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An Informal Newsletter associated with Collaborative Computational Project No. 5 on Molecular Dynamics and Monte Carlo Simulations of Macroscopic systems.

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Contents

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Editorial

Welcome readers, to the new format CCP5 Newsletter. As the title suggests, it is our intention to publish the Newsletter every three months and thus bring matters of interest swiftly to the attention of participants in CCP5. Such newsletters are a feature of all the other collaborative computational projects connected with Daresbury and they are clearly popular with their subscribers. Hopefully, the CCP5 Newsletter will follow in this tradition.

A newsletter such as this however is only of value if the participants in the project are willing to contribute towards its production. To this end, we request of our readers that from time to time, they submit written articles to the editor on any subject they would like to bring to the attention of CCP5 participants. Such articles may be of any length and handwritten (provided they are legible). We wish to be both informative and informal in the newsletter and hopefully encourage a lively debate.

It may also be necessary to commission articles from our readers on occasions and we hope that our good natured colleagues will understand this and contribute in the knowledge of assisting a worthy project! All this in the interest of CCP5.

Since this edition is the first of its kind for CCP5 it is largely devoted to an account of the CCP5 Polarisability Meeting that took palce at Birkbeck College from 23rd to 24th March 1981, under the guiding hands of John Finney and his associates. Those who were present at the meeting will recall a stimulating and well-attended meeting and our thanks go to Dr. Finney and his associates for their efforts and their hospitality. The editor would also like to thank all the speakers at the meeting who contributed the abstracts of their lectures to this newsletter and also Dr. Julia Goodfellow for providing a review of the meeting.

W. Smith (Editor)

General News

1) Dr. Stephen Thompson has resigned his post as Secretary of CCP5 to take up a permanent position at Cornell University. We wish him well. We welcome in his place Dr. David Heyes who started work at Royal Holloway College on 1st June 1981. Dr. Heyes will take over the vacant post of Secretary. Dr. Smith of Daresbury will be responsible for program distribution and documentation.

2) Copies of the molecular dynamics programs MDATOM, HMDIAT, MDLIN, MDLINQ, MDTETRA and MDPOLY (all of which were written by Stephen Thompson for the CRAY-1 computer) are available from W. Smith at Daresbury. Applicants are requested to supply a magnetic tape if possible as this greatly increases the efficiency of the transaction. Applicants with specific difficulties however will be dealt with sympathetically.

3) The next CCP5 Meeting will take place at U.M.I.S.T., Manchester, on 10-11th September 1981. The subject of the meeting will be "Non Standard Molecular Dynamics". Readers interested in attending should contact:

Dr. J.M.R. Clarke
Department of Chemistry
U.M.I.S.T.
P.O. Box 88
Sackville Street
Manchester MD60 1QD

4) Further CCP5 meetings proposed for the future include:

SURFACES: Southampton. December 1981.
(Including topics such as nucleation, interfacial properties, phase changes and clustering.)
Organisers: Dr. N. Parsonage (Southampton), Dr. D. Tildesley (Oxford) and Dr. J.H.R. Clarke (U.M.I.S.T.)

TRANSPORT PROPERTIES: Royal Holloway, March 1981.

Organisers: Dr. P. Madden (Cambridge) and
Dr. M. Gillan (Harwell)

Review of CCP5 Polarisability Meeting

J.M. Goodfellow

A meeting on the use of polarisability for describing systems involving cooperative interactions was held recently at Birkbeck College, University of London. This conference was the second in a series organised by the SRC's Collaborative Computing Project Five (CCP5), which is concerned with computational aspects of molecular dynamics and Monte Carlo simulation techniques. The aim of the meeting was to review the systems for which it has been found necessary to include cooperative effects and to compare the various methods used to assess the non-pair-additive contribution.

The use of atomic polarisability to account for non-pair-additive effects was wide ranging from such dense crystals as Uranium oxide to that of water in biological systems. The need for the inclusion of quite complicated polarisation effects was shown by both D. Frankel (Amsterdam) and P. Madden (Cambridge) in their interpretation of collision induced effects in light scattering spectra.

