Strongly correlated Coulomb systems—a path integral Monte Carlo approach

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Gainesville, 22 May 2003
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F. Peeters (Antwerp),
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Outline

1. Introduction: correlated Coulomb systems
2. Journey to the center of Jupiter
3. Path integral Monte Carlo: idea and applications
4. Mesoscopic Coulomb systems. „Artificial atoms“
   *Wigner crystallization* in Quantum dots and heterostructures.
   *Single-electron control* of collective behavior
5. Summary and outlook
Coulomb Systems

$1eV \cong 10^4 K$

Electron density, 1/ccm
Correlation and Quantum effects

Coulomb Interaction:

\[ U_{ab}(r) = \frac{e_a e_b}{r} \]

\[ \Gamma \equiv \langle U \rangle / k_B T \]

\[ r_s \equiv \langle U \rangle / E_F \propto \bar{r} / a_B \]

\[ E_F \] - Fermi Energy \quad \[ a_B \] - Bohr Radius

\[ \chi = n \lambda^3 = 1 \]

Strong Coulomb correlations

\[ \Gamma = 1 \]

\[ r_s = 1 \]

DeBroglie wave length

\[ \lambda = \frac{h}{\sqrt{2\pi m k_B T}} \]

Overlap of wave functions, Spin effects

\[ \lambda = \bar{r} \]

Quantum effects

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Internal Energy of Coulomb Matter

Energy isotherm of hydrogen

\[ E = E^{ideal} + E^{int} \]

Equilibrium phase diagram of Coulomb systems

Universal scaling

length and energy scales: \( a_B, \ E_R \)

Hydrogen semiconductors

\( 0.5 \text{A} \quad 13.6 \text{eV} \)

\( 100 \text{A} \quad 5 \cdot 10^{-3} \text{eV} \)

M. Bonitz, Physik Journal 7/8 2002, p.69

THEORY

Weak U: perturbation theory, Strong U: first-principle simulations

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Mysterious Hydrogen

Insulator-metal transition, Anomalous compressibility (?), plasma phase transition (?)

Filinov, Fortov, Bonitz, Levashev, JETP Letters 74, 384 (2001)

Hydrogen conductivity (experiment) in cm/ohm

- Nellis et al.
+ Fortov et al.

T=3,000-10,000K
(Some of) the animations, shown at this place in the talk can be viewed at my web page: http://elde.mpg.uni-rostock.de/mb

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Correlated electron-hole-plasma in optically excited semiconductors

Quasi-equilibrium

Example: 2-dimensional quantum well

\[ k_B T = 0.2E_R, \]

increase \( n \): \( r_s = 10 \rightarrow 0.5 \)
Rigoros Path integral Monte Carlo simulations

Full account of
- Coulomb interaction,
- Quantum effects
- Spin of electrons and holes

Dots: Electron- (hole-)
Position and extension (Wave function)
in 2D Quantum well

\[ r_s = \bar{r}/a_B \]

V. Filinov, W. Hoyer, S.W. Koch, and M. Bonitz 2001

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Partially ionized e-h-plasma

Excitons
Biexcitons

Trions,
Cluster,
...

$T = 10 \text{ K}, n^{2D} = 1.19 \times 10^8 \text{ cm}^{-2}, r_s^{2D} = 4.158$
Electron-hole-droplets
partially delocalized phase
Correlated delocalized phase

Fermi liquid

$T = 10 \, \text{K}, \; n^{2D} = 5203 \times 10^8 \, \text{cm}^{-2}, \; r_s^{2D} = 0.63$
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Path integral quantum Monte Carlo

Equilibrium: Minimize total Energy
\[ F = -k_B T \ln Z \]

Partition function
\[ Z = \text{Tr} \hat{\rho} \] yields all thermodynamic quantities

N-particle-density operator (canonical ensemble):
\[ \hat{\rho} = e^{-\hat{H}/k_B T}, \quad \hat{H} = \hat{K} + \hat{U} \]

Problem: \( \hat{\rho} \) known only for limiting cases, where \( \langle \hat{U} \rangle \ll \langle \hat{K} \rangle \)

Feynman:
\[ e^{-\hat{H}/k_B T} \equiv \left[ e^{-\hat{H}/(k_B M T)} \right]^M \] Use known result for \( e^{-\hat{H}/(k_B M T)} \)

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First-principle thermodynamics of correlated quantum systems

N-particle density operator: \( \rho = \exp(-\beta H) \)

Path Integral Representation

**Partition function** (distinguishable particles):

\[
Z(\beta) = \int dr_1 \langle r_1 | e^{-\beta \hat{H}} | r_1 \rangle = \int dr_1 \cdots \int dr_M \prod_{\tau=1}^{M} \langle r_{\tau} | e^{-\beta \hat{H}/M} | r_{\tau+1} \rangle
\]

\( \beta = 1/k_B T, r_2, \ldots r_M \) : intermediate coordinates,
\( \rho^{HT} = \exp\{-\beta \hat{H}/M\} \) : high-temperature density matrix

