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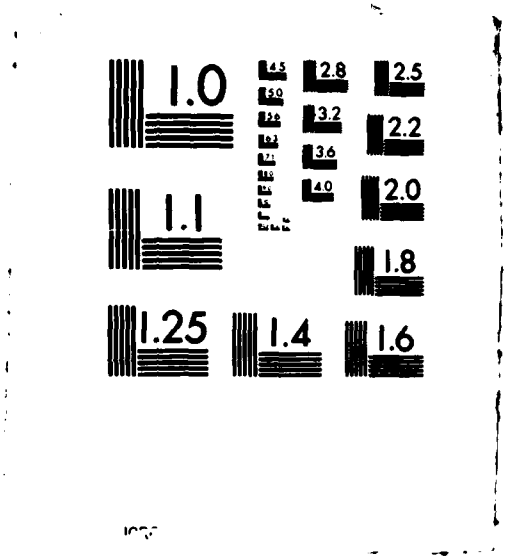
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# Dynamics of a Laser-Irradiated Adatom

by

Sander van Smaalen, André Peremans, Henk F. Arnoldus and Thomas F. George

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The dynamics of an adsorbed atom irradiated by an IR laser in resonance with a single pair of states of the vibrational adbond is studied. Using a non-perturbative treatment for the laser-adbond interaction, a master equation is derived, which governs the time evolution of the populations of the laser-dressed states of the adbond. The effect of resonant heating and laser-induced desorption, as an example of a possible laser-induced surface process, is discussed.

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# Dynamics of a laser-irradiated adatom

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Abstract - The dynamics of an adsorbed atom irradiated by an I.R. laser in resonance with a single pair of states of the vibrational adbond is studied. Using a non-perturbative treatment for the laser-adbond interaction, a master equation is derived, which governs the time evolution of the populations of the laser-dressed states of the adbond. The effect of resonant heating and laser-induced desorption, as an example of a possible laser-induced surface process, is discussed.

## INTRODUCTION

It has been realized for quite a time that laser irradiation can affect or induce chemical reactions [1,2]. Combining this with the common knowledge that surfaces, i.e., catalysts, can influence chemical reactions, then opens the interesting possibility of joining the control by lasers and surfaces, in order to manipulate the occurring chemical reactions in greater detail. A quantitative theoretical treatment of a chemical reaction is very involved. However, the first step in a chemical reaction is the formation of an activated complex. The energy necessary for the formation of the complex can be extracted from the translational and vibrational energy of the reactants or added inert species. Here enters one possibility of applying a laser to modify the reaction, since reacting molecules might acquire the necessary energy to become excited to high vibrational or electronic states from the radiation field. Surfaces alter reactions via the induced modifications of the adsorbed species and by the fact that they restrict the motion of the molecules. The role played by a laser in a surface reaction is more complicated than for gas-phase reactions. The radiation may excite the molecule before adsorption, it may excite the substrate, or it may excite the already adsorbed molecule. We shall only consider the latter mechanism.

Detailed comprehension of laser-induced surface reactions starts with the study of the dynamics of a single adsorbed molecule which is illuminated by a strong coherent field. In this paper we shall focus on recent developments in the theory of the dynamics of vibrationally-excited atoms. Topics to be covered are: How can the laser excite the adatom, which excited states will be populated, and where does the absorbed energy go? Furthermore, the feasibility of laser-induced (resonant) desorption is discussed.

## RELAXATION AND COHERENT EXCITATION

We consider an adsorbed atom on a harmonic crystal, irradiated by a laser, which is tuned into resonance with a single pair of levels of the vibrationally bounded atom [3-5]. Only the motion perpendicular to the surface is taken into account, because the lateral motion (migration over the surface) hardly couples to the field. The adbond is represented by its reduced density operator [6]

$$\sigma(t) = \mathrm{Tr}\ \rho(t) \qquad (1)$$

where the trace is over the quantum states of the crystal and over the radiation states. With standard techniques [6,7], it is easy to show that in absence of a laser, the time evolution of the level populations $P_n(t) = \langle n|\sigma(t)|n\rangle$ is governed by the master equation

$$\frac{d}{dt}P_n(t) = \sum_k \left(a_{kn}\, P_k(t) - a_{nk}\, P_n(t)\right) \qquad (2)$$

where $a_{nk}$ is the rate constant for the transition from level n to level k, and the summation extends over the eigenstates of the adbond. The rate constants are determined by the interaction of the adbond with the substrate. Most extensively studied is the relaxation of the adbond due to the lattice vibrations [3-5]. A transition from state $|n\rangle$ to state $|k\rangle$ is then accompanied by the emission into or the absorption from the substrate of one or

