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We summarize the salient points of the lectures and contributions presented at the Workshop on Statistical Mechanics of Ionic Matter held at the "Centre de Physique des Houches" (France) from March 29th to April 9th, 1982. The summary is organized according to the main topics covered by the workshop and followed by a short list of updated references.

KEY WORDS: Inertial confinement; liquid metals; molten salts; onecomponent plasmas; polyelectrolytes; strongly coupled plasmas; superionic conductors; white dwarfs.

1. INTRODUCTION

The workshop on the Statistical Mechanics of Ionic Matter was devoted to an interdisciplinary confrontation of various aspects of strong Coulomb correlations which play a dominant role in several, seemingly unrelated, fields. The meeting brought together about 60 participants, from 12 countries, with widely different backgrounds, ranging from Statistical Mechanics and Plasma Physics to Condensed Matter Physics, Physical Chemistry, and Astrophysics. Essentially oriented towards theoretical aspects, the workshop did, however, benefit from the presence of a few experimentalists. The two-week activity (from March 29th to April 9th, 1982) included a dozen short cycles of two to three lectures, a dozen invited one-hour seminars,

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shorter contributed seminars, a poster session, and four round table discussions. Details about these various contributions can be found in Section 2, while an outline of the most salient results is presented in Section 3. Some perspectives for future research are given in the concluding section. We have added a list of references, which, although far from exhaustive, is meant to orient the reader towards existing reviews and the more recent literature devoted to this interdisciplinary subject.

2. CONTRIBUTIONS

2.1. Lectures

- 1. B. J. Alder, D. Ceperley, and E. L. Pollock (Livermore)
 - a. Stochastic quantum methods;
 - b. Green's function Monte Carlo: Applications to the one component plasma and hydrogen;
 - c. Computational methods for finite temperature.
- 2. J. L. Bobin (Paris VI): Physics of inertially confined plasmas.
- 3. H. E. De Witt (Livermore): Equation of state of dense plasmas and ionic mixtures.
- 4. W. Dieterich (Konstanz): Superionic conductors.
- 5. H. Gould (Clark): Introduction to kinetic theory of charged particle systems.
- 6. B. Jancovici (Orsay): Two-dimensional Coulomb fluids.
- 7. G. Kalman (Boston): Nonlinear response functions and collective modes in strongly coupled plasmas.
- 8. J. L. Lebowitz (Rutgers): Exact results and approximations in Coulomb systems.
- 9. I. R. McDonald (Cambridge): Computer simulations of the dynamics of ionic crystals and fluids.
- 10. R. More (Livermore): Atomic physics in dense plasmas.
- 11. M. P. Tosi (Trieste): Static and dynamic properties of ionic liquids.
- 12. H. M. Van Horn (Rochester): Dense astrophysical plasmas.

2.2. Seminars

- 1. N. W. Ashcroft (Cornell): Quantum plasmas: The physics of dense metallic hydrogen and deuterium.
- 2. C. Deutsch (Orsay): Heavy ion fusion.
- 3. J. W. Dufty (Gainesville): Kinetic theory of line broadening in dense plasmas.

- 4. R. Evans (Bristol): The surface properties of ionic liquids.
- 5. M. J. Gillan (Harwell): Numerical simulations of superionic conductors.
- 6. R. Klein (Konstanz): Dynamics of charged spherical brownian particles.
- 7. W. D. Kraeft (Greifswald): Statistical mechanics of partially ionized plasmas.
- 8. Ph. A. Martin (Lausanne): Some exact results on charge fluctuations and screening.
- 9. D. J. Stevenson (Cal. Tech.): Planetary interiors.
- 10. P. Turq (Paris VI): Coulomb interactions in polyelectrolyte solutions.
- 11. F. I. B. Williams (Saclay): Crystallization of the two-dimensional electron gas.

