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by

Jui-teng Lin and Thomas F. George

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On the Synergistic Effects of Laser/Phonon-Stimulated Processes: A Master Equation Approach*

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Abstract

The synergistic effects of an adspecies/surface system influenced by a coherent laser field and an incoherent thermal phonon field are theoretically investigated. A generalized master equation is derived in the Schrödinger-Markoff picture and the total stimulated transition rates are decomposed into three parts: thermal phonon, laser and the interference terms. The energy transfer dynamics is pictured via the evolution equation of the average excitation of the active mode. A random phase of the off-diagonal matrix elements induced by the laser-stimulated surface processes is introduced. Finally, possible applications of the laser/phonon-stimulated master equation are discussed.

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I. Introduction

Laser-matter interactions of heterogeneous systems have been recently studied and the possible applications/implications of laser-stimulated transient effects, such as materials processing and evaluation, semiconductor lithography and microelectronics and the development of new lasers, motivate the fundamental studies in this newly-developed field.

Laser-stimulated surfaces processes (LSSP) such as migration, desorption, adsorption, ionization, recombination and predissociation usually combine the laser transient effects and the subsequent energy relaxation of the excited adspecies.[1] Due to the multiple time scales of the heterogeneous system, LSSP may be grouped into several types, e.g., mode-selective, adspecies-selective, migrational-selective and local-heating, etc. These selective and nonselective aspects are characterized not only by the coherent properties of the laser radiation, frequency, polarization, intensity and duration, but also by the pumping rate compared to the relaxation or coupling rates among inter- and intra-modes, e.g., electron-phonon interaction, phonon-phonon coupling and active-mode-phonon coupling.[2]

In a previous paper,[1] we studied the desorption dynamics by means of a laser-stimulated rate equation, where the thermal effects and the laser excitation were assumed to be two independent driving forces, and we focused on a cold surface where the laser excitation plays the dominant role. In the present paper, we shall study the synergistic effects of laser/phonon excitation where both the laser and thermal phonons may play equally important roles.

II. Equation of Motion in the Schrödinger - Markoff Picture

We consider a model system consisting of a group of species adsorbed on a solid surface and subject to laser radiation. The total vibrational Hamiltonian of the system may be expressed in the following form:

$$H = H_A + H_B + H_{AA} + H_{BB} + H_{AB} + H_{AF} + H_{BF}. \qquad (1)$$

H_A and H_B are the unperturbed Hamiltonian of the optically-active (A) mode and the phonon bath (B) modes, respectively; H_{AA} and H_{BB} represent the intramode couplings of the active and phonon bath modes, respectively; H_{AB}, H_{AF} and H_{BF} represent the A and B mode coupling, the A mode and laser field and the B mode and laser field interactions, respectively. For the situation where the vibrational spectrum is partitioned into a high-frequency region consisting of the A modes resonantly excited by a laser, we may neglect H_{BF} since the low-frequency B modes are far off-resonant with respect to the laser radiation. A second-quantization representation of the total Hamiltonian was previously shown in Ref. 3 consisting of the combinations of the ladder operators $a^\dagger(a)$ for the active mode and $b^\dagger(b)$ for the phonon modes. For example, $H_{AF} = V(t)\ (a^\dagger + a)$ where $V(t)$ is proportional to the electric field of the radiation.

In the Schrödinger - Markoff picture (SMP), the equation of motion for the reduced density operator $S = Tr_B \rho$, traced over the phonon modes, with the total Hamiltonian given by eq. (1) is found to be [3,4]

$$\frac{\partial S}{\partial t} = -i[\omega_{eff} a^{\dagger}a, S] - iV(t)\left([a^{\dagger}, S] + [a, S]\right)$$

$$+ \frac{\gamma_1}{2}\left(2aSa^{\dagger} - a^{\dagger}aS - Sa^{\dagger}a\right) + \gamma_1 \bar{n}_j \left(a^{\dagger}Sa + aSa^{\dagger} - a^{\dagger}aS - Saa^{\dagger}\right), \tag{2.a}$$