The relative importance of non-pair-additive terms was shown by J. Murrell (Sussex) who found that the three-body energy terms in quantum mechanical calculations of alkali halides were significant compared with the two-body terms. J. Quinn (Birkbeck) presented data from liquid water simulations showing that although the four-body energies were small, the three-body terms contributed to approximately 10% of the total energy. Although this latter fraction of 10% would appear to be significant and difficult to ignore, the four-body terms were an order of magnitude smaller and hence not so essential to an accurate simulation.

In calculating the energy of formation and migration of defects in solids, C.R.A. Catlow (University College, London) needed to account for multibody interactions as the rigid ion models failed to reproduce satisfactorily experimental energies. Both the use of the shell model and a modification of the rigid ion model to reproduce the known dielectric constant lead to a much closer agreement of theory with experiment. M. Dixon (Oxford) also emphasised the use of the shell model in simulations of molten salts. Although the physical validity of this well-known model was criticised, it does lead to good agreement with known energies whereas the point polarisable ion model leads to a polarisation catastrophe in these dense ionic systems.

Specific algorithms for the inclusion of polarisability were described by H. Berensden (Gronigen) and P. Barnes (Birkbeck College, London). Both speakers have developed models for liquid water in which the need to take into account the cooperative nature of the hydrogen bond is important. Berensden uses a system of point charges which change in value due to the potential acting upon them from all other charges in the liquid. Barnes uses a self-consistent interactive procedure in which the dipole moments of each water molecule is updated due to the electropole field of all surrounding molecules.

The meeting ended with several topics of more general interest for molecular dynamics and Monte Carlo simulations. D. Beveridge (Hunter College, N.Y.) showed data from extensive simulations on liquid water in which convergence of some thermodynamic quantities - especially the specific heat - did not occur until well beyond the usually acceptable number of MC configurations. The rate of convergence was considerably increased by the use of force bias and preferential sampling techniques although at the expense of extra computation. M. Mezei (H. College, N.Y.) and P. Claveire (Paris) showed the importance of establishing that the multipole expansion (often used to describe the electronic configuration of a molecule) does actually converge for the system being studied.

Recent computational improvements have been made through the use of vector processors (like the CRAY at DL) to decrease the time of simulations which involve cooperative effects so that they become comparable with the simpler effective pairwise potentials. Increases in computational speed by factors of three to ten over the CDC 7600 have been found by both Berensden and Barnes by vectorisation of their algorithms. The versatility of computer graphics for displaying complex three-dimensional surfaces was seen in a demonstration by Quinn (Birkbeck College).

This meeting underlines the usefulness of the SRC's collaborative computing projects in that a group of scientists all using very computer intensive simulation techniques were able to compare and discuss the various methods available for the best method of simulating 'reality' in the most efficient manner.

The Three Partial Pair Correlation Functions of Water Determined from Neutron Diffraction

J.C. Dore and J. Reed

Neutron diffraction measurements have been made on D_2O/H_2O mixtures. Despite the large incoherent scattering from the hydrogen atoms, it is possible to measure the small contribution due to coherent scattering. The observed cross-section data may be fitted with a single molecule form-factor and the inter-molecular contributions extracted for each of the measurements. Fourier transformation of the data gives four real-space distribution functions which define the three partial correlation functions $g_{OO}(r)$, $g_{OH}(r)$ and $g_{HH}(r)$. Since these three functions are effectively over-determined by the four measurements, various internal consistency checks can be applied. The preliminary results are surprising and suggest that correlations in the OH and HH extend much further than predicted by any of the current computer simulations. Oscillations in the region 5-8 Å are in good agreement with the earlier separation of Palinkas et al[1] using x-ray, neutron and electron diffraction data.

1. G. Palinkas, E. Kalman and P. Kovaks, Molec. Phys. **34**, 525 (1977).

Polarisability in the Simulation of Solids

C.R.A. Catlow

Methods for the incorporation of polarisability effects in solids in the investigation of the defect formation and migration energies which determine transport properties (e.g. in the UO_2 system) were discussed.

The basic method used was a static lattice model in which electrostatic energy terms were evaluated by Ewald's method and short range interactions were dealt with by using two-body analytic functions. Crystal defects were modelled using a two-region method (c.f. HADES), where Region I consisted of an explicit crystal region surrounding the defect and Region II a more remote region where continuum methods of approximation were used.

