#### Dynamical Jahn-Teller effect in the first excited Con

Zhishuo Huang<sup>3, a)</sup> and Dan Liu<sup>2,1,h)</sup>

<sup>1)</sup> Theory of Nanomaterials Group, KU Leuven, Celestijnenlaan 200F, B-3001 Leuven,

Designation.

<sup>2)</sup>Shaanzi Institute of Flexible Electronics, Northwestern Polytechnical University, 127 West Youyi Road, Xi'an,

710072, Shaann, China (Dated: 28 May 2019)

The Jahn-Teller problems of  $C_{00}$  anions involving  $t_{1g}$  next lowest unoccupied molecular orbital (NLUMO) were theoretically investigated. The orbital vibronic coupling parameters for the  $t_{1g}$  orbitals were derived from the Kohn-Sham orbital levels with hybrid B3LYP functional by the frozen phonon approach. With the use of these coupling parameters, the vibronic states of the first excited  $C_{00}$  were calculated, and were analyzed. The dynamical Jahn-Teller stabilization energy of the first excited  $C_{00}$  is stronger than that of the

ground electronic states, resulting in two times larger splitting of vibronic levels than those of the ground state  $C_{00}$ . The present coupling parameters prompt us to understand more about the excited  $C_{00}$ .

#### I. INTRODUCTION

Highly symmetric  $C_{00}$  exhibits complex Jahn-Teller dynamics characterized by orbital-vibration entanglement in various charged and excited states<sup>1–3</sup>. Among these states, negatively charged  $C_{60}$  is one of the most interesting cases because it often serves as a building brick of materials<sup>4–10</sup>. In order to comprehend thoroughly the role of the building brick, many properties of negatively charged  $C_{60}$  should be understood clearly, especially about JT effect involved properties. Though JT effect, including dynamic JT effect, of  $C_{60}$  anions have been intensively investigated<sup>11–34</sup>, it is only last years that the actual situation in the ground electronic states of  $C_{60}^{\infty}$  molecule (n = 1 - 5) has been established with accurate coupling parameters, which showed the importance of dynamic JT effect.<sup>32,53</sup>.

So far, the works about the dynamic JT effect in negatively charged  $C_{00}$  have been almost always in the ground electronic configuration populating only the lowest unoccupied molecular orbitals, which is the  $t_{1n}$  orbital. However, to our knowledge, neither the vibronic coupling parameters for excited electronic configuration, say t<sub>1s</sub> next lowest unoccupied molecular orbial (NLUMO), nor the relevant JT effect has been theoretically investigated much. While it is believed that the nature of excited Con anions involving the next lowest unoccupied molecular orbital is of fundamental importance to interpret absorption spectra of isolated  $C_{00}^{-72.28.35-41}$ , electron transfer process of fullerene  $^{62.43}$ , and excitation spectra of alkali-doped fullerides 64-36, and the JT effect involving the NLUMO must be significant in highly alkali doped 46 and alkali-earth/rage-earth doped fullerides 47-53. Furthermore, it might also be important 24 in recently reported light induced superconductivity of alkali-doped fullerides 53.56. Recently, bound excited states of C\_{60} have been theoretically investigated<sup>57-43</sup>, and the stability of the first excited  ${}^2T_{1g}$  states of  $C_{60}^-$  has been confirmed, nevertheless, the vibronic problem has not been investimated.

In this work, we address the dynamical JT effect of first excited  $C_{90}$  anison populating the  $t_{1g}$  NLUMO. The vibronic coupling parameters are derived from the data obtained by density functional theory (DFT) calculations with hybrid B3LYP exchange-correlation functional. Using these coupling parameters, the vibronic states are obtained by numerically diagonalizing the dynamical JT Hamiltonian matrix, and are analyzed.

#### II. JAHN-TELLER EFFECT

#### A. Model Hamiltonian

The  $t_{1g}$  next LUMO of neutral  $C_{60}$  with  $I_h$  symmetry is triply degenerate and separated from the other orbital levels<sup>1</sup>. According to the selection rule, the  $t_{1g}$  orbitals couple to totally symmetric  $a_g$  and five-fold degenerate  $h_g$  representation as in the case of  $t_{1g}$  orbitals<sup>62</sup>:

$$[t_{1g} \otimes t_{1g}] = a_g \oplus h_g.$$
 (1)

In this work, we take the equilibrium structure of  $C_{60}$  as the reference. Therefore, besides the  $h_g$  modes, the vibronic couplings to the  $a_g$  modes are nonzero. The linear vibronic Hamiltonian of  $C_{60}$  in the first excited electronic  $(t_{1g}^1)$  configuration resembles to that for the ground  $t_{1g}^1$  electronic configuration  $t_{1,1,10,62}^1$ .