2.3. Contributed Seminars and Posters

- 1. A. Alastuey (Orsay): Density functional theory applied to surface problems.
- 2. B. Bernu (Paris): Thermoconduction in two-component plasmas and ionic liquids.
- 3. D. Boercker (Livermore): Electrical conductivity in plasmas.
- 4. R. Cauble (Springfield): Plasma polarization shifts of spectral lines.
- 5. D. K. Chaturvedi (Konstanz): Transport properties of a Coulomb lattice gas.
- 6. T. Gaskell (Sheffield): Ionic dynamics in the strongly coupled onecomponent plasma.
- 7. L. Lantto (Oulu): Metallic hydrogen as a two-component fluid.
- 8. M. Lavaud (Orléans): Recherche de resommations optimales de graphes à partir de majorants.
- 9. D. Mac Gowan (Glasgow): Comment on the MSA for the one-component plasma.
- 10. D. Merlini (Bochum): Boundary conditions and thermodynamics of the 2d one-component plasma.
- 11. G. Pizzimenti (Messina): Generalized MSA for charged hard sphere systems.
- 12. Y. Protasov (Moscow): Transport properties of nonideal plasmas (experimental results).
- 13. D. Quitmann (Berlin): Dynamics of concentration fluctuations in an almost ionic liquid alloy.
- 14. S. Rogde (Bergen): The validity of the HNC and a PY-like approximation as a function of the range of the interparticle potential in ionic systems.

- 15. F. Rogers (Livermore): Plasma pseudopotentials and HNC calculations of the equation of state of multicomponent plasmas.
- 16. G. Senatore (Trieste): Optimized RPA for the structure of liquid alkali metals as electron-ion plasmas.
- 17. J. Wallenborn (Brussels): Kinetic theory of the transport properties of a strongly coupled one-component plasma.

3. SUMMARY

In view of the broad spectrum of topics, it is clearly a difficult task to summarize the large amount of information which was presented at the workshop in a well-balanced fashion. Important omissions are unavoidable and our sole responsibility. To proceed, we have subdivided the program, somewhat arbitrarily, into the following twelve topics.

3.1. Rigorous results^(1,2)

The conditions of stability and of existence of the thermodynamic limit for Coulomb systems are by now well understood. A crucial role in the proof of these properties is played by Newton's theorem (implying screening) and by the Fermi–Dirac statistics.⁽³⁾

Among the more recent rigorous results with potential applications the following were discussed at the workshop:

(a) Multipolar sum rules, generalizing the conditions of charge neutrality and of perfect screening, as well as their consequences for the charge fluctuations and the screening in the vicinity of surfaces.⁽⁴⁾

(b) Upper bounds and optimal diagram resummations for the virial expansion. $^{(5)}$

(c) The equilibrium properties (thermodynamics, correlation functions) of the two-dimensional one-component plasma (OCP) for a particular value of the coupling $(\Gamma = e^2/k_BT = 2)$.⁽⁶⁾

3.2. Quantum Plasmas^(7,8)

"Exact" results for small periodic systems ($N = 10^2 - 10^3$ particles) can be obtained by computer simulations. For the ground state (T = 0) of a *N*-boson system, the Schrödinger equation can be solved "exactly" by a stochastic Monte Carlo method (diffusion and branching process in configuration space). The method can be extended so as to treat fermions⁽⁸⁾ (by introducing nodal surfaces) and finite temperatures (by solving the Bloch

equation for the density matrix). A number of variations of the stochastic Monte Carlo method have been successfully explored: the Green's function method for the ground state⁽⁷⁾ and a convolution method starting from the classical limit.⁽⁹⁾ Concrete applications include the computation of the ground-state energy of small molecules, the precise location of the Wigner transition of the electron gas (jellium),⁽¹⁰⁾ and a few preliminary results for the ground state of the hydrogen plasma (electrons + protons).

