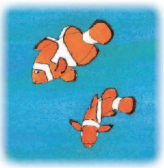
Random Notes
（ランダム ノート）



研究ノート

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Key Words: Surfaces/Interfaces, Elementary Processes, Dynamics, Hydrogen, Steering, Dynamical Quantum Filtering, Design

RESEARCH NOTES
（研究ノート）

Only one way to do this.
Forget about conventions . . . style . . . form . . .
Jut down all the ideas that come to mind . . .
Don't worry about the rest . . .
Just the usual research notes . . .

SURFACES
（表面）

Surfaces . . . Ubiquity . . . synonymous.
The motivation is plain and simple.

PIE IN THE SKY
（うまい話）

Elimination of noxious gases in automobile exhausts . . . Production of petrochemicals . . . The list of examples of vastly important chemical reactions goes on, and on.

And all these reactions are catalyzed on the surfaces of appropriate powdered metals and oxides.

This is an industry worth hundreds of billions of yen, having direct or indirect influence on ～ 20% of the world economy. [1]

If it were possible to gain a detailed, atomic-level understanding of surface reactions, then it might be possible to design cheaper, more efficient (. . . , more user-and environment-friendly, . . .) commercial catalysts (catalytic substrates).

SIMPLE IS BEST, BUT NEVER EASY
（シンプル イズ ベスト　でも簡単じゃない）

The pie in the sky has remained thus —
A Pie in the Sky. We have yet to hear of the development of a single industrially significant, new catalyst material directly resulting from the forty-some years of investigation on surfaces. [2]

The reason for this "failure" is, again, plain and simple.

Catalysis on surfaces is a very complex affair, and even the elementary processes that together comprise a catalytic reaction, such as the dissociation and sticking of a molecule to a surface, are not very well understood.

Thus, researches have turned to the study of elementary molecule-surface interactions. [3, 4]

HYDROGEN
（水素）

"To understand hydrogen is to understand all of physics!" or
"To understand hydrogen, one must understand all of physics". [5]

Hydrogen is the lightest of all the elements and has the simplest structure, a single electron orbiting a nucleus consisting of a proton. It is by far the most abundant element in the universe. On the Earth, it occurs chiefly combined with Oxygen as Water.
With WATER as the only EMISSION from hydrogen

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combustion, hydrogen is being promoted as the POWER SOURCE of the future.

Hydrogen is also involved in many industrially relevant processes, and its role in determining the physical properties of various materials is well recorded. Considering the special role of Hydrogen in the history and development of Science, it would not be an exaggeration to say -

"Whatever (mechanism) works for Hydrogen should work for others as well."

Although Oxygen needs no further introduction, it is worth mentioning that oxygen plays an important role in determining what would make or break a good material, and it also plays a crucial role in determining the success or failure of body metabolism.

DYNAMICS

（ダイナミクス）

"A slight change (e.g., induced by light irradiation) in the electronic states making up a solid surface interacting with atoms or molecules gives rise to a slight change in the position of atoms and molecules, which in turn initiates further a marked change in the electronic states. Eventually, the coupled electronic and atomic processes proceed to the final state on the surface. This maybe properly regarded as characteristic of all dynamical processes occurring on surfaces, including chemical reactions.

It is also a characteristic of all dynamical processes occurring around us, starting from those involving life forms and extending to other complex systems.

Thus, in order to deepen our understanding of the dynamics of complex systems, it is necessary to clarify the fundamental mechanism behind the dynamics of each of the elementary processes. The most important breakthrough we can achieve towards a thorough understanding of the dynamics occurring around us would come from an accurate understanding of the elementary processes, in which mass, charge, and energy transport play important roles." [6]

This is the basic rationale underlying the need to understand the dynamical nature of surfaces.

PLAYGROUND

（校庭）

One area where significant progress has been made is in the development of tools to determine the arrangements of atoms at a surface. As a result of this progress, the atomic arrangements of quite a variety of crystal surfaces are now known.

With recent advances in experimental techniques in the field of Surface Science, we are now at the stage where we can prepare well-characterized solid surfaces, which, in a sense, serve as Playgrounds for Physicists.[3, 4]

The solid surface provides us with a stage to study the dynamics of complex systems, where state transitions of the corresponding electron system are closely connected with changes of atomic and molecular motion on various scales of magnitude with respect to time, space, and energy.