with

$$\omega_{eff}(t) = \omega_0 - \varepsilon^*(a^{\dagger}a+1) + \phi(t), \tag{2.b}$$

$$\gamma_1 \approx 2\pi \sum_{\nu} |K_{\nu}|^2 n_{\nu}\, \delta(\omega_0 - 2\varepsilon^* n - \sum_j \omega_j), \tag{2.c}$$

$$n_{\nu} = \prod_j (\bar{n}_j + 1) - \prod_j \bar{n}_j , \tag{2.d}$$

where $(\omega_0 - \varepsilon^*)$ and ω_j are the fundamental frequency of the active mode and the j-th phonon mode with the vibrational quantum number n and $\bar{n}_j$, respectively. The important features of the above equation of motion in SMP are: (i) the many-body effects of the thermal phonon modes are effectively reduced to a multiphonon damping factor γ_1 which is characterized by the mean occupation number $\bar{n}_j$ (at temperature T), the coupling strength K_{ν} and the delta function $\delta(\omega_0, \omega_j)$ for conserving energy; (ii) the active mode is treated as a stochastic anharmonic quantum oscillator with an effective frequency ω_{eff} given by an anharmonic correction $\varepsilon^*(a^{\dagger}a+1)$ and a random phase or frequency shift $\phi(t)$;[5] (iii) the dephasing effects of the active mode may be caused by a laser/surface-induced mechanism such as thermal fluctuation of the active dipole moment, migration or dislocalization of the adspecies, phase difference between the laser (coherent) and phonon (incoherent) fields coupled to the active mode, random imperfection of the solid surface and finally, the effects of the intramode couplings H_{AA} and H_{BB}.

The evolution equation of the density matrix elements for the vibrational states (m,n),[4,6]

$$\frac{\partial S_{mn}}{\partial t} = \frac{1}{i\hbar} \sum_k (H_{mk} S_{kn} - S_{mk} H_{kn}) - \gamma_{mn} (S - \bar{S})_{mn} , \qquad (3)$$

can be written for our system, described by the operator equation [eq.(2)], as

$$\begin{aligned}\frac{\partial S_{mn}}{\partial t} = &-i \; [(\omega_0-\varepsilon^*+\phi)(m-n)-\varepsilon^*(m^2-n^2)-i\gamma_0] \; S_{mn} \\ &-i\frac{\mu' E(t)}{\hbar} \Big[\sqrt{m}\, S_{m-1,n} - \sqrt{n+1}\, S_{m,n+1} + \sqrt{m+1}\, S_{m+1,n} - \sqrt{n}\, S_{m,n-1} \Big] \\ &+ \frac{\gamma_1}{2} \Big[2 \sqrt{(m+1)(n+1)}\, S_{m+1,n+1} - (m+n) S_{m,n}) \Big] \\ &+ \gamma_1 \bar{n}_j \Big[\sqrt{mn}\, S_{m-1,n-1} + \sqrt{(m+1)(n+1)}\, S_{m+1,n+1} - (m+n+1) S_{m,n} \Big], \qquad (4)\end{aligned}$$

where μ' is a quantity proportional to the derivative of the dipole transition matrix element between ground and first-excited states, $E(t) = E_0 \cos\omega t$ is the electric field of the linearly polarized laser, and $\gamma_0 = \frac{1}{2}(\gamma_m+\gamma_n)$ is the mean natural width for the radiative decay from state m to state n.