$$H = H_a + H_h$$
, (2)

$$H_a = \frac{1}{2} (p_a^2 + \omega_a^2 q_a^2) + V_a q_a,$$
 (3)

$$H_h = \sum_{\gamma=\theta,\epsilon,\xi,\eta,\zeta} \frac{1}{2} \left(p_{h\gamma}^2 + \omega_h^2 q_{h\gamma}^2\right)$$

$$+V_{h}$$

$$\begin{pmatrix}
\frac{1}{2}q_{h,\theta} - \frac{\sqrt{2}}{2}q_{h,\eta} & \frac{\sqrt{2}}{2}q_{h,\eta} & \frac{\sqrt{2}}{2}q_{h,\eta} \\
\frac{\sqrt{2}}{2}q_{h,\eta} & \frac{1}{2}q_{h,\theta} + \frac{\sqrt{2}}{2}q_{h,\eta} & \frac{\sqrt{2}}{2}q_{h,\xi} \\
\frac{\sqrt{2}}{2}q_{h,\eta} & \frac{1}{2}q_{h,\theta} + \frac{\sqrt{2}}{2}q_{h,\xi} & -q_{h,\theta}
\end{pmatrix}$$
(4)

<sup>\*\*</sup>Electronic mail: phisbuolesang@gmail.com

b) Electronic mail: iamdlig@aveps.edu.cn

# **The Dynamical Jahn Teller Effect In Localized Systems**

Isaac B. Bersuker, Victor Z. Polinger

## The Dynamical Jahn Teller Effect In Localized Systems:

The Dynamical Jahn-Teller Effect in Localized Systems ÎUriĭ Evgen'evich Perlin, Max Wagner, 1984 Vibronic Interactions and the Jahn-Teller Effect Mihail Atanasov, Claude Daul, Philip L.W. Tregenna-Piggott, 2011-11-23 The concepts of the Jahn Teller effect and vibronic coupling are being applied to more and more systems in both chemistry and physics Aspects of structural chemistry such as the distortion of the nuclear framework to a lower symmetry conformation have received an increasing attention as well as the dynamics on the coupled potential energy surfaces The Jahn Teller intersections are now recognized as prototype cases of conical intersections where the nuclear motion is known to be inherently nonadiabatic in nature and interchanges freely between the different potential energy surfaces In the condensed phase especially the significance of the Jahn Teller effect has been increasingly appreciated following the discovery of superconductivity in the fullerides and of very large colossal magnetoresistance in the manganite perovskites Indeed these materials are particularly challenging since the Jahn Teller interaction competes with electronic correlation effects Vibronic Interactions and the Jahn Teller Effect Theory and Applications provides an in depth discussion of the Jahn Teller effect and vibronic interactions as reflected by the contributions presented at the XX International Conference on the Jahn Teller effect Fribourg Switzerland 2010 The following topics have been treated in a clear and concise way Complex topologies of Jahn Teller effect and conical intersections Multi state vibronic interactions on strongly coupled potential energy surfaces Interplay of vibronic and spin orbit coupling Strain in Jahn Teller systems and cooperative Jahn Teller effect Orbital ordering and its relation to ferromagnetism ferroelectricity and molecular magnets The Jahn Teller effect in icosahedral systems The Jahn Teller effect and high temperature superconductivity This book is of interest to a wide audience including academic and industrial theoretical and experimental physicists chemists spectroscopists and crystallographers The Theory of the Jahn-Teller Effect Arnout Ceulemans, 2022-09-29 This book provides a comprehensive discussion of the Jahn Teller Effect TTE focusing on the boson fermion interaction While current research is concerned with measuring and calculating ever more sophisticated and complex manifestations of the JT effect the present volume takes away the epicycles of the theory and focuses on the symmetry dilemma at its core When fermions and bosons meet they get entangled and form a new dynamic reality According to the rules of Molecular Symmetry this reality is limited to a small set of patterns with degeneracy cardinalities 2 3 4 5 and 6 The novelty of the book is that it adopts a unique mathematical technique known as the Bargmann Fock representation and treats all degeneracies in detail So far this method was only applied to the simplest doublet case therefore its extension to the entire range of cases offers a new unified perspective This volume will help the reader acquire a clear understanding of the JT effect discover its universal mechanism and it will be a great tool for researchers and graduates working on this topic Electron-phonon Dynamics And Jahn-teller Effect - Proceedings Of The Xiv International Symposium N Terzi, G Bevilacqua, L Martinelli, 1999-08-31 The Jahn Teller effect is a consequence of the

