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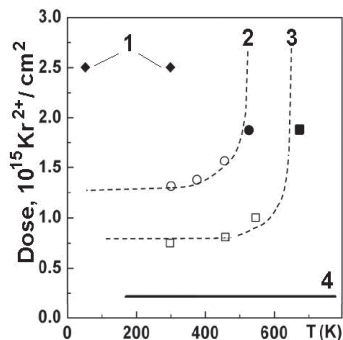
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Radiation induced disordering in crystalline actinide hosts

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Incorporation of transuranic actinides into durable crystalline phases is most appropriate way for management with long-lived nuclear wastes. Decay of the isotopes leads to lattice disordering (amorphization) and influences their properties. Optimal route for determination of stability of waste forms during irradiation is their bombardment by heavy ions. It allows to obtain critical doses (D_c) and temperatures (T_c) of crystalline actinide host phases. We have discussed new data on the behavior of oxides with pyrochlore- and garnet-type structure during irradiation by 1 MeV Kr^{2+} . Damages of the phases were observed in situ by TEM method and HRTEM after runs. Strong dependence between phases composition and T_c (D_c) values were revealed for the pyrochlore-structure compounds, while various garnet-type hosts have a close characteristics. These data are compared with the results obtained in experiments with Cm-doped phases of same compositions [1,2]. The results are used for choice of the actinide hosts with enhanced radiation stability.

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Critical dose vs temperature at pyrochlore irradiation by 1 MeV Kr^{2+} . 1 – $A_2(Zr_{1.93}U_{0.07})O_7$, 2 – $A_2(SnZr_{0.93}U_{0.07})O_7$, 3 – $A_2(Sn_{1.93}U_{0.07})O_7$, 4 – $Gd_2Ti_2O_7$, “A” = 0.26La + 0.51Ce + 0.24Pr + 0.79Nd + 0.14Sm + 0.04Eu + 0.03Gd. Open symbols – amorphous, filled – crystalline.

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Structure and vibrations in disordered systems

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It is largely accepted that disordered systems are characterized by a short range order usually very similar to that of the corresponding crystalline phase at the same density. It is less clear to what extent dynamic properties of disordered systems and crystals can be compared. In particular, high-frequency collective excitations reminiscent of phonons in solids exist as well in disordered systems. They are traditionally discussed in terms of relaxation processes characteristic of the disordered state [1].

We discuss here a quantitative comparison of the collective excitations in liquid and polycrystalline sodium [2]. We show that liquid sodium exhibits acoustic excitations of both longitudinal and transverse polarization at frequencies strictly related to those of the

corresponding crystal. The only relevant difference between the liquid and the polycrystal appears in the broadening of the excitations: an additional disorder-induced contribution comes into play in the case of the liquid, which we show to be related to the distribution of local structures around the average one. This is then the microscopic regime for the dynamics of a disordered system, and it corresponds to the lengthscale characteristic of the short range order.

On the opposite side of the lengthscale, in the macroscopic regime, the dynamic properties of a disordered system can be described treating it as an isotropic continuum. This regime is well known in many details [1]. It is clear that the macroscopic and microscopic regimes must be separated by an intermediate regime: the mesoscopic one. We present here an experimental [3] and numerical [4] study of this mesoscopic regime looking at the acoustic dynamics of glasses. We study in particular how, on decreasing the acoustic wavelength, the continuum, Debye approximation for the acoustic dynamics breaks down. In crystals, this takes place when the wavelength approaches the interatomic distance. We show here that in glasses this instead takes place on the larger lengthscale characteristic of the medium range order. Specifically, we find that the acoustic excitations with nanometer wavelengths show the clear signature of being strongly scattered. This crossover region is accompanied by a characteristic softening of the acoustic excitations that marks the breakdown of the Debye approximation.

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Keywords: disordered systems, x-ray scattering, vibrational properties

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Structure and dynamics of hydrogen bonded systems from inelastic scattering

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We have combined neutron scattering and a range of numerical simulations, including phonon calculations and molecular dynamics, to study hydrogen bonds in condensed matter. Two examples from recent and current theses will be presented. The first concerns proton transfer with increasing temperature in short inter-molecular hydrogen bonds. These bonds have unique physical and chemical properties and are thought to play a fundamental role in processes like enzymatic catalysis. By combining elastic and inelastic neutron scattering results with *ab initio*, lattice dynamics and molecular dynamics simulations, low frequency lattice modes are identified which modulate the potential energy surface of the hydrogen bond proton and drive proton transfer. New experimental data confirm and give further insight into the mechanism of proton transfer.

The second example concerns phonons of DNA. In particular acoustic phonons give direct access to the elastic properties of DNA which are important for its interaction with proteins and more generally its function. Inelastic X-ray and neutron scattering measure different modes relating to the interaction between base-pairs and the way this is modulated by the helix structure. The experimental data is interpreted with the aid of large scale, all-atom, phonon calculations. The combination of simulations and inelastic scattering data give insight into the stiffness of DNA and its structural integrity on different length scales.

Keywords: molecular_crystal, DNA, molecular_dynamics