DEPARTMENT OF CHEMISTRY

Charlotte Secher
Secretary

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Direct tel: +45 8942 3884
Direct fax: +45 8619 6199
Email: cha@chem.au.dk
Web: chem..au.dk

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SCIENTIFIC REPORT

Scientific Report on the European Science Foundation Exploratory Workshop:

New Information from Modern Charge Density Studies, held 26 - 29 June 2003, at Sandbjerg Estate, Denmark.

1. Executive summary.

The charge density (CD) of a molecular system is probably the most information rich observable, which is available for the system. It contains much information about the chemical bonding as well as the physical and chemical properties of molecular systems. It can be determined experimentally from elaborate X-ray diffraction measurements at short wavelength and low temperature. In some cases it can also be calculated from theory. The CD defines the electronic ground state of a system, and this fundamental insight has lead to the illustrious successes of Density Functional Theory in computational chemistry and physics. Due to its fundamental importance, the CD is used across many disciplines in physics, chemistry, biology and the life sciences.

The workshop had the ambitious aim of bringing together scientists from disciplines using CD concepts with scientists specialized in measuring, calculating and interpreting the CD. In this way, the workshop would attempt to build common ground for the many fields drawing on the information contained in the CD, but also, in the bridging process, provide a forum in which new directions for future CD studies can be envisioned. A potential important European added value of the workshop would be the formation of strong new networks across Europe. We believe the workshop fulfilled all these diverse goals, and set the stage for an exciting European future in charge density analysis.

Department of Chemistry
University of Aarhus
Langelandsgade 140
DK-8000 Aarhus C
Denmark
Tel: +45 8942 1111
Fax: +45 8619 6199
Web: www.chem.au.dk

A direct outcome of the workshop is
1) the formation of a new European network spanning 11 countries of research groups active in charge density studies as well as
2) publication of proceedings of the PESC Workshop on New Information from Modern Charge Density Studies in a special volume of the international journal Acta Crystallographica Section A.

The workshop took place at the Aarhus University conference centre Sandbjerg Estate, which over the years has gained much experience in hosting international workshops and conferences. We believe the facilities provided an excellent setting for stimulating scientific discussion. In the genuine southern Denmark tradition no participants were allowed to loose a gram of weight during the meeting and indeed the food service and housing lived well up to expectations.

The PESC workshop was organised together with the third European Charge Density Meeting (ECDM-III), which preceded the workshop (June 24 to 26). The rationale of this construction was to offer a full-scale meeting in which both the established charge density community and newcomers from related fields could meet. The back-to-back organisation also allowed the meeting to be cost-effective, and provide opportunity for a substantial number of young researchers to participate. The PESC workshop had 22 invited lecturers, some of which, at own expense, also took part in the preceeding ECDM-III meeting, like many ECDM-III participants stayed on for the PESC workshop. There were a total of 65 participants involved in the arrangement. The joint organisation of the two meetings also allowed better time for breaking the ice, and it gave a wonderful chance for younger researcher to interact in an informal atmosphere with the established experts.

Appended with this report please find a copy of the **programme and abstract book** for the total meeting arrangement.

Furthermore, the ECDM-III webpage (**www.chem.au.dk/ECDM-III**) with details of the meeting will remain active for the rest of the year.

2. Scientific content of the event.

The scientific program consisted of four half-day sessions, an opening lecture and poster presentations. The opening lecture was given by Professor Coppens, who is the founder of the field of X-ray charge density analysis. Professor Coppens set a perfect stage for the rest of the PESC talks by discussing the complementary nature, but also mutual interdependence, of experimental and theoretical charge density methods.

In the first session, the main theme was chemical bonding and reactivity determined by experimental charge density studies. The session was opened by Professor Destro, a pioneer of helium temperature crystallography, who discussed the advantages of very low temperature measurements. The talk gave an immediate insight for newcomers and theoreticians to the challenges and rewards of first class experiments. During the rest of the meeting, Professor Destro was often the experimental reference point, and he furthermore served as an exemplary initiator of new discussions. In the following talk, Dr. Iversen was the first of three young scientists. In his talk he pushed the application of CD methods into new fields by discussing comparative results on a series of model complexes for enzymatic active sites. Dr. Macchi followed up by showing the new frontiers that can be reached by application of Bader topological methods in analysis of charge densities of coordination complexes. The session was closed by Dr. Espinosa, who discussed the field of quantitative crystal engineering through correlations of topological indices of hydrogen bonds.