These "exact" results constitute important tests for the predictions of approximate theories such as the recent results obtained for the pair distribution functions of the electron gas and of the hydrogen plasma (T=0) by the variational method on the basis of the FHNC integral equation.⁽¹¹⁾

Such "exact" results will also be very valuable for the study of the transition between the metallic and molecular phases of compressed hydrogen (P > 1 Mbar); at low temperatures the metallic phase could present many different physical phenomena (pairing, superconductivity, superfluidity in the case of deuterium).⁽¹²⁾

3.3. Simulations of Dense Classical Plasmas^(2,13)

If the development of Monte Carlo simulation methods for degenerate systems is a fairly recent field of research, the simulation of classical systems is by now a well-developed subject. The two domains of applications of these methods are as follows:

(a) The computation of static properties (thermodynamics, equilibrium distribution functions) using the Monte Carlo method of Metropolis *et al.* Very accurate results have recently been obtained both for the solid and fluid phases of the OCP (one-component plasma); they have located the fluid-solid transitions at a value of the coupling parameter $\Gamma = e^2/ak_BT \simeq 170 \ [a = (\frac{3}{4}\pi n)^{1/3}, n = N/V].^{(14)}$ These simulations are being extended to the case of binary ionic mixtures (BIM) embedded in a neutralizing background.

(b) The method of "molecular dynamics" (MD), which amounts to a numerical integration of the coupled equations of motion of N particles and which enables one to obtain information on the dynamic properties of dense plasmas: correlation functions, fluctuation spectra, transport coefficients. It has been successfully applied to the OCP model (dispersion and damping of the plasma mode, the transport properties), to the semiclassical hydrogen plasma (TCP model); the latter results have disclosed the very different behavior of the autocorrelation function of the individual velocities (rapid decay) and of the electric current (slow decay).⁽¹⁵⁾ The thermo-

electric transport coefficients of the hydrogen plasma for $\Gamma \simeq 1$ have also been computed using this method.⁽¹⁶⁾

3.4. Integral Equations^(2,17)

There has been a renewed interest in the classical integral equations for calculating the pair distribution functions. For Coulomb systems, the hypernetted chain (HNC) and the mean spherical approximations (MSA) are the most adequate equations since they automatically satisfy the conditions of charge neutrality and of perfect screening. The more recent investigations have attempted to improve upon these equations and to render them thermodynamically consistent.

(a) The modified HNC schemes attempt to include by a semiempirical⁽¹⁸⁾ or diagrammatic⁽¹⁹⁾ procedure the so-called bridge diagrams. These models yield excellent results for the case of the three-^(18,19) and⁽²⁰⁾ two-dimensional OCP.

(b) The generalizations of the MSA equation (GMSA) impose thermodynamic consistency of the MSA by modifying the closure relation. This scheme yields good results for the OCP when the effective hard-sphere diameter is suitably adapted.⁽²¹⁾ An extension of this method allows for a precise determination of the structure factor of liquid metals.⁽²⁰⁾ For the OCP, the effective hard-sphere diameter of the MSA model can be further optimized.⁽²³⁾ The GMSA has also been applied successfully to the case of oppositely charged hard spheres of different diameters.⁽²⁴⁾ An alternative approach consists in a modification of the Percus–Yevick equation (PY) suitable for the case of long-range potentials.⁽²⁵⁾

3.5. Metallic and Ionic Liquids and Crystals, Superionic Conductors⁽²⁶⁻²⁸⁾

These topics fall broadly into the fields of Chemical Physics and of Condensed Matter Physics, but they have many connections with the physics of dense plasmas.

(a) The alkali metals have a simple electronic structure which allows them to be assimilated, in first approximation, to the OCP which reproduces the experimental structure factor very well without adjustable parameters; the correct small-k behavior can be obtained within a generalized RPA method.⁽²²⁾ The observed universality of the melting and freezing criteria can be interpreted within a liquid-phase based theory of freezing with one or more order parameters.⁽²⁹⁾

(b) This theory of freezing can be extended to simple molten salts (alkali halides) on the basis of their charge structure factor. Computer

simulations have shown that the corresponding dynamic charge structure factor exhibits a well-defined "optic" peak;⁽³⁰⁾ the damping of this optic mode presents interesting theoretical features, but unfortunately clear experimental evidence for it is still lacking.⁽³¹⁾ Experimental neutron scattering results for the concentration fluctuations in the almost ionic Li–Pb alloy have recently become available.⁽³²⁾

(c) Metal-salt solutions (alkali-alkali halides) present an abrupt transition from an ionic to a metallic structure for high metal concentrations $(X \simeq 0.9)$ with a corresponding rapid variation of the electrical conductivity (Mott transition).⁽³³⁾

