Spin-spin interaction and magnetic state of 2-D Wigner crystal

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It is demonstrated that there is anti-ferromagnetic spin-spin interaction between nearest electrons in the 2D Wigner crystal. This is also valid for the Wigner liquid - the state with destroyed long-range order but preserved short-range one. We calculate the value of the anti-ferromagnetic interaction (both analytically and numerically) and discuss a possible magnetic state of the Wigner crystal. This state can be strongly influenced by the spin-Peierls mechanism.

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We consider a two dimensional electron gas (1/r repulsion) at zero temperature in the presence of a uniform neutralizing background. It was shown a long ago by Wigner that at sufficiently low density the electron gas (or electron fluid) undergoes a transition into a crystal state. This is because at low density the Coulomb interaction dominates the kinetic energy and the correlated state becomes energetically favorable. Analysis of the lattice dynamics shows that the stable crystal structure in 2D is the triangular lattice.

The Wigner crystallization has been observed for electrons at the surface of liquid helium. Another 2D system for which the electron density can be easily controlled is an inversion layer at a semiconductor surface.

Theoretically the Wigner crystallization has been studied using Monte Carlo simulations, see e.g. Refs. These calculations are pretty reliable as far as the critical density is concerned. However there is some controversy about possible Fermagnetic Fermi liquid at a density slightly higher than the crystallization density, see Refs.

Interest in Wigner crystallization has been renewed recently after observation of the insulator-conductor transition in dilute 2D electron systems. Although this transition probably takes place in the liquid phase it is pretty close to the point of crystallization. A very interesting feature of the transition is suppression of the conducting phase by in-plane magnetic field which influences only spin degrees of freedom.

In the present work we calculate effective spin-spin interaction in 2D Wigner crystal. This calculation is also valid for the Wigner liquid - the state with destroyed long-range order but preserved short-range order.

To avoid misunderstanding let us note that our calculation does not show any magnetic phase transition in the liquid state (i.e. there is no ferromagnetic Fermi liquid between normal Fermi liquid and Wigner crystal). The state which we call Wigner liquid is just strongly renormalized normal Fermi liquid. Nevertheless magnetic properties of the Wigner liquid (and Wigner crystal) are quite unusual and somewhat similar to that of cuprate superconductors. There is competition between superexchange (electron correlation) which gives antiferromagnetic interaction between electron spins and the usual exchange Coulomb interaction which gives ferromagnetic contribution. The superexchange is proportional to t^2/U where t is the parameter which describes hopping of an electron to a nearby site and U is the Coulomb repulsion for two electrons sitting on the same site. As a result, both the superexchange and exchange are proportional to the squared overlap between electron wave functions centered on the different sites of the Wigner crystal. Therefore, simple estimates can not answer the question about the sign of spin-spin interaction and we need more accurate calculations. To provide better understanding and reliability of the results we have performed these calculations twice: analytically and numerically.

Hamiltonian of the system under consideration is

\[ H = \sum_i p_i^2 / 2 + \sum_{i<j} \frac{1}{|r_i - r_j|} + \text{const}, \]  

where \( p_i \) and \( r_i \) are 2D momentum and coordinate respectively. We use effective atomic units which means that all distances are measured in units of the effective Bohr radius \( a_B = \hbar^2 / m^* \epsilon e^2 \), and energies in units of \( \hbar^2 \epsilon e^2 / \pi m^* a_B \). Here \( m^* \) is the effective electron mass, and \( \epsilon \) is the dielectric constant which we assume to be independent of frequency. The number density of the electrons \( n \) is fixed by condition of electroneutrality. An average distance \( r_s \) between the electrons is defined by \( \pi r_s^2 = 1/n \). It is well established that the crystallization to the triangular lattice occurs when \( r_s \approx 37 \). In the presence of “disorder” further localization of the electrons stabilizes the Wigner crystal at higher densities \( r_s \approx 10 \), see \( \text{[2]} \). The distance between the nearest sites in the lattice is equal to \( a = \sqrt{2\pi / \sqrt{3} r_s} \approx 1.90 r_s \).