The second session, which was on theoretical charge density studies, involved 6 eminent theoreticians of the highest international status. Professor Frenking started by showing the latest advances on understanding the energetics of chemical bond formation. As a newcomer to the charge density community, who has nevertheless a distinguished career based on interpretation of theoretical densities, Professor Frenking engaged in stimulating discussions of both his own results as well as those of many other contributors. Next, Dr. Gatti explained the merits of a completely new theoretical tool: the source function. Based on the still few available results of application of the source function, Dr. Gatti educated everyone and provided the community with an exciting new tool for obtaining chemical information from charge densities. Before the coffee break, Professor Blaha reviewed the present state of the art in density functional theory exemplified with the WIEN2K code and applications to solid state physics. After the break, Professor Grin took the audience on a journey of rational materials design based on application of the Electron Localisation Function to novel

semiconducting materials. Professor Popelier then changed the theme to biological applications and presented thought-provoking new ideas on how to develop optimal force fields for application in molecular dynamics simulations. The session was wrapped up by Professor Gatteschi, who opened our eyes to potential new applications of CD methods in the field of molecular magnetism – a highly visible area of modern nanoscience.

The Saturday morning session was dedicated to magnetism in relation to charge densities. The session was intended to further break the ice to new frontiers of CD research. Dr. Winpenny opened the session by presenting the rich chemistry of high spin cages and single molecule magnets. A number of very interesting structures were discussed, and key problems which may be attacked with charge density methods were pointed out. Dr. Lelievre-Berna then swept the audience away with a presentation on the latest advances in polarised neutron science with focus on spin density applications. Once again, the amazing interdisciplinarity of CD concepts was brought to fore with discussions of the complimentary nature of different experimental techniques. After the coffee break two talks provided further insight into the chemistry and physics of molecular magnetism as seen from the charge density point of view. Dr. Goeta and Dr. Pillet then presented X-ray charge density analysis of organic radicals and various organometallic systems. Since these studies bridge various fields of chemistry and physics, both talks demonstrated beautifully the critical interdisciplinary element in modern molecular science.

The concluding session consisted of six talks intended to show the road to the future with presentation of novel experimental techniques. Dr. Cole opened by discussing the mind boggling opportunities of obtaining charge density information on excited state structures through use of third generation synchrotron radiation facilities. Huge progress in instrumentation and crystallographic concepts has now brought this goal within reach. Professor Hansen then showed results on charge density of systems in external electric fields. The minute changes in bonding electron distributions can now be measured experimentally, and this gives hope for developing better theories of the atomic origin of various field induced phenomena. Before the coffee break, Professor Katrusiak explained about the considerable experimental challenges of single crystal diffraction measurements of crystals under pressure. After the break, Professor Takata provided a wonderful glimpse of the emerging new possibilities for obtaining detailed charge densites from powder diffraction data. Exciting applications to novel nanoporous materials revealed that charge density analysis has a bright future in providing fundamental insight into key issues

of nanoscience. Professor Luger then showed the state of the art in experimental single crystal studies at synchrotron sources. A series of results on biomolecules were presented as well as the very latest results from the new helium temperature diffractometer at the German synchrotron at Hasylab. The meeting was closed by Professor Lecomte who reminded everyone that all the technical advances in X-ray radiation, computer power, detectors, interpretation tools etc. must not let us loose attention of traditional merits. Random application of charge density methods in emerging fields must not completely replace continued methodological research on the basic development of the X-ray method.

3. Assessment of the results, contribution to the future direction of the field.

In conclusion, the scientific programme lived up to all expectations with lively sessions and vigorous discussions all in an informal and constructive atmosphere. The scientific discussions extended far beyond the scheduled sessions, and it was exciting to walk through the halls of the conference centre even late at night and experience continued discussions showing the genuine interest of the participants. An extra ad-hoc session was organised on Saturday evening to prepare the formation of a new European network on charge density analysis. The direct result of this meeting was selection of an 11 members steering committee (representatives from most of the participating European countries), which will write the application to ESF for the formal formation of a network.

