2009己醜年

謹賀新年

大连化物所
2007-2008年1101组总结报告

韩克利

中科院大连化物所1101组

一、人员情况

- 院士：1人（何国钟研究员）
- 研究员：2人（韩克利、王利）
- 副研究员：4人 （徐大力、孙巨龙、尹鸿鸣、刘建勇）
- 助理研究员：2人（赵广久、王艳秋）
- 博士后3人，研究生36人

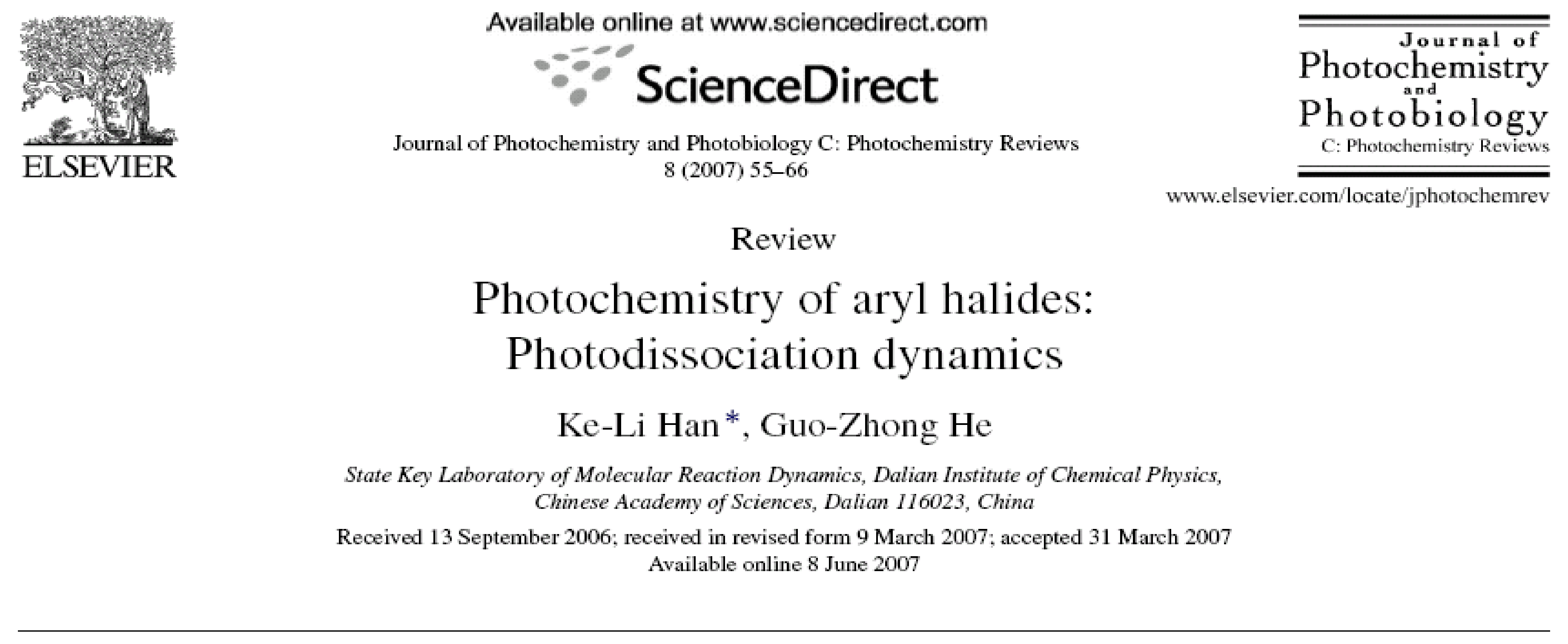
二、承担的项目

- 973项目
- 国家自然科学基金重点项目
- 国家自然科学基金创新群体
- 国家自然科学基金面上项目
- 分子反应动力学国家重点实验室项目

卤代芳烃的光解动力学

代表性工作之一

(IF=8.12)



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Journal of Photochemistry and Photobiology C: Photochemistry Reviews
8 (2007) 55–66

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Journal of Photochemistry and Photobiology
C: Photochemistry Reviews

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Review

Photochemistry of aryl halides: Photodissociation dynamics

Ke-Li Han*, Guo-Zhong He

State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China

Received 13 September 2006; received in revised form 9 March 2007; accepted 31 March 2007
Available online 8 June 2007

这篇综述文章总结了我们课题组在卤代芳烃光解动力学方面的系列研究工作，同时总结国际同行对我们这项工作的引用和评价.

2009己醜年

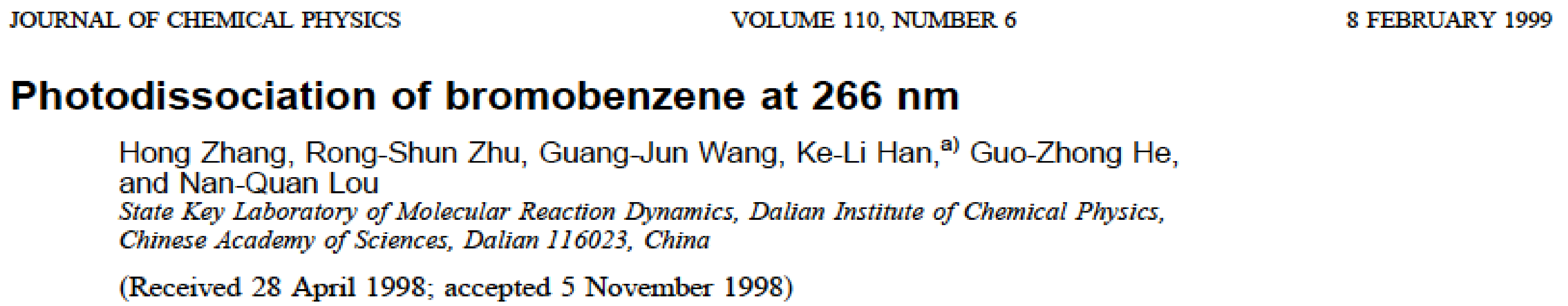
Ke-Li Han was born in 1963 in Shandong, China. He received his BS degree in Physics from Shandong University in 1983. He received his PhD degree in Physical Chemistry from Dalian Institute of Chemical Physics, Chinese Academy of Sciences in the early time of 1990. Then he was appointed as an Assistant Professor at the same Institute to study the reaction dynamics of alkaline atoms with aryl halides by using laser induced fluorescence in a beam-gas arrangement. He was a post-Doc at University of California at Davis to study the collision dynamics of C_{60} with electron and noble gas and also at Emory University to measure the rate constants of the $NO + NH_2$ reaction in 1993–1995. He was promoted to be an Associate Professor at Dalian Institute of Chemical Physics, Chinese Academy of Sciences, in 1993 and a Professor of Chemical Physics in 1995. In 1996, he started to study the photodissociation dynamics of aryl halide molecules by using translational photofragment spectroscopy on the universal crossed beam apparatus. His current interests lie in experimental and theoretical reaction dynamics including photodissociation and reactive dynamics in gas phase, ultrafast dynamics in solution phase, nonadiabatic reaction dynamics, and so on.

Guo-Zhong He was born in Nanhai, Guangdong Province in China in May, 1933. Enrolled in the Department of Chemical Engineering, Tsinghua University, Beijing, China in 1951, and graduated from Beijing Institute of Petroleum in 1955. He was the Vice Chairman of Scientific Committee of Dalian Institute of Chemical Physics (1992–1999) and the first Director of State Key Laboratory of Molecular Reaction Dynamics (1990–1996), and the Vice Chairman or Chairman of Scientific Committee of the State Key Laboratory of Molecular Reaction Dynamics (1996–2006) in China. Now he is the Professor. Since 1979, He spent about 6 years as a visiting scholar or visiting professor at the Department of Chemistry, University of California at Berkeley; the Department of Physics, the University of Kaiserslautern at Germany; the Department of Chemistry, the Iowa State University; the Department of Chemistry, University of Illinois at Chicago; Lawrence Berkeley Laboratory at USA; the University of Hong Kong and Hong Kong University of Science and Technology at Hong Kong. He studied basic properties of solid particle fluidization and fluidized coking of heavy oil, the combustion theory and experiments and the HF chemical laser in his early career. Since 1978, he began his study of the reaction dynamics by using molecular beam. Since 1981, he has published about 180 papers in scientific journals. He was elected as Member of the Chinese Academy of Sciences in 1991.

the photoexcitation of the benzene ring leads to a rich variety of reactions. Consequently, now the photochemistry of aryl halides has become a major area [11–49].

Experimentally, the photodissociation dynamics of aryl halides have widely been studied in the gas phase by photofragment translational spectroscopy (PTS) [50–65], in real time by ultrafast pump-probe spectroscopy combined with time-of-flight (TOF) [66–71], by time-resolved Fourier-transform spectroscopy [35,71,72], by resonance enhanced multiphoton ionization (REMPI) technique [73–76], and by multimass ion imaging techniques [36,48,49,77–80]. Fox example, Bersohn and coworkers [50,51] were firstly to study the photodissociation dynamics of the aryl halides at 193 and 248 nm. Their work indicated that the dissociation pathway depends on the competition between intersystem crossing and internal conversion, which is a function of both ring size and halogen substituent. Recently, photodissociation of chlorobenzene and *o*-, *m*-, *p*-chlorotoluene at 193 or 248 nm have been reported by Ichimura et al. [54–57]. They suggest that photodecomposition of the C–Cl bond in chlorobenzene or *o*-, *m*- and *p*-chlorotoluene may take place through three different channels, i.e., a direct dissociation channel, a channel via vibrationally excited triplet levels and a channel via highly excited vibrational levels of the ground electronic state. Molecular beam studies [52,53] also indicate that UV photodissociation of iodobenzene can take place via two different types of dissociation processes due to the presence of the π, π^* electronic system in the phenyl ring in addition to the n, σ^* electronic system in the C–I bond. Cheng et al. [66] recently performed femtosecond real-time photodissociation dynamical experiments of iodobenzene at 278 nm and directly determined the dissociation times of the two recoil velocity components of the I fragment. Kadi et al. [67–69] have measured the decay time constants of aryl halides in the excited state. Theoretically, quantum chemical calculations of potential energy surfaces have previously been performed for aryl halides [81–85]. All the studies mentioned above predict that photodissociation mechanism of aryl halides is very complicated because multiple dynamical channels are involved.

