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In this work ab-initio calculations have been used to explain and predict structural, electronic, mechanical, vibrational and thermodynamical properties of the transition metal and rare-earth nitrides. It has been shown that the computer simulations with ab-initio accuracy are a perfect tool for testing structures and measuring properties on them that can directly be compared to experiments.

Starting out from the total energy and electronic structure calculation using the first principles plane wave pseudo potential implemented in Quantum ESPRESSO (PWSCF) and ABINIT packages, we have comprehensively studied the structural, electronic and dynamical properties of platinum nitrides in possible three structures are racksalt, zincblende, fluorite and pyrite. The calculated total energy shows that the platinum nitride is stabilized in pyrite structure and named as platinum perniride defined by nitrogen dimmers as N\(_2^{2-}\) or N\(_2^{4-}\) units. We notice during our studies on this material, is that the phonon frequencies and its pressure dependence in the zincblende and racksalt structure do not compare well with the Raman spectroscopic data and hence the platinum nitride in these structures are ruled out. The lattice constant, bulk modulus, electronic band structure, phonons at ambient and high pressure, which have been calculated in racksalt, zincblende and pyrite structures agree best with experimental data only for pyrite structure. For pyrite structured platinum nitride (PtN\(_2\)), the calculated lattice constant agree well for both LDA and GGA exchange correlation functional implemented in both packages, while the bulk modulus agrees exceptionally well with experimental value only in the case of LDA. The bulk modulus calculated in the present calculation using GGA is better than earlier calculations in the same group. The lattice dynamical calculations reveal that the pyrite PtN\(_2\) is dynamically stable. Our study further showed that the PWSCF predicts better the
frequencies of the characteristic mode $E_g$ and higher modes $T_g$ and $A_g$. The ABINIT predicts the higher Raman active phonon modes reasonably well but fails in predicting the frequency of characteristic mode. The pressure variation of Raman active phonon modes is in good agreement with experiment up to 40GPa. Electronic band structure calculation indicates this compound to be semiconducting. Finally, such reinvestigation is very much useful to understand the observed data of platinum pernitride and of further help to synthesize other noble metal nitrides.

Using phonon density of states, we have determined thermal properties such as Helmholtz free energy, internal energy, entropy and constant volume specific heat for HfN and ZrN at high pressure and temperature in the framework of density functional theory. The calculated phonon densities of states at both ambient and high pressures are in reasonably qualitative agreement with experimental Raman spectra measured at various pressures up to 32GPa. The red shift is observed with the variation of hydrostatic pressure within the optical frequency region. The thermodynamical functions behave normally up to the considered pressures.

We systematically investigated the electronic band structure and phonon properties of NbN and MoN in rocksalt phases. The main results from the band structure are (i) the NbN and MoN shows metallic nature in rocksalt phase. (ii) band structure and electronic density of states deduced that the $2p$-N and $4d$-TM states in both rocksalt NbN and MoN structure hybridized strongly, and hybridization increases with increase in atomic number. Phonon dispersion calculations show that the optimized structure of both compounds is dynamically instable because of imaginary frequencies at X and W-points of the Brillouin zone. However, the increase of smearing value $\sigma$ results in to the finite temperature effect and increases the frequency of the soft phonon modes at X and W-points and instabilities disappear. Thus we could conclude that a finite temperature
lead to the desired stabilization effect in the phonons of rocksalt NbN and MoN. We observe from phonon calculations at ambient pressure the rocksalt phase cannot exist as a ground state phase. The increase in acoustic phonon frequencies is due to the lowering of Fermi energy resulting from the increase of smearing value $\sigma$.

We have also characterised the cadmium nitride (CdN) using first principles calculations in the framework of density functional theory. For this, CdN has been studied in three different crystal structures: rocksalt, NiAs and CuS ($B_{18}$). We found that the rocksalt phase is energetically and dynamically more stable than the other two crystal structures. The phonon calculations show that that the CdN has positive phonon frequencies throughout the Brillouin zone and hence indicates dynamical stability for CdN in rocksalt phase. Finally, we have calculated the thermal properties like Debye temperature ($\theta_D$) and specific heat at constant volume ($C_v$) using quasi harmonic approximation. The simulated Debye temperature has been found in quite good agreement with the earlier theoretical and experimental findings.

