

14-Diffraction Physics and Optics

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14.02 - Electron Densities at the Forefront of Chemistry and Solid State Physics

OCM-14.02.01

POSITIVE AND NEGATIVE SIDES TO CHARGE DETERMINATION. By Kersti Hermansson, Department of Chemistry, Uppsala University, Box 531, S-75121 Uppsala, Sweden

Molecular modelling - molecular mechanics, Monte Carlo and molecular dynamics calculations - is gaining ground in all branches of condensed matter chemistry. The success of these methods stands and falls with the quality of the force field used. For polar and van der Waals systems the electrostatic interaction is a major contributor to the overall interaction. The derivation of reliable electrostatic parameters - atomic charges, distributed charges, dipoles etc. - is a difficult enterprise. The atomic charges built into different commercial molecular modelling packages vary widely and give rise to large differences in the computed electrostatic interaction energies for a pair of molecules.

In the talk I will discuss:

- the role of atomic charges in computer simulations
- methods to derive atomic charges (from experiment and theory)
- models "beyond atomic charges", e.g. distributed multipoles, polarisable atoms, etc.

OCM-14.02.02 CHARGE DENSITY STUDIES WITH THE MAXIMUM ENTROPY METHOD. By R. Y. de Vries¹, W. J. Briels and D. Feil. *Chemical Physics Laboratory, University of Twente, PO Box 217, 7500 AE Enschede, The Netherlands.*

In many fields, the Maximum Entropy Method (MEM) has become the standard method to obtain images from incomplete and noisy data. In crystallography, there are only a few studies for which the MEM has been applied to obtain charge density maps from Fourier data. Sakata & Sato (*Acta Cryst.* (1990). A46, 263-270) applied the MEM to the very accurate *Pendellösung* data on silicon, where they showed that the method handles the series termination effect very well.

We have developed a computer program to maximize the entropy subject to $\chi^2 = \sum_k^M \frac{(F_k - D_k)^2}{\sigma_k^2} = M$, where D_k is the measured structure factor, F_k is the structure factor calculated from the electron density, σ_k is the standard deviation of the measured reflection and M is the total number of reflections. The program was based on the work of Bryan & Skilling (*Mon. Not. R. Astr. Soc.* (1984) 211,111-124).

We present a study of the MEM on hypothetical crystals. Structure factors were calculated from *ab initio* wave functions. Noise was added to these structure factors to simulate the experimental situation. We discovered that when less accurate data are available problems arise due to the fact that the constrained optimization of the entropy does not lead to a proper χ^2 -distribution. In the obtained Electron Density Distribution (EDD) the calculated value of (a few) low order reflections tend to deviate much from the measured value causing the high order reflections of the obtained EDD to be exactly equal to their measured values. The resulting EDD deviates uncomfortably much from the original distribution. A weighting scheme is suggested which leads to a better χ^2 -distribution and a much improved EDD.

Whereas in the MEM deviations from a flat distribution are only allowed as far as is necessary to satisfy the constraints imposed by the observations, in the Minimum Cross-Entropy Principle the deviations from a *a priori* distribution are minimized. The results of the two methods will be compared.

OCM-14.02.03

MAGNETIC COMPTON SCATTERING, PROGRESS AND PROSPECTS.

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It is only comparatively recently that studies of HoFe₂, which has dominant orbital magnetisation, have confirmed that magnetic Compton scattering, in the impulse approximation, is indeed only sensitive to the spin magnetisation. The experiment yields a projection of the momentum distribution of the unpaired spin component and this magnetic Compton profile is characteristic of the electron's atomic origin. Thus for example in HoFe₂ the 4f and 3d contributions can be easily separated and their relative contribution to the spin moment deduced. It is also possible to study the nature of the diffuse component which in this material appears to be associated with the Ho site; it does not change in magnitude significantly between room and low temperature in contradiction to unpublished neutron data. Unfortunately appropriate band calculations are not yet available to be tested by these data. The establishment of an absolute scale, for example by measuring a standard ferromagnet such as Fe then allows the individual moments to be assigned; furthermore combination of these data with bulk magnetisation measurements allows the orbital moments to be determined. In the same material the magnetic Compton profile has been measured at temperatures down to 10K and an estimate of the spin compensation point (200 ± 10K) made. This work clearly shows that magnetic Compton scattering provides a large amount of detailed information data with which models of the electron density distribution can be tested, even in a material for which the net magnetic scattering effect is only 0.05% of the charge scattering. Other experiments, in which high resolution magnetic profiles have been obtained and the spin sub-band momentum distributions separated, will be reviewed and the implications for future work considered.

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HIGH RESOLUTION COMPTON SCATTERING FOR MOMENTUM DENSITY AND FERMI SURFACE DETERMINATION. By A. Bansil, Department of Physics, Northeastern University, Boston, Mass. 02115, USA. and N. Shiotani, Tokyo University of Fisheries, Kounan, Minato, Tokyo 108, Japan.

The energy profile of the Compton scattered x-rays is well known to probe the electron distribution of the target in momentum space, i.e. the momentum density $n(p)$ of the electron system. For investigating bulk electronic states, the Compton experiment does not suffer the drawback of surface sensitivity of UV photoemission, or of defect sensitivity of the dHvA and positron annihilation spectroscopies. Further, especially in complex unit cell materials (e.g. the high-T_c's), the signals associated with different pieces of the Fermi surface can be suppressed or enhanced in various other spectroscopies as a result of matrix element and other effects. On the other hand, the Compton scattering samples all the electron states with equal weight. These facts make the Compton scattering a valuable complement to other k-resolved spectroscopies. In principle thus, the Compton technique can provide unique information concerning the Fermi surface parameters and spectral properties of the electron states at and near the Fermi energy in ordered as well as disordered crystals.