謹賀新年

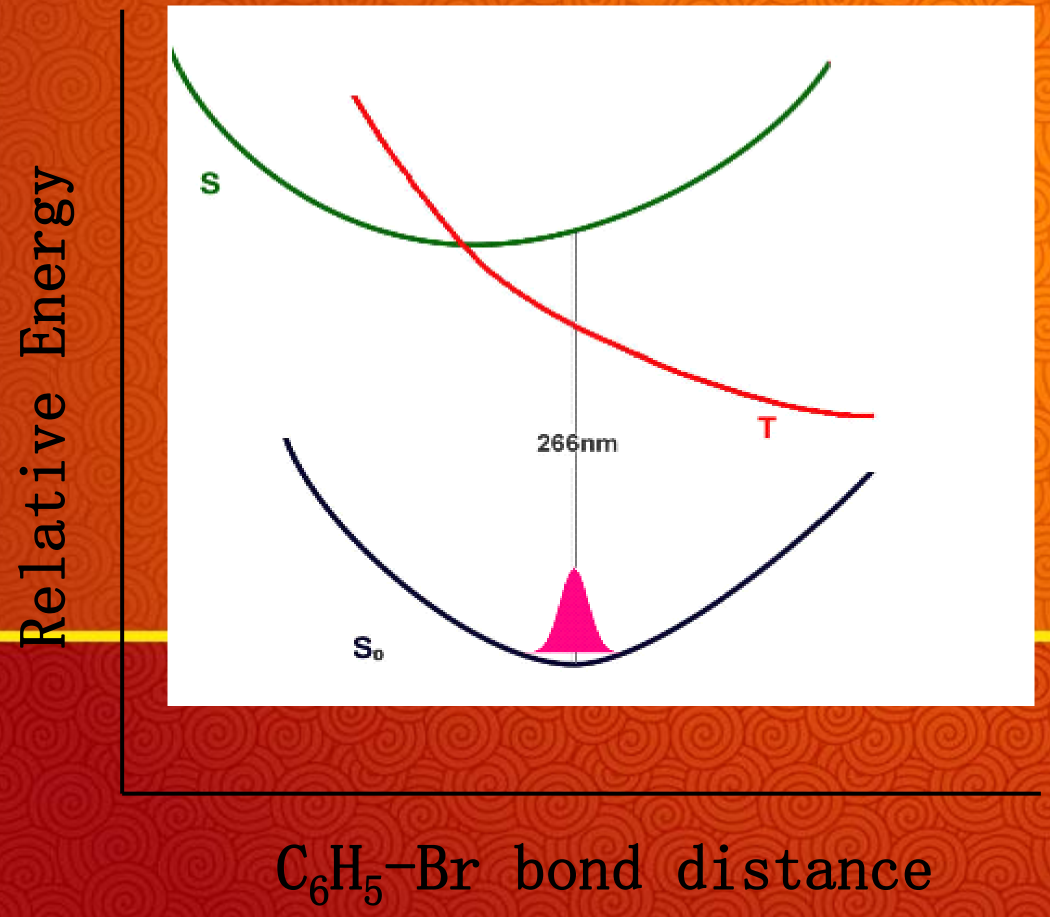


JOURNAL OF CHEMICAL PHYSICS VOLUME 110, NUMBER 6 8 FEBRUARY 1999

Photodissociation of bromobenzene at 266 nm

Hong Zhang, Rong-Shun Zhu, Guang-Jun Wang, Ke-Li Han,[a] Guo-Zhong He, and Nan-Quan Lou
State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China

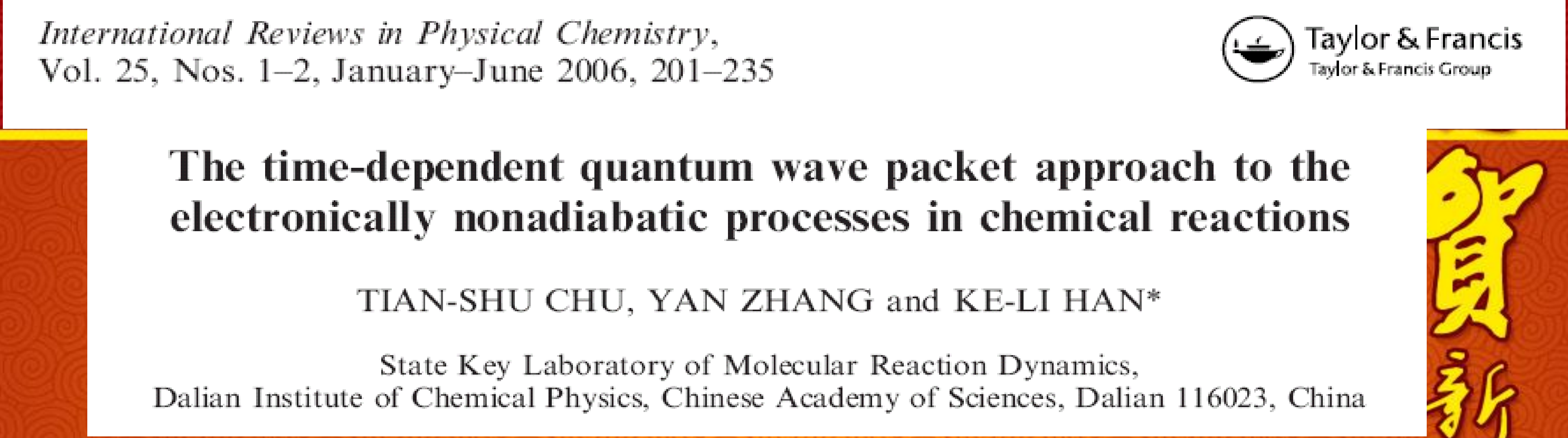
(Received 28 April 1998; accepted 5 November 1998)



根据实验和量子化学计算，首先建议了一个单重态和三重态交叉的光解机理，后来国际同行的理论和实验广泛引证这个机理。

代表工作之二：非绝热量子动力学

(IF=6.02)



International Reviews in Physical Chemistry,
Vol. 25, Nos. 1–2, January–June 2006, 201–235

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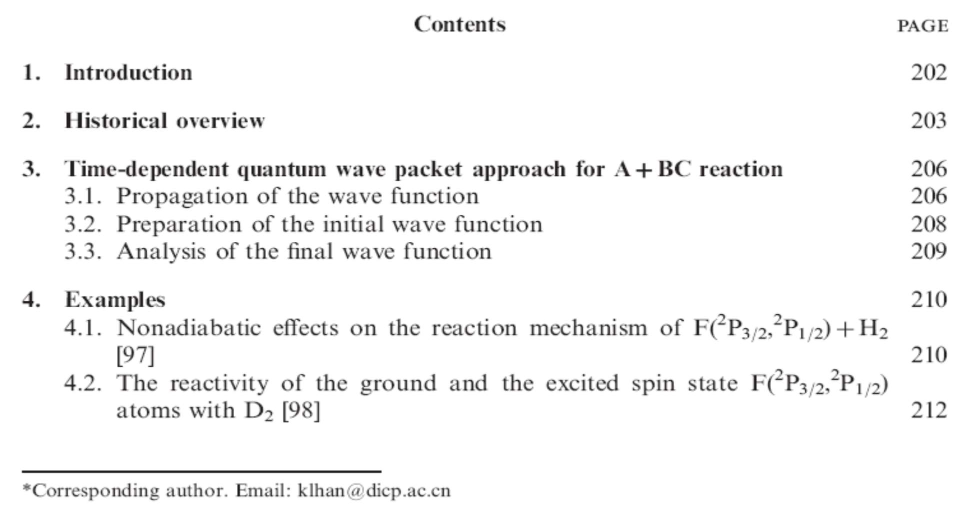
The time-dependent quantum wave packet approach to the electronically nonadiabatic processes in chemical reactions

TIAN-SHU CHU, YAN ZHANG and KE-LI HAN*

State Key Laboratory of Molecular Reaction Dynamics,
Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China



Contents

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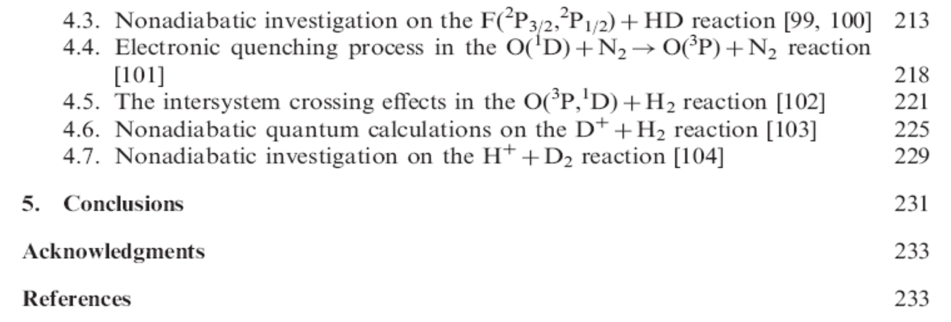
*Corresponding author. Email: klhan@dicp.ac.cn



Dear Dr Han,

I am pleased to let you know that your article The time-dependent quantum wave packet approach to the electronic ally nonadiabatic processes in chemical reactions, which appeared in Volume 25 issue 1-2 of our journal International Reviews in Physical Chemistry, was one of the most accessed articles of the journal in 2006. I have set up a link in the 'aims and scope' section of the journal website www.informaworld.com/trpc to the full list of the top 10 most accessed articles, in which your article features.

I do hope that this evidence of appreciation for your work will encourage you to submit more articles to the journal in the future.

Best Regards,

Anna

Anna West - Publishing Editor, Physical Sciences

Taylor & Francis

4 Park Square, Milton Park, Abingdon, OX14 4RN

phone: 0207 017 7474

email: anna.west@tandf.co.uk

2009己醜年

謹賀

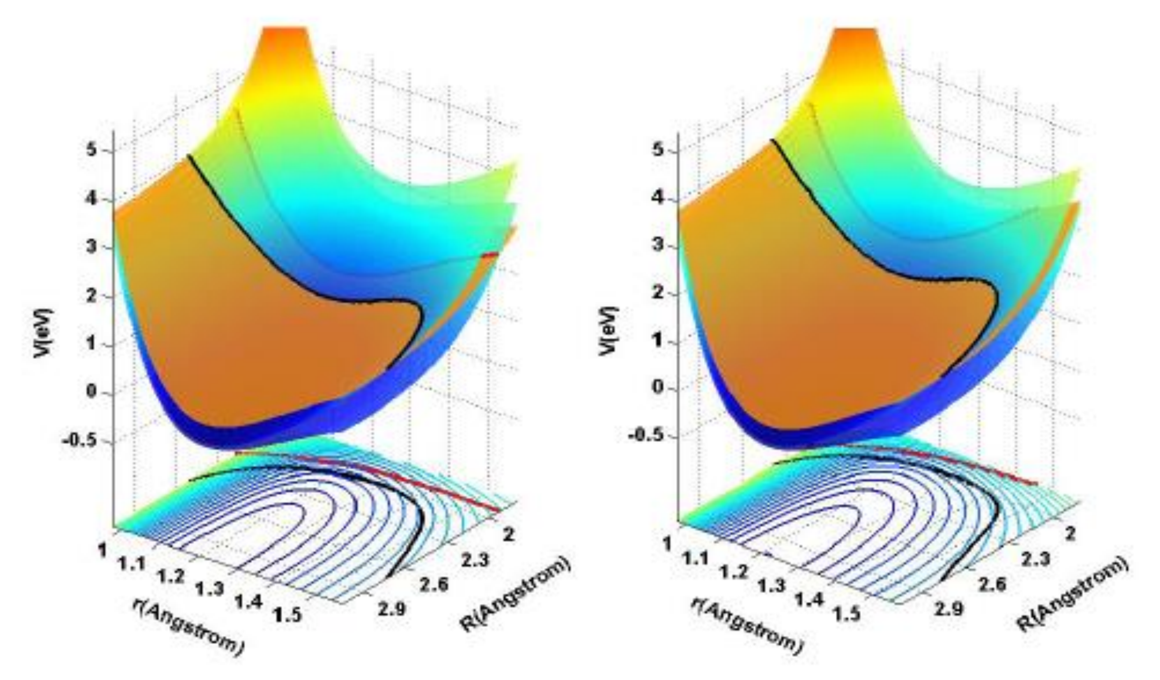
Chu TS, Zhang Y, Han KL

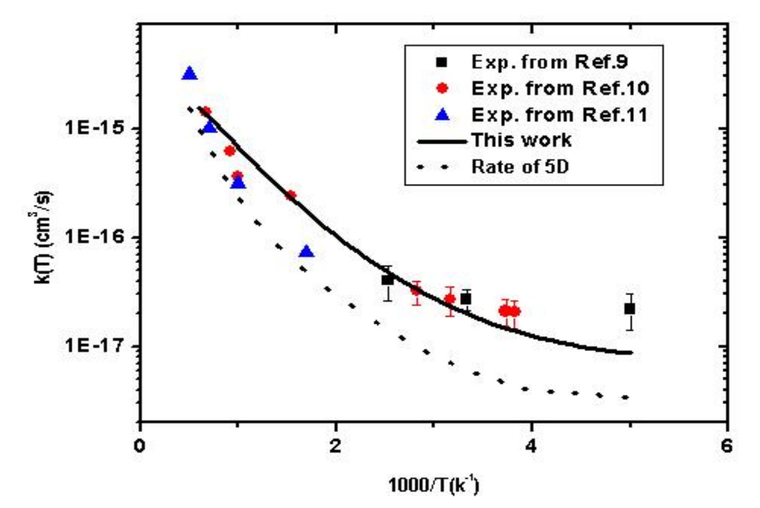
The time-dependent quantum wave packet approach to the electronically nonadiabatic processes in chemical reactions