Electrostatic potential acting on the electron near equilibrium position in the lattice is

\[ U_1(r) \approx \text{const} + \frac{\gamma r^2}{2 a^3}, \]  

where \( \gamma \) is the dielectric constant.
where $r \ll a$ is deviation from the equilibrium position. To find $\gamma$ let us freeze all other electrons in their equilibrium positions and calculate $U_1(r)$. Accounting for the six nearest sites gives $\gamma = 3$, and summation over entire lattice gives

$$
\gamma = 3 \sum_{n=1}^{\infty} \sum_{k=0}^{n-1} \frac{1}{(n^2 + k^2 - kn)^{3/2}} = 5.5171. \tag{3}
$$

Ground state electron wave function in the potential (2) is

$$
\psi(r) = \frac{1}{\sqrt{\pi c}} e^{-r^2/2c^2}, \quad c = \frac{a^{3/4}}{\gamma^{1/4}} \tag{4}
$$

In the above calculation we assume that size of the wave function is much smaller than the lattice spacing, $c \ll a$, or $(a\gamma)^{1/4} \gg 1$. For the crystallization point this parameter equals $(a\gamma)^{1/4} = 4.4$, and therefore the approximation is well justified in the crystal state. We stress that the parameter appears in the exponent and therefore 4.4 is a very large value. Moreover, the approximation is justified in the liquid phase as soon as $(a\gamma)^{1/4} \gg 1$. The matter is that the sum (3) is saturated at 2-3 coordination circles and it is independent of the presence or absence of the long-range order. For conditions of the experiments this parameter equals $(a\gamma)^{1/4} = 3.1$.

To find the magnitude of spin-spin interaction constant (the constant $J$, which can be substituted to the Heisenberg Hamiltonian $J \sum_{(i,j)} \vec{S}_i \cdot \vec{S}_j$) we have to solve a two-particle problem, freezing all the electrons except the nearest two ones which are shown by crosses at Figure 1.

FIG. 1. Boundary conditions for two-particles problem.

Hamiltonian of the problem is

$$
\hat{H} = \frac{\vec{p}_1^2}{2} + \frac{\vec{p}_2^2}{2} + U(\vec{r}_1) + U(\vec{r}_2) + \frac{1}{|\vec{r}_1 - \vec{r}_2|}, \tag{5}
$$

where $U(\vec{r})$ is potential of all frozen electrons (dots at Fig. 1). The splitting between the ground states for total spin $S = 1$ and $S = 0$ gives us the constant $J$:

$$
J = (E_g^{S=1} - E_g^{S=0}) \equiv (E_A - E_S), \tag{6}
$$

$$
\hat{H} \Psi_S = E_S \Psi_S, \tag{7}
$$

$$
\hat{H} \Psi_A = E_A \Psi_A. \tag{8}
$$

Because of the Fermi statistics the two-electron wave function is antisymmetric with respect to permutation. Therefore, the symmetric coordinate wave function corresponds to spin $S = 0$ and the antisymmetric one corresponds to $S = 1$.

Of course the accurate solution of the problem can only be found (and have been found) numerically. However, to find the sign and the basic dependence $J$ on a distance parameter $a$ we performed an approximate analytical
calculation. To this end, we will follow the procedure, which was suggested long time ago by Gorkov and Pitaevskii for the calculation of the term splitting in hydrogen molecule [11].