(d) Molecular ionic crystals (KCN, NaCN, NaNO₂) show interesting modifications of their orientational order with temperature. Computer simulations have shown the importance of the quadrupole moment in the interpretation of the experimental results.⁽³⁴⁾

(e) Superionic conductors constitute a phase intermediate between an ionic crystal and a molten salt. Their high electrical conductivity ($\sigma \simeq 1(\Omega \text{ cm})^{-1}$) reflects the great mobility of one of the ionic species. The transition to the superionic state is discontinuous for AgI but continuous for the chlorides and the fluorides.⁽²⁸⁾ Two types of theoretical models have been used to interpret the transport properties of these systems: lattice gas models with independent or correlated particles,⁽³⁵⁾ and fluid models using a description in terms of collective variables. Molecular dynamics simulations (MD) of CaF₂ have shown that the one-body density of the F⁻ ions remains essentially concentrated around the lattice sites and that anion diffusion is essentially due to a hopping mechanism.⁽³⁶⁾

3.6. Macromolecular lonic Systems⁽³⁷⁾

The Coulomb correlations play an important role in the determination of the structure and transport of solutions of polyelectrolytes. The basic phenomena of condensation of counterions can be understood on the basis of simple models while the correlations between the polyions and the counterions can be described by the Poisson–Boltzmann equation.⁽³⁸⁾

Light scattering experiments have revealed that solutions of charged spherical brownian particles exhibit a liquidlike structure. The dynamical structure factor and the diffusion coefficient of these interacting brownian charges have been computed from the mode-coupling theory and good agreement with the experiments has been obtained.⁽³⁹⁾

3.7. Interfacial and Surface Properties of Ionic Systems⁽⁴⁰⁾

The density profiles at the liquid-gas interface of molten salts and liquid metals can be computed within the density functional theory. The

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results obtained for the surface tension within the square gradient approximation of Van der Waals are in good agreement with the experiments.⁽⁴¹⁾ A similar method has been applied to the two-dimensional $OCP^{(42)}$ for which the density profile is exactly known at the particular value of the coupling $\Gamma = 2$.⁽⁴³⁾

3.8. Two-Dimensional Plasmas^(2,44)

Two-dimensional systems of charges can interact through the twodimensional $(\ln r)$ or the three-dimensional (1/r) Coulomb potential.

(a) The static properties of the two-dimensional OCP (ln r) have been evaluated by means of several integral equations (HNC, STLS, TI)⁽⁴⁵⁾ and by computer simulations which have localized the freezing transition at $\Gamma = 140$. Several exact results have been obtained for $\Gamma = 2.^{(6,43)}$ The dynamical properties are very similar to those of the three-dimensional OCP.^(46,47)

(b) A two-dimensional system of charges embedded in a neutralizing background and interacting through a 1/r potential is a good model for the (classical) electron layers bound to a liquid helium surface and for the (quantal) electron layers in MOS devices.⁽⁴⁴⁾ The dynamic properties of this model have been studied by computer simulations⁽⁴⁸⁾ and analyzed theoretically.⁽⁴⁹⁾ The oscillations observed in the velocity autocorrelation function have been interpreted in terms of a coupling of the self-motions to the plasma mode.⁽⁵⁰⁾ The theoretical predictions for the freezing transition $(\Gamma \simeq 125)$ are in good agreement with the laboratory experiments which emphasize moreover the coupling of the electrons to the ripplons of the He surface.^(51,44) The nature (order) of the transition has not yet been firmly established by the laboratory experiments, but these seem to be compatible with the dislocation unbinding mechanism.⁽⁵²⁾ The dependence of the static properties of the two-dimensional electron gas on the number of particles (N) and on the boundary conditions have been examined in some detail.⁽⁵³⁾