III. Laser/Phonon-Stimulated Master Equation

The evolution equation of the energy population is given by the diagonal term, $P_n \equiv S_{nn}$ in eq. (4),

$$\begin{aligned}\frac{dP_n}{dt} = &\gamma_1 \Big[(n+1)(\bar{n}_j+1)\, P_{n+1} + n\bar{n}_j\, P_{n-1} - (2n\bar{n}_j+n+\bar{n}_j)\, P_n \Big] - \gamma_n P_n \\ &- \big(i\mu' E(t)/\hbar\big) \Big[\sqrt{n} (S_{n-1,n} - S_{n,n-1} - \sqrt{n+1}\, (S_{n,n+1} - S_{n+1,n}) \Big] , \qquad (5)\end{aligned}$$

where $P_{n\pm 1} = S_{n\pm 1, n\pm 1}$ and the off-diagonal terms $S_{n,\, n\pm 1}$ can be calculated iteratively by means of the equation[6]

$$\frac{\partial S^{(j)}_{mn}}{\partial t} = \frac{1}{i\hbar}\,[H,\, S]^{(j-1)}_{mn} - \gamma_{mn}\,(S-\bar{S})^{(j)}_{mn} \,, \tag{6}$$

with the condition $S^{(0)}_{mn}(t) = S^{(0)}_{mn}(t)\,\delta_{mn}$ and $S^{(0)}_{mn}=0$ for $m\neq n$, we obtain

$$S^{(1)}_{n,n+1}(t) = -\left(\frac{i}{\hbar}\right)\sqrt{n+1}\int_0^t u'E(t')\exp\left[\int_0^{t'} \Omega(t'')(t'-t'')dt''\right] \times$$

$$[P^{(0)}_{n+1}(t')-P^{(0)}_{n}(t')]\;dt' \,, \tag{7.a}$$

where

$$\Omega(t)=i[\omega_0-2\varepsilon^*(n+1) +\phi(t)] - (2n+1-4\bar{n}n+4\bar{n})\gamma_1/2 -\gamma_0 \,. \tag{7.b}$$

Note that $\Omega(t)$ contains a random phase factor $\phi(t)$. By assuming a "white noise", $<\phi(t)>=0$, and a Markoffian correlation, $<\phi(t)\phi(t')> = 2\gamma_2\delta(t-t')$, we may evaluate the ensemble-averaged quantity [over the random variable $\phi(t)$], as

$$< \exp\left[i\int_0^{t'} \phi(t'')(t'-t'')dt''\right]> \approx \sum_{n=0}^{\infty} \frac{(-\gamma_2 t)^n}{n!} = e^{-\gamma_2 t} \,, \tag{8}$$

which is governed by the dephasing (T_2) factor γ_2. Equation (7.a) then takes the form

$$<S^{(1)}_{n,n+1}(t)>=-\left(\frac{i}{\hbar}\right)\sqrt{n+1}\int_0^t u'E(t')\exp[\bar{\Omega}_1][<P^{(0)}_{n+1}(t')>-<P^{(0)}_{n}(t')>]dt' \,, \tag{9.a}$$

$$\bar{\Omega}_1(t-t') = \Big(i[\omega_0-2\varepsilon^*(n+1)]-\Gamma_1 \Big)(t-t') \tag{9.b}$$

$$\Gamma_1 = \gamma_1(2n+1+4n\bar{n}+4\bar{n})/2 + \gamma_2 + \gamma_0 \,. \tag{9.c}$$

By the same procedure, we find

$$<S^{(1)}_{n-1,n}(t)> = \left(\frac{i}{\hbar}\right)\sqrt{n} \int_0^t \mu' E(t') \exp[\bar{\Omega}_2] [<P^{(0)}_{n-1}(t')> - <P^{(0)}_n(t')>] dt', \quad (10.a)$$

$$\Omega_2(t-t') = \Big(i[\omega_0 - 2\varepsilon^* n] - \Gamma_2\Big)(t-t') , \quad (10.b)$$

$$\Gamma_2 = \gamma_1 (2n-1+4n\bar{n})/2 + \gamma_2 + \gamma_0 . \quad (10.c)$$

Substituting eqs. (9) and (10) into eq. (5), we obtain the ensemble-averaged generalized maser equation (GME), neglecting the ensemble-averaged notation <...>, for the first-order solution,

$$\frac{dP_n}{dt} = \gamma_1 \ [(n+1)(\bar{n}_j+1)P_{n+1} + n\bar{n}_j P_{n+1} - (2n\bar{n}_j + n + \bar{n}_j)P_n] - \gamma_n P_n$$