INTERNATIONAL REVIEWS IN PHYSICAL CHEMISTRY 25 (1-2): 201-235 JAN-JUN 2006

Times Cited: 34

O2(*a* 1Δ)+O2(*a* 1Δ) → O2(*b* 1Σ)+O2(*b* 1Σ) 电子传能过程的非绝热动力学计算 (第一个六维的非绝热量子散射研究) JCP Communication (2008)

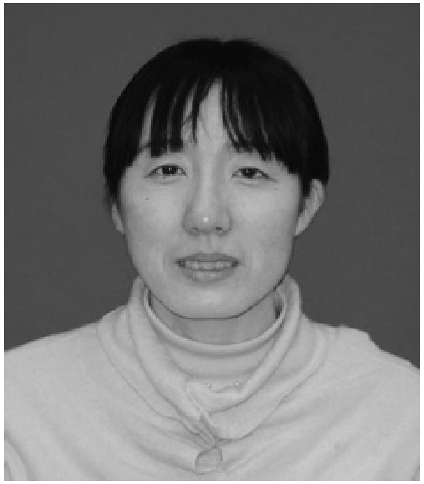




Effect of Coriolis coupling in chemical reaction dynamics

Tian-Shu Chu[ab] **and Ke-Li Han***[a]

Received 2nd October 2007, Accepted 30th January 2008
First published as an Advance Article on the web 5th March 2008
DOI: 10.1039/b715180b



Tian-Shu Chu received her PhD in physical chemistry at the Dalian Institute of Chemical Physics, Chinese Academy of Sciences under the supervision of Professor Keli Han in 2005. To date, she is a visiting scholar at the Dalian Institute of Chemical Physics. Her current research interests focus on studying nonadiabaticity in chemical reactions by quantum dynamics methods.

Ke-Li Han is a professor at the Dalian Institute of Chemical Physics, Chinese Academy of Sciences. He received his PhD in physical chemistry at the Dalian Institute of Chemical Physics, Chinese Academy of Sciences in 1990. In the years 1993–1995, he worked as a postdoctoral researcher at the University of California, USA. In 1995, he became Professor at the Dalian Institute of Chemical Physics. Research interests of his group include experimental and theoretical studies of nonadiabatic processes in reaction dynamics, investigation on molecular dynamics with attosecond resolution, simulation of enzyme dynamics, and study of hydrogen-bond dynamics in intermolecular electron transfer.

代表工作三：阿秒分辨的量子动力学

PRL 95, 123001 (2005) PHYSICAL REVIEW LETTERS week ending 16 SEPTEMBER 2005

Correlation Quantum Dynamics between an Electron and $D_2{}^+$ Molecule with Attosecond Resolution

Jie Hu, Ke-Li Han,* and Guo-Zhong He

State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China

(Received 28 April 2005; published 12 September 2005)

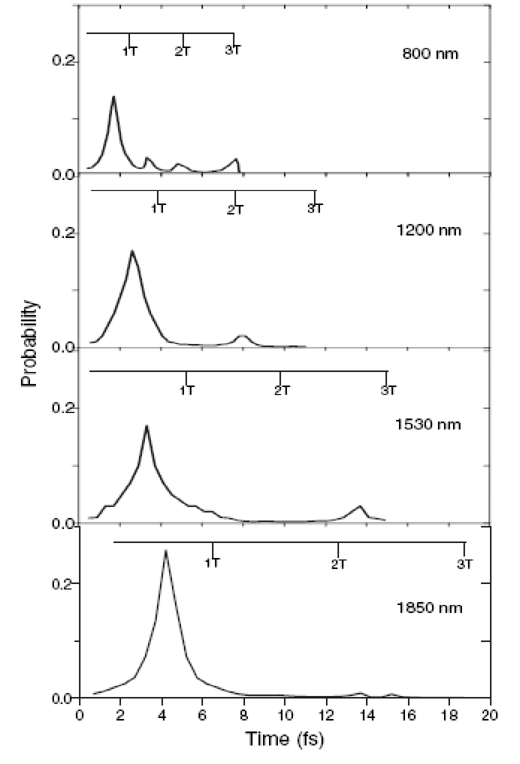
$$i\hbar\frac{\partial}{\partial t}\psi_{\rm I}(\rho,z,\theta,t)=\left[-\frac{\hbar^2}{2\mu}\left(\frac{\partial^2}{\partial\rho^2}+\frac{1}{\rho}\frac{\partial}{\partial\rho}+\frac{\partial}{\partial z^2}+\frac{\partial}{\partial\theta^2}\right)+V(R,\rho,z,t)\right]\psi_{\rm I}(\rho,z,\theta,t), \quad (1)$$

$$i\hbar\frac{\partial}{\partial t}\psi_{\rm II}(R,t)=\left[-\frac{\hbar^2}{m_p}\frac{\partial^2}{\partial R^2}+V(R,t)\right]\psi_{\rm II}(R,t). \quad (2)$$

$$\psi(t+\delta t,\rho,z)=\exp\left(-\frac{iH_1}{2\hbar}\delta t\right)\exp\left(-\frac{iH_2}{\hbar}\delta t\right)\times\exp\left(-\frac{iH_1}{2\hbar}\delta t\right)\psi(t,\rho,z)+O(\delta t^3) \quad (5)$$

where

$$H_1=-\frac{\hbar^2}{2\mu}\left(\frac{\partial^2}{\partial\rho^2}+\frac{1}{\rho}\frac{\partial}{\partial\rho}+\frac{\partial^2}{\partial z^2}\right),\qquad H_2=V_c+V(R,t)-2\Gamma\hbar\frac{\partial^2}{\partial t^2}. \quad (6)$$



2009己醜年

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Hu J, **Han KL**, He GZ

Correlation quantum dynamics between an electron and D-2(+) molecule with attosecond resolution

PHYSICAL REVIEW LETTERS 95 (12): Art. No. 123001 SEP 16 2005

Times Cited: 34

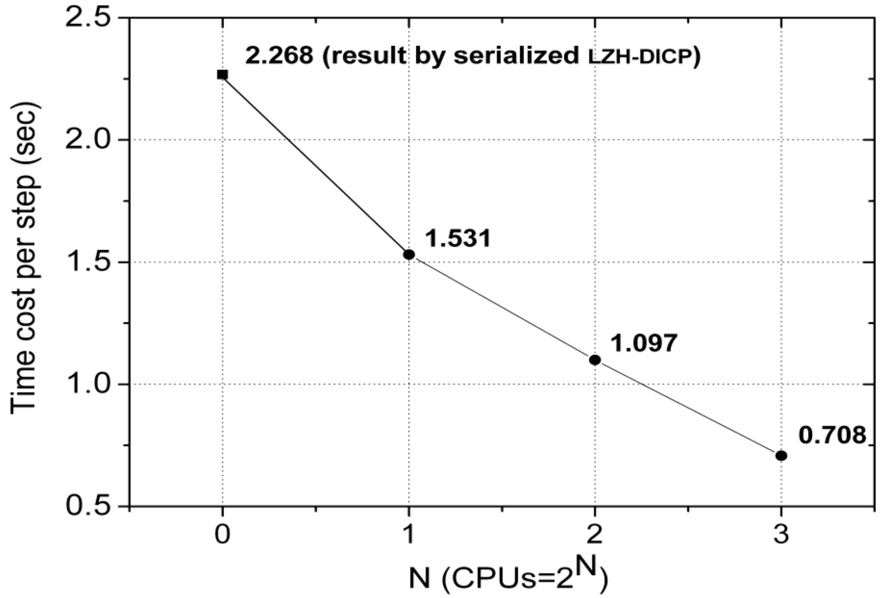
- ➢LZH-DICP(Lu-Zhang-Han)
- ➢Application
  - ✓ Hydrogen atom
  - ✓ Molecular hydrogen ion
  - ✓ Carbon monoxide cation
  - ✓ High-Harmonic Generation

Lu, Zhang, Han, Phys Rev E 2008, 77, 066701.

LZH-DICP: Test on 3.20 GHz Pentium 4D

	H原子			1D模型H_2^+			
	Δr	Δt	T_{CPU}	ΔR	Δz	Δt_e	T_{CPU}
sine-DVR	0.2	0.1	2 *h* 47 *m* 29 *s*	0.1	0.4	0.2	5 *h* 2 *m* 33 *s*
CN	0.15	0.05	3 *d* 7 *h* 20 *m* 6 *s*	0.05	0.2	0.1	8 *d* 12 *h* 11 *m* 57 *s*

Intel(R) Xeon(R) CPU 2.60GHz
四核处理器上并行测试



Molecular Hydrogen ion

<u>Theory</u>

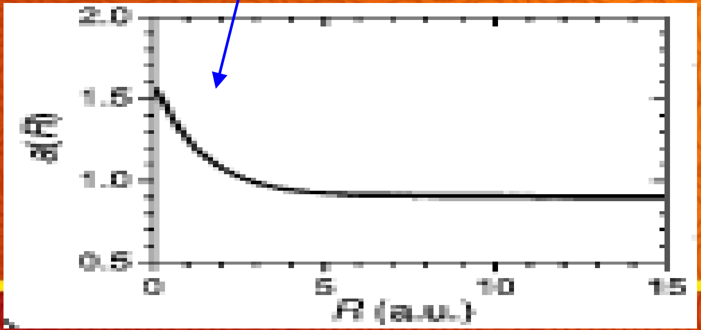
!!! Dissociation

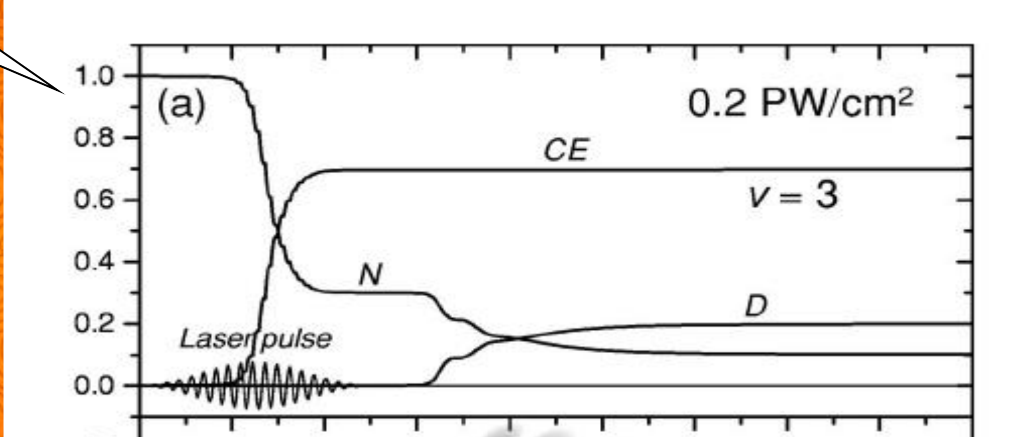
Ionization

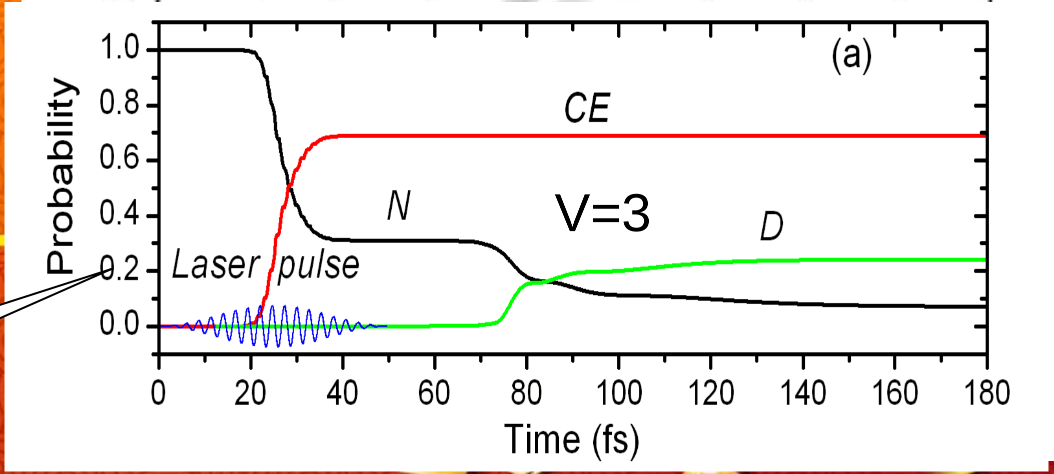
B. Feuerstein and U. Thumm,
Phys. Rev. A, 2003, 65, 043405