3.9. Plasma Kinetic Theory^(2,54)

The so-called "renormalized" kinetic theories, initially developed for studying the spectra of fluctuations around thermodynamic equilibrium in simple liquids, have been successfully applied to the OCP,⁽⁵⁵⁾ and more recently also to the TCP,⁽⁵⁶⁾ for intermediate and strong couplings. Very satisfactory results have been obtained for the shear viscosity of the OCP but difficulties still subsist for the thermal conductivity and the diffusion coefficient when $\Gamma \gg 1$.⁽⁵⁷⁾ A detailed analysis of the velocity autocorre-

lation function of the OCP has drawn attention to the importance of the coupling of the self-motions to the transverse collective modes.⁽⁵⁸⁾ A quantum-mechanical kinetic theory for the electrical conductivity of the TCP yields a formula interpolating between the Ziman formula for degenerate plasmas and a renormalized Spitzer formula for quasiclassical plasmas.⁽⁵⁹⁾ Experimental measurements of the (nonlinear) electrical conductivity of dense cold plasmas are available.⁽⁶⁰⁾

The quadratic nonlinear response function formalism yields another approach to the study of the collective modes of dense plasmas. This method has been used to study the dispersion of the plasmon mode of the OCP.⁽⁶¹⁾ Sum rules have been established for the longitudinal and transverse fluctuations of an OCP in a strong magnetic field.⁽⁶²⁾

3.10. Astrophysical Applications⁽⁶³⁾

Many astrophysical situations involve dense plasmas, for example the interiors of giant planets, white dwarfs, and neutron star crusts. A precise estimate of the structure of the luminosity and of the evolution paths of dense astrophysical objects requires a good knowledge of the equation of state, the phase diagram, and the transport coefficients of dense plasmas and in particular of the OCP and BIM model. Among the objects which have received much attention recently one can quote the pulsating white dwarfs⁽⁶⁴⁾ and the X-ray burst sources (accretion at the surface of neutron stars).

The relatively low temperatures ($T \simeq 10^4$ K) encountered in the interior of the giant planets are responsible for the strong Coulomb coupling phenomena encountered there. The properties of these planetary interiors are strongly influenced by the transition between the molecular and metallic phases of hydrogen and by the solubility of helium.⁽⁶⁵⁾

3.11. Inertial Confinement Fusion⁽⁶⁶⁾

The physics of dense plasmas plays an essential role in inertial confinement thermonuclear fusion experiments where highly compressed pellets are brought to high temperatures by means of laser, electron, or ion beams so as to reach physical conditions comparable to those of stellar interiors. The energy gain of this implosion process has been shown to obey simple scaling laws. The center of the compressed pellet has to remain thermally isolated while the corona is heated by the beam. An essential but unsolved problem concerns the heat flux limitation. The origin of this inhibition of the thermal transport could be due to various physical processes (turbulence, intense magnetic fields) or else could be revealed by adapting the kinetic theory description to the nonlinear, nonequilibrium situations prevailing in this finite medium.⁽⁶⁷⁾

The stopping power of heavy ions by a partially degenerate electron plasma plays a crucial role in heavy ion fusion devices. A calculation of this stopping power within the RPA, with account of the contribution from bound electrons, is now available.⁽⁶⁸⁾

3.12. Plasma Atomic Physics⁽⁶⁹⁾

The dense plasma obtained in a laser implosion experiment is a very complex medium mainly because of the presence of heavy, partially ionized, atoms and because of the important deviations from thermodynamic equilibrium. The structure and interaction of heavy, partially ionized atoms can be rather well studied within Thomas–Fermi theory and its extensions.⁽⁷⁰⁾ The ionization equilibria can be described by a generalized Bohr model with screened charges.⁽⁷¹⁾ The nonequilibrium effects require a formulation of thermodynamics with different electron and ion temperatures ($T_e \neq T_i$). The equation of state for such a plasma can then be obtained within a generalized Debye–Hückel theory.⁽⁷¹⁾

Most of the information on dense plasmas is based on spectroscopic diagnostics. The broadening by the plasma of the linewidths of the atomic spectra can be studied in detail within a kinetic theory.⁽⁷²⁾ The observed shifts of the atomic lines due to polarization effects are well reproduced by simple models.⁽⁷³⁾

The equation of state and the degree of ionization of dense plasmas can be obtained by diagrammatic methods within a Green's function formulation of quantum statistical mechanics.⁽⁷⁴⁾ An alternative approach is based on the use of pseudopotentials taking into account the bound states via the Planck-Larkin formula.⁽⁷⁵⁾

4. CONCLUSIONS

During the discussions and round tables a certain number of conclusions and perspectives for future investigations have been brought up. They can be tentatively summarized as follows.