We multiply Eq. (7) by $\Psi_A$ and Eq. (8) by $\Psi_S$, take the difference between the results and calculate the integral over some region in configuration four dimensional space of the electrons. We choose the integration volume in which $x_1 \leq x_2$ (i.e. to the left of the hyperplane $\Sigma(x_1 = x_2)$). Using the Hamiltonian (7) we obtain

$$
\left( E_S - E_A \right) \int \int_\Omega \Psi_A \Psi_S \, dr_1 \, dr_2 = \int_\Sigma \left( \nabla_x \Psi_A - \nabla_x \Psi_S \right) \, d\Sigma.
$$

(9)

The kinetic energy term in the right-hand side is reduced to the surface integral using

$$
\nabla_x \Psi_A - \nabla_x \Psi_S = \nabla_x \left( \Psi_S - \Psi_A \right)
$$

and an integration by parts.

Now we introduce combinations of the functions $\Psi_{1,2} = 1/\sqrt{2} (\Psi_S \pm \Psi_A)$. They correspond to the states of "distinguishable" particles, when, e.g. for $\Psi_1(r_1, r_2)$, the first electron is principally located near its equilibrium position $x = -a/2$ and the second electron near position $x = a/2$. A simple calculation gives

$$
\int \int_\Omega \Psi_S \Psi_A \, dr_1 \, dr_2 = \frac{1}{2} \int \int_\Omega \left( \nabla_r \Psi_1^2 - \nabla_r \Psi_2^2 \right) \, dr_1 \, dr_2 \approx \frac{1}{2}.
$$

Substituting the wave functions $\Psi_{1,2}$ into Eq. (8) and taking into account that under $r_1 \leftrightarrow r_2$ permutation the wave functions $\Psi_1 \leftrightarrow \Psi_2$, we obtain

$$
J = -4 \int \left[ \nabla \frac{\partial \Psi_1}{\partial x_1} \right]_{x_1=x_2} \, dx_2 \, dy_1 \, dy_2.
$$

(10)

The formula (10) shows that the main contribution to the exchange constant is given by the region where the electrons are close to each other. Indeed, the $x$ coordinates of both electrons coincide ($x_1 = x_2$), however, the $y$ coordinates may be different. In this case there are strong correlations between the positions of the electrons due to Coulomb repulsion. This means that we should go beyond the approximation where the two-particle wave function of the electrons is represented as a product of single-particles wave functions.

It is easy to take into account the effect of the correlations in the quadratic approximation.

Assuming that the particles are distinguishable and oscillate near their equilibrium positions, we write the Hamiltonian in the following form

$$
\hat{H} = -\frac{\Delta_1}{2} - \frac{\Delta_2}{2} + \frac{\omega^2}{2} \left( (x_1 + a/2)^2 + y_1^2 + (x_2 - a/2)^2 + y_2^2 \right) + \left\{ \frac{1}{|r_1 - r_2|} - \frac{1}{|r_1 - a/2|} - \frac{1}{|r_2 + a/2|} \right\}.
$$

(11)

Here the frequency $\omega = \sqrt{\gamma/a^3}$.

The Hamiltonian (11) is valid at small displacements $x_i$ and $y_i$ from their equilibrium positions. Expanding the last term in the curly brackets near $(\tilde{x}_{1,2} = x_{1,2} \mp a/2)$ we finally get the following Hamiltonian in the quadratic approximation:

$$
\hat{H} = -\frac{\Delta_1}{2} - \frac{\Delta_2}{2} + \frac{\omega^2}{2} \left( \tilde{x}_1^2 + y_1^2 + \tilde{x}_2^2 + y_2^2 - \frac{4}{\gamma} \tilde{x}_1 \tilde{x}_2 + \frac{2}{\gamma} y_1 y_2 \right) + O(\tilde{x}^3/a^4).
$$

(12)

Using an obvious change of variables

$$
u, v = \frac{y_1 \pm y_2}{\sqrt{2}}, \quad \xi, \eta = \frac{x_1 \mp x_2}{\sqrt{2}},
$$