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more phonons. For one-phonon processes the rate constants are given by [4]

$$a_{nk} = |S_{nk}|^2 f(\omega_{nk}) \qquad (3)$$

$$f(\omega_{nk}) = \frac{4\pi^2}{M\hbar} \frac{g(|\omega_{nk}|)}{\omega_{nk}} (\bar{n}(\omega_{nk}) + 1) \qquad (4)$$

where $\omega_{nk} = \omega_n - \omega_k$ denotes the transition frequency, M is the mass of a surface atom, $\bar{n}(\omega) = [\exp(\hbar\omega/kT) - 1]^{-1}$, and $S_{nk}$ is the matrix element of the derivative of the adbond potential. The function $g(|\omega_{nk}|)$ is the density of phonon states. For a transition frequency which is larger than the Debye frequency, $g(|\omega_{nk}|)$ is vanishingly small, and multiphonon transitions have to be taken into account [8]. On metals the coupling with electronic transitions in the conduction band provide another possible channel for relaxation [9,10].

The interaction between the laser and the adbond can be treated analogously to the phonon interactions [5], provided that the intensity is not too high. This yields a similar master equation as Eqn. (2). The transition rates then divide into a phonon part (Eqns. (3) and (4)) and a term due to the interaction with the laser, which represents radiative stimulated transitions. The latter only occurs in the rate constants $a_{ge}$ and $a_{eg}$ for the coupled levels $|g\rangle$ and $|e\rangle$. It is found to be proportional to the laser intensity or, equivalently, to the square of the Rabi frequency

$$\Omega = \hbar^{-1}\underline{\mu}\cdot\underline{E} \qquad (5)$$

where $\underline{\mu}$ is the transition-dipole matrix element, and $\underline{E}$ denotes the amplitude and polarization of the electric field of the laser.

The successive approximations involved can easily be studied with the Zwanzig projection technique [7]. For the time evolution of the reduced density matrix we find the exact equation

$$\frac{d\sigma}{dt} = \frac{1}{i\hbar}[H_s, \sigma(t)] - Tr_b \int_0^t dt' \, K(t,t') \, \rho_b \sigma(t-t') \qquad (6)$$

where the memory kernel K(t,t') contains the Hamiltonian of the adatom, $H_s$, the bath, and their interaction. Here $\rho_b$ equals the density matrix of the bath in thermal equilibrium. The first approximation is to retain in K(t,t') only the lowest-order (second-order) terms in the interaction. This is assumed to be accurate for the phonon coupling and is also expected to be a good approximation for the laser interaction [10]. The second assumption is the Markov approximation [6], which asserts that we can replace $\sigma(t-t')$ by $\sigma(t)$ in Eqn. (6). This can be justified if $\sigma(t)$ does not change much on a timescale on which K(t,t') decays to zero as a function of t' for fixed t. For phonons, this is the characteristic time of the displacement autocorrelation function, which is sufficiently short [11]. For the laser interaction the decay time of K(t,t') is the correlation time of the laser, which is essentially infinite for monochromatic radiation. It was found that for reasonable laser intensities the Markov approximation gives poor results [10]. Then an integrodifferential equation (Eqn. (6)) has to be solved, rather than a master equation.

A non-perturbative approach to the laser-adbond interaction was developed recently [4]. Here the Hamiltonian of the adbond, the laser field and the interaction was diagonalized, and subsequently the phonon damping was taken into account in the usual way. Note that with this alternative treatment the problems with the Markov approximation do not arise. Besides that, the method applies to arbitrary strong irradiances.

Let us denote the eigenstates of the adbond by $|k\rangle$, and in particular by $|g\rangle$ and $|e\rangle$ the two states which are coupled by the laser. Then the eigenstates of the adbond Hamiltonian, the laser field and the interaction are given by $|k\rangle$ for $k \neq e,g$, but $|g\rangle$ and $|e\rangle$ are superposed to yield the so-called laser-dressed states [4,12]. Explicitly we obtain

$$|+\rangle = \sin(\theta/2)|g\rangle + \cos(\theta/2)|e\rangle \qquad (7)$$

$$|-\rangle = \cos(\theta/2)|g\rangle - \sin(\theta/2)|e\rangle \qquad (8)$$

with $\theta = \arctan(\Omega/\Delta)$ and $\Delta$ the detuning of the laser from resonance. Subsequent coupling to the phonon reservoir then results in a master equation for the dressed-state populations. The transition rates assume a more complicated form then given by Eqn. (3). For example, the transition from $|+\rangle$ to $|k\rangle$ with $k \neq e,g$ is given by the rate constant [4]

