Trapped Ion Liquids and Crystals

John Bollinger
NIST-Boulder
Ion storage group

Wayne Itano, David Wineland, Joseph Tan, Pei Huang, Brana Jelenkovic, Travis Mitchell, Brad King, Jason Kriesel, Marie Jensen, Taro Hasegawa, Dan Dubin – UCSD (theory)
Outline:

- **Introduction:**
  Penning traps, thermal equilibrium, one-component plasmas, strong correlation

- **Observation of crystalline structure:**
  Bragg scattering, real imaging, structural phase transitions

- Can we observe the predicted liquid-solid phase transition?

- **Modes**

  ➡️ No quantum mechanics; everything in this talk uses classical physics

  ➡️ Please ask questions!!

  ➡️ Ref: Dubin and O’Neil, Rev. Mod. Physics **71**, 87 (1999)
Types of traps in atomic physics

Rf or Paul Trap
RF & DC Voltages

Penning Trap
(or Penning-Malmberg trap)
DC Voltages & Static B-Field

(see Wineland lectures)

good for tight confinement
and laser cooling smaller
numbers of particles

good for laser cooling larger
numbers of particles
Penning–Malmberg traps

\[ ^9\text{Be}^+ \]
\[ \nu_c \sim 7.6 \text{ MHz} \]
\[ \nu_z \sim 800 \text{ kHz} \]
\[ \nu_m \sim 40 \text{ kHz} \]

- g-factors, atomic phys.  
  U of Wash., Mainz, Imperial College, ...

- mass spectroscopy  
  U of Wash., MIT, Harvard, ISOLDE/CERN, ...

- cluster studies  
  Mainz/Griefswald

- non-neutral plasmas  
  UCSD, Berkeley, Princeton, NIST, ...

- anti-matter  
  UCSD, Harvard, CERN, Swansea, ...
NIST Penning trap

4.5 Tesla
Super Conducting Solenoid

Quartz
Vacuum envelope
$P < 10^{-10}$ Torr
Non-neutral plasmas in traps evolve into bounded thermal equilibrium states

thermal equilibrium, Hamiltonian and total canonical angular momentum conserved

\[ f(r, v) \propto \exp[-(h + \omega_r p_\theta)/k_B T] \]
where \( h = \frac{mv^2}{2} + e\phi(r) \) and \( p_\theta = mv_\theta r + \frac{eB}{2c} r^2 \)

\[ f(r, v) \propto n(r, z) \exp[-\frac{m}{2k_B T}(v + \omega_r r \hat{\theta})^2] \]

- density distribution
- plasma rotates rigidly at frequency \( \omega_r \)

\[ n(r, z) \propto \exp\left\{-\frac{1}{k_B T}[e\phi_p(r, z) + e\phi_T(r, z) + m\omega_r(\Omega_c - \omega_r) \frac{r^2}{2}]\right\} \]

\( \Omega_c = \frac{eB}{mc} = \) cyclotron frequency
- plasma potential
- trap potential
- Lorentz force potential
- centrifugal potential

Lorentz-force potential gives radial confinement !!
Equilibrium plasma properties

- thermal equilibrium $\Rightarrow$ rigid rotation $\omega_r$

- $T \sim 0 \Rightarrow$ constant plasma density,
  
  $n_o = 2\varepsilon_o m\omega_r (\Omega_c - \omega_r)/e^2$,
  
  $\Omega_c$ = cyclotron frequency
  
  plasma density $\rightarrow 0$ over a Debye length $\lambda_D = [k_B T/(4\pi n_o e^2)]^{1/2}$

- quadratic trap potential, $e\phi_T \sim m\omega_z^2 (z^2 - r^2/2) \Rightarrow$ plasma shape is a spheroid

  aspect ratio $\alpha \equiv z_o/r_o$

  determined by $\omega_r$

  $\frac{\omega_z^2}{2\omega_r (\Omega_c - \omega_r)} = Q_1^0 \left[ \frac{\alpha}{(\alpha^2 - 1)^{1/2}} \right] / (\alpha^2 - 1)^{1/2}$

  associated Legendre function
Plasma aspect ratio determined by $\omega_r$

$$\frac{\omega_z^2}{2\omega_r(\Omega_c - \omega_r)} = Q_1^0 \left[ \frac{\alpha}{(\alpha^2 - 1)^{1/2}} \right]$$

- Simple equilibrium theory describes the plasma shapes
- Experimental measurements of plasma shape vs $\omega_r$
- $\alpha = z_0/r_0$
Ions in a trap are an example of a one component plasma