(13)

we separate Hamiltonian (12) into four independent oscillators with frequencies $\omega_{u,v} = \sqrt{(\gamma \pm 1)/a^3}$ and $\omega_{\xi,\eta} = \sqrt{(\gamma + 2)/a^3}$. Thus, the ground state wave functions are
To find \( J \) is of the form

\[
\Psi_1(u, v, \xi, \eta) = \frac{(\omega_u \omega_v \omega_\xi \omega_\eta)^{1/4}}{\pi} \exp \left(-1/2[\omega_u u^2 + \omega_v v^2 + \omega_\xi \xi^2 + \omega_\eta (\eta + a/\sqrt{2})^2] \right),
\]

\[
\Psi_2(u, v, \xi, \eta) = \Psi_1(u, v, \xi, -\eta).
\]

Substituting (14) in Eq. (10) we obtain

\[
J = +2\omega_\eta a \int [\Psi_2 \Psi_1]_{x_1 = x_2} dx_2 dy_1 dy_2 = (\gamma + 2)^{3/4} \sqrt{\frac{2}{\pi}} a^{-5/4} e^{-\sqrt{(\gamma + 2)a}/2}.
\]

This formula is presented in the atomic units. In regular units it can be written as

\[
J = \frac{e^2}{\epsilon a} \left[ \frac{a_B^*}{a} \frac{4(\gamma + 2)^{3/4}}{a_B} \right]^{1/4} \exp \left( -\sqrt{\frac{\gamma + 2}{4a_B} \frac{a}{a_B} \frac{a}{a_B}} \right) = 3.62 \frac{e^2}{\epsilon a} \left[ \frac{a_B^*}{a_B} \right]^{1/4} \exp \left( -1.37 \sqrt{\frac{a}{a_B}} \right)
\]

The plus sign in the exchange constant shows that the system is anti-ferromagnetic. It is worthwhile to note that the exponent in (15) is different from \( e^{-\sqrt{m}/2} \), which appears, if states \( \Psi_{1,2} \) are represented by a product of independent single-particle wave functions.

In order to check the importance of correlations and find the correct exponent for \( J \) we have performed numerical calculations of the problem over a rectangle area (see Fig. 1). To be absolutely correct we have to impose periodic boundary conditions in the rectangle. However, the wave function is very small at the boundary and so the results are not sensitive to the boundary condition. It is much more convenient to make the wave function vanish at the boundary and this is the condition which we use in the present work.

The single particle basis set is given by (see Fig. 1)

\[
\phi_i(r) \equiv \phi_{nm}(x', y') = \frac{2}{\sqrt{AB}} \sin \left( \frac{\pi}{A} nx' \right) \sin \left( \frac{\pi}{B} my' \right)
\]

\[
\varepsilon_i \equiv \varepsilon_{nm} = \frac{\pi^2}{2} \left[ \frac{n^2}{A^2} + \frac{m^2}{B^2} \right],
\]

where \( A = 3a \) and \( B = \sqrt{3}a \). Hence, for the two-electron problem the set is

\[
|i\rangle \equiv |i_1 i_2\rangle = C_{i_1 i_2} [\phi_{i_1}(r_1)\phi_{i_2}(r_2) \pm \phi_{i_1}(r_2)\phi_{i_2}(r_1)].
\]

\[
E_i = \varepsilon_{i_1} + \varepsilon_{i_2}
\]

The sign \( + \) corresponds to \( S = 0 \) (anti-ferromagnetic), and the sign \( - \) corresponds to \( S = 1 \) (ferromagnetic). The normalization coefficient \( C_{i_1 i_2} = 1/2 \) if \( i_1 = i_2 \) otherwise it equals \( 1/\sqrt{2} \). The matrix element of the Hamiltonian is of the form

\[
\langle i | \hat{H} | j \rangle = E_i \delta_{i j} + \langle i | \hat{V}^{(1)} | j \rangle + \langle i | \hat{V}^{(2)} | j \rangle,
\]

where \( \langle i | \hat{V}^{(1,2)} | j \rangle \) are matrix elements of the single particle potential and the two-particle interaction correspondingly.

