

increase of bulk modulus and a red shift in the optical gap. The electronic density of states (DOS) and charge density calculations revealed that the magnetic transition was due to the pressure-induced Fe 3d electron delocalization.

Keywords: FeCO<sub>3</sub>, first-principles, high pressure

## MS.19.5

*Acta Cryst.* (2008). A64, C43

### Predicting crystal structures by random searching

Chris J Pickard<sup>1</sup>, R J Needs<sup>2</sup>

<sup>1</sup>University of St Andrews, School of Physics and Astronomy, North Haugh, St Andrews, Fife, KY16 9SS, UK, <sup>2</sup>University of Cambridge, Cambridge, CB3 0HE, UK, E-mail: cjp10@st-andrews.ac.uk

We predict crystal structures by relaxing randomly chosen starting configurations within density functional theory (DFT)[1]. We combine this with constrained searches incorporating knowledge derived from the random searches and other sources. Applications to various systems are described, including hydrogen,[2] silane,[1] and aluminum hydride.[3] Our study of solid hydrogen includes harmonic proton zero point motion and leads to a radical revision of the DFT phase diagram up to 400 GPa. The most stable phases remain insulating to very high pressures, in agreement with experiment, and we find a strong candidate for phase III. We predict that the equilibrium structure of silane is insulating up to about 200 GPa. Our stable structure in the pressure range 50-250 GPa has high symmetry, each Si site is equivalent and each H site is equivalent, and it is held together by electron-deficient three-center-two-electron bonds. This structure has been observed in a recent x-ray diffraction study.[4] For aluminum hydride we find a transition from the insulating low-pressure alpha phase to an insulating layered structure and then to a metallic phase. Our metallic structure has been observed in a recent X-ray diffraction study.[5]

[1] C. J. Pickard and R. J. Needs, *Phys. Rev. Lett.* 97, 045504 (2006).

[2] C. J. Pickard and R. J. Needs, *Nature Physics* 3, 473 (2007).

[3] C. J. Pickard and R. J. Needs, *Phys. Rev. B* 76, 144114 (2007).

[4] M. I. Eremets, I. A. Trojan, S. A. Medvedev, J. S. Tse, Y. Yao, *Science* 319, 1506-1509 (2008).

[5] I. Goncharenko, M. I. Eremets, M. Hanfland, J. S. Tse, M. Amboage, Y. Yao, and I. A. Trojan, *Phys. Rev. Lett.* 100, 045504 (2008).

Keywords: *ab-initio* calculations, high-pressure phase transformations, hydride compounds

## MS.20.1

*Acta Cryst.* (2008). A64, C43

### Protein folding dynamics by time resolved SAXS and single molecule fluorescence spectroscopy

Satoshi Takahashi<sup>1,2</sup>, Tsuyoshi Konuma<sup>1</sup>, Yuji Goto<sup>1</sup>, Tetsuro Fujisawa<sup>3</sup>, Masahito Kinoshita<sup>1</sup>, Kiyoto Kamagata<sup>1,2</sup>

<sup>1</sup>Institute for Protein Research, Osaka University, Yamadaoka, 3-2, Suita, Osaka, 565-0871, Japan, <sup>2</sup>CREST, JST, Kawaguchi, Saitama, 332-0012, Japan, <sup>3</sup>Graduate School of Engineering, Gifu University, Gifu, 501-1193, Japan, E-mail: st@protein.osaka-u.ac.jp

To understand the mechanism of protein folding, the changes in protein compactness in protein folding were investigated by time-resolved small angle X-ray scattering (SAXS). The system is based on the continuous-flow mixing device and the beamline at

SPRING8 designed for X-ray scattering of biological macromolecules (BL45XU-SAXS). By using the device, we observed the folding dynamics of  $\beta$ -lactoglobulin, which forms a burst phase intermediate possessing a significant amount of non-native helices. We characterized that the intermediate is collapsed. Furthermore, the collapse is accelerated in the presence of trifluoroethanol, which increases the helical content of the intermediate. The results suggested that the non-native helix formation is coupled to the collapse in the folding of  $\beta$ -lactoglobulin. To observe the heterogeneity in protein folding, we next developed a new method to detect fluorescence from single molecules. Proteins labeled with a fluorescent dye were injected into a capillary at a low concentration for the observation of one molecule at a time. A laser was introduced into the capillary, and the fluorescence was imaged as traces on EMCCD camera. Thus, the traces reflect the time-dependent changes of the fluorescence signals from single proteins. We monitored the dynamics of yeast iso-1-cytochrome c labeled by Alexa532 at the single molecule level. By analyzing the autocorrelation function of the traces, we could detect characteristic time constants for the fluctuating motions of the sample.

Keywords: small angle X-ray scattering, single molecule fluorescence spectroscopy, protein folding

## MS.20.2

*Acta Cryst.* (2008). A64, C43

### XPCS studies of slow, non-diffusive dynamics in glassy soft materials

Robert L Leheny

Johns Hopkins University, Physics & Astronomy, 3400 N. Charles St., Baltimore, MD, 21218, USA, E-mail: leheny@pha.jhu.edu

For many disordered soft solids such as gels and concentrated emulsions, the solid phase is separated from a fluid state by an ergodic-nonergodic transition that leaves the system in an out-of-equilibrium configuration. Often these out-of-equilibrium materials display a protracted evolution of their dynamical properties that bears resemblance to behavior seen in molecular and polymer glasses. In order to understand these dynamics at a microscopic scale we have investigated such disordered soft solids through x-ray photon correlation spectroscopy (XPCS), a technique uniquely suited for elucidating the nanoscale motions in glassy materials. After a brief introduction to XPCS, the talk will describe our findings from studies on a set of materials that includes clay suspensions, sheared depletion gels, and concentrated nanoemulsions [1-3]. In short, the dynamics we observe are distinct from the cooperative molecular diffusion in liquids near the glass transition. Rather, they are a type of exceedingly slow, non-diffusive motion that can be modeled as strain in response to heterogeneous local stress and that are apparently universal to a range of disordered materials.

[1] R. Bandyopadhyay, D. Liang, H. Yardimci, D. A. Sessoms, M. A. Borthwick, S. G. J. Mochrie, J. L. Harden and R. L. Leheny, *Phys. Rev. Lett.* 93, 228302 (2004).

[2] H. Guo, J. N. Wilking, D. Liang, T. G. Mason, J. L. Harden, and R. L. Leheny, *Phys. Rev. E*, 75, 041401 (2007).

[3] B. Chung, S. Ramakrishnan, R. Bandyopadhyay, D. Liang, C. F. Zukoski, J. L. Harden, and R. L. Leheny, *Phys. Rev. Lett.* 96, 228301 (2006).

Keywords: XPCS, soft glassy materials, stress relaxation