Breakdown of Fermi Degeneracy in the Simplest Liquid Metal



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Summary

We report the experimental observation of the breakdown of Fermi degeneracy in fluid metallic deuterium at $T = 0.4 T_F$

- We have studied the optical conduction of shocked deuterium as a function of temperature up to its Fermi temperature, T_F
- At 0.4 $T_{\rm F}$, we observed the quantum degenerate to classical crossover in dense fluid deuterium, challenging the standard convention in dense plasma literature, which assumes $T = T_{\rm F}$ demacrates such boundary
- Our data provide an invaluable benchmark to dense plasma transport models over an order of magnitude in *T*





Collaborators

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In 1926, Fermi and Dirac independently introduced the quantum statistics describing indistinguishable systems with antisymmetrical eigenfunctions (fermions)



On the Theory of Quantum Mechanics. By P. A. M. DIRAC, St. John's College, Cambridge.

(Communicated by R. H. Fowler, F.R.S.-Received August 26, 1926.)

"The solution with symmetrical eigenfunctions must be the correct one when applied to light quanta, since it is known that the Einstein-Bose statistical mechanics lead to Planck's law of black-body radiation. The solution with antisymmetrical eigenfunctions, though, is probably the correct one for gas molecules, since it is known to be the correct one for electrons in the atom."

P. M. Dirac*



CONSIDERAZIONI SULLA QUANTIZZAZIONE DEI SISTEMI CHE CONTENGONO DEGLI ELEMENTI IDENTICI.

Nota di ENRICO FERMI.

E. Fermi**

Le regole date da Sommerfeld per la determinazione delle orbite quantiche dei sistemi che ammettono la separazione delle variabili, e che, come si sa, si riducono ad imporre che per tali orbite gli integrali delle fasi $(\int) p \, dq$, siano tutti multipli interi della costante h di Planck, si sono mostrate





*P. A. M. Dirac, Proc. Roy. Soc. A. <u>112</u>, 661 (1926). ** E. Fermi, Rend. Lincei 3, 145 (1926).

The Fermi–Dirac statistics are a direct manifestation of the Pauli exclusion principle

- This gives rise to the structure of the periodic table
- The electron conduction in metals and semiconductor
- The quantum hall effect
- Degeneracy pressure in compact astrophysical objects, e.g., neutron stars and white dwarfs





These Fermi–Dirac statistics, at high enough *T*, assume the classical Maxwellian behavior

• The key energy scale that dictates the relevant thermodynamic statistics (Maxwellian or Fermi–Dirac) is the Fermi energy

In the Fermi–Dirac limit ($T \ll T_F$) The average number of fermions in a single-particle state *i*

$$n_i = \frac{1}{\mathbf{e}(\epsilon_i - \mu)/k_{\mathrm{B}}T + 1}$$

 ϵ_i is the energy of this single state, μ is the chemical potentional $\sim E_{\rm F,}$ and $k_{\rm B}$ is the Boltzmann constant

In the classical Boltzmann limit ($T \gg T_F$)

$$n_i = \frac{1}{\mathbf{e}(\epsilon_i - \mu)/k_{\mathrm{B}}T}$$



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The conduction carrier's velocity distribution differs substantially between classical and quantum statistics

In the classical Maxwellian statistics (ideal plasmas)

In the Fermi–Dirac statistics (electrons in metals)







 $v_{\rm F} = (2E_{\rm F}/m_{\rm e})^{1/2}$

All carriers contribute to electronic conduction

Only the carriers close to the Fermi surface traveling at the Fermi velocity contribute to electronic conduction





Degeneracy and Fermi energy demarcate regimes of classical and quantum statistics of Fermi matter



 $\Theta = T / T_F$





*ICF: inertial confinement fusion

In fluids and plasmas, another key energy scale is the strength of ion-ion interaction (interionic coupling)

- This is the ratio of the potential energy to the kinetic energy, which describes the correlation of the fluid/plasma
- $\Gamma_{\rm ii} = e^2/\alpha k_{\rm B}Ts$,

where $\alpha = (3/4\pi n)^{1/3}$ is the ion sphere radius, *e* is the electron charge, and *k*_B is the Boltzmann constant

- Low, dense, high-temperature plasmas are weakly coupled $\Gamma_{ii} \ll$ 1 (gas-like)
- Dense, low-temperature plasmas are strongly coupled $\Gamma_{ii} \gg$ 1 (liquid-like)
- At sufficiently high Γ_{ii} , plasma will crystalize into a solid-like state (Wigner crystallization) $\Gamma_{ii} \sim 17$



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Shocked (deuterium) is a unique system to investigate these different states of dense matter



 The fluid transforms from a strongly coupled, highly degenerate metal to weakly coupled, classical plasma

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Shocked (deuterium) is a unique system to investigate these different states of dense matter

- Above the insulator metal transition, there are no core electrons to screen the electron-ion interaction or introduce bound (ionization) states; accordingly, the interactions are purely Coulombic
 - in its simplest picture: electrons are scattering off ions
- Above the maximum compression $\rho/\rho_0 \sim$ 4.2 to 4.5, the density plateaus as a function of increasing shock velocities
 - all of the pressure increase is thermal pressure





Line-imaging velocimetry and streaked optical pyrometry (SOP) were used to measure the shock velocities, reflectance, and temperature

CD shells filled with liquid D₂ $\rho_0 = 0.172$ g/cm³



- Experimental observables
 - shock velocities, reflectance, and temperature
- The range of velocities studied is 20 to 65 km/s