$$\int_0^t dt' \Big(W_{n,n-1}(t-t')[P_{n-1}(t') - P_n(t')] + W_{n,n+1}(t-t')[P_{n+1}(t') - P_n(t')] \Big), \quad (11)$$

with the time dependent transition rates

$$W_{n,n+1}(t-t') = 2(n+1)(\mu'/\hbar)^2 E(t)E(t') e^{-\Gamma_1(t-t')} \cos[(\omega_0 - 2\varepsilon^* n - 2\varepsilon^*)(t-t')], \quad (12.a)$$

$$W_{n,n-1}(t-t') = 2n(\mu'/\hbar)^2 E(t)E(t') e^{-\Gamma_2(t-t')} \cos[(\omega_0 - 2\varepsilon^* n)(t-t')]. \quad (12.b)$$

The above GME may be reduced to the Pauli master equation (ME) by using the fact that the population inversion $[P_{n\pm1}(t) - P_n(t)]$ is usually a slowly varying function[4] in the time of $\Gamma^{-1}_{1,2}$ and hence can be factored outside the integral in eq. (11). Taking an average over the cycle of of the field, $2\pi/\omega$, and assuming a near resonance condition such that the integrated transition rates $W_{n,n\pm1}(t)$ become time-independent, and we obtain a simple laser/phonon-stimulated master equation (LPME)

$$\frac{dP_n}{dt} = \left(W^P_{n,n+1}+W^{LP}_{n,n+1}\right)\left(P_{n+1}-P_n\right) + \left(W^P_{n,n-1}+W^{LP}_{n,n-1}\right)\left(P_{n-1}-P_n\right)$$

$$+ \left(R_{n,n+1}P_{n+1} - R_{n,n-1}P_n\right) - \gamma_n P_n \,, \qquad (13)$$

where the stimulated transition rates are given by

$$W^P_{n,n+1} = \gamma_1 \bar{n}_j (n+1) \,, \qquad (14.a)$$

$$W^P_{n,n-1} = \gamma_1 \bar{n}_j n \,, \qquad (14.b)$$

$$W^{LP}_{n,n+1} = \frac{(n+1)}{2}\left(\frac{\mu' E_0}{\hbar}\right)^2 g_1(\bar{\Delta}) \,, \qquad (14.c)$$

$$W^{LP}_{n,n-1} = \frac{n}{2}\left(\frac{\mu' E_0}{\hbar}\right)^2 g_2(\bar{\Delta}) \,, \qquad (14.d)$$

$$g_{1,2}(\bar{\Delta}) = \Gamma_{1,2} / (\bar{\Delta}^2_{1,2}+\Gamma^2_{1,2}) \,, \qquad (14.e)$$

$$\bar{\Delta}_1 = \omega_0 - 2\varepsilon^*(n+1) - \omega \,, \qquad (14.f)$$

$$\bar{\Delta}_2 = \omega_0 - 2\varepsilon^* n - \omega \,. \qquad (14.g)$$

The phonon-induced relaxation rates $R_{n,n+1}=(n+1)\gamma_1$ and $R_{n,n-1}=n\gamma_1$ describe the irreversible energy dissipative processes for excited-species on a cold surface. The final term in eq.(13) describes the spontaneous radiative decay of the n-th vibrational levels.

IV. Discussion and Conclusion

It is shown in the LPME [eq.(10)] that the active mode is influenced by a coherent laser field and an incoherent thermal phonon field with total stimulated transition rates ($W^P_{n,n\pm1} + W^{LP}_{n,n\pm1}$) which may be further decomposed into three components ($W^P_{n,n\pm1}+W^L_{n,n\pm1}-W^I_{n,n\pm1}$), corresponding to phonon-stimulated, laser-stimulated and the "interference" transition rates, respectively. $W^P_{n,n\pm1}$ is given in eq.(14) and