Note: different
soft-core Coulomb potential

$$-\frac{1}{\sqrt{(z-\frac{R}{2})^2+a(R)}}-\frac{1}{\sqrt{(z+\frac{R}{2})^2+a(R)}}$$



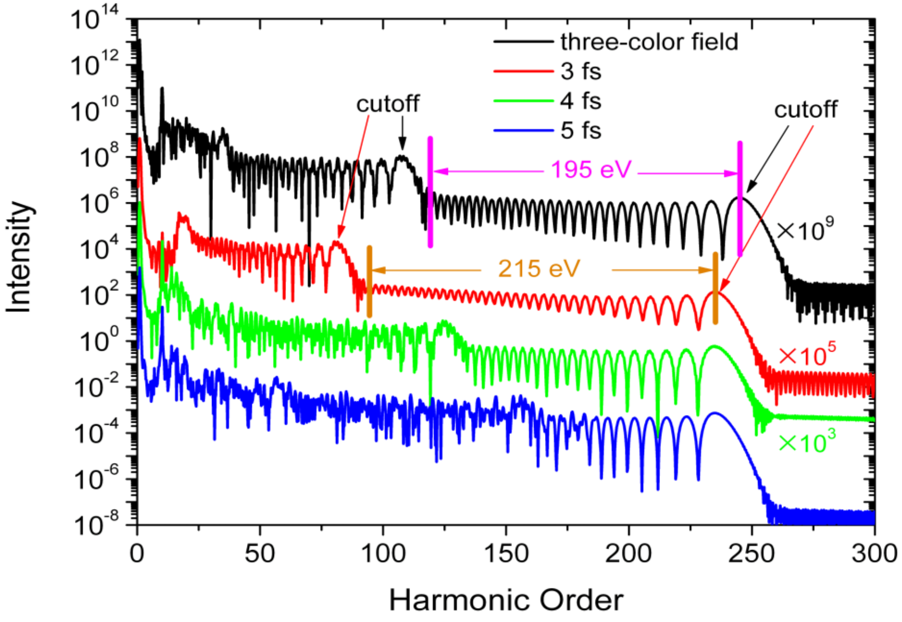


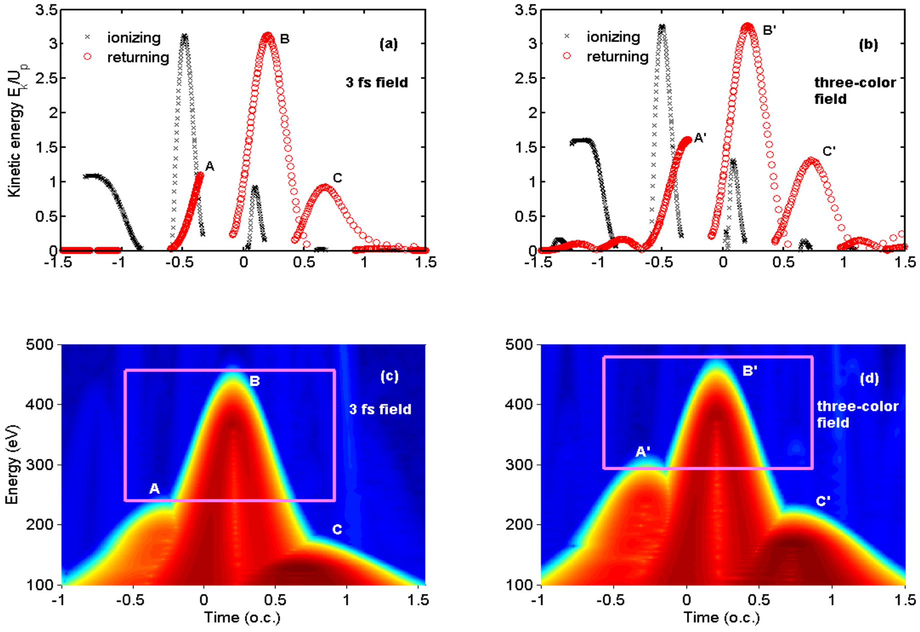


Our calculations

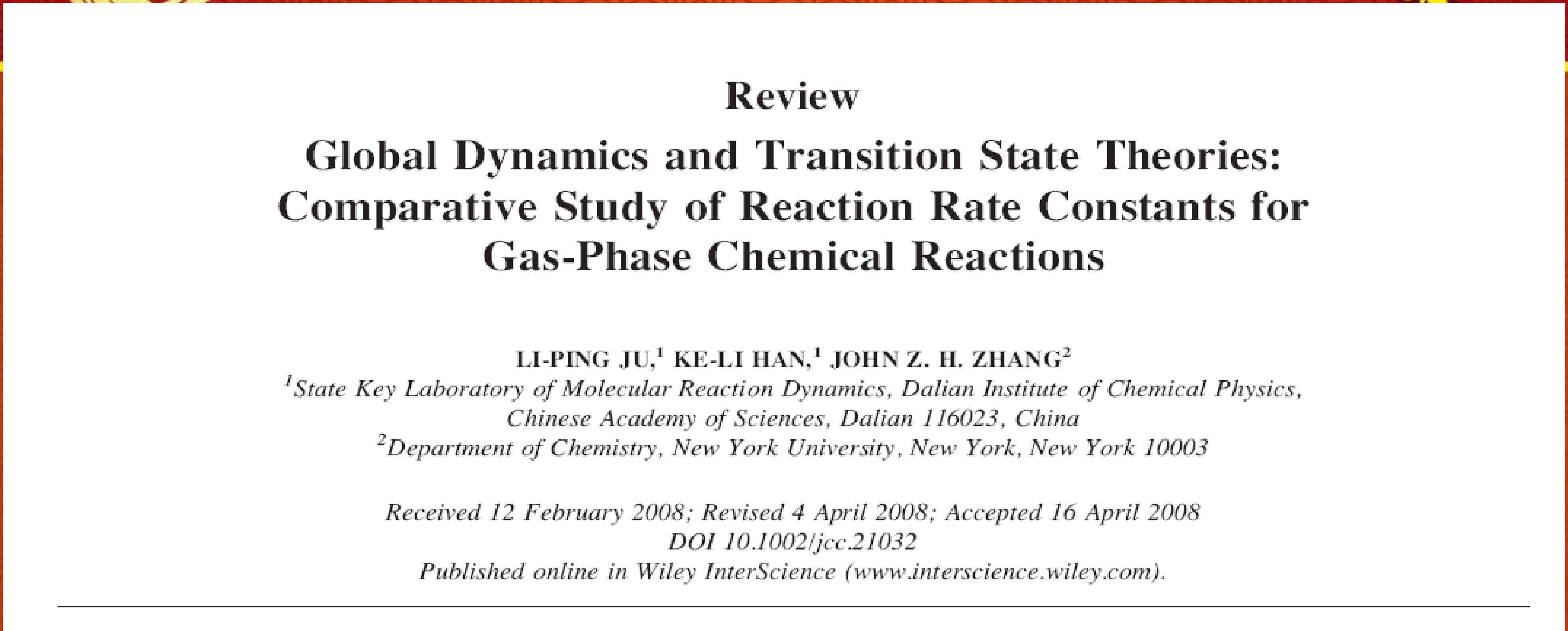
200

谨賀新年


Intensity
Harmonic Order
10^{14}
10^{12}
10^{10}
10^{8}
10^{6}
10^{4}
10^{2}
10^{0}
10^{-2}
10^{-4}
10^{-6}
10^{-8}
0
50
100
150
200
250
300
three-color field
3 fs
4 fs
5 fs
cutoff
cutoff
195 eV
215 eV
×10^{9}
×10^{5}
×10^{3}


Kinetic energy E_k/U_p
ionizing
returning
(a)
3 fs field
A
B
C
(b)
three-color field
A'
B'
C'
3.5
3
2.5
2
1.5
1
0.5
0
-1.5
-1
-0.5
0
0.5
1
1.5
Energy (eV)
500
400
300
200
100
(c)
3 fs field
(d)
three-color field
Time (o.c.)
-1
-0.5
0
0.5
1
1.5