\[
\langle i | \hat{V}^{(1)} | j \rangle = 2 C_{i_1 i_2} C_{i_3 i_4} \left[ V_{1_1 1_3}^{(1)} \delta_{i_2 i_4} \pm V_{1_1 1_3}^{(1)} \delta_{i_2 i_4} + V_{1_1 1_3}^{(1)} \delta_{i_2 i_4} + V_{1_1 1_3}^{(1)} \delta_{i_2 i_4} \right]
\]

\[
\langle i | \hat{V}^{(2)} | j \rangle = 2 C_{i_1 i_2} C_{i_3 i_4} \left[ V_{1_1 1_3 1_4} \pm V_{1_1 1_3 1_4} \right].
\]

To find \( J \), which is exponentially small, we need a very large basis set. The most time consuming part is the computation of the two-particle matrix element \( \langle i | \hat{V}^{(2)} | j \rangle \), which formally is a 4-dimensional integral. Fortunately this integral can be reduced to an integral which is effectively one dimensional. This reduction, which is demonstrated in the Appendix, allowed us to perform computations with the size of Hilbert space up to \( N = 1380 \).

Numerical solution of the problem was performed for two different cases. Firstly, we considered all the frozen electrons (points at Fig. 1) as point-like charges, which means that the mean-field potential in this case is just the sum of the Coulomb potentials:

\[
U(x, y) = \sum_{kl} u_0(|r_{kl} - r|),
\]

\[
u_0(r) = 1/r.
\]
The sum runs over sites of the triangular lattice.

Secondly, we considered the density of the frozen electrons to be distributed according to \( U(x, y) = \sum_{kl} u_1(|r_{kl} - r|) \), and hence

\[
u_1(r) = \sqrt{\frac{\pi}{c}} e^{-r^2/2c^2} I_0(v^2/2c^2),
\]

where \( I_0(x) \) is the modified Bessel function. In both cases all the results are very close and therefore we present plots only for the second case.

The matrices \( (19) \) for ferromagnetic and anti-ferromagnetic cases were calculated and diagonalized. The Hilbert space was truncated at some high energy state. To be confident that the ground state was found with reasonable accuracy we used two basis sets with dimensions \( N = 975 \) and \( N = 1380 \), where \( N \) is the total number of two-particle states.

The plots of ground state electron density

\[
\rho(r) = \langle 0 | \delta(r - r_1) + \delta(r - r_2) | 0 \rangle
\]

for \( a = 45 \), which corresponds to the limit of our calculations, are given in Figure 2. Similar plots for \( a = 15 \), which corresponds to the conditions of the experiments \( (9,10) \), are given in Figure 3. The fact that the maximums coincide with the lattice sites tells us about the self-consistency of the method. The shape of the density operator near the equilibrium positions also corresponds to the expected Gaussian electron density, obtained from the combinations of the wave functions in \( (14) \):

\[
\rho(x,y)_{S,A} = N_{S,A} \frac{2}{\pi} \sqrt{\frac{\omega_u \omega_v \omega_\xi \omega_\eta}{(\omega_u + \omega_v)(\omega_\xi + \omega_\eta)}} e^{-\tilde{\omega}_y y^2} \left[ e^{-\tilde{\omega}_x (x + a/2)^2} + e^{-\tilde{\omega}_x (x - a/2)^2} \pm 2 e^{-\omega_a a^2/2} e^{-\tilde{\omega}_x x^2} \right].
\]

Here \( N_{S,A} = |1 \pm e^{-\omega_a a^2/2}|^{-1} \) is the normalization coefficient due to the nonorthogonality of the functions \( \Psi_1 \) and \( \Psi_2 \), and \( \tilde{\omega}_y = 2 \omega_u \omega_v / (\omega_u + \omega_v) \), \( \tilde{\omega}_x = 2 \omega_\xi \omega_\eta / (\omega_\xi + \omega_\eta) \).