$$W^L_{n,n+1} = \left(\frac{n+1}{2}\right)\left(\frac{\mu' E_0}{\hbar}\right)^2 \mathcal{L}_1 \,, \qquad (15.a)$$

$$W^L_{n,n-1} = \left(\frac{n}{2}\right)\left(\frac{\mu' E_0}{\hbar}\right)^2 \mathcal{L}_2 , \qquad (15.b)$$

$$\mathcal{L}_{1,2} = \gamma_0 \Big/ (\bar{\Delta}^2_{1,2} + \gamma_0^2), \qquad (15.c)$$

are simply the surface-free laser pumping rates characterized by the Rabi frequency and a Lorentzian with FWHM equal to the natural level width γ_0. The interference term is related to W^L and W^P by

$$W^I_{n,n\pm1} \approx \left(W^L_{n,n\pm1}\right)^2 \left(\gamma_2 + W^P_{n,n\pm1} N_{1,2} \Big/ \bar{n}_j (n+1)\right) , \qquad (16)$$

for $\gamma_0 >> N_{1,2}\, \gamma_1 + \gamma_2, \bar{\Delta}_{1,2}$.

For a monatomic lattice with phonon frequency ≈ Debye frequency (ω_D), the multiphonon coupling factor [eq. (2.c)] may be approximated as[7] $\gamma_1 \approx \gamma_1(0) T^{N-1} [\exp(\hbar\omega_0 /kT) - 1]$, with the zero temperature factor $\gamma_1(0) \sim \alpha^{2N}$, where $N \approx (\omega_0 - 2\varepsilon^* n)/\omega_D$ in the order parameter and $0<\alpha<<1$. For a highly excited active mode, the multiphonon order parameter, N, decreases as the anharmonic correction $2\varepsilon^* n$ increases and in turn results a stronger coupling factor γ_1. A stronger coupling factor γ_1 results in a significant interference term W^I as shown in eqs. (16). Therefore, the total transition rate in the LPME can be approximated by the sum of W^P and W^L only when W^I is negligible, i.e., $(N_{1,2}\gamma_1 + \gamma_2) << \gamma_0$.

To give a clearer picture of the synergistic effects of laser/phonon excitation processes, we consider the average excitation of the active mode, $<n_A> \equiv <<a^\dagger a>> = <\mathrm{Tr}(S a^\dagger a)>$, where $<< \cdots >>$ denotes an ensemble average over the A and B modes. The evolution of $<n_A>$ can be obtained from eq. (2) as

$$<n_A> = [i\mu' E(t)/\hbar] [<<a>> - <<a^\dagger>>] + \gamma_1 \bar{n}_j - \gamma_1 <n_A> , \qquad (17)$$

which shows that the active mode is pumped by two forces, a laser field $\mu' E(t)$ and a thermal phonon field $\gamma_1 \bar{n}_j$, while it is damped by a multiphonon relaxation rate γ_1, the last term in eq. (17). For the case of no laser field, $E(t)=0$, $\langle n_A \rangle$ reaches the steady-state value $\bar{n}_j$ and is in thermal equilibrium with all the other modes. With laser radiation, a nonequilibrium state where the active mode has a higher vibrational energy than the others is provided, and selective excitation is possible in a time scale shorter than that of the multiphonon relaxation γ_1^{-1}.

In conclusion, we propose some possible applications of LPME [eq. (13)] where the laser and the surface play equally important roles: (i) solving the LPME for P_n by, e.g., generating function method, enables us to calculate physical quantities such as the desorption probability[1] and the desorption rate (or the mean first passage time);[8] (ii) the adspecies/surface spectrum governed by the correlation function of the off-diagonal matrix elements [eqs. (9) and (10)] will provide us surface dynamical information such as the surface coverage and the migration rate of the adspecies through the dephasing broadening of the spectrum γ_2 which reflects the collisional effects; (iii) replacing the vibrational population function by the lattice site probability, the LPME in eq. (13) then may be used to describe the dynamical motion of the adspecies on the solid surface in which the vibrational pumping rates may be regarded as the lattice hopping rates.[9]

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