

Accurate description of surface temperature effects for the quantum dynamics of H₂ on a metal surface

Bauke Smits and Mark F. Somers

Theoretical Chemistry, Leiden Institute of Chemistry
Leiden University, The Netherlands



Universiteit
Leiden
The Netherlands

Introduction

- H₂ dissociation on a Cu(111) surface is an important benchmark system.¹
- The specific reaction parameter (SRP) approach to DFT yields dissociation probabilities to within chemical accuracy (< 43 meV error) using a 0 K static surface.²
- The static corrugation model (SCM) was designed to include surface temperature effects using an extra potential on the static, but distorted, surface.^{3,4}
- The SCM was expanded with an embedded-atom method potential (EAM) to generate highly accurate surface slabs (EAM-SCM).^{5,6}
- EAM potential also allows for a moving surface: dynamic corrugation model (DCM).⁶
- EAM-SCM and EAM-DCM reaction probabilities obtained with quasi-classical dynamics (QCD) reproduce experimental values.⁶

Goals

Include surface temperature effects in the quantum dynamical treatment of H₂ dissociating on a metal surface.

- Thermally distorted Cu(111) surface with EAM-SCM.
- Quantum dynamics with time-dependent wave packet approach.

Dynamic Corrugation Model (DCM)

Static Corrugation Model (SCM)

BOSS

V_{Coup}

$V_{\text{Strain}} / V_{\text{EAM}}$

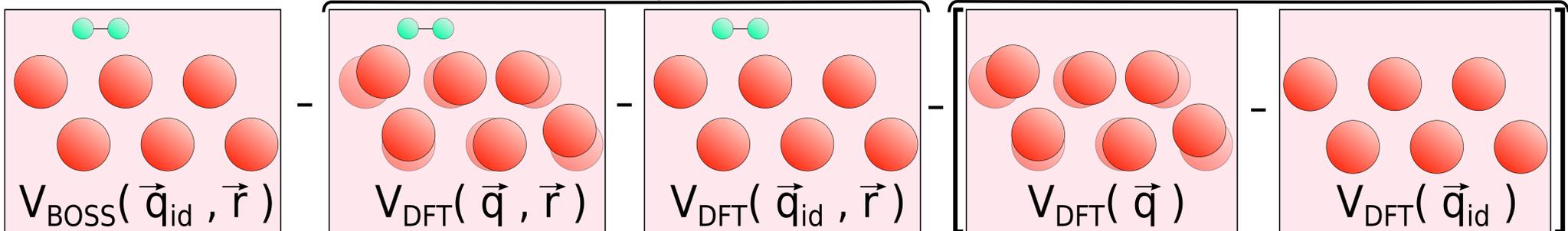


Figure 1. Schematic overview of the SCM coupling potential and its relation to system configurations. Here \vec{q} and \vec{r} denote the surface and the hydrogen degrees of freedom, respectively.

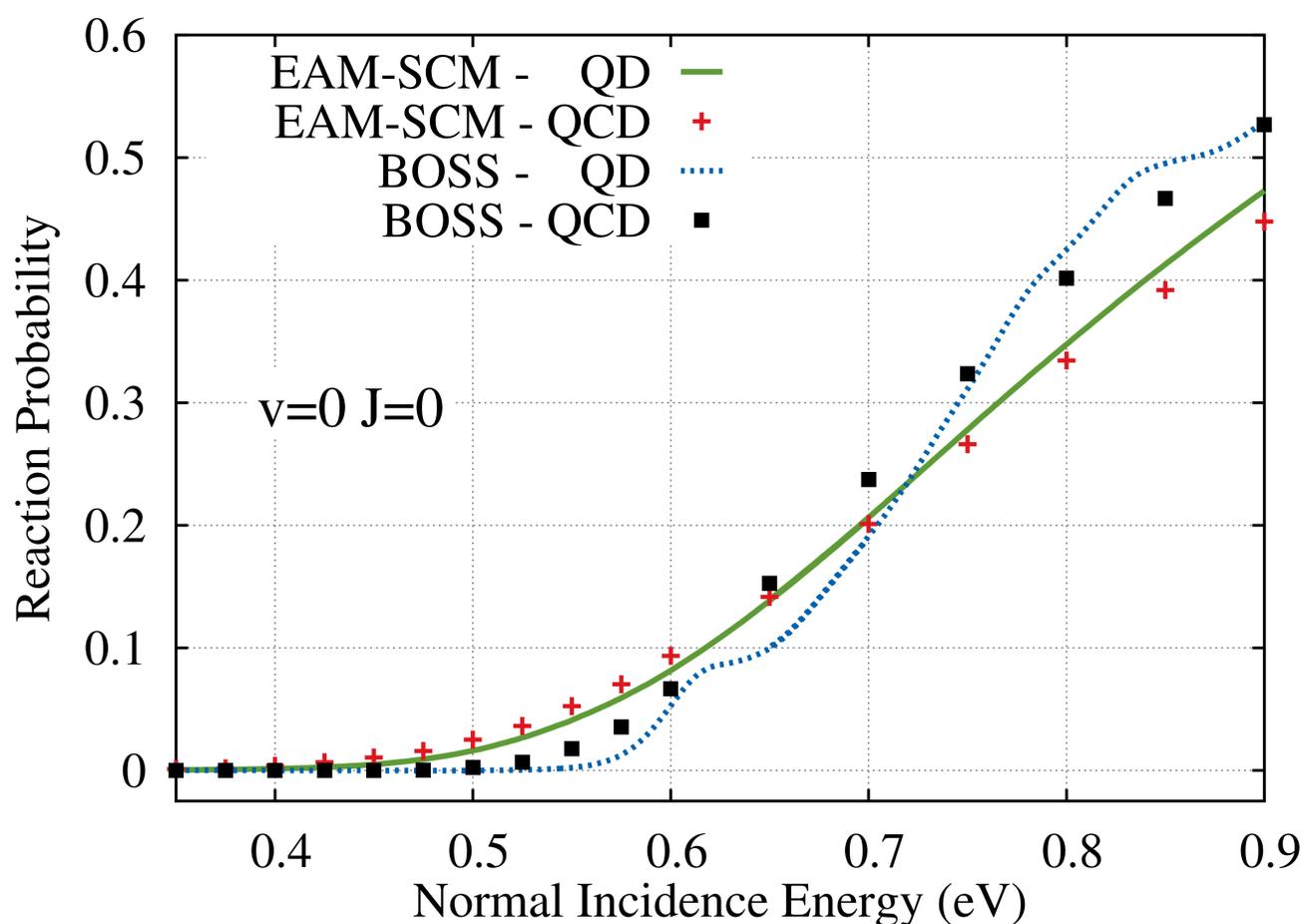


Figure 2.⁷ Quasi-classical and quantum dynamical reaction probabilities for H₂ in the rovibrational ground state on a Cu(111) surface at a modelled surface temperature of 925 K.

Method

- Quantum dynamics (QD) of H₂ on 104 unique Cu(111) surface traces using the EAM-SCM.
- 50.000 QCD simulations per incidence energy on randomly selected surface slabs with EAM-SCM.
- Total of 25.000 distorted surface slabs obtained with EAM potential.⁶
- Compare to BOSS results for QD and QCD.

Results

- EAM-SCM reaction probability curves show slightly more broadness than the BOSS results.
- Characteristic of surface temperature effects.
- Very good agreement between QD and QCD.

b.smits@lic.leidenuniv.nl

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