- **FIG. 2.** Profiles of the density operator \( \rho(\vec{r}) \) for the antiferromagnetic (two left plots) and ferromagnetic (right plots) ground states for \( a = 45 \) \( (r_s = 24) \). The dot-dashed lines show the analytical results in quadratic approximations (formula \( (24) \)).
FIG. 3. Profiles of the density operator $\rho(\vec{r})$ for the antiferromagnetic (two left plots) and ferromagnetic (right plots) ground states for $a = 15$ ($r_s = 7.9$). The dot-dashed lines show the analytical results in quadratic approximations (formula (24)).

We found that for the whole region of the strength parameter $a$ the anti-ferromagnetic ground state is always below the ferromagnetic one. We obtained the following values of the constant $J(a)$ for the parameters of experiments [9,10] ($\epsilon = 8$, $m^* = 0.19 m_e$)

\[
J(15) = 6.66 \cdot 10^{-4} = 0.6 K,
J(45) = 1.48 \cdot 10^{-6} = 1.4 \cdot 10^{-3} K.
\]

(25)

Experiments [9,10] correspond to $a \simeq 15$. The behavior of $J$, as expected, has an exponential dependence $\sim e^{-\delta \sqrt{a}}$. The plot of the dependence $\ln(J)$ vs. $\sqrt{a}$ summarizes our results in Figure 4. Diamonds and crosses show the magnitude for different basis sets and nicely depict the truncation effects for large $a$ and for small number of basis states. The dot-dashed line represents the theoretical curve (15) and the solid line is the best fit.

FIG. 4. Dependence $\ln(J)$ on $\sqrt{a}$. Diamonds and crosses represent data for large ($N = 1380$) and shortened ($N = 975$) bases correspondingly. The dot-dashed line shows the theoretical curve of Eq. (15). The solid line is the best fit by curve $y = C - \delta \sqrt{a} - \beta \ln(a)$. 

6
We fitted our data by two different functions. For the first one we fixed the power of $a$ in the preexponential factor (the analytical formula \[13\] gives $a^{-5/4}$):

$$\ln(J) = C - \delta \sqrt{a} - 5/2 \ln(\sqrt{a})$$

and got $C = 2.25, \delta = 1.595$. Let us note that the constant $\delta$ is slightly larger than the predicted analytical value $\delta_{an} = \sqrt{\gamma + 2/2} = 1.37$. This fact shows that the higher order terms in the expansion of the Hamiltonian \[13\] are important for getting the correct magnitude of the exponent.

In the second case we looked for the best parameters for

$$\ln(J) = C - \delta \sqrt{a} - \beta \ln(\sqrt{a}).$$

We obtained $C = 2.02, \delta = 1.74, \beta = 1.88$.

The analytical formula for $J$ in Eq. \[13\] and more accurate fits of the numerical calculation data allows one to estimate the value of $J$ for the Wigner crystal and Wigner liquid states in the large region of densities.

Due to geometric frustration collinear long-range antiferromagnetic order is, strictly speaking, not possible on a triangular lattice. The possible solution in this case is a system spin wave function which in the zero approximation consists of spin zero pairs. The antiferromagnetic interaction increases when the distance between the electrons decreases. Therefore, there should be a tendency for nearby electrons coupled to spin zero pairs to move slightly toward each other. This phenomenon is usually called the “Spin-Peierls” mechanism.

It is interesting that the value of the spin-spin interaction $J = J(15)$ is comparable with the energy $\mu H$ in the experiments \[10\], where $H$ is the critical magnetic field destroying conductivity. We can speculate that this field effectively transforms the system to a ferromagnetic state. In the ferromagnetic state the conductivity should be smaller than in the antiferromagnetic state. Indeed, hopping of the electrons with opposite spins from one site to another is allowed by the Pauli principle. The magnetic field rearranges spins in the same direction. In this case such hopping is suppressed by Pauli blocking. This possibly destroys the conductivity.