The polarisation models available were:

i) Neglect of polarisation altogether (Rigid ion model)
ii) Point polarisable ion model (PPI)
iii) Core-shell model
iv) Breathing shell model

The breathing shell model involves essentially three-body effects and while often necessary for studying dynamical properties, are not essential for studying defect properties. The results of these simulations provide the following salient points.

The core-shell model gave best agreement with experiment. This probably results from the coupling between the short range forces and ionic dipole forces inherent in the model. The PPI model was unable to converge on a solution in all cases. The rigid ion model does not produce good results in the standard implementation but significant improvement occurs if the model is first parameterised to give the correct dielectric constant.

Non Pair-additive Terms for Alkalai Halides

J.N. Murrell

SCFMO calculations on lithium fluoride clusters both within a crystal environment and as isolated species confirm an earlier conclusion of Bounds and Hinchliffe[1] that 3-body energies are as important as the non-Coulombic 2-body energies but 4-body energies are unimportant. It can be shown by perturbation theory that this result is expected for induction (polarization) and charge-transfer energies and calculations with varying basis sets confirm the importance of both[2]. The fact that charge-transfer energy is important has implications for any model which introduces non pair additive energies only through polarization terms.

1. D.G. Bounds and A. Hinchliffe, Molec. Phys. 40, 989 (1980).
2. J.N. Murrell, J. Tennyson and M.A. Kamel, Molec. Phys. in press.

Non-additivity in Water-ion-water Interactions[1]

S. Romano, E. Clementi, H. Kistenmacher and W. Kolos

The three-body system $Li^+(H_2O)_2$ was analyzed to study that non-additive part of the interaction potential which can be obtained by the Hartree-Fock approximation.

For long and intermediate distances the three-body correction was found to be well represented by the induction energy where bond dipoles are induced on each water molecule by point charges located on the (unpolarizable) lithium ion and on the other molecule respectively: for shorter distances this approximation was corrected by means of an exponential repulsive term. Such a potential model for non-additive interactions was extended to the more general situation $Li^+(H_2O)_n$, and Monte-Carlo calculations were carried out on clusters containing up to six water molecules: comparison with other simulation results and with available data showed a significantly improved agreement with experiment. Tentative values for ΔH are presented for n = 7, 8, ..., 20, where experimental data are not available.

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[1] Theoret. Chim. Acta (Berl.) 55, 257-266 (1980).

Light Scattering and Birefringence Studies of Molecular Liquids

P.A. Madden

Methods for obtaining the orientational correlation parameter (g_2) of a liquid from depolarised light scattering intensities and from electrically and magnetically induced birefringence data were assessed in terms of recent molecular theories of 'local field' effects.

Experimental values of g_2 for several molecular liquids can be correlated with the molecular structure and the thermodynamic state of the fluid. The relationship between g_2 and the collective and single molecule reorientation times for these fluids was discussed.

Information from computer simulation experiments on the validity of the assumptions used in the molecular 'local field' theories was presented.

MD Calculations on Collision-Induced Effects in Molecular Fluids

D. Frenkel

Collision induced effects in molecular fluids have traditionally been regarded as a nuisance because they complicate the interpretation of light scattering experiments. On basis of extensive molecular dynamics calculations on (atom-atom L.J.) N_2, O_2, Cl_2 and CO_2, it is argued that the ad hoc procedures that have been used to disentangle collision-induced and orientational light scattering in molecular fluids may not be based on realistic assumptions. In particular, it seems that for small molecules like the ones studied, the total light-scattering spectrum cannot be decomposed unambiguously into an orientational and a collision-induced part. Nor does there seem to be a clear cut separation in time-scales between collision-induced and orientational fluctuations.

The MD simulations suggest a number of experiments that could be performed in order to test whether our explanation of collision-induced effects in terms of the molecular polarizability tensor is correct. Most important among these experiments are:

1) a measurement of the frequency dependence of deviations from the Lorentz-Lorenz relation in the refractive index of molecular fluids;
2) measurement of the isotropic collision-induced light-scattering of molecular fluids;
3) measurement of the 2nd spectral moment of Raman bands.

