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**Pressure-induced amorphizations of silica analogues: a probe of the
relationship between order and disorder.**

Final Report:

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Final Report

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The purpose of my high pressure investigations is to determine the relationship between order and disorder in amorphous materials using high pressure techniques. Specifically, the experimental techniques used are high pressure x-ray diffraction, electron transmission microscopy, and Raman scattering. This grant has resulted in twelve (12) publications plus numerous conference proceedings.

There are several principal results of the work : (1) We have shown that the structural ordering in pressure-amorphized solids can be described as defects in curved-space.² (2) For CaSiO_3 -based minerals the pressure-induced amorphizations occur because of an impeded phase transition and the amorphous phase may be viewed as a defective long-period modulated phase.³ (3) We discovered a new phase of GaPO_4 , which was recently predicted using a molecular dynamics calculation.⁴ This phase transition occurs at 15.9 GPa. This novel phase contains tetrahedral and octahedral coordination shells of oxygen atoms around the phosphorus and gallium atoms respectively. (4) We have shown that pressure-amorphized silica is amorphous at the molecular level.¹ (5) Using Raman scattering and energy-dispersive X-ray diffraction we studied a series of sulfate and chromate salts. These measurements reveal that the sulfate and chromate tetrahedra in the monopotassium salts become disordered, while those in the tripotassium salts stay ordered. This disordering occurs without a significant change in the structural arrangement of the cations. We suggested that the disordering results from an impeded ferroelastic phase transition, which causes

the zone boundaries, and therefore the tetrahedra, to be distorted.⁵ (6) Energy-dispersive and angle-dispersive X-ray-diffraction (EDXD and ADXD) measurements show that when compressed by pressures above 10.0 GPa isostructural lead, barium, and strontium nitrate crystal transform reversibly to highly disordered structures. Raman measurements under compression suggest that the transformation occurs via reorientation of the nitrate ions to multiple sites and movement of the metal ions from the crystal positions. Our main conclusion is that EDXD and ADXD measurements give different meanings to the notion "X-ray amorphous". Thus, whenever this phrase is used, the method used and the conditions that prevail must be specified. We also suggest that our results show that Raman spectroscopy can be used to distinguish nonrandom nitrate and cation environments in metal nitrate glasses.⁶

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