayer with polar molecules of arbitrary polarization angle therefore deserve further investigation.

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