The combination of experimental techniques and computation power now available renders surfaces an ideal venue in which to study the mechanisms of elementary dynamical processes and their concatenation into descriptions of processes of fundamental and technological importance, e.g., chemical reactions.

HOLY GRAIL

（聖杯）

One of the most exciting challenges of present-day science is the task of developing a detailed (microscopic) picture of surface reactions. This would involve understanding the intra- and inter-molecular motions of the reactants, as they undergo changes at a surface, and understanding the related issues of energy requirements, energy flow, and energy disposal for these microscopic interactions. To make any headway in understanding such surface reactions, the most fundamental surface reactions must be understood. This entails carrying out systematic and comprehensive studies of —

(1) the elementary processes involved;
(2) the mechanisms (esp., quantum effects) behind the processes;
(3) the role of the various internal degrees-of-freedom (including that of the electron charge- and spin degrees-of-freedom); and
(4) how Nature was able to induce the emergence of a complex catalytic reactivity from the synergy between components.

We then gain a basic knowledge of Elementary Dynamical Quantum Processes in Excitations and Reactions at Surfaces/Interfaces.

Basic knowledge comes in the form of the relevant potential energy (hyper-) surfaces (PESs) corresponding to particular reactions (with reaction partners explicitly taken in account), and knowing how the (quantum) dynamics of the corresponding relevant processes would evolve under the influence of these PESs.

ORIENTATION, STEERING, FILTERING
（配向、舵取り、フィルタリング）

Without collisions, there is no surface reaction.

Molecular collisions are an inherent part of surface reactions. They cause molecular and geometrical rearrangements, induce energy transfer to specific quantum states, or simply cause a molecule to break apart.

The stereodynamics of the reactants (molecules), the orientation and the movement of molecule in three-dimensional space, plays an important role in surface reactions.

As with astronauts approaching their space station, some maneuvers (steering[3, 4]) are more effective than others in successful docking.

One could also say that the pre-determined positions of the docking bays in the space stations has limited (filtering[3, 4]) the maneuvers possible for successful docking.

CONTROL
（制御）

The inherent orientation dependence of gas-surface reaction dynamics can be explored, e.g., to increase the ortho-para(o-p) H_2 conversion yield (cf., e.g., [4]), utilizing dynamical concepts such as Quantum Filtering [3, 4], Steering [3, 4], and Steric Effects [3, 4].

Furthermore, as shown theoretically [7] (and later confirmed experimentally [8]), not only is the $H_2(D_2)$-surface interaction (viz., the $H_2(D_2)$ dissociative adsorption dynamics) strongly dependent on the orientation of the impinging H_2 (D_2), but (that the orientation effects are) also strongly sensitive to the kinetic (translational) energy of the impinging $H_2(D_2)$.

Considering the reverse process of associative desorption, the angular distribution of the desorbing $H_2(D_2)$ would also be dependent on the corresponding kinetic energy. These results indicate/hint at possible further means of probing local surface reactivity[9].

MANIPULATION
（操作）

Carbon-based nano-materials continue to attract much attention because of their suggested material potentials, e.g., as hydrogen storage devices in fuel-cell-powered electric vehicles [10, 11]. However, the results have somehow remained contentious [12]. This indicates not only a need for more systematic investigations, but also a microscopic picture of the mechanism underlying $H(H_2)$ adsorption onto, absorption into, and desorption from carbon-based materials.

Systematic investigation on the $H(H_2)$ interaction with the surface plane of a graphene sheet [13]; possible stable $H(H_2)$ configurations [14], once found inside/between graphite layers; $H(H_2)$ interaction with the armchair [15] and zigzag [16] edges of graphite layers; and their significance in relation to experimental findings [17] were thus carried out.

Finally, based on the Surface Science insights gained, the carbon-based material was manipulated to efficiently store $H(H_2)$[18], with the predicted

structure subsequently observed by more recent experimental findings [19].

Other examples have been reported. [20–23]

SPICE

（スパイス）

" . . . He who controls the spice, controls the universe . . . " [24]

What is " the Spice " ?

Food . . .　Clothing . . .　Shelter . . .

Water . . .　Oil . . .　Electricity . . .

Resources . . .　Materials . . .

Energy . . .　Life . . .

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