- One component plasma (OCP) – consists of a single species of charged particles immersed in a neutralizing background charge

- Ions in a trap are an example of an OCP (Malmberg and O’Neil PRL 39, (77))

\[
n(r, z) \propto \exp\left\{-\frac{1}{k_B T} \left[ e \phi_p(r, z) + e \phi_T(r, z) + m \omega_r (\Omega_c - \omega_r) \frac{r^2}{2} \right] \right\}
\]

Looks like neutralizing background

\[
\nabla^2 \{ \phi_T(r, z) + \frac{1}{e} m \omega_r (\Omega_c - \omega_r) \frac{r^2}{2} \} = -4\pi e n_{\text{bkgnd}}
\]

\[
n_{\text{bkgnd}} = -\frac{m \omega_r (\Omega_c - \omega_r)}{2\pi e^2}
\]

- Thermodynamic state of an OCP determined by:

\[
\Gamma \equiv \frac{q^2}{a_{WS} k_B T}, \quad \frac{4}{3} \pi a_{WS}^3 n \equiv 1 \quad \Gamma \approx \frac{\text{potential energy between neighboring ions}}{\text{ion thermal energy}}
\]

\[
\Gamma > 1 \Rightarrow \text{strongly coupled OCP}
\]
Why are strongly coupled OCP’s interesting?

- Strongly coupled OCP’s are models of dense astrophysical matter – example: outer crust of a neutron star

- For an infinite OCP, \( \Gamma > 2 \Rightarrow \) liquid behavior
  \( \Gamma \sim 173 \Rightarrow \) liquid-solid phase transition to bcc lattice
  - Brush, Salin, Teller (1966) \( \Gamma \sim 125 \)
  - Hansen (1973) \( \Gamma \sim 155 \)
  - Slatterly, Doolen, DeWitt (1980) \( \Gamma \sim 168 \)
  - Ichimaru; DeWitt; Dubin (87-93) \( \Gamma \sim 172-174 \)

- Coulomb energies/ion of bulk bcc, fcc, and hcp lattices differ by < \( 10^{-4} \)

- with trapped ions, \( n_0 \sim 10^9 \text{ cm}^{-3} \)
  \( \Gamma > 500 \)

\( T < 5 \text{ mK} \Rightarrow \Gamma > 500 \)
Plasmas vs strongly coupled plasmas

Increasing Correlation $\Gamma$

$\Gamma = 2$

$\Gamma = 175$

Laser-cooled ion crystals

White Dwarf Interiors

Neutron Star Crusts

Number Density (Charged Particles / m$^3$)

Temperature (K)
Are there other laboratory strongly coupled OCP’s?

- rf traps - Drewsen (Aarhus) – rf micromotion limits plasma size

- ion storage rings  \( T_{||} \sim \) few mK→K,  \( T_{\perp} >> T_{||} \)
  - ion strings observed in GSI (200 MeV/\( \mu \), fully stripped ions)
  -1 eV ion crystals observed in PALLAS (Schramm, Nature 2001)

- ultra-cold neutral plasmas  \( n \geq 10^9 \text{ cm}^{-3}, T \sim 10 \mu \text{K} \) before photo-ionization
  Rolston (NIST); Raithel (Mich.); Killian (Rice); Eyler/Gould (UConn);
  Bergerson (BYU); Gallagher (UVA); ...

other strongly coupled (screened) Coulomb systems

- dusty plasma crystals
  Melzer, et al., PRE 53, 2757 (96)
  Thomas et al., Nature 379, 806 (96)
  Pieper, et al., PRE 54, 5636 (96)

- colloidal suspensions
  Murray & Grier, American Scientist 83, 238 (95)
  Vos et al., Langmuir 13, 6004 (97)

3-d dusty plasma crystal; from Goree, U. of Iowa
Strongly coupled plasma work in ion traps

- 1987 – Coulomb clusters in Paul traps
  MPI Garching (Walther)
  NIST (Wineland)

- 1988 – shell structures in Penning traps
  NIST group

- 1992 – 1-D periodic crystals in linear Paul traps
  MPI Garching
  *Nature* **357**, 310 (92)

- 1998 – 1-D periodic crystals with plasma diameter
  > 30 $a_{WS}$
  Aarhus group
  *PRL* **81**, 2878 (98)
How large must a plasma be to exhibit a bcc lattice?