The pressure dependent structural, electronic and phonon properties for PdN$_2$ in pyrite crystal structure have been also investigated in the present thesis using first principles calculations. The calculated properties have been reproduced accurately and consistently with available experimental data. The electronic phase transition from insulator to metallic state, multivalue EOS, drastic change in Bond length of Pd-N and N-N, and dynamical instability as well as anomalous behavior of zone centre phonon frequencies at 11GPa clearly suggest that PdN$_2$ pyrite crystal structure decomposes at 11GPa sharply. The mechanical and dynamical stability clearly shows the PdN$_2$ pyrite crystal structure can be recovered in high crystalline form at ambient pressure.

The non-spin and spin polarized calculations of total energy, electronic band structure and lattice dynamical properties of FeN$_2$ have been performed under the framework of
density functional theory to confirm structural stability of FeN$_2$. The electronic band structure and density of states suggest a metallic nature and hint the role of electrons near the Fermi level in establishing the directional bonding between Fe-d and N-p states and electronic stability for SP-FeN$_2$. A spin polarised FeN$_2$ hexagonal $Rar{3}m$ structure possessing nitrogen dimmers gives a dynamically stable structure and a high bulk modulus. The lattice dynamical calculations show that the frequencies of phonon modes are positive throughout the Brillouin zone for FeN$_2$, when the magnetic moment is switched on and makes SP-FeN$_2$ dynamically stable under ambient conditions. The total and partial phonon densities of states are also calculated and the role of each species in the total phonon density of states of FeN$_2$ is analyzed. The total phonon density of states along with the partial phonon density of states would be a working guidance to the future neutron scattering or any similar study on the phonon dynamics of FeN$_2$. A group theoretical analysis is also presented for vibrational modes of FeN$_2$ structure, which shows that the $E_g$ and $A_{1g}$ are Raman active and mainly due to the N-N dimmer vibrations only. The calculated temperature dependent lattice specific heat and Debye temperature do not show any anomalous behaviour across the studied temperature range.

Using an ab-initio pseudopotential method within local density approximation and dual-space Gaussian pseudopotential (HGH) of the density functional theory, we have calculated the structural, electronic, elastic and phonon properties of lanthanum nitride (LaN) in the rocksalt and high pressure CsCl structures. The calculated results for lattice constant, bulk modulus and elastic properties compare well with the available experimental and other theoretical calculations. The present band structure calculation suggests that the LaN is semi-metallic in its stable phase in contrast to many other rocksalt structured phase which are metallic. The LDA calculations give stable phonons
in complete Brillouin zone for both zero and high pressure phases. The HGH pseudopotential approximation seems more successful in predicting the elastic, structural and phase transition pressure while LDA gives the complete dynamical stability. In conclusion, the present calculations clearly reveal that the NaCl phase is more probable for LaN.

The first-principles plane wave self consistent calculations on structural, electronic and vibrational properties for lanthanum nitride in ThC₂ and pyrite phases have also been performed in present thesis. According to our calculations, the lanthanum pernitride (in pyrite phase) contains nitrogen dimers and shows high mechanical strength reflected from the higher bulk modulus than their constituent elements. The total energy calculations show that the lanthanum nitride in pyrite phase with La₂N stoichiometry exhibits more favorable thermodynamic stability. The absence of nitrogen dimers decreases the bulk modulus and corresponding mechanical properties. The present results also indicate that the electronic spectra of both LaN₂ and La₂N exhibits non-zero value of the density of states at the Fermi level, i.e. both these materials have a metallic conductivity. Our analysis reveals that bonding in LaN₂ have a covalent metallic character while La₂N exhibits ionic bonding. The covalent bonding is formed due to hybridization effect of N–N states (for N₂ dumbbells) and N₂–La states in LaN₂ while La-La hybridization is responsible for the ionic character in La₂N phase. In addition, between the lanthanum atoms and N₂ dumbbells, ionic bonds emerge owing to the charge transfer from La to N₂ Fermi delocalized La-La d states. Finally, the phonon calculations for both stoichiometric LaN₂ and La₂N phases reveal that the La₂N phase is dynamically unstable while La₃N shows positive phonon modes throughout the Brillouin zone clearly indicating dynamical stability.
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Finally, based on our results, we have provided at the end of each chapter of present thesis work same hints that regard the perspective for further that this work has opened, which among others include: the possibility of synthesizing new superhard materials containing nitrogen dimmers. It is due to conclusions like the ones reported along this chapter, that we can now see clearly the importance of continuing such a fruitful interplay between theory and experiment in high pressure research, which we hope will continue to be present and in an increasingly strong way, in the years to come.