

## Crystalline Phase of Strongly Interacting Fermi Mixtures

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We show that the system of weakly bound molecules of heavy and light fermionic atoms is characterized by a long-range intermolecular repulsion and can undergo a gas-crystal quantum transition if the mass ratio exceeds a critical value. For the critical mass ratio above 100 obtained in our calculations, this crystalline order can be observed as a superlattice in an optical lattice for heavy atoms with a small filling factor. We also find that this novel system is sufficiently stable with respect to molecular relaxation into deep bound states and to the process of trimer formation.

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The use of Feshbach resonances for tuning the interaction in two-component ultracold Fermi gases of <sup>6</sup>Li or <sup>40</sup>K has led to remarkable developments, such as the observation of superfluid behavior in the strongly interacting regime through vortex formation [1], and Bose-Einstein condensation of weakly bound molecules of fermionic atoms on the positive side of the resonance (the atom-atom scattering length  $a > 0$ ) [2]. Being highly excited, these extremely large diatomic molecules are remarkably stable with respect to collisional relaxation into deep bound states, which is a consequence of the Pauli exclusion principle for identical fermionic atoms [3].

Currently, a new generation of experiments is being set up for studying mixtures of different fermionic atoms, with the idea of revealing the influence of the mass difference on superfluid properties and finding novel types of superfluid pairing. Weakly bound heteronuclear molecules on the positive side of the resonance are unique objects [4,5], which should manifest collisional stability and can pave a way to creating ultracold dipolar gases.

So far it was believed that dilute Fermi mixtures should be in the gas phase, like Fermi gases of atoms in two different internal states. In this Letter we find that the system of molecules of heavy (mass  $M$ ) and light (mass  $m$ ) fermions can undergo a phase transition to a crystalline phase. This is due to a repulsive intermolecular potential originating from the exchange of light fermions and inversely proportional to  $m$ . As the kinetic energy of the molecules has a prefactor  $1/M$ , above a certain mass ratio  $M/m$  the system can crystallize.

We show that the interaction potential in a sufficiently dilute system of molecules is equal to the sum of their pair interactions and then analyze the case where the motion of heavy atoms is confined to two dimensions, whereas the light fermions can be either 2D or 3D [6]. We calculate the zero-temperature gas-crystal transition line using the diffusion Monte Carlo (DMC) method and draw the phase

diagram in terms of the mass ratio and density. This phase transition resembles the one for the flux lattice melting in superconductors, where the flux lines are mapped onto a system of bosons interacting via a 2D Yukawa potential [7]. In this case the Monte Carlo studies [8,9] identified the first order liquid-crystal transition at zero and finite temperatures. Aside from the difference in the interaction potentials, a distinguished feature of our system is related to its stability. The molecules can undergo collisional relaxation into deep bound states, or form weakly bound trimers. We analyze resulting limitations on the lifetime of the system.

We first derive the Born-Oppenheimer interaction potential in the system of  $N$  molecules. In this approach the state of light atoms adiabatically adjusts itself to the set of heavy-atom coordinates  $\{\mathbf{R}\} = \{\mathbf{R}_1, \dots, \mathbf{R}_N\}$  and one calculates the wave function and energy of light fermions in the field of fixed heavy atoms. Omitting the interaction between light (identical) fermions, it is sufficient to find  $N$  lowest single-particle eigenstates, and the sum of their energies will give the interaction potential for the molecules. For the interaction between light and heavy atoms we use the Bethe-Peierls approach [10] assuming that the motion of light atoms is free everywhere except for their vanishing distances from heavy atoms.

The wave function of a single light atom then reads

$$\Psi(\{\mathbf{R}\}, \mathbf{r}) = \sum_{i=1}^N C_i G_{\kappa}(\mathbf{r} - \mathbf{R}_i), \quad (1)$$

where  $\mathbf{r}$  is its coordinate, and the Green function  $G_{\kappa}$  satisfies the equation  $(-\nabla_{\mathbf{r}}^2 + \kappa^2)G_{\kappa}(\mathbf{r}) = \delta(\mathbf{r})$ . The energy of the state (1) equals  $\epsilon = -\hbar^2 \kappa^2 / 2m$ , and here we only search for negative single-particle energies (see below). The dependence of the coefficients  $C_i$  and  $\kappa$  on  $\{\mathbf{R}\}$  is obtained using the Bethe-Peierls boundary condition:

$$\Psi(\{\mathbf{R}\}, \mathbf{r}) \propto G_{\kappa_0}(\mathbf{r} - \mathbf{R}_i); \quad \mathbf{r} \rightarrow \mathbf{R}_i. \quad (2)$$