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APPENDIX A: MATRIX ELEMENTS OF THE INTERACTION

The eigenfunctions and eigenvalues for the single-electron problem in the two-dimensional rectangle with sides $A = 3a$ and $B = \sqrt{3}a$ are given by Eq. \[17\]. The matrix element of the Coulomb interaction between two unfrozen electrons is given by the integral over the rectangle’s area

$$V_{i_1i_2j_1j_2} = \int \phi_{i_1}(r_1')^* \phi_{i_2}(r_2')^* \frac{1}{|r_1' - r_2'|} \phi_{j_1}(r_1') \phi_{j_2}(r_2') d^2r_1' d^2r_2'$$  \hspace{1cm} (A1)

It is convenient to change variables (see Fig. 1):

$$u = x_1' - x_2', \hspace{0.5cm} v = y_1' - y_2', \hspace{0.5cm} x_2'' = x_2', \hspace{0.5cm} y_2'' = y_2'$$  \hspace{1cm} (A2)

Introducing notations $\tilde{u} = \frac{u}{\pi}, \tilde{v} = \frac{v}{\pi}$, and

$$F_{n_1n_2n_3n_4}(\tilde{x}_1, \tilde{x}_2) = 4 \sin(\tilde{x}_1 n_1) \sin(\tilde{x}_1 n_3) \sin(\tilde{x}_2 n_2) \sin(\tilde{x}_2 n_4),$$

$$W_{n_1n_2n_3n_4}(\tilde{u}) = \int_{0}^{\pi - \tilde{u}} F_{n_1n_2n_3n_4}(\tilde{x}_2 + \tilde{u}, \tilde{x}_2) d\tilde{x}_2$$

$$J_{n_1n_2n_3n_4} = \frac{1}{2} \left(1 + (-1)^{n_1+n_2+n_3+n_4}\right),$$  \hspace{1cm} (A3)

the matrix element \[A1\] can be rewritten as:

$$V_{i_1i_2j_1j_2} = J_{n_1n_2n_3n_4} J_{m_1m_2m_3m_4} \frac{4}{AB\pi^2} \int_{0}^{A} \int_{0}^{B} W_{n_1n_2n_3n_4}(\tilde{u}) W_{m_1m_2m_3m_4}(\tilde{v}) \frac{du \ dv}{\sqrt{u^2 + v^2}}.$$  \hspace{1cm} (A4)
In this transformation we use the following relation
\[
\int_0^\pi \frac{\pi-\hat{u}}{F_{n1n2n3n4}(x,x+\hat{u}) \ dx} = (-1)^{n_1+n_2+n_3+n_4} \int_0^{\pi-\hat{u}} F_{n1n2n3n4}(x+\hat{u},x) \ dx.
\]

It is convenient to calculate the double integral \(\text{(A4)}\) using polar coordinates \(\hat{u} = \frac{\pi u}{A} = \frac{\pi}{\sqrt{3}} \cos t, \ \hat{v} = \frac{\pi v}{A} = r \sin t\). Taking into account that \(\tan^{-1}(B/A) = \pi/6\) we find that
\[
V_{i1j2k3l4} = J_{n1n2n3n4} J_{m1m2m3m4} \frac{4}{3n^3} [V_1 + V_2]
\]
\[
V_1 = \int_0^{\pi/6} \int_0^{\pi\sqrt{3}/\cos(t)} W_{n1n2n3n4}(r \cos(t)/\sqrt{3}) W_{m1m2m3m4}(r \sin(t)) \ dr,
\]
\[
V_2 = \int_0^{\pi/3} \int_0^{\pi/\cos(t)} W_{n1n2n3n4}(r \sin(t)/\sqrt{3}) W_{m1m2m3m4}(r \cos(t)) \ dr.
\]