Such experiments could tell us whether polarizability fluctuations in molecular fluids can be adequately explained in terms of the DID (dipole-induced dipole) mechanism.

Polarisability in Ionic Liquids

J.H.R. Clarke and S.M. Hayne

We are concerned with the importance of non-pairwise-additive interactions in determining properties of pure ionic liquids. Bounds and Hinchliffe[1] have shown (using ab initio calculations) that although such effects contribute only a small fraction of the total energy of small ionic clusters they can have a very large effect on the electronic polarizability. We have used molecular dynamics computer simulation of liquid KCl at 1300 K and near zero pressure to calculate the depolarisation ratio (ρ) of light scattering (5-500 cm^{-1}) which is determined by the relative magnitudes of fluctuations in the trace and anisotropic parts of the interaction induced polarisability. Using only the pair-additive interaction terms given by Bounds and Hinchliffe the predicted value of ρ is an order of magnitude smaller than as experimentally observed[2] to an extent that is outside the error limits of the computations. Arguments were presented that this may be a result of strong cancellation effects on the additive two-body trace polarisability by three-body terms. The uncertainties in computing properties dependent on 3- and 4-body correlations using computer simulation of small periodic samples was emphasised[3].

1. Bounds and Hinchliffe, Molec. Phys. 40, 989 (1980); J. Chem. Phys. 72, 298 (1980).
2. Clarke and Woodcock, J. Chem. Phys. 57, 1006 (1972).
3. Clarke and Woodcock, Chem. Phys. Lett. 78, 121 (1981).

Molten Salt Simulations with Polarizable Ions

M. Dixon

Ionic materials are polarized by electric fields through two mechanisms. Firstly the entire ions move relative to one another. The second mechanism arises from the distortion of the electron density of the ions by their local fields[1]. The susceptibility to polarization is related to the frequenccy dependent dielectric constant $\varepsilon(\omega)$ which depends on the transverse optic frequency ω_T:

$$\varepsilon(\omega) = \varepsilon(\infty) + \frac{(\varepsilon(o) - \varepsilon(\infty))}{(\omega^2_T - \omega^2)} \omega^2_T$$

The longitudinal optic frequency,

$$\omega_L = (\varepsilon(O)/\varepsilon(\infty))^{1/2}\, \omega_T \,. \quad \text{(Ref. 2)}$$

If a model fails to give a satisfactory description of the dielectric properties of a material it must fail also to describe the collective charge oscillations which correspond to the optic phonons in the crystal. The dispersion curves for NaI illustrate clearly the failure of rigid ion models; $\varepsilon(\infty)$ is always unity for such a model[3]. Point polarizable ion models, for which the polarization of an ion is proportional to the electric field at the ion, are unsatisfactory. Point polarizable ion models overestimate the polarization of the ions and are liable to polarization catastrophes[4,5,6]. The class of shell models all couple the polarization of an ion to the short range repulsion between the electron densities of that ion and its neighbours. This class of models has been extremely successful when used for predicting the phonon dispersion curves of crystals[3]. It has also been successful for predicting rumpling of crystal surfaces and defect energies[6,7] for which rigid ion and point polarizable ion models have proved very inadequate.

A method is presented for implementing the shell model within a molecular dynamics simulation. A simple steepest descent method is used for the adiabatic relaxation of the zero mass shells in the fields arising from neighbouring ions. This method proves to be an order of magnitude more expensive in computing time than a corresponding rigid ion simulation. The method is applied to the simulation of molten salts and the results show better agreement with experiment than the corresponding rigid ion model for the partial radial distribution functions, the diffusion coefficients, and the longitudinal currents.

1. B. Donovan and J.F. Angress 1971, Lattice Vibrations (Chapman & Hall)
2. R.H. Lyddane, R.G. Sachs, E. Teller, Phys. Rev. 59, 673 (1941).
3. M. Dixon and M.J.L. Sangster, J. Phys. C. 9, 909 (1975).
4. K.M. Diller, D. Phil. Thesis, Oxford (1975).
5. G. Jacucci, I.R. McDonald and A. Rahman, Phys. Rev. A13, 1581 (1976).
6. C.R.A. Catlow, this conference.
7. P. Tasker, DL/SCI/R15, p86 (1980).