Up to a normalization constant,  $G_{\kappa_0}$  is the wave function of a bound state of a single molecule with energy  $\epsilon_0 = -\hbar^2 \kappa_0^2 / 2m$  and molecular size  $\kappa_0^{-1}$ . From Eqs. (1) and (2) one gets a set of  $N$  equations:  $\sum_j A_{ij} C_j = 0$ , where  $A_{ij} = \lambda(\kappa) \delta_{ij} + G_{\kappa}(R_{ij})(1 - \delta_{ij})$ ,  $R_{ij} = |\mathbf{R}_i - \mathbf{R}_j|$ , and  $\lambda(\kappa) = \lim_{r \rightarrow 0} [G_{\kappa}(r) - G_{\kappa_0}(r)]$ . The single-particle energy levels are determined by the equation

$$\det[A_{ij}(\kappa, \{\mathbf{R}\})] = 0. \quad (3)$$

For  $R_{ij} \rightarrow \infty$ , Eq. (3) gives an  $N$ -fold degenerate ground state with  $\kappa = \kappa_0$ . At finite large  $R_{ij}$ , the levels split into a narrow band. Given a small parameter

$$\xi = G_{\kappa_0}(\tilde{R}) / \kappa_0 |\lambda'_{\kappa}(\kappa_0)| \ll 1, \quad (4)$$

where  $\tilde{R}$  is a characteristic distance at which heavy atoms can approach each other, the bandwidth is  $\Delta \epsilon \approx 4|\epsilon_0| \xi \ll |\epsilon_0|$ . It is important for the adiabatic approximation that all lowest  $N$  eigenstates have negative energies and are separated from the continuum by a gap  $\sim |\epsilon_0|$ .

We now calculate the single-particle energies up to second order in  $\xi$ . To this order we write  $\kappa(\lambda) \approx \kappa_0 + \kappa'_{\lambda} \lambda + \kappa''_{\lambda\lambda} \lambda^2 / 2$  and turn from  $A_{ij}(\kappa)$  to  $A_{ij}(\lambda)$ :

$$A_{ij} = \lambda \delta_{ij} + [G_{\kappa_0}(R_{ij}) + \kappa'_{\lambda} \lambda \partial G_{\kappa_0}(R_{ij}) / \partial \kappa] (1 - \delta_{ij}), \quad (5)$$

where all derivatives are taken at  $\lambda = 0$ . Using  $A_{ij}$  (5) in Eq. (3) gives a polynomial of degree  $N$  in  $\lambda$ . Its roots  $\lambda_i$  give the light-atom energy spectrum  $\epsilon_i = -\hbar^2 \kappa^2(\lambda_i) / 2m$ . The total energy  $E = \sum_{i=1}^N \epsilon_i$  is then given by

$$E = -(\hbar^2 / 2m) \left[ N \kappa_0^2 + 2 \kappa_0 \kappa'_{\lambda} \sum_{i=1}^N \lambda_i + (\kappa \kappa'_{\lambda})'_{\lambda} \sum_{i=1}^N \lambda_i^2 \right]. \quad (6)$$

Keeping only the terms up to second order in  $\xi$  and using basic properties of determinants and polynomial roots we find that the first order terms vanish, and the energy reads  $E = N \epsilon_0 + \frac{1}{2} \sum_{i \neq j} U(R_{ij})$ , where

$$U(R) = -\frac{\hbar^2}{m} \left[ \kappa_0 (\kappa'_{\lambda})^2 \frac{\partial G_{\kappa_0}^2(R)}{\partial \kappa} + (\kappa \kappa'_{\lambda})'_{\lambda} G_{\kappa_0}^2(R) \right]. \quad (7)$$

Thus, up to second order in  $\xi$  the interaction in the system of  $N$  molecules is the sum of binary potentials (7).