In order to write down an analytical expression for the function \(W_{n1n2n3n4}(\hat{u})\) let us introduce the following notations
\[
\begin{align*}
n &= |n_3 - n_1|, \quad n = 0, 1, 2, \ldots \\
m &= |n_4 - n_2|, \quad m = 0, 1, 2, \ldots \\
l &= n_3 + n_1, \quad l = 2, 3, \ldots \\
k &= n_4 + n_2, \quad k = 2, 3, \ldots
\end{align*}
\]

Using \(\text{(A3)}\) one can find that
\[
W_{n1n2n3n4}(\hat{u}) = \begin{cases} 
   f_1(n,m,l,k;\hat{u}) & \text{if } n = m 
eq 0 \\
   f_2(n,l,k;\hat{u}) & \text{if } l = k \\
   -f_2(n,m,l;\hat{u}) & \text{if } n = k \\
   -f_2(m,n,k;\hat{u}) & \text{if } m = l \\
   f_3(l,k;\hat{u}) & \text{if } n = m = 0 \\
   f_4(n,l;\hat{u}) & \text{if } n = m \neq 0 \text{ and } l = k \\
   f_5(l;\hat{u}) & \text{if } n = m = 0 \text{ and } l = k.
\end{cases}
\]

where
\[
\begin{align*}
f_1(n,m,l,k;u) &= \frac{n \sin(nu) (k^2 - m^2)}{(n^2 - k^2) (n^2 - m^2)} + \frac{m \sin(um) (l^2 - n^2)}{(m^2 - l^2) (m^2 - n^2)} - \frac{l \sin(lu) (k^2 - m^2)}{(l^2 - m^2) (l^2 - k^2)} - \frac{k \sin(uk) (l^2 - n^2)}{(-n^2 + k^2) (k^2 - l^2)} \\
f_2(n,m,l;u) &= \frac{n - u}{2} \cos(nu) + \frac{\sin(nu)}{2n} \left(1 + 2 \frac{n^4 - m^2 l^2}{(n^2 - m^2) (n^2 - l^2)} + \frac{m \sin(um) (n^2 - l^2)}{(m^2 - l^2) (m^2 - n^2)} + \frac{l \sin(lu) (n^2 - m^2)}{(l^2 - n^2) (l^2 - m^2)} \right) \\
f_3(n,m;u) &= \frac{n - u}{2} \frac{\sin(um)n^2}{(m^2 - n^2) m} - \frac{\sin(nu)m^2}{(n^2 - m^2) n} \\
f_4(n,m;u) &= \frac{n - u}{2} \cos(nu) + \frac{\sin(nu)}{2n} \left(3 \frac{n^2 + m^2}{n^2 - m^2} + \frac{m \sin(um) 3}{2m} \frac{m^2 + n^2}{m^2 - n^2} \right) \\
f_5(n;u) &= \frac{n - u}{2} \cos(nu) + \frac{3}{2n} \sin(nu)
\end{align*}
\]

This completes the description of the calculation procedure for the two-electron Coulomb matrix element. The advantage is that each of the two integrals in \(\text{(A3)}\) require numerical work equivalent only to the computation of a 1D integral.

Calculation of the single-particle matrix element of the external potential \(U(\mathbf{r})\) is much simpler. It is convenient to use \(x, y\) instead of \(x', y'\) (see Fig. 1). Then
\[
V_{i1k3}^{1(1)} = \frac{4}{AB} (-1)^{n+m} J_{n1n3} J_{m1m3} \int_0^{A/2} \int_0^{B/2} U(x,y) \times \left\{ \cos(\frac{\pi}{A}nx) - (-1)^{n_2} \cos(\frac{\pi}{A}lx) \right\} \left\{ \cos(\frac{\pi}{B}my) - (-1)^{m_2} \cos(\frac{\pi}{B}ky) \right\} dx \ dy.
\]

\(\text{(A9)}\)
where

\[
\begin{align*}
    n &= |n_3 - n_1| \\
    m &= |m_3 - m_1| \\
    l &= n_3 + n_1 \\
    k &= m_3 + m_1
\end{align*}
\]

(A10)