1989 - Dubin, planar model  PRA 40, 1140 (89)
result: plasma dimensions $\geq 60$ interparticle spacings required for bulk behavior
$N > 10^5$ in a spherical plasma $\Rightarrow$ bcc lattice

2001 – Totsji, simulations, spherical plasmas, $N \leq 120$ k
PRL 88, 125002 (2002)
result: $N > 15$ k in a spherical plasma $\Rightarrow$ bcc lattice
Doppler laser cooling

$^9\text{Be}^+$

$P_{3/2}$

$S_{1/2}$

(neglecting hyperfine structure)

$\nu_0 (\lambda=313 \text{ nm})$

$+3/2$

$+1/2$

$-1/2$

$-3/2$

$+1/2$

$-1/2$

$T_{\text{min}}(^9\text{Be}^+) \sim 0.5 \text{ mK}$

$T_{\text{measured}} < 1 \text{ mK}$

Laser torque

The laser beam position and frequency control the torque and $\omega_r$

With the laser beam directed as shown, increasing torque $\Rightarrow$ increasing $\omega_r \Rightarrow$ decreasing radius
NIST Bragg scattering set-up

\[ f_{\text{rotation}} = 240 \text{ kHz} \]
\[ n = 7.2 \times 10^8 / \text{cm}^3 \]

Bragg scattering from spherical plasmas with N~ 270 k ions

Evidence for bcc crystals
Rotating wall control of the plasma rotation frequency

Huang, et al. (UCSD), PRL 78, 875 (97)
Huang, et al. (NIST), PRL 80, 73 (98)
Phase-locked control of the plasma rotation frequency


time averaged Bragg scattering

camera strobed by the rotating wall

- $N > 200,000$ ions $\Rightarrow$ always observe bcc crystalline patterns

- $100,000 > N > 20,000$ $\Rightarrow$ observe fcc, hcp?, in addition to bcc
Real space imaging

Mitchell. et al., Science 282, 1290 (98)
Top-view images in a spherical plasma of ~180,000 ions

\[ \omega_r = 2\pi \times 120 \text{ kHz} \]

**bcc (100) plane**
- Predicted spacing: 12.5 \( \mu \text{m} \)
- Measured: 12.8 \( \pm 0.3 \) \( \mu \text{m} \)

**bcc (111) plane**
- Predicted spacing: 14.4 \( \mu \text{m} \)
- Measured: 14.6 \( \pm 0.3 \) \( \mu \text{m} \)
Real-space images with planar plasmas

with planar plasmas all the ions can reside within the depth of focus
Planar structural phases can be ‘tuned’ by changing $\omega_r$.”

**top-views**

- 65.70 kHz
- 66.50 kHz

**side-views**

- 1 lattice plane, hexagonal order
- 2 planes, cubic order
Top- (a,b) and side-view (c) images of crystallized $^9\text{Be}^+$ ions contained in a Penning trap. The energetically favored phase structure can be selected by changing the density or shape of the ion plasma. Examples of the (a) staggered rhombic and (b) hexagonal close packed phases are shown.
Theoretical curve from Dan Dubin, UCSD

Stick-slip motion of the crystal rotation

- not a true phase lock!
- frequency offset ($\omega_r - \omega_{\text{wall}}$) due to creep of 2 -18 mHz
- regions of phase-locked separated by sudden slips in the crystal orientation
- stick-slip motion due to competition between ? laser and rotating wall torques
- mean time between slips $\sim 10$ s; what triggers the slips?
Summary of crystal observations

spherical plasmas
  ● bcc crystals observed with N > 200 k ions
  ● other crystal types (fcc, hcp) observed for 20 k < N < 200 k
  ● shell structure observed for N < 20 k ions

planar plasmas
  ● structural phase transitions between rhombic planes (bcc-like) and hexagonal planes (fcc-like or bcc-like)
  ● good agreement with the predicted T=0 minimum energy lattice for plasmas < 10 lattice planes thick

Can we observe the predicted thermodynamic phase transition at Γ~172?
Phase transition can be determined from the specific heat

- details of specific heat at the phase transition appear to only weakly depend on $N$ and the type of structure.  Schiffer, PRL 88, 205003 (2002)

molecular dynamics simulation with a spherical plasma with 10 000 ions in shells
Measure specific heat with a constant (or known) dE/dt

- Our dominant heating rate appears to be due to background collisions
  ⇒ constant energy input dE/dt to the plasma, independent of liquid or solid state