With respect to the rigorous results, the existence of the thermodynamic limit of two-dimensional 1/r systems remains to be shown. One would also like to have rigorous results concerning the large r asymptotic behavior of the direct correlation function c(r) both for short-ranged and for long-ranged Coulomb potentials.

Concerning the computer simulations of quantal plasmas most efforts will be directed to extensions of the present Monte Carlo methods towards

finite temperatures and many-fermion systems, in particular for partially degenerate plasmas (TCP). In the long run one would also like to see quantum dynamics be incorporated into these computer simulation methods.

At present, the simulation of dense classical plasmas is mainly concerned with the extension towards nonequilibrium situations, in particular the relaxation of a two-temperature hydrogen plasma. The simulations of the collective modes of dense plasmas (OCP, TCP) in the presence of strong magnetic fields would also yield valuable "experimental" information against which the recent theoretical developments could be tested.

The structure and dynamics of simple molten salts (alkali halides) are by now fairly well understood, but clear experimental evidence (neutron scattering) concerning the optic mode of ionic liquids is still missing. The results obtained for solid KCN and NaNO₂ indicate their richness in physical behavior and the usefulness of the extensions of the simulation technique to molecular ionic crystals and liquids. A theoretical analysis of the conductor–insulator transition of metal-salt solutions would also be very instructive. In the field of superionic conductors one would like to have quantitative theoretical results in order to interpret the simulation and neutron scattering results.

The macromolecular ionic liquids (polyelectrolytes, colloids, etc.) constitute another very rich class of systems for which the experimental and theoretical studies undergo at present rapid developments. The extension of the integral equations for the structural studies of nonspherical objects and the use of kinetic theories to obtain corrections to the limiting laws are certainly promising directions for future investigations.

The present interest in the surface properties of ionic liquids should result in rapid progress in this field in the near future. The theoretical study of the long-range correlations along interfaces of ionic liquids is presently under way.⁽⁴⁰⁾

The study of two-dimensional plasmas has experienced enormous progress in recent years. Nevertheless the study of the transport properties and of the critical behavior near the collapse temperature of two-dimensional two-component plasmas remains still to be done.⁽⁷⁶⁾

The present renewed interest in kinetic theory in relation with the computer simulation results is very promising. A detailed study of the transition from the weak-coupling Landau-Spitzer regime ($\Gamma \ll 1$) to the intermediate-coupling regime ($\Gamma \simeq 1$) would be very valuable. The computations of the transport coefficients of the two-component plasma in this region ($\Gamma \simeq 0.1$) are inaccessible by computer simulations but of great interest to fusion plasmas. The study of the dynamics of dense plasmas in

magnetic fields⁽⁷⁷⁾ should also be tractable within the present-day kinetic theories.

The properties of dense plasmas play an important role in a large variety of astrophysical problems. Among the problems raised during the meeting one should mention the relation between the coefficients of selfdiffusion and of mutual diffusion of strongly charged species and their evaluation for a large variety of physical situations. Another open problem concerns the demixing properties of different ionic species in the solid phase and the solubility of partially ionized atoms in metallic hydrogen.

Inertial confinement fusion raises a broad spectrum of complex problems, many of which do not belong to the realm of statistical mechanics. Among the more basic problems one should mention the correct interpretation of the observed heat flux limitation. A rigorous formulation of the statistical mechanics of dense partially ionized plasmas should become available in the near future. The preliminary results concerning twotemperature plasmas should be extended to the case of intermediate couplings. An active area of research is concerned with the calculation of pseudopotentials for dense partially degenerate plasmas in the presence of bound states. The influence of the plasma environment should result in a density dependence of these pseudopotentials and in the appearance of three-body effects. Calculations in this direction should be very valuable in relation to the current fusion experiments.

All participants agreed that many connections between apparently remote disciplines have been revealed very convincingly during the workshop. The features common to many problems arising in a wide variety of fields, which result from the presence of strong Coulomb correlations, will certainly continue to be the object of intense research in the near future.

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