\( r_i \equiv \{ r_i^1, r_i^2, \ldots, r_i^N \} \)

**Partition function for Fermions:**

\[
Z(\beta)_F = \int dr_1 \cdots \int dr_M \frac{1}{N!} \sum_P (-1)^{\delta_P} \langle r_1 | e^{-\beta \hat{H}/M} | r_2 \rangle \cdots \langle r_{M-1} | e^{-\beta \hat{H}/M} | \hat{P} r_M \rangle,
\]

\( \hat{P} : N\)-particle permutation operator (fermionic exchange).
Illustration: Snapshots of closed electron „paths“

„Spin-less“ particles

Exchange included

5 Electrons in 2D simulation box, Path ends labeled by thick dots

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Applications of Path integral Monte Carlo

→ *first-principle calculations of equilibrium properties:*

a) *many-body* systems: high-order correlations, partial ionization, bound state renormalization, Bose condensation etc.

b) *few-body* systems: binding energies of atoms, molecules/excitons, bi-excitons, trions and larger complexes follow non-perturbatively

c) „Exact“ effective quantum *pair potentials*: input for DFT simulations or semiclassical molecular dynamics

d) *mesoscopic* systems (N=10...50): Fermi liquid behavior, Wigner crystallization etc.
Electron-hole bound states in quantum wells

- Quasi-2D e-h-plasma, finite well width $z$
- renormalized in-plane (well) interaction, e-h-wave function overlap varies (non-monotonically) with $z$

Largest e-h overlap $\rightarrow$ largest exciton binding energy
Exciton and biexciton binding energies as function of quantum well width $L$

$\rightarrow$ PIMC simulations very accurate, avoid any basis expansion, applicable to finite temperature, geometry of minor importance

Effective Quantum pair potentials

1. Exact pair potential from exact 2-particle density matrix (numer.)

\[ U_{pair}^{ab}(r) = -k_B T \ln \rho_{ab}(r, r) \]

2. Derive analytical potential (1 fit parameter)

Result:

Drastic improvement of previous potentials (Kelbg, Deutsch etc.)

<table>
<thead>
<tr>
<th>applicable to strong coupling including bound states</th>
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\[ U, \text{ units of Ryd} \]

\[ T=30,000 \text{K} \]
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Mesoscopic Electron clusters in Quantum dots („artificial atoms“)

Model: N=1...100 Electrons in spherical harmonic „trap“

quasi-2-dim confinement by external Fields or Heterostructures

Classical ground state: \( E_{\text{kin}} = 0 \)
- Shell structure, hexagonal and spherical Symmetry
- strong N-dependence

At high density:
- strong Coulomb interaction,
- Quantum and spin effects

\[ \hat{H} = - \sum_{i=1}^{N} \frac{\hbar^2 \nabla_i^2}{2m_i} + \sum_{i=1}^{N} \frac{m_i^+ \omega_i^2 r_i^2}{2} + \sum_{i<j}^{N} \frac{e^2}{\epsilon_0 |\mathbf{r}_{ij}|} \]

\( N=19 \) \hspace{1cm} \( N=42 \) \hspace{1cm} \( N=82 \)

\( \rightarrow \) Challenge for Theory!

Path integral Monte Carlo

Bedanov/Peeters

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Coulomb (Wigner) crystal

Ground state of the electron gas in metals

E. Wigner, Physical Review 46, 1002 (1934):

→ exchange and correlation energy of the electron gas

„If the electrons had no kinetic energy, they would settle in configurations which correspond to the absolute minima of the potential energy. These are close-packed lattice configurations, with energies very near to that of the body-centered lattice....“
Experimentally observed Coulomb (Wigner) crystals

- electrons on helium droplets,
- dusty plasmas,
- ions in traps or storage rings
- predicted: in White dwarf stars
\( \rightarrow \text{All classical systems (} B = 0 \text{)} \)

Ca\(^{+} \) ions in Paul trap
G. Werth, Uni Mainz

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Wigner crystallization of quantum electron clusters

Variation of temperature or density (confinement)

Wigner crystal expected for: \( \frac{U_{\text{Coulomb}}}{E_{\text{Kin}}} \geq 40 \) (Coupling Parameter \( \Gamma, r_s \))

Result: two crystal-“phases“: Intra-shell- and Inter-shell order [Lozovik, Bedanov/Peeters]

Increase density: \( \rightarrow \) growing overlap of electrons:
- OO-crystal \( \rightarrow \) RO-crystal \( \rightarrow \) „liquid“

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Phase diagram of the mesoscopic Wigner crystal

Classical Liquid

Temperature

Confinement Strength

RM - Radial Melting, OM – Rotational Melting

Particle Number

Quantum Liquid („Wigner-Molecule“)

Wigner Crystal strongest correlations

\[ \Gamma \equiv \langle U \rangle / k_B T \]

\[ r_s \equiv \langle U \rangle / E_F \propto \bar{r} / a_B \]