代表工作之四：全维动力学与过渡态理论的比较

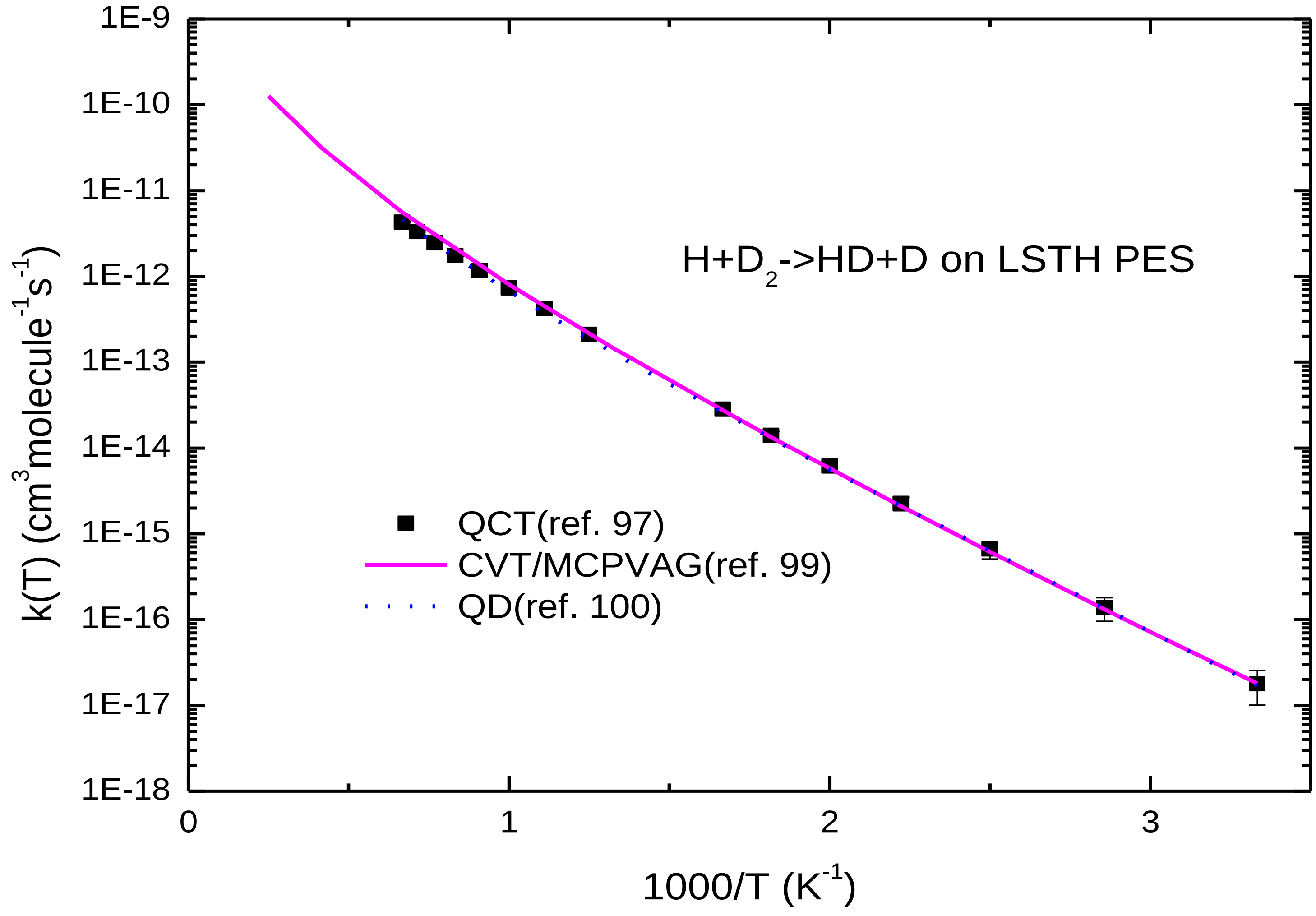


Review

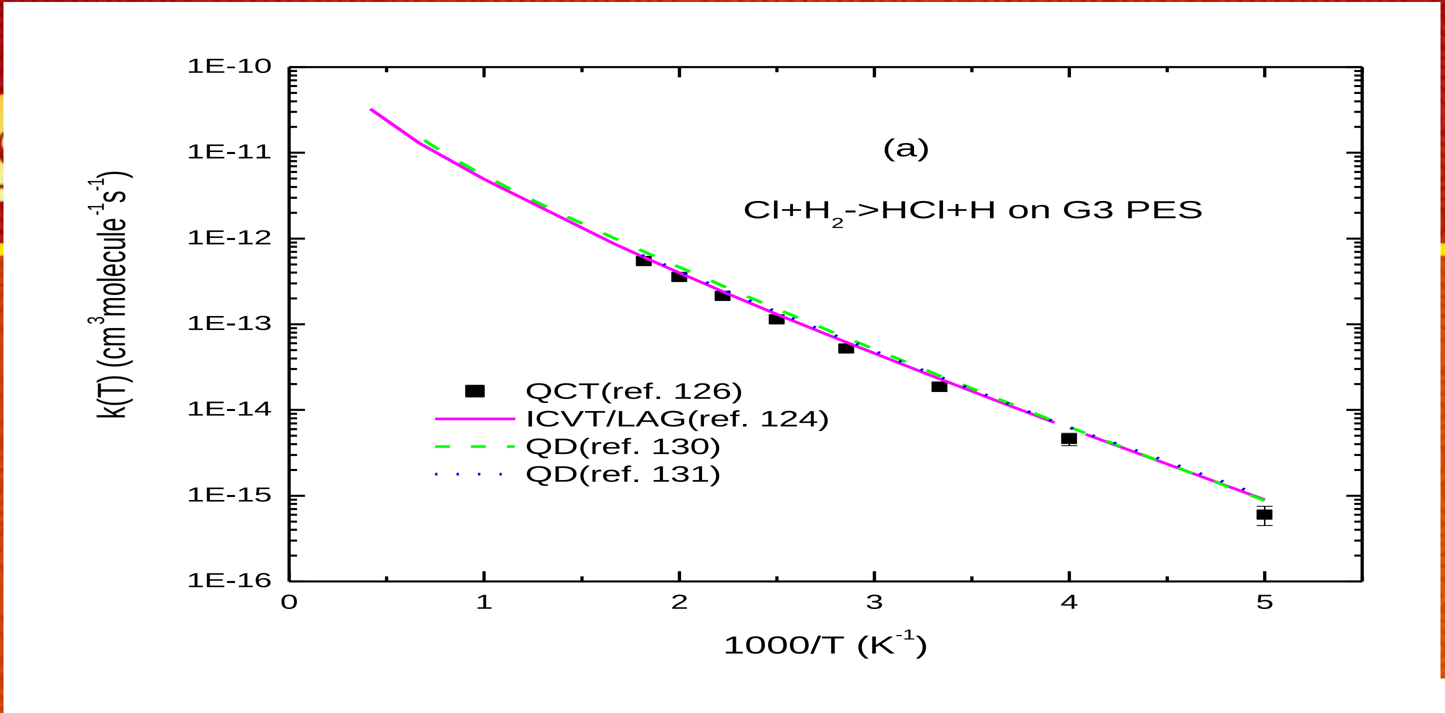
Global Dynamics and Transition State Theories: Comparative Study of Reaction Rate Constants for Gas-Phase Chemical Reactions

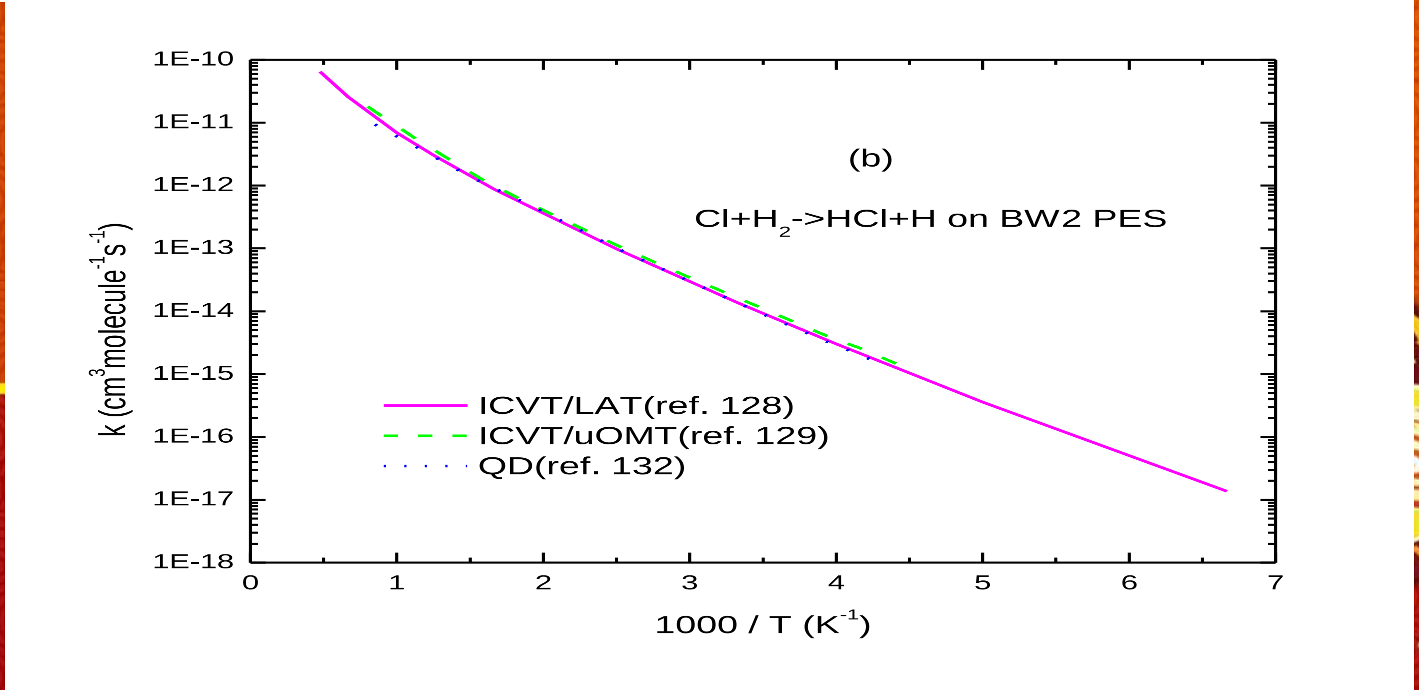
LI-PING JU,[1] KE-LI HAN,[1] JOHN Z. H. ZHANG[2]
[1]*State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics,*
Chinese Academy of Sciences, Dalian 116023, China
[2]*Department of Chemistry, New York University, New York, New York 10003*

Received 12 February 2008; Revised 4 April 2008; Accepted 16 April 2008
DOI 10.1002/jcc.21032

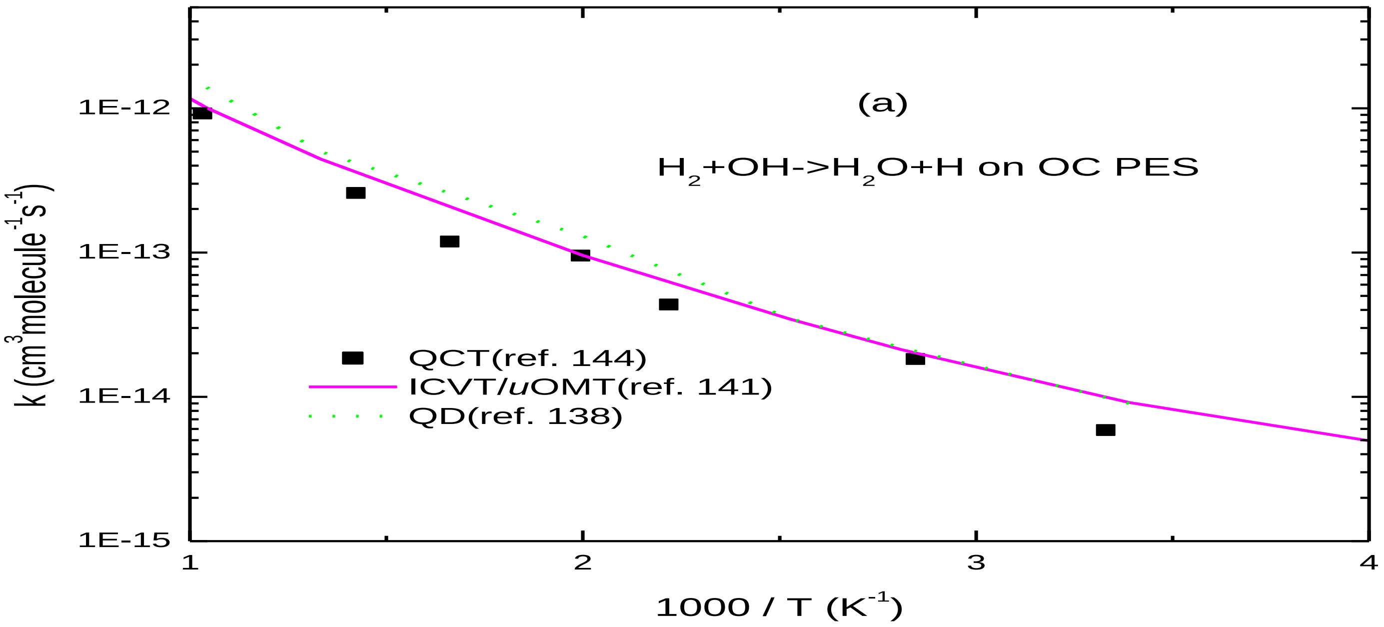

H+D$_2$->HD+D on LSTH PES
k(T) (cm^{3}molecule^{-1}s^{-1})
1000/T (K^{-1})
1E-9
1E-10
1E-11
1E-12
1E-13
1E-14
1E-15
1E-16
1E-17
1E-18
0
1
2
3
QCT(ref. 97)
CVT/MCPVAG(ref. 99)
QD(ref. 100)

