Dissociation Dynamics of Unstable Molecular Systems

A Thesis

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MOHANLAL SUKHADIA UNIVERSITY

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by

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DECLARATION

I, **Mr. Amrendra Kumar Pandey**, S/O Mr. Ram Bihari Pandey, resident of Room No:101, PRL Thaltej Hostel, Thaltej, Ahmedabad-380054, hereby declare that the work incorporated in the present thesis entitled, **"Dissociation Dynamics of Unstable Molecular Systems"** is my own and original. This work (in part or in full) has not been submitted to any University for the award of a Degree or a Diploma. I have properly acknowledged the material collected from secondary sources wherever required. I solely own the responsibility for the originality of the entire content.

Date : May 26, 2014

(Amrendra Kumar Pandey)

CERTIFICATE

I feel great pleasure in certifying that the thesis entitled, **"Dissociation Dynamics of Unstable Molecular Systems"** embodies a record of the results of investigations carried out by Mr. Amrendra Kumar Pandey under my guidance. He has completed the following requirements as per Ph.D. regulations of the University.

(a) Course work as per the university rules.
(b) Residential requirements of the university.
(c) Regularly submitted six monthly progress reports.
(c) Presented his work in the departmental committee.
(d) Published minimum of one research paper in a refereed research journal.

I am satisfied with the analysis of data, interpretation of results and conclusions drawn. I recommend the submission of thesis.

Date : May 26, 2014

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Abstract

A molecular system is generally viewed as a composite of two sub-systems of electrons and nuclei which are drastically different on the basis of their dynamical time scales. These two sub-systems, however are coupled by the Coulomb interactions. The assumptions and approximations based on the inherent differences in the dynamical properties of the electrons and nuclei are used to construct the picture of a molecule. The resulting model of the molecular system explain and predict many molecular processes very successfully. However, there are processes in which their predictions do fail. Dissociation processes of highly charged molecular systems provide a unique opportunity to examine and explore their nature in this regard. Such studies are employed as a probe to investigate the regime and conditions of the validity of assumptions and approximations that are otherwise remain valid in the molecular processes.

This thesis is concerned with experimental as well as theoretical investigations to explore some of the fragmentation properties of the doubly ionized molecular ions of N_2 and CO, which are isolectronic molecules with different symmetries but nearly identical total ionization cross-sections. By combining ab initio calculations with the experimental investigation of the kinematics of dissociative double ionization of these molecules, we have identified various transient states contributing to the dissociation process. Further we have quantified the BO and non-BO processes therein. It has emerged as an important result because of the striking differences between their observed proportions for these dications. The work reported in this thesis empasize the relevance of such comparative studies.

List of Publications

1. *Charge symmetric dissociation of doubly ionized N_2 and CO molecules*
   A. Pandey, B. Bapat, K.R. Shamasundar, The Journal of Chemical Physics, **140, 3** (2014)

2. *Effect of transmission losses on measured parameters in multi-ion coincidence momentum spectrometers*
   A. Pandey, and B. Bapat, International Journal of Mass Spectrometry, **361, 0** (2014)

3. *A comparative study of Dissociative Ionization of N_2 and CO*
   A. Pandey, B. Bapat, K.R. Shamasundar, Journal of Physics: Conference Series, **488, 5** (2014)

Acronyms and Abbreviations

AO	Atomic Orbital
CE	Coulomb Explosion
CI	Configuration Interaction
CSD	Charge Symmetric Dissociation
CAD	Charge Asymmetric Dissociation
CSF	Configuration State Function
DI	Dissociative Ionization
DLD	Delay Line Detector
FWHM	Full Width Half Maximum
GTO	Gaussian Type Orbital
HF	Hartree Fock
KER	Kinetic Energy Release
LCAO	Linear Combination of Atomic Orbitals
MCP	Multi Channel Plate
MCSCF	Multi Configuration Self-Consistent-Field
MO	Molecular Orbital
MRCI	Multi Reference Configuration Interaction
PEC	Potential Energy Curve
RIMS	Recoil Ion Momentum Spectrometer
SCF	Self Consistent Field
STO	Slater Type Orbital
TOF/ToF	Time Of Flight

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