*VISAR: velocity interferometer system for any reflector

In a highly degenerate regime ($0.07 < T/T_F \sim 0.35$), reflectance plateaus are ~0.4 to 0.43, consistent with the resistivity saturation in Fermi-liquid behavior





Reflectance was normalized to the stationary Au cone



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This reflectance saturation value corresponds to the minimum metallic conductivity criterion derived by Mott–loffe–Regal for disordered or fluid metals

Increasing temperatures \rightarrow diminishing mean free path \rightarrow decreasing τ

Mean free path cannot be less than interatomic spacing

$$au_{\text{MIR}} = \frac{lpha}{V_{\text{f}}} = 1/\hbar (4\pi^2 n^2)^{1/3}$$

Assuming full ionization, and at $\rho \sim 0.741$ (4.5 compression) $R_{\text{MIR}} = 0.38$



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A. F. loffe and A. R. Regal, Prog. Semicond. <u>4</u>, 237 (1960);

In the classical regime, reflectance starts rising continuously up to 0.7 at 65 km/s





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The rise in the reflectance occurs at $T > 0.4 T_F$ and $\Gamma_{ii} \sim 2.5$



DFT: density functional theory QMD: quantum molecular dynamics M. Zaghoo *et al.*, "Breakdown of Fermi Degeneracy in the Simplest Liquid Metal," to be submitted to Physical Review Letters.





The reflectance increase above 0.4 T_F can occur because of either increased density, ionization at the same density, or increased relaxation times

- Increased density
- Increased ionization $n_{\rm e}/n_{\rm i}$
- Increased relaxation time

$$\mathbf{R}(\boldsymbol{\omega}) = \left(\frac{\sqrt{\boldsymbol{\varepsilon} - \mathbf{1}}}{\sqrt{\boldsymbol{\varepsilon} + \mathbf{1}}}\right)^{\mathbf{2}}$$

$$\varepsilon(\omega) = 1 + \frac{i}{\omega \varepsilon_0} \frac{n_e e^2 \tau}{m(1 - i\omega \tau)}$$



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LLE

An increase in density or ionization fraction cannot account for the observed R



*PBE: Perdew–Burke-Ernzerhof functional **QMC: quantm Monte Carlo

[†]L. Caillabet, S. Mazevet, and P. Loubeyre, Phys. Rev. B <u>83</u>, 094101 (2011). N. M. Tubman et al., Phys. Rev. Lett. 115, 045301 (2015).







Experimentally inferred relaxation times above $T \sim \alpha 5$ eV reveal $\tau \sim \alpha T^{1.55}$, which is consistent with the characteristic Landau–Spitzer dependence $\tau \sim T^{1.50}$



The metallic plasma has transformed from a system where relaxation time is described by Fermi degeneracy to one where it is better described by Maxwellian statistics.

M. Zaghoo *et al.*, "Breakdown of Fermi Degeneracy in the Simplest Liquid Metal," to be submitted to Physical Review Letters.







The data provide an invaluable benchmark to dense plasma transport models over an order of magnitude in T



 Both Lee–More and *Purgatorio* models underestimate the conductivity of the deuterium plasma across the whole range, but *Purgatorio* does better in the degenerate regime

> Y. T. Lee and R. M. More, Phys. Fluids 27, 1273 (1984); P. A. Sterne et al., High Energy Density Phys. 3, 278 (2007).





Our experimental and computational data challenge the standard convention in dense plasma literature, which assumes that T/T_F delineates quantum and classical regimes

 Ab initio DFT-QMD results show that the onset of the crossover remains unchanged at increasing densities









DFT calculations by S. X. Hu

Our results extend studies of degeneracy to new fermionic species (electrons) in a dense liquid system



Density (cm ⁻³)	Fermions	Mass of the fermionic species (kg)	7 _F (Κ)	Exp
$\textbf{2.33}\times\textbf{10^{23}}$	Electrons	9.1 × 10 ⁻³¹	157,000	Opt
~10 ¹²	K ⁴⁰ atoms	$6 imes 10^{-26}\ (K^{40})$	~10 ⁻⁶	Den di c

M. Zaghoo *et al.*, "Breakdown of Fermi Degeneracy in the Simplest Liquid Metal," to be submitted to Physical Review Letters;
B. DeMarco and D. S. Jin, Science <u>285</u>, 1703 (1999);
B. DeMarco, S. B. Papp, and D. S. Jin, Phys. Rev. Lett. <u>86</u>, 5409 (2001).

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perimental probe

tical conductivity

nsity, momentum istribution, and compressibility

The breakdown of degeneracy will have key consequences on the thermodynamic and transport properties of the plasma

For
$$\Theta \ll 1$$
, $k \propto T^{-1}$, $\eta \propto T^{-2}$, $C_v \sim \Theta$, and $\mu > 1$

while for $\Theta \gg 1$, $k \propto T^{5/2}$ and $\eta \propto T^{5/2}$, $C_v \sim 3R$, and $\mu < 1$

- C_v is the heat capacity, μ is the chemical potentional
- *k* is the electronic thermal conductivity, η is the shear viscosity





Our conventional interpretation of the adiabat (P/P_F) might require a revision







Summary/Conclusions

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Backup





Our results are consistent path-integral Monte Carlo (PIMC) calculations, which confirm the crossover from the degenerate to the classical limit at 0.3 to 0.4 $T_{\rm F}$