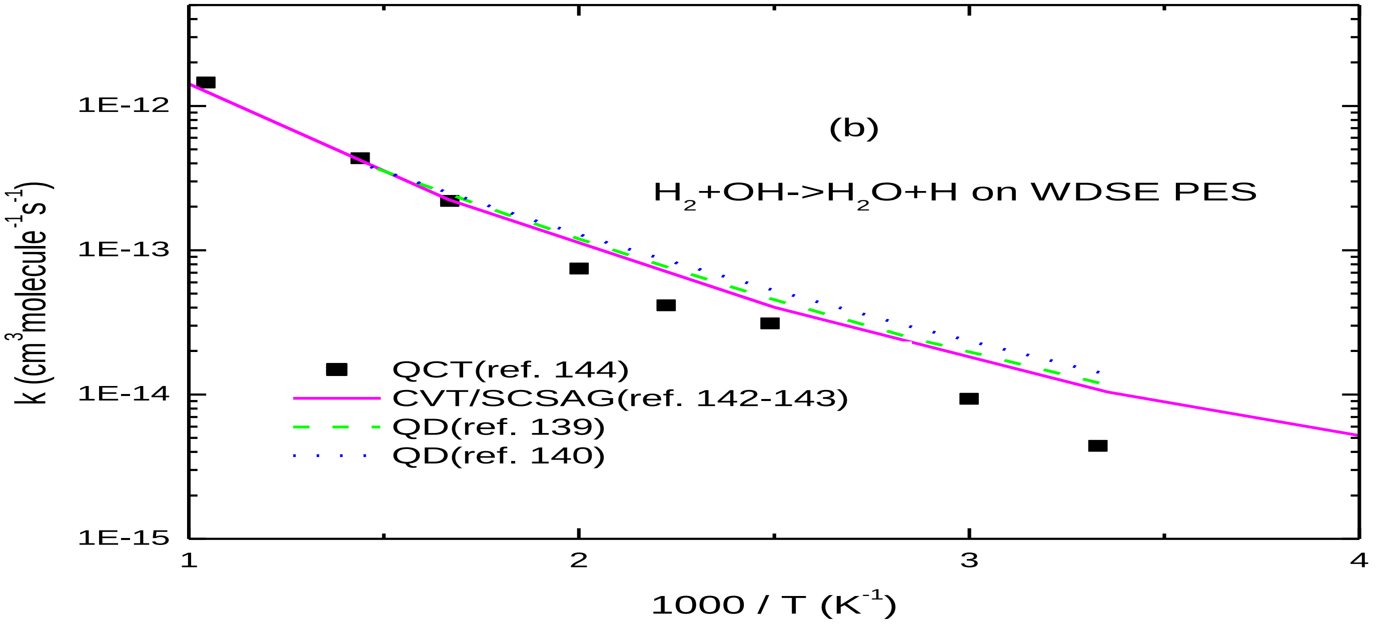
謹賀新年

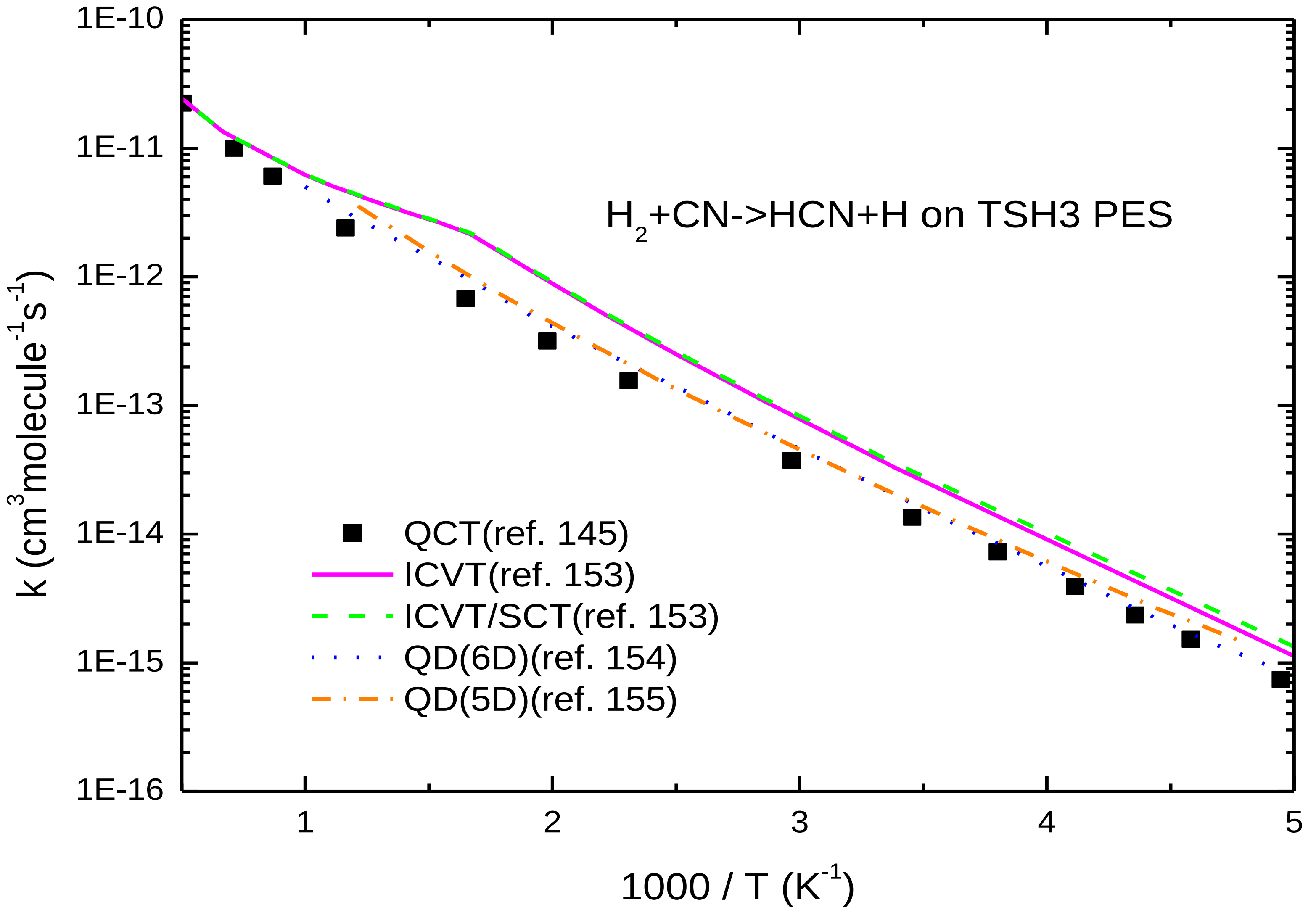

(a)
Cl+H$_2$->HCl+H on G3 PES
QCT(ref. 126)
ICVT/LAG(ref. 124)
QD(ref. 130)
QD(ref. 131)
k(T) (cm^{3}molecule^{-1}s^{-1})
1E-10
1E-11
1E-12
1E-13
1E-14
1E-15
1E-16
0
1
2
3
4
5
1000/T (K^{-1})

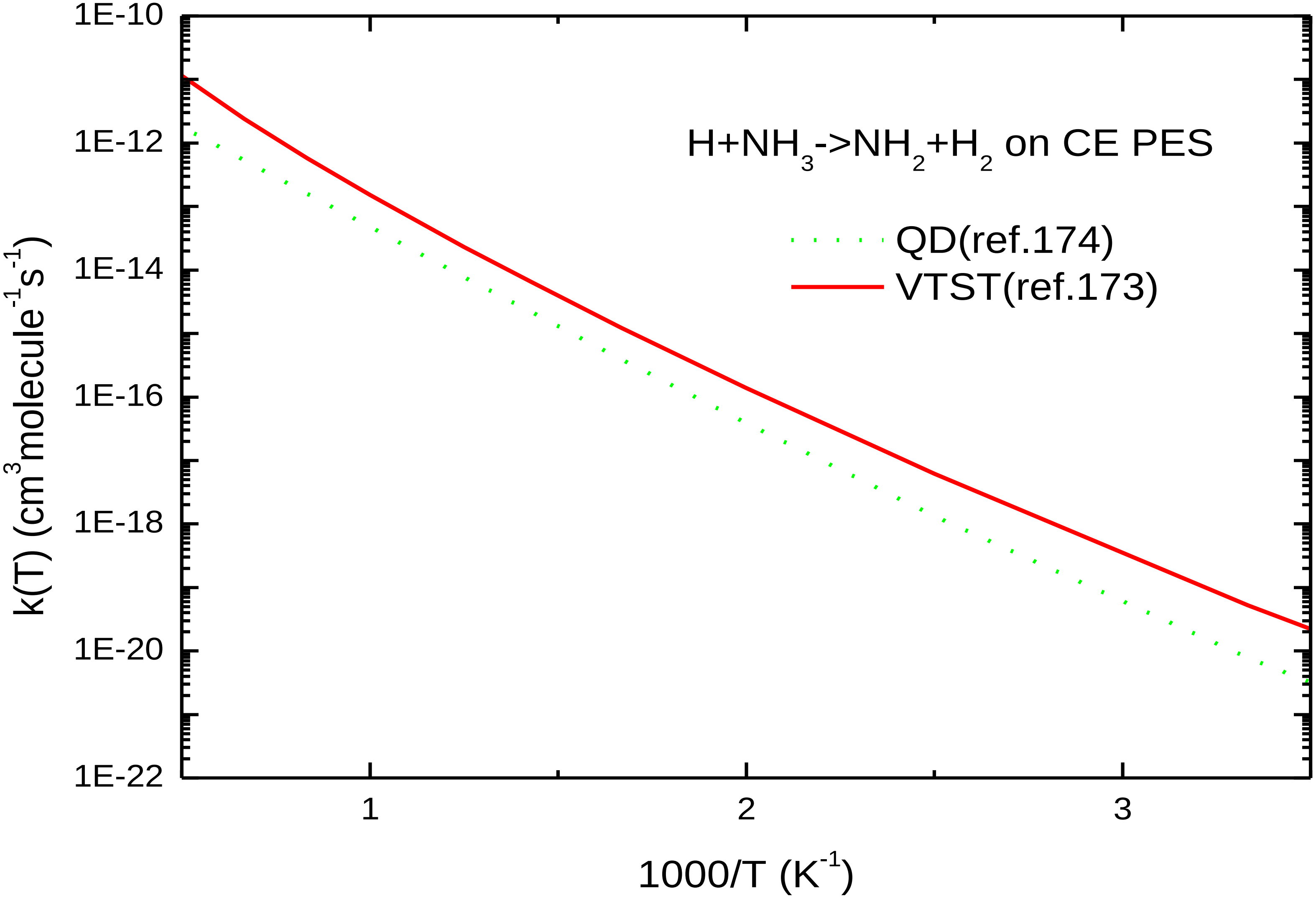

(b)
Cl+H$_2$->HCl+H on BW2 PES
ICVT/LAT(ref. 128)
ICVT/uOMT(ref. 129)
QD(ref. 132)
k (cm^{3}molecule^{-1}s^{-1})
1E-10
1E-11
1E-12
1E-13
1E-14
1E-15
1E-16
1E-17
1E-18
0
1
2
3
4
5
6
7
1000 / T (K^{-1})