$$a_{+k} = g_- f(\omega_{+k})|S_{ak}|^2 + g_+ f(\omega_{+k}-\omega_L)|S_{gk}|^2 \qquad (9)$$

with $g_- = \cos^2(\theta/2)$, $g_+ = \sin^2(\theta/2)$, and $\omega_L$ is the laser frequency. A particularly transparent interpretation arises if two energy levels are assigned to a single dressed state. Then $\omega_+$ and $\omega_- - \omega_L$ can be regarded as the eigenvalues of $|+\rangle$, and similarly $\omega_-$ and $\omega_- + \omega_L$ represent the positions of $|-\rangle$, as is illustrated in Fig. 1. The transitions from $|+\rangle$ to $|k\rangle$ then

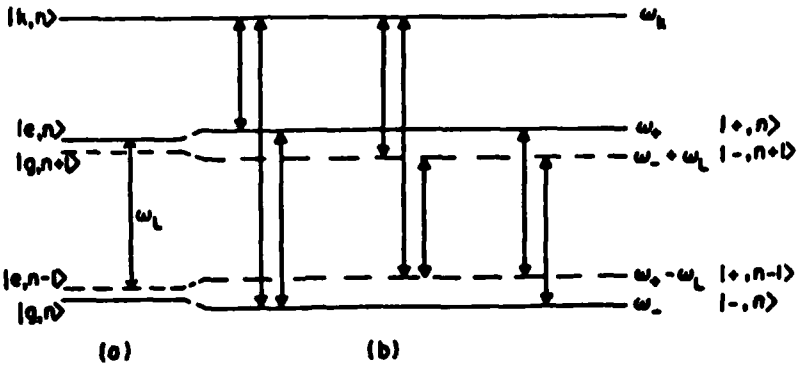


Fig. 1. The energy levels of the adbond plus the laser are represented by the diagram (a) on the left-hand side. The number of photons in the laser mode is indicated by n, and $\omega_L$ is the laser frequency. Diagram (b) shows the energy levels of the eigenstates of the adbond and the laser, including the interaction. The adbond states $|g\rangle$ and $|e\rangle$ are then replaced by the dressed states $|-\rangle$ and $|+\rangle$. The occurring phonon transitions are indicated by arrows. The left-most three transitions persist when the laser is turned off. The other transitions occur only in the presence of the laser. Apart from the change in adbond state and in the number of phonons, they also involve the absorption or emission of photons.

contain transitions from level $\omega_+$ to $\omega_k$, and from level $\omega_+ - \omega_L$ to $\omega_k$. The phonon energies involved are exactly the level distances, as depicted in Fig. 1.

Transitions between either one of the levels $\omega_+$ or $\omega_-$ and a level $\omega_k$ and transitions between $\omega_k$ and $\omega_\ell$ ($k,\ell \neq +$ or $-$) survive when the laser is turned off. Hence they can be interpreted as pure-phonon transitions. The additional transitions also require the absorption or emission of photons. For a low intensity the rate constants for these transitions are proportional to the laser power (one-photon process) or its square (two-photon process). For high intensities they assume saturation values, corresponding to the value $\theta = \pm \pi/2$ in the parameterization of the dressed states.

The coherences between the dressed states evolve independently from the populations. It can be shown that they vanish exponentially in time [4]. The inverse of Eqns. (7) and (8) can be used to derive an equation for the populations of the bare states. It then follows that the equations for the populations are coupled to those for the coherences $P_{ee}$ and $P_{gg}$. This result is different from perturbation theory, which yields a master equation for the bare-level populations, regardless of the time evolution of the coherences.

Sufficiently long after the switch-on of the laser, the system will reach a steady state, for which the reduced density matrix of the adbond remains constant in time. For $dP_{kk}/dt = 0$, the coherences between the bare states can then be eliminated, and a genuine master equation for the bare-level populations arises [4,13]. We find

$$A'_n P_n(\infty) = \sum_k a'_{kn} P_k(\infty). \qquad (10)$$

Here, $A'_n = \sum_k a'_{nk}$ and $a'_{nk} = a_{nk}$, except for $a'_{eg}$ and $a'_{ge}$, which are given by

$$a'_{eg} = a_{eg} + \frac{A_e + A_g}{(A_e + A_g)^2 + 4\Delta^2}\,\Omega^2 \qquad (11)$$

$$a'_{ge} = a_{ge} + \frac{A_e + A_g}{(A_e + A_g)^2 + 4\Delta^2}\,\Omega^2. \qquad (12)$$

Note that in the steady state the Markov approximation for the laser field gives the same result as the non-perturbative approach [10].