If the motion of light atoms is 3D, the Green function is  $G_{\kappa}(R) = (1/4\pi R) \exp(-\kappa R)$ , and  $\lambda(\kappa) = (\kappa_0 - \kappa) / 4\pi$ , with the molecular size  $\kappa_0^{-1}$  equal to the 3D scattering length  $a$ . Equation (7) then gives a repulsive potential

$$U_{3D}(R) = 4|\epsilon_0| (1 - (2\kappa_0 R)^{-1}) \exp(-2\kappa_0 R) / \kappa_0 R, \quad (8)$$

and the criterion (4) reads  $(1/\kappa_0 R) \exp(-\kappa_0 R) \ll 1$ . For the 2D motion of light atoms we have  $G_{\kappa}(R) = (1/2\pi) K_0(\kappa R)$  and  $\lambda(\kappa) = -(1/2\pi) \ln(\kappa/\kappa_0)$ , where  $K_0$  is the decaying Bessel function, and  $\kappa_0^{-1}$  follows from [6]. This leads to a repulsive intermolecular potential

$$U_{2D}(R) = 4|\epsilon_0| [\kappa_0 R K_0(\kappa_0 R) K_1(\kappa_0 R) - K_0^2(\kappa_0 R)], \quad (9)$$

with the validity criterion  $K_0(\kappa_0 R) \ll 1$ . In both cases, which we denote  $2 \times 3$  and  $2 \times 2$  for brevity, the validity criteria are well satisfied already for  $\kappa_0 R \approx 2$ .

The Hamiltonian of the many-body system reads

$$H = -(\hbar^2 / 2M) \sum_i \Delta_{\mathbf{R}_i} + \frac{1}{2} \sum_{i \neq j} U(R_{ij}), \quad (10)$$

and the state of the system is determined by two parameters: the mass ratio  $M/m$  and the rescaled 2D density  $n \kappa_0^{-2}$ . At a large  $M/m$ , the potential repulsion dominates over the kinetic energy and one expects a crystalline ground state. For separations  $R_{ij} < \kappa_0^{-1}$  the adiabatic approximation breaks down. However, the interaction potential  $U(R)$  is strongly repulsive at larger distances. Hence, even for an average separation between heavy atoms  $\bar{R}$  close to  $2/\kappa_0$ , they approach each other at distances smaller than  $\kappa_0^{-1}$  with a small tunneling probability  $P \propto \exp(-\beta \sqrt{M/m}) \ll 1$ , where  $\beta \sim 1$ . We extended  $U(R)$  to  $R \lesssim \kappa_0^{-1}$  in a way providing a proper molecule-molecule scattering phase shift in vacuum and checked that the phase diagram for the many-body system is not sensitive to the choice of this extension.

Using the DMC method [11] we solved the many-body problem at zero temperature. For each phase, gaseous and solid, the state with a minimum energy was obtained in a statistically exact way. The lowest of the two energies corresponds to the ground state, the other phase being metastable. The phase diagram is displayed in Fig. 1. The guiding wave function was taken in the Nosanow-Jastrow form [12]. Simulations were performed with 30 particles and showed that the solid phase is a 2D triangular lattice. For the largest density we checked that using more particles has little effect on the results.

For both  $2 \times 3$  and  $2 \times 2$  cases the (Lindemann) ratio  $\gamma$  of the rms displacement of molecules to  $\bar{R}$  on the transition

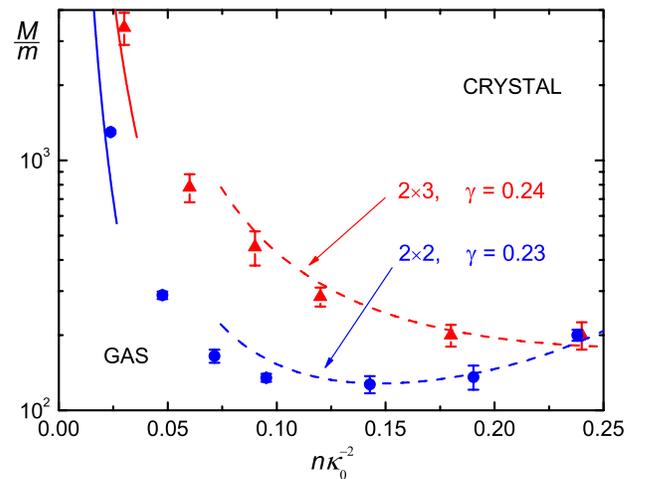


FIG. 1 (color online). DMC gas-crystal transition lines for 3D (triangles) and 2D (circles) motion of light atoms. Solid curves show the low-density hard-disk limit, and dashed curves the results of the harmonic approach (see text).