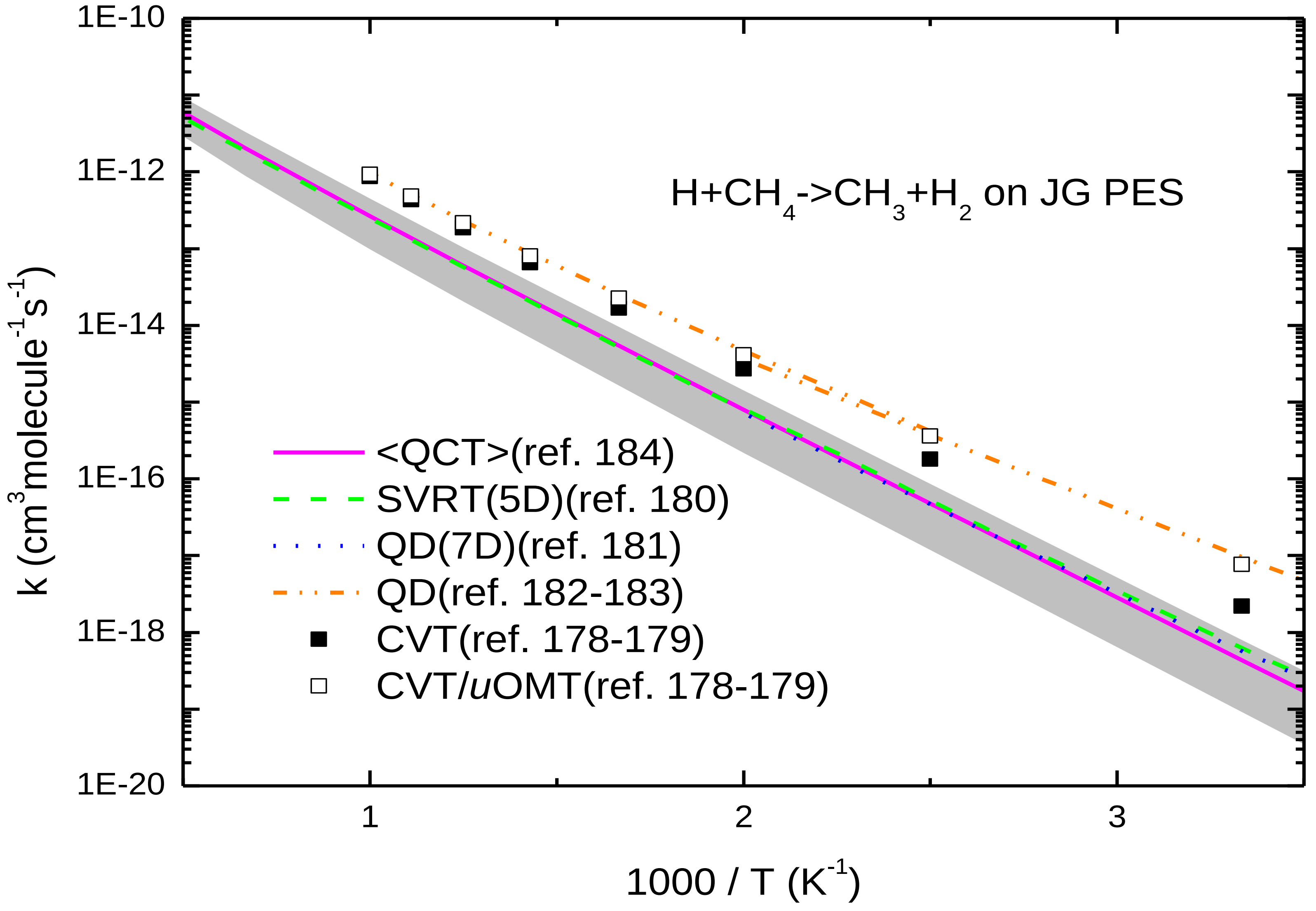
謹賀新年


(a)
H_2+OH->H_2O+H on OC PES
QCT(ref. 144)
ICVT/*u*OMT(ref. 141)
QD(ref. 138)
k (cm^{3}molecule^{-1}s^{-1})
1E-12
1E-13
1E-14
1E-15
1
2
3
4
1000 / T (K^{-1})

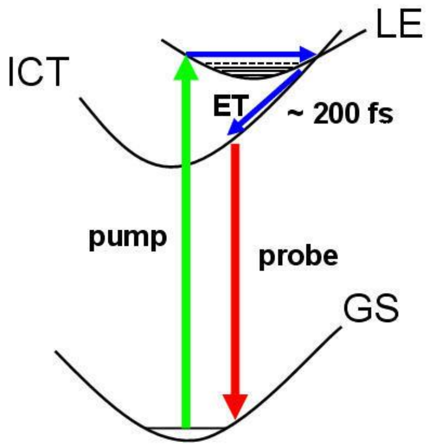

(b)
H_2+OH->H_2O+H on WDSE PES
QCT(ref. 144)
CVT/SCSAG(ref. 142-143)
QD(ref. 139)
QD(ref. 140)
k (cm^{3}molecule^{-1}s^{-1})
1E-12
1E-13
1E-14
1E-15
1
2
3
4
1000 / T (K^{-1})


H_2+CN->HCN+H on TSH3 PES
k (cm^{3}molecule^{-1}s^{-1})
1E-10
1E-11
1E-12
1E-13
1E-14
1E-15
1E-16
1
2
3
4
5
1000 / T (K^{-1})
QCT(ref. 145)
ICVT(ref. 153)
ICVT/SCT(ref. 153)
QD(6D)(ref. 154)
QD(5D)(ref. 155)


H+NH$_3$->NH$_2$+H$_2$ on CE PES
QD(ref.174)
VTST(ref.173)
k(T) (cm^{3}molecule^{-1}s^{-1})
1E-10
1E-12
1E-14
1E-16
1E-18
1E-20
1E-22
1
2
3
1000/T (K^{-1})


H+CH$_4$->CH$_3$+H$_2$ on JG PES
k (cm^{3}molecule^{-1}s^{-1})
1000 / T (K^{-1})
1E-10
1E-12
1E-14
1E-16
1E-18
1E-20
1
2
3
<QCT>(ref. 184)
SVRT(5D)(ref. 180)
QD(7D)(ref. 181)
QD(ref. 182-183)
CVT(ref. 178-179)
CVT/*u*OMT(ref. 178-179)

代表工作之五：氢键促进的光诱导电子转移猝灭荧光



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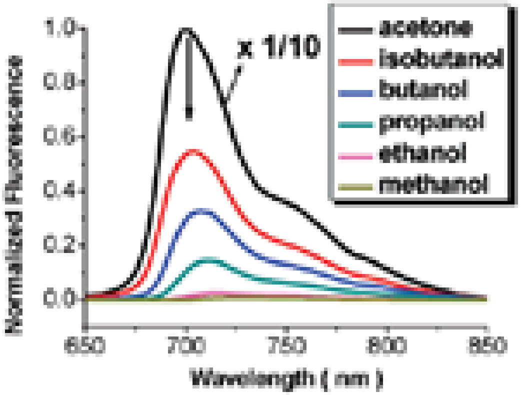
PET



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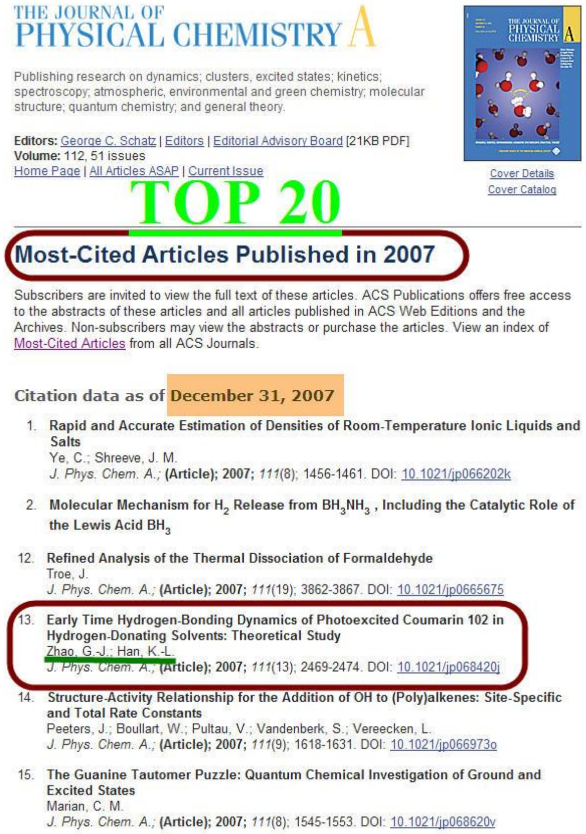

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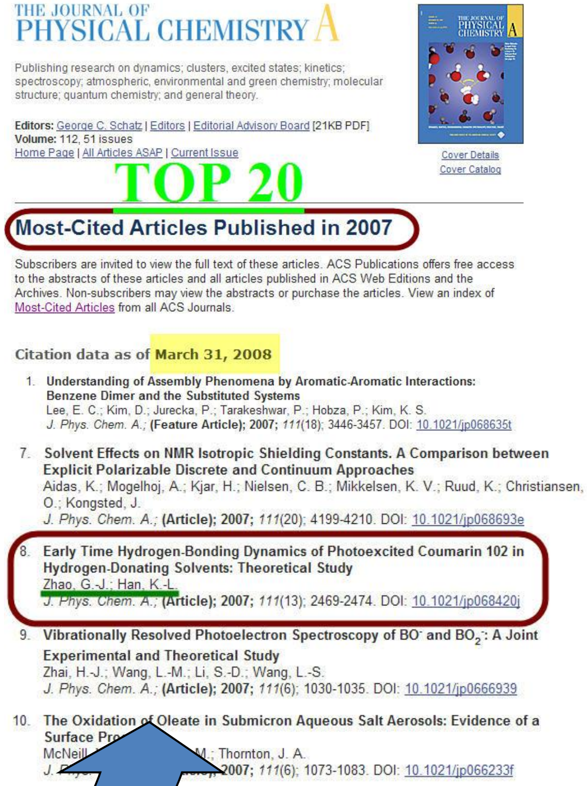

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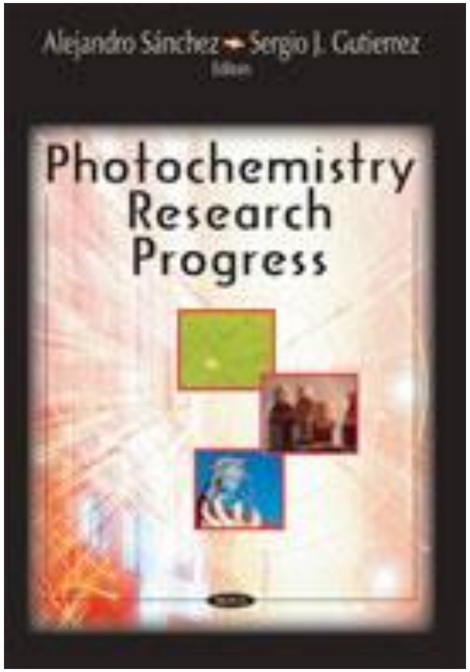

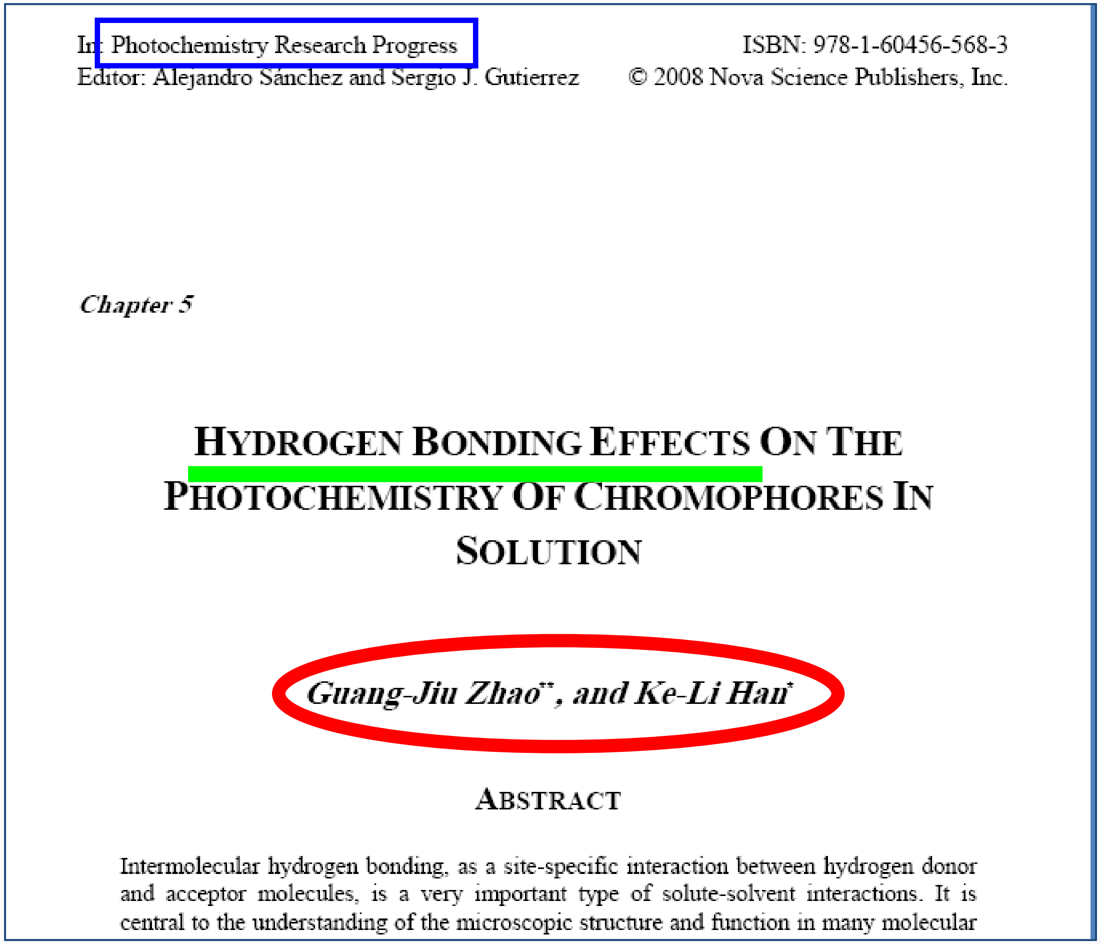

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Chapter 5

HYDROGEN BONDING EFFECTS ON THE PHOTOCHEMISTRY OF CHROMOPHORES IN SOLUTION

Guang-Jiu Zhao*,**, and Ke-Li Han*

ABSTRACT

Intermolecular hydrogen bonding, as a site-specific interaction between hydrogen donor and acceptor molecules, is a very important type of solute-solvent interactions. It is central to the understanding of the microscopic structure and function in many molecular