Finally, we are happy to announce that we have succeeded in obtaining a go-ahead for publishing the proceedings of the PESC Workshop as a Special Issue of the prestigious international journal Acta Crystallographica A.

4. Final programme for

European Science Foundation Exploratory Workshop:

New Information from Modern Charge Density Studies, held
26 - 29 June 2003, at Sandbjerg Estate, Denmark.

Thursday, June 26

20.30	Opening by Bo Brummerstedt Iversen	
	Introductory talk by *Philip Coppens*, State University of New York at Buffalo, USA THE INTERPLAY BETWEEN THEORY AND EXPERIMENT IN CHARGE DENSITY STUDIES.	**T1**

Friday, June 27

	Chair: Finn Krebs Larsen	
8.45-9.00	*Judith Howard*, University of Durham, England ***Presentation of the sponsoring European Science Foundation.***	
Session on	***Chemical bonding and reactivity determined by experimental charge density studies.***	
9.00-9.40	*Riccardo Destro*, Università di Milano, Italy GAIN FROM CHARGE-DENSITY-QUALITY X-RAY DIFFRACTION EXPERIMENTS.	**T2**
9.40-10.20	*Bo Brummerstedt Iversen*, University of Aarhus, Denmark THE ELECTRON DENSITY DISTRIBUTIONS OF REDOX ACTIVE MIXED VALENCE CARBOXYLATE BRIDGED TRINUCLEAR IRON COMPLEXES.	**T3**
10.20-10.40	Coffee	
10.40-11.20	*Piero Macchi*, Università di Milano, Italy THE ELUSIVE NATURE OF THE METAL-METAL BOND IN ORGANOMETALLIC CLUSTERS.	**T4**
11.20-12.00	*Enrique Espinosa*, Université de Bourgogne, France FROM WEAK VAN DER WAALS TO STRONG COVALENT INTERACTIONS: A COMPARATIVE ANALYSIS OF THE TOPOLOGICAL AND ENERGETIC PROPERTIES OF THE ELECTRON DENSITY DISTRIBUTION INVOLVING X-H - - FY SYSTEMS.	**T5**

12.00-13.30 Lunch
Chair: Philip Coppens

Session on ***Theoretical charge density calculations.***

13.30-14.10 *Gernot Frenking*, Philipps-Universität Marburg, Germany **T6**
THE ROLE OF COMPUTATION IN THE INTERPRETATION OF THE CHARGE DENSITY DISTRIBUTION.

14.10-14.50 *Carlo Gatti*, CNR-ISTM, Milano, Italy **T7**
CHEMICAL INFORMATION FROM THE SOURCE FUNCTION.

14.50-15.30 *Peter Blaha*, Technische Universität Wien, Austria **T8**
THEORETICAL CHARGE DENSITY STUDIES IN CON-DENSED SYSTEMS.

15.30-16.00 Coffee

16.00-16.40 *Yuri Grin*, Max-Planck-Institut, Dresden, Germany **T9**
INTERPRETATIVE TOOLS FOR STUDIES IN MATERIALS RESEARCH.

16.40-17.20 *Paul Popelier*, The University of Manchester, England **T10**
ATOMIC PROPERTIES OF AMINO ACIDS: COMPUTED ATOM TYPES AS A GUIDE FOR FUTURE FORCE FIELD DESIGN.

17.20-18.00 *Dante Gatteschi*, University of Florence, Italy **T11**
ON MOLECULAR MAGNETS.

18.00 Dinner

Saturday, June 28

Chair: Judith Howard

Session on ***Magnetism in Relation to Charge Density Studies.***

9.00-9.40 *Richard Winpenny*, The University of Manchester, England **T12**
STUDIES OF HIGH SPIN CAGES AND SINGLE MOLECULE MAGNETS.

9.40-10.20 *Eddy Lelievre-Berna*, Institut Laue-Langevin, Grenoble, France **T13**
ADVANCES IN POLARIZED NEUTRON DIFFRACTION FOR SPIN DENSITY DETERMINATION.

10.20-10.40 Coffee

10.40-11.20 *Andres E. Goeta*, University of Durham, England **T14**
CHARGE DENSITY ANALYSES FOR THE STUDY OF MAGNETIC EXCHANGE PATHWAYS.