Algorithms for Polarization through Charge Modification

H.J.C. Berendsen

Polarisability can be introduced by inducing point dipoles, by displacing charges or by modifying the magnitude of charges. The latter method requires at least four charges to be present in the model.

In order to formulate the equations for charges, energy and forces, we write the energy U as the sum of the electrical interaction energy U_{el} and in internal polarization energy U_{pol}: $U = U_{el} + U_{pol}$. The electrical energy equals the total electrical energy of the polarized system (<u>not</u> including a factor 1/2 for the energy due to the induced charges as is usually encountered in polarization energies); the polarization energy is a quadratic term in the induced polarization. The charges adjust themselves in such a way that the partial derivative of U with respect to the polarization vanish. The matrix of coefficients for the quadratic term are related to the polarisability tensor of the molecule and the geometry of the point charge model. It turns out that U_{pol} exactly equals −1/2 times the electrical energy of the induced charges. The model implies that the charges on each molecule are modified by the potentials on the charges of the same molecule. The relation is a simple 4 × 4 matrix multiplication. The forces are gradients of the energy U and become equal to the (polarized) charge times the electric field at the charge. The energy can be reworked to a very simple expression: one half the <u>un</u>polarized charge times the potential, summed over all charges and particles. The forces are not simple gradients of this expression for the energy due to the dependence of polarization on co-ordinates.

Applied to a four-point charge model off liquid water, the algorithm shows an exponential convergence, needing only 2 iterations per time step and increasing computer time by a factor 2 to 2.5. No polarisability catastrophe or unreasonable dipole fluctuations occur.

Supercomputer Tests (Additional Contribution)

Tests of a vectorisable MD program on various computers, including the CRAY 1, CYBER 203 and CYBER 205, were reported. The relative speed depends on the vectorisability of the routines tested. The CYBER 205 was about 15% faster than the CRAY 1 for the overall time step, 40% faster on the force calculation, 70% faster on a well-vectorisable routine, but only half as fast on a non-vectorisable routine.

A full report will become available and will be sent to those who indicate their interest in writing to Prof. H.J.C. Berendsen, Laboratory of Physical Chemistry, Nijenborgh 16, 9747 AG Groningen, The Netherlands.

Algorithms for Polarisation using Multipole Expansions

P. Barnes

The introduction of polarisation into potentials used in Monte-Carlo simulations is important for certain fluids such as water[1]. Although polarisation algorithms are not really suitable for the Monte Carlo method, they are necessary for example in estimating the free energy of water in specific active sites of biological molecules. One must therefore consider possible simplifications and their likely effect.

A solution by iterative methods for polarisation would entail programming loops of the following kind;

<u>Dipole updating for all N</u> $\underline{\Delta\mu}^{N}_{k} = \alpha^{N}_{\alpha\beta}\cdot\underline{\Delta F}^{N}_{k}$ (1)

($\alpha^{N}_{\alpha\beta}$ = polarisability tensor, $\underline{\Delta F}^{N}_{k}$ = field increment on molecule N and equals the initial (dipole + quadrupole) field for k = 1)

<u>Field updating for all N</u> $\underline{\Delta F}^{N}_{k+1} = \sum_{M} {}^{M}g^{N}_{\alpha\beta}\cdot\underline{\Delta\mu}^{M}_{k}$ (2)

(${}^{M}g^{N}_{\alpha\beta}$ = a geometrical tensor fixed for any molecular pair M-N)

iterative
k
cycle

With implementation on Cray computers, full advantage of vectorisation cannot be made w.r.t. eq. (2) and so the polarisation cycles become the main computing time user. The following table examines some possible shortcuts:

Code Name	Method	Timing Ratios	Errors E	Errors ΔE	Periodic updating
MC2	Full polarisation (eqs.(1) and (2)) every M-C step	1	0% (nominal)	0%	Every MC-step
MC3	Full polarisation cycles only within "cut-off" radius of MC - displaced molecule	0.66	0.01%	14%	circa 1 in 40 MC-steps
MC3 $_{1/2}$	As MC3 but treating new configuration as perturbation of old one	0.35	-	-	-
MC4	No polarisation cycles <u>after</u> initial configuration except periodic updating	0.21	0.03%	45%	circa 1 in 40 MC steps

Accuracy of energy calculations are for 216 water molecules at STP, with 7 to $7^1/_2$ Å cut-off, 99% polarisation convergence, and CRAY1 timings. E refers to the internal energy and ΔE its average MC-change for 50% overall success.