- For a sufficiently large plasma where surface effects can be neglected, $E_R = E_R^{(0)} + U$
  where $E_R$ = energy of the plasma in the rotating frame
  $E_R^{(0)}$ = zero-temperature mean-field plasma energy (no correlations, fluid description depends only on $\omega_r$ and trap parameters)
  $U = 3NkT/2 + U_{corr}$ = energy of infinite OCP

- Assume constant $\omega_r$, $dE/dt = (3\times k\times 0.1 \text{ K/s})/2$, theoretical expression for $U$

\[\begin{array}{c}
\text{Temperature (K)} \\
\begin{array}{c}
T_{final} \\
T_{ideal gas}
\end{array}
\end{array}\]

\[\begin{array}{c}
\text{Time (s)} \\
0.1 \\
0.05 \\
2\times 10^{-4}
\end{array}\]

\[\begin{array}{c}
\text{ideal gas} \\
\text{strongly coupled OCP} \\
\text{latent heat}
\end{array}\]
Energy level diagram for $^9\text{Be}^+$ in high magnetic field

$B_{\text{high}} \Rightarrow (m_I, m_J)$ basis

Temperature measurement

- Monitor cooling laser fluorescence
- Depopulation laser resonant $\Rightarrow$ decrease in fluorescence
- Measure Doppler broadening $\Rightarrow T$

$\lambda = 313 \text{ nm}$

- $2p\,^2P_{3/2}$
  - $F = 0, 1, 2, 3$
  - $(m_I, +3/2)$
  - $(m_I, +1/2)$
  - $(m_I, -1/2)$
  - $(m_I, -3/2)$

- $2p\,^2P_{1/2}$
  - $F = 1, 2$
  - $(m_I, -1/2)$
  - $(m_I, +1/2)$

- $2s\,^2S_{1/2}$
  - $F = 1, 2$
  - $(m_I, m_J)$

- $\Rightarrow$ fluorescence cooling depopulation

$F = 0, 1, 2, 3$
Measurement of the ion temperature

Fluorescence measurements

Cooling beams

Signal = $C_{after}/C_{before}$

$t_{delay} = 0$ ms

$w_D = 5 \pm 2$ MHz

$T = 1.6 \pm 0.6$ mK

$t_{delay} = 1000$ ms

$w_D = 215 \pm 5$ MHz

$T = 2.47 \pm 0.05$ K
Heating rate measurements

Slow heating at short times: 50-100 mK/s

Miniature RF-traps: ~50 mK/s
Onset of abnormal heating is at $T = 10 \text{ mK}$
- the temperature of the solid–liquid phase transition
- but what is the explanation?
Pressure dependence

Clear correlation between pressure and heating!

Pressure increase ⇒ “step” occurs at earlier time
larger step
Heating rate measurements on a clean cloud of Be\(^+\) ions

- no anomalous heating at the solid-liquid phase transition!!
- anomalous heating requires heavy mass ions
Sympathetic cooling of impurity ion cyclotron motion

- Impurity ions heated by residual gas collisions; sympathetically cooled by laser-cooled Be\(^+\) ions

\[
\frac{dT_i}{dt} = r(T_i - T_{Be}) + h \\
T_i = \text{impurity ion temperature} \\
\therefore \quad 0.1 \text{ K/s}
\]

- Phase-locked control of \(\omega_r\) \(\Rightarrow\) heavy ions crystallized \(\Rightarrow\) \(T_i < 10 \text{ mK}\)

- But large magnetic field \(\Rightarrow\) \(r_c = \langle v \rangle / \Omega_c \ll a_{WS}\) \(\Rightarrow\) cyclotron motion decouples from parallel

- Sympathetic cooling rate in crystalline state approximately given by (Dan Dubin, UCSD):

\[
r_\perp \sim \frac{w_{Pe}^4}{\Omega_c^4} \gamma \sim 5 \times 10^{-4} \gamma \quad \text{for impurity ion cyclotron motion}
\]

\[
r_{||} \sim \gamma
\]

\(\gamma\) = laser damping rate for Be\(^+\) < 1 kHz for current experimental work

- \(h \sim 0.1 \text{ K/s, } r_\gamma < 0.1 \text{ Hz } \Rightarrow\) \(T_{\gamma,i} > 1 \text{ K}\) which is the correct order of magnitude to account for the observed heating
Impurity ion cyclotron motion drive increases the heating

Rapid heating explained by a release of the impurity ions cyclotron energy when the parallel ion temperature increases above 10 mK, the temperature of the solid-liquid phase transition.