electron phonon coupling in high symmetry systems Its influence covers a wide range of physical and chemical properties and systems As the biannual Jahn Teller symposia bring together experimental and theoretical physicists and chemists from all over the world this proceedings volume reports the latest scientific news on the effect The contents of the volume range from the general aspects to some special topics such as ultrafast processes fullerenes point defects cooperative phenomena HTSC and oxide properties Some contributions are dedicated to the memory of Mary O Brien a globally honored specialist in the theory of the Jahn Teller effect Throughout personal reminiscences of O Brien's enormous contributions to the subject the recent history of the effect is summarized The Jahn-Teller Effect Horst Köppel, David R. Yarkony, Heinz Barentzen, 2009-12-08 The Jahn Teller effect continues to be a paradigm for structural instabilities and molecular dynamical processes This volume provides a survey of the current Jahn Teller interactions at the interface of quantum chemistry and condensed matter physics Vibronic Interactions in Molecules and Crystals Isaac B. Bersuker, Victor Z. Polinger, 2012-12-06 Vibronic interaction effects constitute a new field of investigation in the physics and chemistry of molecules and crystals that combines all the phenomena and laws originating from the mixing of different electronic states by nuclear displacements This field is based on a new concept which goes beyond the separate descriptions of electronic and nuclear motions in the adiabatic approximation Publications on this topic often appear under the title of the lahn Thller effect although the area of application of the new approach is much wider the term vibronic interaction seems to be more appropriate to the field as a whole The present understanding of the subject was reached only recently during the last quarter of a century As a result of intensive development of the theory and experiment it was shown that the nonadiabatic mixing of close in energy electronic states under nuclear displacements and the back influence of the modified electronic structure on the nuclear dynamics result in a series of new effects in the properties of molecules and crystals The applications of the theory of vibronic in of spectroscopy including visible ultraviolet in teractions cover the full range frared Raman EPR NMR nuclear quadrupole resonance NQR nuclear gam ma resonance NOR photoelectron and x ray spectroscopy polarizability and magnetic susceptibility scattering phenomena ideal and impurity crystal physics and chemistry including structural as well as ferroelectric phase transitions stereochemistry and instability of molecular including biological systems mechanisms of chemical reactions and catalysis Cooperative Phenomena in Jahn—Teller Crystals Michael D. Kaplan, Benjamin G. Vekhter, 2012-12-06 This book by Kaplan and Vekhter brings together the molecular world of the chemist with the condensed matter world of the physicist Prior to the collapse of the Soviet Union chemists in the West devoted lit to relationships between molecular electronic structure and tle attention solid state vibronic phenomena Treating quantum mechanical problems wherein the adiabatic Born Oppenheimer approximation fails was done by brute force With bigger and better computers available in the West molecular orbital calculations were done on observed and conceived static structures with little concern for any cooperativity of vibrational behavior that might connect these states While it had long been

understood in the West that situations do occur in which different static structures are found for molecules that have identical or nearly identical electronic structures little attention had been paid to understanding the vibrational states that could connect such structures It was easier to calculate the electronic structure observed with several possible distortions than to focus on ways to couple electronic and vibrational behavior In the former Soviet Union computational power was not as accessible as in the West Much greater attention therefore was devoted to conserving computational time by considering fundamental ways to han dle the vibrational connectivity between degenerate or nearly degenerate electronic states Jahn-Teller Effect in C60 and Other Icosahedral Complexes C. C. Chancey, M. C.M. O'Brien, 2021-02-09 Because of the high symmetry involved the Jahn Teller effect is the natural starting point for considering electron phonon or vibronic interactions in icosahedral molecules This work is the first comprehensive theoretical analysis of the Jahn Teller interaction in C60 and other icosahedral complexes The importance of this research derives in part from the increasing widespread interest in C60 and other molecular clusters and their application in science and industry The electrical and spectroscopic properties of fullerene and fulleride compounds depend intimately on the coupling between the electronic and vibrational modes of these systems and this book addresses the fundamental theoretical questions In particular a chapter is devoted to the connection between the theory and experimental observations such as ESR electron spin resonance effects and molecular spectra Earlier books have discussed the theory of Jahn Teller interactions in lower symmetry structures cubic tetrahedral tetragonal trigonal this is the first that focuses on the new icosahedral systems whose most famous example is Buckminsterfullerene C60 The book s authors have over fifty years of combined research experience into the theoretical aspects of the Jahn Teller Charge Density Waves in Solids L.P. Gor'kov, G. Grüner, 2012-12-02 The latest addition to this series covers a effect field which is commonly referred to as charge density wave dynamics. The most thoroughly investigated materials are inorganic linear chain compounds with highly anisotropic electronic properties. The volume opens with an examination of their structural properties and the essential features which allow charge density waves to develop The behaviour of the charge density waves where interesting phenomena are observed is treated both from a theoretical and an experimental standpoint The role of impurities in statics and dynamics is considered and an examination of the possible role of solitons in incommensurate charge density wave systems is given A number of ways to describe charge density waves theoretically using computer simulations as well as microscopical models are presented by a truely international board of authors

Solitons S.E. Trullinger, V.E. Zakharov, V.L. Pokrovsky, 2012-12-02 In the twenty years since Zabusky and Kruskal coined the term soliton this concept changed the outlook on certain types of nonlinear phenomena and found its way into all branches of physics The present volume deals with a great variety of applications of the new concept in condensed matter physics which is particularly reached in experimentally observable occurrences. The presentation is not centred around the mathematical aspects the emphasis is on the physical nature of the nonlinear phenomena occurring in particular situations.

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