11.20-12.00 *Sebastien Pillet*, Université Henri Poincaré, Nancy, France **T15**
MOLECULAR MAGNETIC SYSTEMS, WHAT CAN BE EXPECTED FROM CHARGE DENSITY ANALYSIS?

12.00-13.30 Lunch

Chair: Bo Brummerstedt Iversen

Session on ***Challenging Charge Density Studies. The Future.***

13.30-14.10 *Jacqueline M. Cole*, University of Cambridge, England **T16**
"IN SITU" CHARGE DENSITY STUDIES OF PHOTO-INDUCED PHENOMENA: POSSIBILITIES FOR THE FUTURE?

14.10-14.50 *Niels K. Hansen*, Université Henri Poincaré, Nancy, France **T17**
STUDIES OF ELECTRIC FIELD INDUCED STRUCTURAL AND ELECTRON DENSITY MODIFICATIONS BY DIFFRACTION.

14.50-15.30 *Andrzej Katrusiak*, Adam Mickiewicz University, Poznan, Poland **T18**
CHALLENGES OF CHARGE-DENSITY DISTRIBUTION FROM HIGH-PRESSURE STUDIES.

15.30-16.00 Coffee

16.00-16.40 *Masaki Takata*, JASRI/SPring-8, Japan **T19**
A DIRECT OBSERVATION OF GAS MOLECULES IN NANO-CHANNEL METAL ORGANIC SOLID BY THE MEM/RIETVELD METHOD.

16.40-17.20 *Peter Luger*, Freie Universität Berlin, Germany **T20**
HIGH RESOLUTION SYNCHROTRON DATA COLLECTIONS FOR CHARGE DENSITY WORK AT 100 K AND 20 K.

17.20-18.00 *Claude Lecomte*, Université Henri Poincaré, Nancy, France **T21**
SOME FRONTIER PROBLEMS IN CHARGE DENSITY STUDIES.

18.00 Closing Dinner

Sunday, June 29

Ad Hoc meetings and discussions

12.00 Lunch and departure from Sandbjerg

Professor Vladimir Tsirelson, Mendeleev University, Moscow, Russia, gave his PESC lecture: ON FUNCTIONS AND QUANTITIES DERIVED FROM THE ELECTRON DENSITY, on Wednesday, June 25.

Professor Tsirelson had to give his lecture at an earlier time than first planned, because he had received an invitation to meet with the German president Rau in Berlin, where recent Humboldt awardees were given an official reception on June 26.

5. FINAL LIST OF PARTICIPANTS

European Science Foundation Exploratory Workshop:
New Information from Modern Charge Density Studies, held
26 - 29 June 2003, at Sandbjerg Estate, Denmark

Convenors:

1. Bo Brummerstedt Iversen
Department of Chemistry
University of Aarhus
DK-8000 Aarhus C
Denmark
E-mail: bo@chem.au.dk

2. Finn Krebs Larsen
Department of Chemistry
University of Aarhus
DK-8000 Aarhus C
Denmark
E-mail: kre@chem.au.dk

ESF representative:

3. Judith A.K. Howard
University of Durham
Department of Chemistry
Science Laboratories
South Road
Durham DH1 3LE
United Kingdom
Email:J.A.K.Howard@durham.ac.uk

Participants:

4. Peter Blaha
Institute of Materials Chemistry
Technische Universität Wien
Getreidemarkt 9
A-1060 Vienna
Austria
E-mail:
pblaha@theochem.tuwien.ac.at

5. Jacqueline Cole
University of Cambridge
Department of Chemistry
Lensfield Road
Cambridge CB2 1EW
U. K.
E-mail: jmc61@cus.cam.ac.uk

6. Philip Coppens
732 NSM Building
Chemistry Department
SUNY/BUFFALO
Buffalo, NY 14260-3000
U. S. A.
E-mail: coppens@acsu.buffalo.edu

7. Riccardo Destro
Dipartimento di Chimica fisica
ed Elettrochimica
University of Milano
Via Golgi 19
I-20133 Milano
Italy
E-mail: riccardo.destro@unimi.it