Is there a theory for why the impurity ion cyclotron energy is released for T > 10 mK?

Can we get rid of the heating?
Can we get rid of the heating?

- work with clean plasmas with no impurity ions
- stay in the liquid phase for as long as possible

Plasma spends less than 100 ms in the solid phase prior to measurement

⇒ No additional energy is released at the solid–liquid phase transition
Is there a theory for why the impurity ion cyclotron energy is released for $T > 10 \text{ mK}$?

- need a theory of perp/parallel energy equipartition
- such a theory exists in the absence of correlations ($\Gamma < 1$) 
  O’Neil, Glinsky, Rosenbluth, Ichimaru,… (1992)
  \[
  \frac{T_\parallel}{dt} = n_o \bar{v} b^2 I(\bar{\kappa})(T_\perp - T_\parallel)
  \]
  \[
  \bar{b} = \frac{2e^2}{k_BT_\parallel}, \bar{v} = \sqrt{\frac{2k_BT_\parallel}{m}},
  \]
  \[
  \bar{\kappa} = \frac{\Omega_c \bar{v}}{\bar{v}} = \text{distance of closest approach} / \text{cyclotron radius}
  \]
- O’Neil/Glinsky theory is more than $10^7$ too small to explain our observed heating rate
- rare close collisions between energetic particles are responsible for the equipartition
- close collisions can be enhanced in a correlated plasma where the Coulomb repulsion is screened by the neutralizing background


nuclear reaction rate ($\Gamma > 1$) $\sim \exp(\Gamma)\mathcal{L}$

nuclear reaction rate($\Gamma<<1$)
Summary of can we observe the phase transition?

- the first-order liquid-solid phase transition at $\Gamma \sim 170$ has never been directly observed in an experiment.

- this should be possible by measuring the latent heat or possibly through Bragg scattering.

- perp/parallel energy equipartition studies can measure the enhancement of close collisions for $\Gamma > 1$ for the first time.
Plasma waves excited by laser radiation pressure

Push on the top of a rotating ion crystal with a laser beam.

The waves which are excited interfere to form a “wake”.

100 μm

$\omega_{rot}$

top-view camera

side-view camera

parallel cooling beam
Variations in image intensity (shown with a false color scale) correspond to variations in the axial motion of the ions in the crystal.

A large spectrum of modes are excited, which interfere to form a wake that is stationary in the source (lab) frame.
Wakes are Stationary in the frame of the source (ship).

Analogous to wakes in water

\[ \omega = \sqrt{gk} \]

Dispersion curve for gravity waves
Analyze image to obtain $\lambda$ and $\omega$

Analyze wake pattern in an annular region that is directly “behind” the push beam.

Fit to damped sinusoid to get $\lambda$:

$$y = C_0 + C_1 \sin(C_2 x + C_3) e^{-C_4 x}$$

$$C_2 = k = 2\pi/\lambda$$

Directly behind the beam, the stationary phase condition gives:

$$V_{source} = \omega_{Rot} r_{source} = \omega/k$$

For this case:

$\lambda = 180 \, \mu m$ \quad $\delta v_z < 1 \, \text{m/s}$

$\omega/2\pi = 500 \, \text{kHz}$ \quad $\delta z < 0.3 \, \mu m$
The data agrees with the theoretical dispersion relationship for drumhead waves in a plasma slab of thickness $2Z_p$. 

$$\tan \left( \frac{kZ_p}{\sqrt{\omega_p^2 / \omega^2 - 1}} \right) = \sqrt{\omega_p^2 / \omega^2 - 1}$$

Dispersion relationship
Theory replicates experiment

Side View

Top View

Experiment

Theory - Dubin

$\delta v_z > 0$

$\delta v_z < 0$
Future areas of work

- Observation of the liquid-solid phase transition
- Studies of the enhancement of the perp/parallel equilibration due to strong correlation
- Shear modes

Application:
- Entangled states of trapped ions

Deterministic entanglement through an $\exp(i\chi J_z^2t)$ interaction

Application:
- Single plane of $^{9}{\text{Be}}^+$ ions

Graphs showing
- $T_{\text{final}} / T_{\text{ideal gas}}$
- $T$ vs. $t_{\text{Delay}}$

Parameters:
- $N_{\text{ion}} = 440,000$
- $V_{\text{trap}} = 500$ V
- $f_{\text{rot}} = 64$ kHz