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MAXIMUM LIKELIHOOD DENSITY MODIFICATION UNDER MAXIMUM ENTROPY CONTROL

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We present a method to improve the electron density for missing atoms of macromolecular structures, using Likelihood maximization [1,2,4] with entropy loss constraints [1,2]. Both the model for the structure factor offset F_h and the one for its associated uncertainty can incorporate: 1. a partial structure, as a PDB file; 2. a bulk solvent envelope; 3. a real-space distribution q(x) for the missing atoms, initially set to a prior-prejudice m(x) [1,3].

An experimental phase distribution can be combined with the model-generated one. When neither 1. nor 2. are available, the method represents an alternative to conventional solvent flattening of the experimental electron density. The log-likelihood of the model, together with its gradient and Hessian, is computed as a sum of log-Rice functions [1,2]. Its maximization drives the scaling of data to model, and of components 1.-3. to one another. q(x) depends on Lagrange multipliers λ_h: the latter are varied to increase likelihood, while keeping the entropy of q(x) relative to m(x) at a constrained maximum. The log-likelihood gain for the free-set reflections is monitored to validate the extra detail introduced in q(x). The method has aided the completion of two macromolecular complexes starting from Molecular Replacement phases [5,6].

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MAXIMUM ENTROPY METHOD ANALYSIS OF THERMAL MOTION AND DISORDER IN THERMOELECTRIC CLATHRATE $Ba_8Ga_{16}Si_{30}$

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Inorganic clathrates are open framework structures, which have recently attracted much attention due to their potential applications as thermoelectric materials. The transport properties in these materials are heavily influenced by structural characteristics such as framework or guest atom disorder and anharmonicity. We have carried out a multi-temperature (15, 100, 150, 200, 300, 450, 600, 900 K) single crystal neutron diffraction study of the type I clathrate $Ba_8Ga_{16}Si_{30}$. The measurements were carried out at the SCD beam line at the Intense Pulsed Neutron Source, USA. The guest atom displacement parameters obtained from structure factor fitting in reciprocal space are analyzed with semi-anharmonic Einstein models. Subsequently, the structure factors are used in maximum entropy method calculations to obtain direct space nuclear densities. Nuclear difference density maps are used for investigating structural disorder. The guest atom nuclear densities are modeled with anharmonic one-particle potential models to fourth order. Even at elevated temperatures anharmonicity is limited indicating that the low thermal conductivity of the clathrate has a different origin

Keywords: MAXIMUM ENTROPY, THERMOELECTRICS, NEUTRON DIFFRACTION

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LOW-TEMPERATURE HRTEM ANALYSIS OF CHARGE- AND ORBITAL-ORDERED STRUCTURES IN PEROVSKITES

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Charge-orbital ordered structures have intensively been investigated by low-temperature electron diffraction and low-temperature HRTEM, due to their close relation to characteristic electro-magnetic properties such as colossal magneto-resistance (CMR) in manganites1). Recently, superstructure models including sinusoidal lattice distortions have been proposed for commensurately doped layered-perovskites, $Nd_{1-x}Sr_{1+x}MnO_4$ ($x = 2/3, 3/4$)[2]). Here, we present our latest data of HREM investigation of $Nd_{1-x}Ca_{1+x}MnO_4$ which show charge-orbital ordering at low-temperature, not only in commensurate but also in incommensurate doping levels ($0.55 < x < 0.75$). Polycrystalline samples of $Nd_{1-x}Ca_{1+x}MnO_4$ were prepared by the standard ceramic method. The HREM observation was conducted for crushed and ion-milled specimen, by a Hitachi HF-3000S analytical TEM (300 kV), equipped with a cold FEG and liq.N_2 as well as liquid He sample holders (Oxford Instruments). Low-temperature electron diffraction measurements revealed the formation of superstructures accompanied by charge-orbital ordering transition in the composition range of $0.55 < x < 0.75$. The wave vector of structural modulation in the system can be described as ks = (1-x) a*. The low-temperature HRTEM images, taken at 80K, show long-period superlattice fringes and sinusoidal transverse modulation of the crystal structure, with the period of a/(1-x): i.e. 3a for x = 0.67. Such sinusoidal modulations are in good agreement with absence of high-order superlattice reflections in electron diffraction patterns. Also, effects of double-reflection should be taken into account for detailed interpretation of HRTEM images.

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ADVANCED TRANSMISSION ELECTRON MICROSCOPY & THE NANOWORLD: WHAT CAN BE DONE THESE DAYS TO SOLVE MATERIALS PROBLEMS?

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Transmission electron microscopes (TEM) are nowadays rapidly improved mostly by industry. The development is driven by an equally fast evolution of nano- and semiconductor technology. Most noticeable, commercially available electron microscopes are now at the threshold to allow routinely for imaging with sub Angstrom spatial resolution together with spectroscopy of around 100 meV of energy resolution. In addition, computational abilities have improved to a point where materials properties can be predicted from computer models that contain a similar number of atoms as is observable by High Resolution TEM. On this level, theory and experiment merge these days. It is crucial to understand strength' and limits of emerging experimental techniques such as exit wave reconstruction, z-contrast imaging and Cs correction in order to benefit from these unique development. In a series of experiments, data about sensitivities, precision, and resolution of these techniques were produced for particular microscopes and they are reported. A methodology capable of comparing the performance of different microscopes was developed. With this information at hand, examples from semiconductor - and interface science are used to depict the current level of agreement between theory and experiment that can be used to further develop our understanding of physical processes on nano scale.

Keywords: HRTEM, STEM, NAONMATERIALS