Such approximations clearly speed up the simulation (e.g. 1/2M MC-steps for water - MC4 over $7^1/_2$ hours) but it is difficult to assess their true effect. MC4 may be applicable for certain biological situations (e.g. isolated "water pockets"). However one is less confident using MC4 to say bulk water where it is suspected that pair-additive models cannot reproduce the long range oscillations in the OH and HH-RDFs[2]. On the other hand method MC2 does have the propensity to propagate long-range indirect interactions across "polarisation bridges"[1] as a consequence of the many body effect resulting from polarisation[3].

1. Barnes P., Finney J.L., Nicholas J.D. and Quinn J.E., Nature, <u>282</u>, 459 (1979).
2. Dore J., reported in this meeting.
3. Quinn J.E. and Gellatly B.J., reported in this meeting.

Multipole Expansion up to Order 14 Based on the Maxwell Poles of Spherical Harmonics. Convergence of the Multipole Series for Water and Ice.

M. Mezei

A comprehensive program for approximating the electrostatic energy of interaction between two charge distributions is described based on the maxwell poles of spherical harmonics. The procedure involves:

(a) obtaining the moments of the charge distributions involved up to the desired order;
(b) determining the corresponding Maxwell poles;
(c) computing the interaction energy of the interacting charge distributions as represented by their respective Maxwell poles;
(d) for lattice energy calculations perform the Ewald summation using the formulae developed by E.S. Campbell.

This procedure has been implemented for water molecule interactions up to order 14. Results will be discussed on:

(a) the convergence of the multipole series;
(b) the effect of various density partitioning schemes;
(c) the convergence of lattice sums for ices Ih, II and IX;
(d) the ability of the polarization model using induced dipoles to represent the multibody contribution to the energy of interaction, based on results for small clusters and Ice Ih.

Three and Four Body Interactions in the P.E. Model

J.E. Quinn

As an aid to understanding the role of three and four body effects in polarisable fluids, calculations have been carried out using the polarisable electropole potential for water to estimate the magnitude of three and four body contributions to the total interaction energy.

Two classes of calculation were performed:

(i) using randomly chosen triplets and quartets;
(ii) using triplets and quartets picked from model systems (bulk water, ice Ih, ice VIII).

Type (i) showed that the three body contributions were typically 5-10% of the total while four body energies were an order of magnitude smaller.

Type (ii) for bulk water showed that three body energies were again of order 5-10% but generally more negative, while four body effects were < 1%. Considerable scatter in the results indicated that there is no correlation between large total energies and large three/four body energies but rather that the partitioning is highly dependent on configuration. The effect of symmetry in the ices was seen to reduce the scatter considerably.

These results lend weight to the idea that many body interactions may be included in computer models by analytic two and three N-body terms and that such series may be very quickly convergent.

The P^3M Code

R.W. Hockney

The particle-particle/particle-mesh or P^3M algorithm is a method of force calculation in which the number of arithmetic operations can be made proportional to the number of particles, N, rather than to its square. This is achieved by obtaining the long range interaction from the solution of the overall potential on a regular three-dimensional mesh using fast Fourier transform techniques (FFT). The correct interaction is obtained at short range by adding on a correction from nearby particles only. This is done pairwise out to 3 or 4 mesh distances. Neighbours are found by a very rapid "linked-list" technique (ref. 2) that requires only about 1,000 stores for a head of chain table, and one extra link coordinate per particle. A program P3M3DP (Ref. 7) is available through the CPC Program Library at Queens University, Belfast. Some examples of computing timesteps on the IBM 360/195 are:

$N = 512$	1s
$N = 4{,}096$	6s
$N = 10{,}648$	17s
$N = 17{,}576$	30s
$N = 21{,}952$	33s

A useful rule of thumb is ~ 1.5s per 1,000 particles. The method is published in the following papers.