lines ranges from 0.23 to 0.27. At low densities  $n$  the de Broglie wavelength of molecules is  $\Lambda \sim \gamma \bar{R} \gg \kappa_0^{-1}$ , and  $U(R)$  can be approximated by a hard-disk potential with the diameter equal to the 2D scattering length. Then, using the DMC results for hard-disk bosons [13], we obtain the transition lines shown by solid curves in Fig. 1. At larger  $n$ , we have  $\Lambda < \kappa_0^{-1}$  and use the harmonic expansion of  $U(R)$  around equilibrium positions in the crystal, calculate the Lindemann ratio, and select  $\gamma$  for the best fit to the DMC data points (dashed curves in Fig. 1).

The mass ratio above 100, required for the observation of the crystalline order (see Fig. 1), can be achieved in an optical lattice with a small filling factor for heavy atoms. Their effective mass in the lattice  $M_*$  can be made very large, and the discussed solid phase should appear as a superlattice. There is no interplay between the superlattice order and the shape of the underlying optical lattice, in contrast to the recently studied solid and supersolid phases in a triangular lattice with the filling factor of order one [14]. Our superlattice remains compressible and supports two branches of phonons.

The gaseous and solid phases of weakly bound molecules are actually metastable. The main decay channels are the relaxation of molecules into deep bound states and the formation of trimer states by one light and two heavy atoms. A detailed analysis of scattering properties of these molecules will be given elsewhere, and here we focus on their stability in an optical lattice.

For a large effective mass ratio  $M_*/m$ , the relaxation into deep states occurs when a molecule is approached by another light atom and both light-heavy separations are of the order of the size of a deep state,  $R_e \ll \kappa_0^{-1}$  [15]. The released binding energy is taken by outgoing particles which escape from the sample. The rate of this process is not influenced by the optical lattice.

We estimate this rate in the solid phase and near the gas-solid transition to the leading order in  $(\kappa_0 \bar{R})^{-1}$ . At light-heavy separations  $r_{1,2} \ll \kappa_0^{-1}$  the initial-state wave function reads  $\tilde{\Psi} = B(\kappa_0^{-1}, \bar{R})\psi(\mathbf{r}_1, \mathbf{r}_2)$ . Writing it as an antisymmetrized product of wave functions (1), for the  $2 \times 3$  case ( $\kappa_0^{-1} = a$ ) we find  $B \approx (1/\bar{R}a^2)\exp(-\bar{R}/a)$ . The quantity  $W = B^2 R_e^6$  is the probability of having both light atoms at distances  $\sim R_e$  from a heavy atom, and the relaxation rate is  $\nu_{3D} \propto W$ . As the short-range physics is characterized by the energy scale  $\hbar^2/mR_e^2$ , we restore the dimensions and write

$$\nu_{3D} = C(\hbar/m)(R_e/a)^4(1/\bar{R}^2)\exp(-2\bar{R}/a), \quad (11)$$

where  $\bar{R}^{-2} \approx n$ . The coefficient  $C$  depends on a particular system and is  $\sim 1$  within an order of magnitude. The relaxation rate  $\nu_{3D}$  is generally rather low. For the K-Li mixture where  $R_e \approx 50 \text{ \AA}$ , even at  $na^2 = 0.24$  (see Fig. 1) the relaxation time exceeds 10 s for  $n = 10^9 \text{ cm}^{-2}$  and  $a = 1600 \text{ \AA}$ . In the  $2 \times 2$  case, for the same  $n$  and  $\kappa_0^{-1}$  the probability  $W$  is smaller and the relaxation is slower.

The formation of trimer bound states by one light and two heavy atoms occurs when two molecules approach each other at distances  $R \lesssim \kappa_0^{-1}$ . It is accompanied by a release of the second light atom. The existence of the trimer states is seen considering a light atom interacting with two heavy ones. The lowest energy solution of Eq. (3) for  $N = 2$  is the gerade state ( $C_1 = C_2$ ). Its energy  $\epsilon_+(R)$  introduces an effective attractive potential acting on the heavy atoms, and the trimer states are bound states of two heavy atoms in this potential.