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Coulomb Bilayers

Two mesoscopic 2D clusters at fixed distance $d$

\[
\hat{H} = -\sum_{i=1}^{N} \frac{\hbar^2 \nabla_i^2}{2m_i^*} + \sum_{i=1}^{N} \frac{m_i^* \omega_0^2 r_i^2}{2} + \sum_{i=1}^{2} \sum_{i<j}^{N_1} \frac{e^2}{\epsilon_b |\mathbf{r}_{ij}|} + \frac{1}{2} \sum_{i=1}^{N_1} \sum_{j=1}^{N_2} \frac{e^2}{\epsilon_b \sqrt{r_{ij}^2 + d^2}}.
\]

New effect: influence of \textit{inter-layer} correlations on ground state, crystal

\[d = 0.3 \quad \text{and} \quad d = 0.5\]

$N_1 = N_2 = 19$

$T = 0, \quad B = 0$

$d$ in units of interparticle distance

→ Change of crystal symmetry with $d$

Reduction of $d$: change from Coulomb to dipole interaction at small $x$:

$$U_{\text{cor}}(a, x) = \frac{\mu^2(d, x)}{a^3} - 2 \frac{e^2}{d} + O(x^6)$$

$$\mu(d, x) = ed\sqrt{1 - \frac{3}{4}x^2}$$

→ Interaction of two excitons
→ Binding energy/interaction tuneable
→ Exciton crystals possible

Ground state exciton distance in trap:

$$a(\omega, d) \approx a_0(\omega, d) \sqrt{1 - \frac{5}{4} \frac{d^2}{a_D^2(\omega, d)}}$$

$$a_D(\omega, d) = \frac{3\mu^2}{m\omega^2}$$

*P. Ludwig, A. Filinov, M. Bonitz (2002)*

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Wigner crystallization in quantum e-h bilayers

Result: two crystal phases: with/without inter-layer ordering

- n=0.07
- Exciton liquid
- 3 electrons and 3 holes, fixed layer distance d

- n=0.15
- Exciton crystal

- n=0.2
- Decoupled e/h crystals

- n=0.25
- Decoupled e/h liquids

probability to find individual electrons/holes (one e is fixed): red=0 → pink=max

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Phase diagram of mesoscopic e-h bilayers

Ne=Nh=8, d in units of exciton Bohr radius, Ha=2 Ryd

A. Filinov, P. Ludwig, V. Golubnychiy, M. Bonitz, Yu. Lozovik, phys. stat. sol. (b) 2003, accepted
Particle number dependence of melting parameters in single dot

<table>
<thead>
<tr>
<th></th>
<th>$N$</th>
<th>$\Gamma_0$</th>
<th>$\Gamma_r$</th>
<th>$r_{SO}$</th>
<th>$r_{sr}$</th>
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<td>19</td>
<td>330</td>
<td>154</td>
<td>400</td>
<td>64</td>
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<tr>
<td></td>
<td>20</td>
<td>$3.4 \cdot 10^{11}$</td>
<td>83</td>
<td>$3.0 \cdot 10^{11}$</td>
<td>51</td>
</tr>
<tr>
<td>Bulk</td>
<td>$\infty$</td>
<td>—</td>
<td>137</td>
<td>—</td>
<td>37</td>
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„Dusty Plasmas“

Alternative road to strong correlations: → very high particle charge $Q$

Experiments of A. Piel, A. Melzer and co-workers (Univ. Kiel)

**Dust particles:** $Q=(5000...10000)$ e

d $≈ 0.0095$mm

Charging in HF-discharge in plasma chamber

Vertical E-field compensates gravitation → dust particles „float“

Web page [http://www.ieap.uni-kiel.de/plasma/ag-piel](http://www.ieap.uni-kiel.de/plasma/ag-piel)
Response to external excitation

Experiments of A. Piel, A. Melzer and co-workers (Univ. Kiel)

Two-dimensional mesoscopic dusty plasma crystal

Tangential excitation by 2 laser pulses

\[ \Gamma^{20}_{OM} \gg \Gamma \gg \Gamma^{19}_{OM} \]

N=19

N=20

Web page: http://www.ieap.uni-kiel.de/plasma/ag-piel
Potential Applications

Crystallization/melting without change of density and temperature:
→ by addition/removal of a single electron

„switch“ between insulator (crystal) and conductor

Single electron- „transistor“

$T = \frac{1}{\Gamma}$

$n \propto \frac{1}{r_s^2}$

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Summary and Outlook

I. Coulomb systems: fascinating variety of structures
   - Planets/stars, atoms, molecules, excitons, Wigner crystal...
   - Coulomb interaction important for many fields. Analogies, Overlap...

II. Quantum effects and strong correlations:

<table>
<thead>
<tr>
<th></th>
<th>Correlations</th>
<th>Quantum effects</th>
<th>Dynamics, Transport</th>
<th>Fast processes</th>
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→ there is no universal theory/computational method!

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