Description (2D)

1. Hockney, Goel and Eastwood (1973) "A 10,000 Particle MD Model with Long Range Forces", Chem. Phys. Lett. 21, 589-591.
2. Hockney, Goel and Eastwood (1974) "Quiet High-Resolution Computer Models of a Plasma", J. Comput. Phys. 14, 148-158.

Theory (3D)

3. Eastwood (1975) "Optimal Particle Mesh Algorithms", J. Comput. Phys. 18, 1-20.

   Eastwood (1976) in Computational methods in Classical and Quantum Physics, Advance Publications.

4. "Value for Money in Particle Mesh Plasma Simulations", 196-205.
5. "Optimum P^3M Algorithms for Molecular Dynamics Simulations", 206-228.
6. Hockney and Eastwood (1981) Computer Simulation Using Particles, McGraw-Hill, Advanced Book Program: Theory (8), Galaxy Clustering (11), Mol. Dyn. (12).

Program (3D)

7. Eastwood, Hockney and Lawrence (1980) "P3M3DP - the Three Dimensional Periodic Particle-Particle/Particle-Mesh Program", Comput. Phys. Commun. 19, 215-261.

Applications (2D)

8. Hockney and Brown (1975) "A Lambda Transition in a Classical Electron Film", J. Phys. C. 8, 1813-1822.
9. Hockney and Goel (1975) "Phase Transitions in Two-Dimensional Potassium Chloride", Chem. Phys. Lett. 35, 500-507.

(3D)

10. Amini and Hockney (1979) "Computer Simulation of Melting and Glass Formation in a KCℓ Microcrystal", J. Non-Crystalline Solids, 31, 447-452.
11. Amini, Fincham and Hockney (1979) "A MD Study of the Melting of Alkali-Halide Crystals", J. Phys. C. 12, 4707-4720.
12. Mitra and Hockney (1980) "Distribution of Holes in Simulated SiO_2 Glass", J. Phys. C. 13, L739-L741.
13. Mitra, Amini, Fincham and Hockney (1981) "Molecular Dynamics Simulation of SiO_2 Glass", Philos. Mag. 43, 365-372.
14. Efstathiou and Eastwood (1981) "On the Clustering of Particles in an Expanding Universe", Mon. Not. R. Astron. Soc. 194

Convergence Acceleration in Monte Carlo Computer Simulation on Water and Aqueous Solutions

P.K. Mehrotra, M. Mezei and D.L. Beveridge

The convergence characteristics of standard Metropolis Monte Carlo calculations on liquid water and aqueous solutions was described, and documentation of the need for convergence acceleration procedures was presented. These take the form of importance sampling procedures added to the Metropolis method. Comparative studies on liquid water using the force bias method show that convergence with this procedure is improved by a factor of 2-3 over standard Metropolis. For aqueous solutions, both force bias and preferential sampling procedures were considered. Neither method alone was found to be adequate for describing structural characteristics of solutions in realizations of the order used for simulations on pure liquids. A combination of the Metropolis, force bias and preferential sampling methods was found to be successful in this respect, and makes aqueous solutions accessible to simulation studies at high levels of computational rigor. Implications of this work for theoretical studies of biological water problems was mentioned.

A Multipolar Expansion using Accurate Gaussian Orbitals

P. Claverie

Readers interested in this topic should contact Professor Claverie at:

Institut de Biologie Physico-Chimique
13 Rue Pierre et Marie Curie
7 5005 Paris
FRANCE

Microdynamical Behaviour of Aqueous LiI Solution (A Poster Presentation)

Gy. I. Szász and K. Heinzinger

The properties of a 2.2 m LiI solution have been calculated from a molecular dynamics simulation over 10 picoseconds at a temperature of 305 K. The basic periodic cube contained 200 water molecules and 8 ion pairs. The effective pair potentials are based on the ST2 water model and a single point charge model for the ions.

In addition to the structural properties of the solution, the self-diffusion coefficients, the reorientation times of the dipole moment and the correlation times of the proton-proton vector of the water molecules are calculated separately for the following subsystems: bulk water and hydration water of Li^+ and I^-. These quantities are compared with the available experimental data, and implications with respect to "structure forming" ability of the ions are presented.