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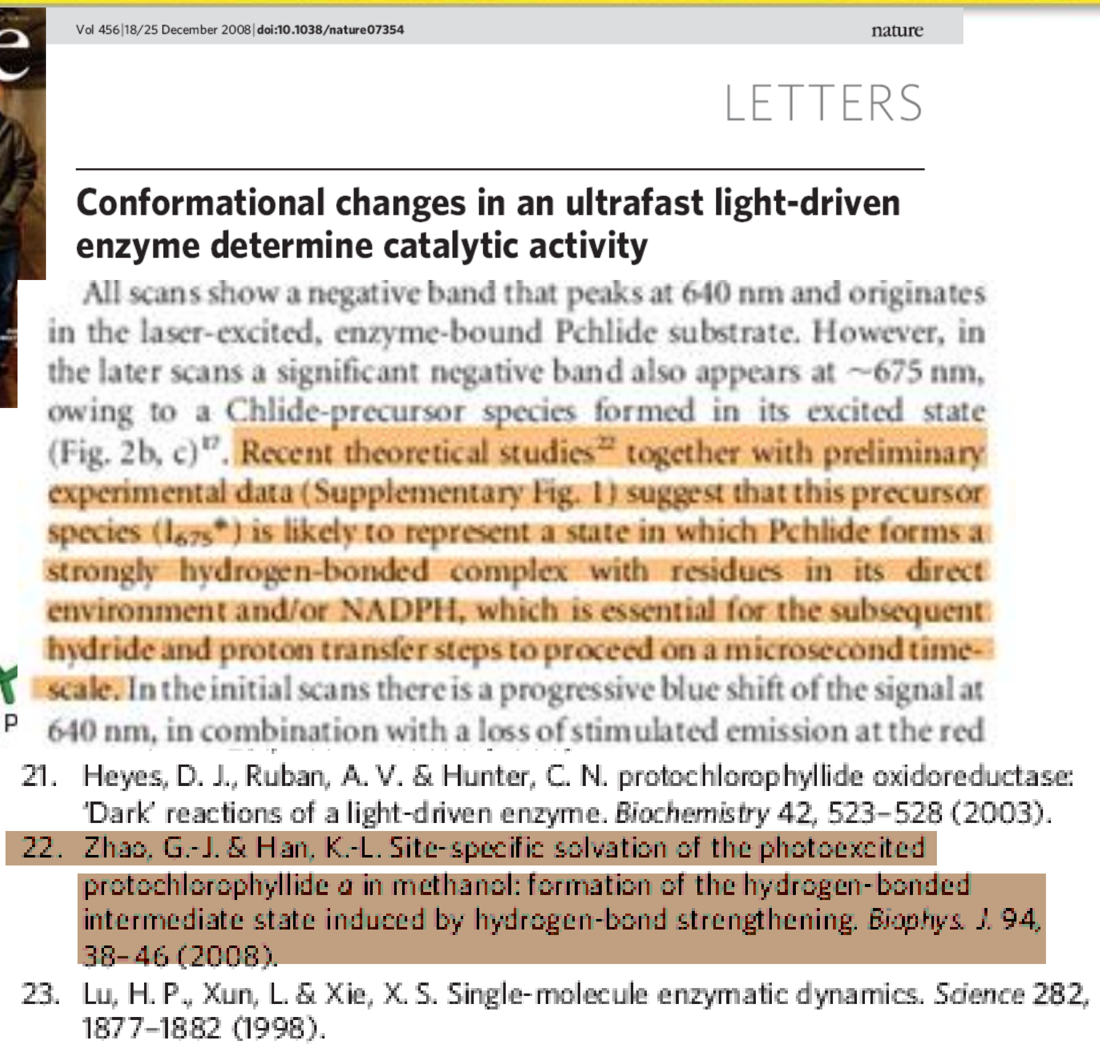

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All scans show a negative band that peaks at 640 nm and originates in the laser-excited, enzyme-bound Pchlide substrate. However, in the later scans a significant negative band also appears at ~675 nm, owing to a Chlide-precursor species formed in its excited state (Fig. 2b, c)[17]. Recent theoretical studies[22] together with preliminary experimental data (Supplementary Fig. 1) suggest that this precursor species (I_{675}*) is likely to represent a state in which Pchlide forms a strongly hydrogen-bonded complex with residues in its direct environment and/or NADPH, which is essential for the subsequent hydride and proton transfer steps to proceed on a microsecond timescale. In the initial scans there is a progressive blue shift of the signal at 640 nm, in combination with a loss of stimulated emission at the red

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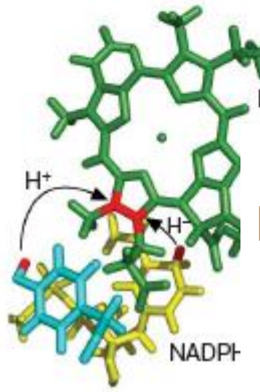

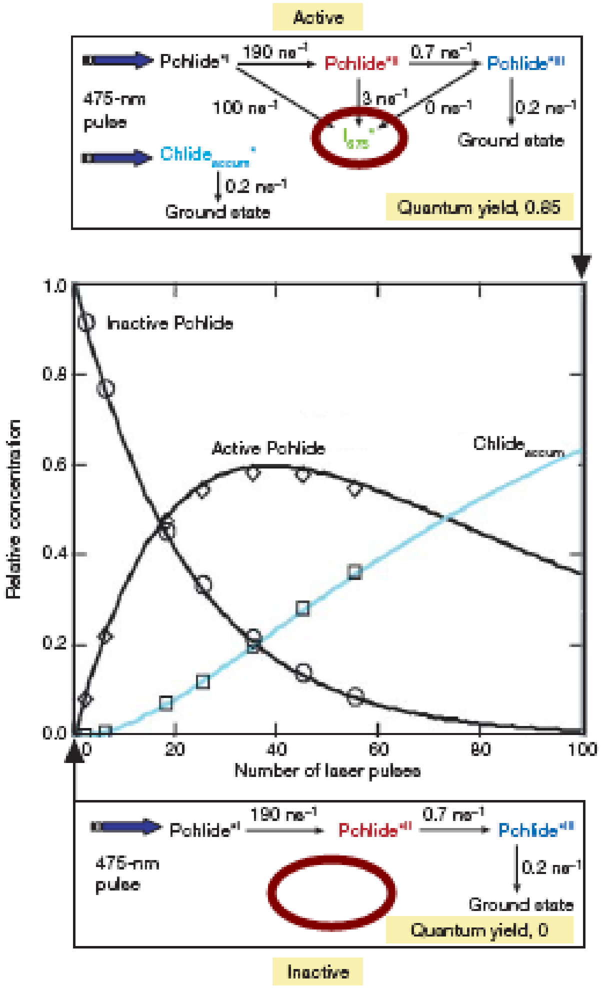

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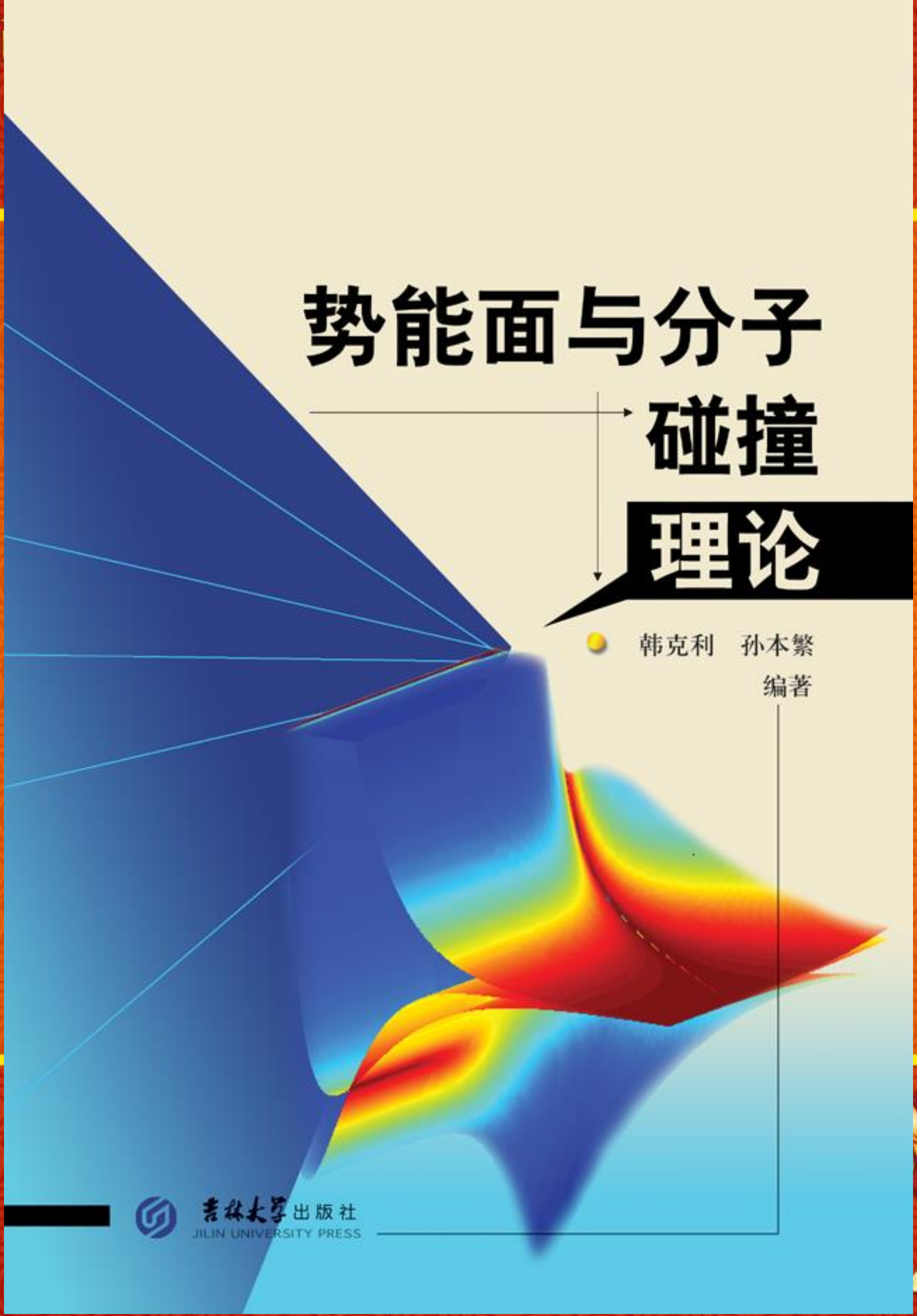

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