## ENERGY FLOW

A phonon transition from state $|k\rangle$ to $|\ell\rangle$ changes the energy of the substrate by an amount of the phonon energy $\hbar|\omega_{k\ell}|$. Phonon absorption lowers the substrate energy, and phonon emission raises its energy content. In equilibrium (and without a laser) the net energy exchange between the adbond and the substrate is zero. In perturbation theory the laser gives rise to transitions between the two levels $|e\rangle$ and $|g\rangle$. With each transition an energy quantum $\hbar\omega_L$ is exchanged between the laser and the adbond. In the non-perturbative approach the absorption/emission of photons is incorporated in the diagonalization, resulting in the appearance of dressed states, and only the phonon transitions remain explicitly present (Fig. 1).

The populations are time independent in the steady state, as is the adbond energy. The only effect of the transitions between the adbond states is then to carry a net energy flow from the laser into the substrate, a process which is called resonant heating [14]. The energy which is absorbed by the crystal per unit of time is given by [13,15,16]

$$\frac{dW}{dt} = \hbar\omega_L \frac{A_e + A_g}{(A_e + A_g)^2 + 4\Delta^2} \times \Omega^2 (P_g(\infty) - P_e(\infty)). \quad (13)$$

For low intensities the populations of $|g\rangle$ and $|e\rangle$ are hardly altered by the radiation, and hence it follows from Eqn. (13) that the energy flow is proportional to the intensity $\Omega^2$. It can be shown from Eqns. (10) and (13) that the quantity $\Omega^2(P_g(\infty)-P_e(\infty))$ becomes independent of $\Omega^2$, and consequently the energy flow saturates. An upper bound for the energy flow is [15]

$$\frac{dW}{dt} \leq \hbar\omega_L A_e P_e(\infty) \quad (14)$$

which exhibits the saturation effect. The equality holds in the low-temperature limit.

From expressions (11) and (12) it follows immediately that we can interpret the second terms on the right-hand side as the rate constants $a_r$ for stimulated photon absorption and emission in the $|e\rangle$ - $|g\rangle$ transition. Then the absorption rate equals $a_r P_g(\infty)$, and stimulated emissions, which accompany an $|e\rangle \rightarrow |g\rangle$ transition, occur at a rate $a_r P_e(\infty)$. The effective number of transitions from $|g\rangle$ to $|e\rangle$ per unit of time then becomes $a_r(P_g(\infty) - P_e(\infty))$, and multiplication by the photon energy $\hbar\omega_L$ then yields the result (13). This identification elucidates the appearance of the various factors in the expression for dW/dt.

## CONCLUSIONS

A theory is presented for the dynamics of an adatom, irradiated by an intense laser, which is in near resonance with a single pair of levels of the vibrational adbond. A master equation for the time evolution of the populations of the laser-dressed states of the reduced density matrix of the adbond is derived. The transitions between the states can be interpreted as phonon transitions between the dressed levels of the adbond. Stimulated radiative transitions are incorporated in the transformation to dressed states.

The phonon transitions between the adbond states give rise to an energy flow from the laser into the substrate. Even in the steady state (where the level populations are time independent) the energy flow assumes a non-zero value. For low intensities the energy flow is proportional to the laser intensity, whereas for high intensities saturation occurs. This is illustrated by the derivation of an upper bound (Eqn. (14)). It follows that the energy flow is limited by the excited-level population, multiplied by its decay constant. This clearly exhibits that, when the laser is used to maintain the adbond in an excited state, a fast heating of the crystal is inevitable. Moreover, the ratio of the excited-level population and the energy flow (considered as a measure of the efficiency of one process over the other) appears to be independent of the laser power.

To be specific, let us compare the efficiency of a laser-induced surface process and the energy flow. A measure of this is the number of photons which is necessary to sustain the desired process, divided by the number of photons which heat the substrate. As an example we have studied laser-induced desorption [15]. It is found that for low temperatures this ratio is independent of the laser power, and much smaller than unity, indicating that laser-induced (resonant) desorption cannot be expected to be a very efficient process.

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