8. Enriques Espinosa
Université de Bourgogne
Faculté des Sciences
LIMSAG, UMR 5633
6, Bd. Gabriel
F-21100 Dijon
France
E-mail: enrique.espinosa@u-bourgogne.fr

9. Gernot Frenking
Phillips-Universität Marburg
Fachbereich Chemie
Hans-Meerwein-Strasse
D-35032 Marburg
Germany
E-mail: frenking@chemie.uni-marburg.de

10. Dante Gatteschi
Department of Chemistry
University of Florence
Via della Lastruccia 3
I-50019 Sesto Fiorentino
Italy
E-mail: dante.gatteschi@unifi.it

11. Carlo Gatti
CNR-ISTM
Istituto di Scienze e
Tecnologie Molecolari
Via Golgi 19
I-20133 Milano
Italy
E-mail: c.gatti@istm.cnr.it

12. Andres E. Goeta
University of Durham
Chemistry Department
Durham DH1 3LE
U K
E-mail: a.e.goeta@durham.ac.uk

13. Yuri Grin
Max-Planck-Institut für Chemische
Physik
Nöthnitzer Strasse 40
D-01187 Dresden
Germany
E-mail: grin@cpfs.mpg.de

14. Niels K. Hansen
LCM3B
Université Henri Poincaré - Nancy I
B.P. 239
F-54506 Vandoeuvre les Nancy
France
E-mail: hansen@lcm3b.u-nancy.fr

15. Andrzej Katrusiak
Adam Mickiewicz University
Department of Crystal Chemistry
ul. Grunwaldzka 6
PL-60-780 Poznan
Poland
E-mail: katran@amu.edu.pl

16. Claude Lecomte
LCM3B
Faculté des Sciences
Université Henri Poincaré, Nancy I
B.P. 239
F-54506 Vandeouvre-les-Nancy
Cédex
E-mail: lecomte@lcm3b.uhp-nancy.fr

17. Eddy Lelievre-Berna
Institut Laue-Langevin
Science Division
6 rue Jules Horowitz
BP 156
F-38042 Grenoble cedex 9
E-mail: lelievre@ill.fr

18. Peter Luger
Freie Universität Berlin
Institut für Chemie/Kristallographie
Takustrasse 6
D-14195 Berlin
Germany
E-mail: luger@chemie.fu-berlin.de

19. Piero Macchi
Dipartimento di Chimica Strutturale
e Stereochimica Inorganica
Via Venezian 21
University of Milano
I-20133 Milano
Italy
E-mail: piero.macchi@istm.cnr.it

20. Sebastien Pillet
UHP Nancy I
Faculté des Sciences
Boulevard des Aiguillettes
F-54506 Vandoeuvre-les-Nancy
France
E-mail: pillet@lcm3b.u-nancy.fr

21. Paul Popelier
University of Manchester
Institute of Science and
Technology (UMIST)
88 Sackville Street
Manchester M60 1QD
U. K.
E-mail: paul.popelier@umist.ac.uk

22. Masaki Takata
JASRI/SPring8
Mikazuki
Sayo
JP-679-5198 Hyogo
Japan
E-mail: takatama@spring8.or.jp

23. Vladimir Tsirelson
Miusskaya Sq., 9
RU-127047 Moscow
Russia
E-mail: tsirel@muctr.edu.ru

24. Richard Winpenny
The University of Manchester
Department of Chemistry
Oxford Road
Manchester M13 9PL
U. K.
E-mail:
richard.winpenny@man.ac.uk

6. Statistical information on participants.

European Science Foundation Exploratory Workshop:

New Information from Modern Charge Density Studies, held 26 - 29 June 2003, at Sandbjerg Estate, Denmark.

The 22 PESC lecturers came from 10 different countries:

Austria (1), Denmark (1), France (5), Germany (3), Italy (4), Japan (1), Poland (1), Russia (1), United Kingdom (4), and U.S.A. (1).

9 of the lecturers were younger than 40 years of age.

Sincerely yours,

Bo Brummerstedt Iversen

Professor, Dr.scient.
P.t. in the U.S.A.

Finn Krebs Larsen

Associate Professor

Department of Chemistry
University of Aarhus
Langelandsgade 140
DK-8000 Aarhus C
Denmark
Tel: +45 8942 1111
Fax: +45 8619 6199
Web: www.chem.au.dk