In an optical lattice the trimers are eigenstates of the Hamiltonian  $H_0 = -(\hbar^2/2M_*)\sum_{i=1,2}\Delta_{\mathbf{R}_i} + \epsilon_+(R_{12})$ . In a deep lattice one can neglect all higher bands and regard  $\mathbf{R}_i$  as discrete lattice coordinates and  $\Delta$  as the lattice Laplacian. Then, the fermionic nature of the heavy atoms prohibits them to be in the same lattice site. For a very large mass ratio  $M_*/m$  the kinetic energy term in  $H_0$  can be neglected, and the lowest trimer state has energy  $\epsilon_{\text{tr}} \approx \epsilon_+(L)$ , where  $L$  is the lattice period. It consists of a pair of heavy atoms localized at neighboring sites and a light atom in the gerade state. Higher trimer states are formed by heavy atoms localized in sites separated by distances  $R > L$ . This picture breaks down at large  $R$ , where the spacing between trimer levels is comparable with the tunneling energy  $\hbar^2/M_*L^2$  and the heavy atoms are delocalized.

In the many-body molecular system the scale of energies in Eq. (10) is much smaller than  $|\epsilon_0|$ . Thus, the formation of trimers in molecule-molecule ‘‘collisions’’ is energetically allowed only if the trimer binding energy is  $\epsilon_{\text{tr}} < 2\epsilon_0$ . Since the lowest trimer energy in the optical lattice is  $\epsilon_+(L)$ , the trimer formation requires the condition  $\epsilon_+(L) \leq 2\epsilon_0$ , which is equivalent to  $\kappa_0^{-1} \geq 1.6L$  in the  $2 \times 3$  case and  $\kappa_0^{-1} \geq 1.25L$  in the  $2 \times 2$  case. This means that for a sufficiently small molecular size or large lattice period  $L$  the formation of trimers is forbidden.

At a larger molecular size or smaller  $L$  the trimer formation is possible. For finding the rate we consider the interaction between two molecules as a reduced 3-body problem, accounting for the fact that one of the light atoms is in the gerade and the other one in the ungerade state ( $C_1 = -C_2$ ). The gerade light atom is integrated out and is substituted by the effective potential  $\epsilon_+(R)$ . For the ungerade state the adiabaticity breaks down at interheavy separations  $R \lesssim \kappa_0^{-1}$ , and the ungerade light atom is treated explicitly. The wave function of the reduced 3-body problem satisfies the Schrödinger equation

$$[H_0 - \hbar^2\nabla_{\mathbf{r}}^2/2m - E]\psi(\{\mathbf{R}\}, \mathbf{r}) = 0, \quad (12)$$

where the energy  $E$  is close to  $2\epsilon_0$ ,  $\{\mathbf{R}\}$  denotes the set  $\{\mathbf{R}_1, \mathbf{R}_2\}$ , and  $\mathbf{r}$  is the coordinate of the ungerade atom. The interaction between this atom and the heavy ones is replaced by the boundary condition (2) on  $\psi$ . The 3-body problem can then be solved by encoding the information on the wave function  $\psi$  in an auxiliary function  $f(\{\mathbf{R}\})$  [16] and representing the solution of Eq. (12) in the form

$$\psi = \sum_{\{\tilde{\mathbf{R}}\}, \nu} \chi_\nu(\{\mathbf{R}\}) \chi_\nu^*(\{\tilde{\mathbf{R}}\}) f(\{\tilde{\mathbf{R}}\}) F_{\kappa_\nu}(\mathbf{r}, \{\tilde{\mathbf{R}}\}), \quad (13)$$

where  $\chi_\nu(\{\mathbf{R}\})$  is an eigenfunction of  $H_0$  with energy  $\epsilon_\nu$ , and  $F_{\kappa_\nu}(\mathbf{r}, \{\tilde{\mathbf{R}}\}) = G_{\kappa_\nu}(\mathbf{r} - \tilde{\mathbf{R}}_1) - G_{\kappa_\nu}(\mathbf{r} - \tilde{\mathbf{R}}_2)$  with  $\kappa_\nu = \sqrt{2m(\epsilon_\nu - E)/\hbar^2}$ . For  $\epsilon_\nu < E$  the trimer formation in the state  $\nu$  is possible. This is consistent with imaginary  $\kappa_\nu$  and the Green function  $G_{\kappa_\nu}$  describing an outgoing wave of the light atom and trimer.

