

Condensed matter physics under pressure

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Some recent high pressure physics work at BARC is described. Both experimental and theoretical aspects are covered.

The use of pressure as a thermodynamic variable for studying condensed matter has become very important in recent years. Its main effect is to reduce the volume of a substance. Thus, in some sense, it mimics the phenomena taking place during the cohesion of solids (e.g. pressure ionization, modifications in electronic properties and phase changes, etc.). High pressures are encountered from deep down the earth to astrophysical objects. Thus, the study of pressure effect on materials helps to unravel the mysteries of the constitution of these planetary bodies. Some of the phase changes under pressure help to synthesize new materials. The recent discovery of high T_c superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_7$ may be indirectly attributed to the pressure effect. In applied fields like simulations of a reactor accident, design of inertial confinement fusion schemes and for understanding the rock mechanical effects of shock propagation in earth due to underground nuclear explosions, the pressure versus volume relations of condensed matter are a vital input.

In the High Pressure Physics Division of BARC, we have been pursuing materials research under pressure for the past two and a half decades. During this period, we have developed a good expertise in the area of experimental production of both static and shock pressures and also in the area of theoretical investigations. Later include capabilities of analysing/predicting pressure-induced phase transitions, frozen phonon calculations to reveal the mechanisms of phase transitions, generation of equations of state from the first principles band structure calculations and molecular dynamics simulations and also the numerical simulation of the effects of shock wave propagation in materials.

For static pressure work, various kinds of diamond anvil cells have been fabricated indigenously. Facilities for the collection of angle dispersive (ADXRD) and energy dispersive powder diffraction (EDXRD) patterns from samples loaded in diamond anvil cells employing a rotating anode X-ray generator and ruby fluorescence technique for pressure measurement have been set up¹. An EDXRD station has also been built on a beam line of the Russian synchrotron, VEPP-3 at Novosibirsk for

performing high pressure studies². The diamond anvil cells have also been adopted for Raman studies³. The measurement of electrical resistance under pressure employing Bridgmann anvil technique is also used for identifying phase transitions.

For generating shock waves, we have set-up a light gas gun facility in our laboratory⁴. This gun, which is the only one existing in our country, is capable of accelerating projectiles to velocities of up to 1 km s^{-1} , which on impacting a target generate shock pressures up to 40 GPa. Instruments for measuring projectile velocity and for recording time-resolved stress wave histories in the shock loaded samples have been developed. Recovery capsules have also been made to retrieve shocked samples, which are then analysed using microscopic techniques like XRD, Raman, electron microscopy, etc.

Recently, we have used this gun to study phase transitions in GeO_2 , Ti and Zr. Polycrystalline samples of the α -quartz phase of GeO_2 (q- GeO_2), recovered after peak shock treatment of 5, 6.8 and 10 GPa have shown that q- GeO_2 irreversibly amorphizes above 6.8 GPa. The estimates of the shear band temperatures suggest that the mechanism of shock-induced amorphization in q- GeO_2 is solid-solid one and not the melt quenched one⁵. In Ti and Zr, our objective has been to understand the causes of the reported scatter in the pressure of α -w transition (6.0–11.9 GPa) under shock conditions. Our experiments show that the initial oxygen content of the sample has a large influence on the transition pressure (e.g. in Zr, α -w transition up to 11 GPa is seen only for oxygen content less than 1600 ppm). Unlike that in static experiments, the effect of shear is found to be small up to 9 GPa in inclined impact experiments in Ti.

The phenomenon of crystalline materials becoming amorphous under compression is only a decade old and presently an area of active research worldwide⁶. We have carried out extensive experimental and theoretical investigations to understand this. The discovery of LiKSO_4 turning amorphous under a hydrostatic pressure of about 13 GPa was amongst the first few in this area⁷. Subsequently, we have found through our diamond anvil cell based X-ray diffraction studies that AlPO_4 , $\text{Ni}(\text{OH})_2$, GeO_2 and LiCsSO_4 also become amorphous under pressure. Further, though AlPO_4 and GeO_2 are isomorphic to quartz SiO_2 , these do not exhibit the

precursor transformation recently observed in α -quartz⁸. Organic compound resorcinol has been shown to amorphize at 4 GPa through Raman scattering studies³. Using the stereo-chemical picture of the solids, we have inferred that the basic driving mechanism of this transformation is steric hindrance coupled with retarded kinetics beyond the limiting compression for a van der Waals contact⁹. We have also carried out extensive molecular dynamical studies to elucidate the nature of structural changes on amorphization¹⁰. Using the results of these simulations, we have shown that the recently observed new phase, preceding amorphization, in α -quartz to be a monoclinic phase and consistent with our proposal of a zone boundary mode softening¹¹. In addition, we find that the pressure induced glass in α -quartz is anisotropic and on release of pressure it is densified with a Si-O coordination number of 4.6. This phase can be brought back to the four coordinated state only on heating beyond 1100 K.

Quantum mechanical electronic structure calculations can be performed accurately, now-a-days, specially for the ground state properties within the local density approximation (LDA). We use these first principles calculations for estimating the structural stability of metals and their compounds, for carrying out frozen phonon calculations and to find the path of a phase transition¹². Recently, we have investigated the high pressure phases of many elements¹³. Excellent agreement with the experimental observations has been obtained for the transition pressures, structural parameters and equations of state. The theoretical total energy calculations have also been recently used by us to predict some new phase transitions¹⁴, which have been detected experimentally elsewhere. We also use BARC parallel processing system to perform such *ab initio* calculations on large systems (e.g. Mg₃₂ Zn₄₉)¹⁵. It is now possible to extend these first principles calculations to investigate the mechanical and defect properties of materials.

The plan for future studies includes efforts to increase the upper pressure limit both in static and dynamic experiments. The experimental set up to measure the optical absorption/reflectivity from compressed samples will be completed shortly. An imaging plate system for X-ray diffraction has been acquired to improve the reliability of diffraction results under pressure. A diamond

anvil cell with Be supports for single crystal X-ray diffraction studies is also under fabrication. Problems related to the study of kinetics of phase transitions under pressure and various amorphization studies are planned.

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