We derive an equation for the function  $f$  in a deep lattice, where the tunneling energy  $\hbar^2/M_*L^2 \ll |\epsilon_0|$ . Then the main contribution to the sum in Eq. (13) comes from the states  $\nu$  for which  $|\epsilon_\nu - \epsilon_+(R_{12})| \lesssim \hbar^2/M_*L^2$ . The sum is calculated by expanding  $\kappa_\nu$  around  $\kappa(R_{12}) = \sqrt{2m(\epsilon_+(R_{12}) - E)/\hbar^2}$  up to first order in  $[\epsilon_\nu - \epsilon_+(R_{12})]/\epsilon_0$  and using the equation  $(H_0 - \epsilon_\nu)\chi_\nu = 0$ . The equation for  $f$  is then obtained by taking the limit  $\mathbf{r} \rightarrow \mathbf{R}_1$  in the resulting expression for  $\psi$  and comparing it with the boundary condition (2). This yields

$$\left[ -(\hbar^2/2M_*) \sum_{i=1,2} \Delta_{\mathbf{R}_i} + U_{\text{eff}}(R_{12}) \right] f(\mathbf{R}_1, \mathbf{R}_2) = 0, \quad (14)$$

where the effective potential  $U_{\text{eff}}(R_{12})$  is given by

$$U_{\text{eff}}(R) = \frac{\hbar^2 \kappa(R)}{m} \frac{\lambda(\kappa(R)) - G_{\kappa(R)}(R)}{(\partial/\partial \kappa)[\lambda(\kappa(R)) - G_{\kappa(R)}(R)]}. \quad (15)$$

At large distances one has  $U_{\text{eff}} \approx U(R) + 2\epsilon_0 - E$ , and for smaller  $R$  where  $\epsilon_+(R) < E$ , the potential  $U_{\text{eff}}$  acquires an imaginary part accounting for the decay of molecules into trimers. The number of trimer states that can be formed grows with the molecular size. Eventually it becomes independent of  $L$  and so does the loss rate.

In this limit, we solve Eq. (14) for two molecules with zero total momentum under the condition that  $f(\mathbf{R}_1, \mathbf{R}_2)$  is maximal for  $|\mathbf{R}_1 - \mathbf{R}_2| = \bar{R} \approx n^{-1/2}$ . We thus obtain  $E$  as a function of the density and mass ratio, and its imaginary part gives the loss rate  $\nu$  for the many-body system. Numerical analysis for  $0.06 < n\kappa_0^{-2} < 0.4$  and  $50 < M_*/m < 2000$  is well fitted by  $\nu \approx (D\hbar n/M_*)(n\kappa_0^{-2}) \times \exp(-J\sqrt{M_*/m})$ , with  $D = 7$  and  $J = 0.95-1.4(n\kappa_0^{-2})$  for the  $2 \times 3$  case, and  $D = 10^2$ ,  $J = 1.45-2.8(n\kappa_0^{-2})$  in the  $2 \times 2$  case. One can suppress  $\nu$  by increasing  $M_*/m$ , whereas for  $M_*/m \lesssim 100$  the trimers can be formed on a time scale  $\tau \lesssim 1$  s.

In conclusion, we have shown that the system of weakly bound molecules of heavy and light fermionic atoms can undergo a gas-crystal quantum transition. The necessary mass ratio is above 100 and the observation of such crystalline order requires an optical lattice for heavy atoms, where it appears as a superlattice. A promising candidate is the  ${}^6\text{Li}$ - ${}^{40}\text{K}$  mixture as the Li atom may tunnel freely in a lattice while localizing the heavy K atoms to reach high

mass ratios. A lattice with period 250 nm and K effective mass  $M^* = 20M$  provide a tunneling rate  $\sim 10^3 \text{ s}^{-1}$  sufficiently fast to let the crystal form. Near a Feshbach resonance, a value  $a = 500$  nm gives a binding energy 300 nK, and lower temperatures should be reached in the gas. The parameters  $n\kappa_0^{-2}$  of Fig. 1 are then obtained at 2D densities in the range  $10^7-10^8 \text{ cm}^{-2}$  easily reachable in experiments. For  $n = 10^8 \text{ cm}^{-2}$  the rate of the trimer formation is of the order of seconds, and these peculiar bound states can be detected optically.

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