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Journal of Photochemistry and Photobiology A: Chemistry 180 (2006) 241–247
Journal of Photochemistry and Photobiology A:Chemistry
www.elsevier.com/locate/jphotochem

Squeezing the ground vibrational state of diatomic molecules

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Available online 24 March 2006

Abstract

We study the squeezing of minimal width vibrational wave packets of diatomic molecules, like Na_2, by using several laser schemes that couple the ground and excited electronic configurations of the molecule. The different schemes imply diabatic and adiabatic laser transformations, or a combination of both, whose efficiency and required physical resources are compared and analyzed.
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Keywords: Quantum control; Femtosecond laser pulses; Molecular squeezing; Wave packet dynamics

1. Introduction

Many molecular properties stem directly from the vibrational wave function, thus the control of the precise position and width of the wave packet [1,2] has played such a fundamental role in quantum control of molecules with laser pulses [3,4]. In this contribution we focus on minimizing the width of the wave packet, what is usually called molecular squeezing. The extreme spatial localization of a quantum motion poses an interesting limit to observe the quantum uncertainty relations, and may have important applications in time-dependent spectroscopy (retrieving potential energy data [5–7]) and quantum dynamics (influencing the cross sections in photodissociation and reactive scattering events [3,4]).

The majority of the physical mechanisms suggested for laser-assisted vibrational squeezing are based on the properties of the quantum dynamics on excited states assuming first-order perturbation regime [8–12], or on the direct application of optimal control theory [13,14]. Recently, we have proposed a set of different physically motivated mechanisms [15–17], which are based on the properties of the following set of adiabatic or diabatic (i.e. sudden) laser-induced transformations: adiabatic squeezing (AS), adiabatic stretching or widening (AW), diabatic or dynamic squeezing (DS) and diabatic widening (DW).

The three basic schemes use one or, two or a sequence of transformations which can be summarized in the following processes:

$$\text{AS scheme}: \ V_1(x) \xrightarrow{\text{AS}} U_s(x) \tag{1}$$

$$\text{ISS scheme}: \ V_1(x) \xrightarrow{\text{DW}} V_2(x) \xrightarrow{\text{DS}} V_1(x) \tag{2}$$

$$\text{ADS scheme}: \ V_1(x) \xrightarrow{\text{AW}} U_w(x) \xrightarrow{\text{DS}} V_1(x) \tag{3}$$

The first scheme, named adiabatic squeezing [15], implies the preparation of a squeezed light-induced molecular potential (LIP) [18], $U_s(x)$, by using several laser fields, where the population is adiabatically transferred. The adiabatic passage via light-induced potentials is a general method devised for population transfer [19] and bond enlargement [20]. In all previous cases the width of the wave function is stretched during the dynamics, whereas in the AS scheme the wave function must be squeezed. The overall effect of the AS transformation is the state selective transition from the initial wave function, $\phi_{0,1}(x)$, to the narrower ground vibrational state of $U_s(x)$, $\phi_{0,s}(x)$. The physical origin of the squeezing is the combined Stark-shift pressure of at least two pulses.

The second scheme, named iterative stretching-squeezing (ISS) [16] implies a two-step process than can be iterated: In the first step (DW), the initial wave function is pumped in the sudden limit to an excited and wider electronic state $V_2(x)$, where it oscillates and breathes; then, when the wave packet is maximally stretched (or widened) in $V_2(x)$, the second step (DS) uses another ultrashort pulse that dumps the population to $V_1(x)$ where it will again oscillate and breathe, leading to dynamical squeezing. Physically, the squeezing is caused by the phase accumulation generated by the mismatch in the

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1010-6030/$ – see front matter © 2006 Published by Elsevier B.V.
doi:10.1016/j.jphotochem.2006.02.006

electronic forces exerted in the different molecular potentials at different iterations.

The third scheme, called adiabatic–diabatic squeezing (ADS) [17], implies a two-step process which cannot be easily iterated: The first step (AW) is similar to the AS scheme except that now the prepared LIP, $U_w(x)$, must be wider or stretched with respect to $V_1(x)$, so that the state-selective population transfer $\phi_{0,1} \rightarrow \phi_{0,w}$ drives all the population to a wider wave function; then the second step involves the second part of the ISS scheme, the sudden dump and the subsequent dynamical squeezing in $V_1(x)$. The physical origin of the squeezing is the same as in the ISS scheme, but here the different electronic forces work on a single step, typically by virtual transitions (Stark-shift), before the wave packet is released.

In both ISS and ADS, each step can be performed by a single pulse or via multi-photon processes using several pulses. The second case is typical of ADS, and in fact is needed if the adiabatic step leads to a wider LIP [17]. However, simpler applications are possible when $U_w(x)$ is just one (wider) excited electronic potential, e.g. $V_2(x)$. Therefore, we distinguish the ADS scheme via LIPs from the ADS via electronic potentials. It can be easily proved that in the ideal harmonic oscillator model, the squeezing in the last case is smaller than that achieved in a single iteration of the ISS scheme. However, this is not necessarily true when using the real potential curves because of the effect of anharmonicity.

There are different pros and cons for preferring one of the schemes. The AS scheme essentially demands intense and (moderately) long pulses, while the ISS scheme requires large bandwidth and time-delay control. The ADS scheme requires intense pulses and time-delay control, especially to determine when the wave packet will be squeezed [16]. All of them need pre-aligned molecules, a requirement which perhaps can be easier to meet using adiabatic transitions [21].

On the other hand, the efficiency of squeezing is quite smaller in AS, but depending on the nature of the molecule (its mass) and the structure of the electronic potentials, ISS can be more efficient (or not) than ADS. Although there are relatively good analytical estimates for the maximal squeezing that can be achieved in harmonic potentials, the effects of anharmonicity are in general too important to neglect, and make very difficult to predict which strategy will provide better results without resorting to numerical simulation.

In this work we provide a numerical analysis of maximal squeezing in Na_2, starting from the ground vibrational state of the ground electronic potential $\phi_{0,1}(x)$ (the minimal width stationary wave function) and using two excited electronic states as a source of squeezing. We show results using all the previous schemes and we find that essentially the same results are obtained by dynamical squeezing in all schemes.

The results of this paper assume that the molecule is oriented along the laser field, so that vibrational squeezing may benefit from recent advances in angular squeezing and molecular alignment [21]. In fact, it is interesting to observe clear analogues between the three proposed mechanisms of vibrational squeezing and the different strategies of molecular alignment [21].

2. Molecular model

For the molecular model we will use the ab initio molecular potentials $X_1\Sigma_g$, $A^1\Sigma_u$ and $1^1\Pi_g$ of the Na_2 [22] (herein V_1, V_2 and V_3, respectively). Expanding the wave function in the three electronic wave packets, the time-dependent Schrödinger equation (TDSE) of the system in the rotating wave approximation (RWA) is

$$\frac{\partial}{\partial t}\begin{pmatrix}\psi_1\\ \psi_2\\ \psi_3\end{pmatrix} = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2} I + \begin{pmatrix} V_1(x)+\hbar w_1(t) & \frac{-\mu_1\varepsilon_1(t)}{2} & 0 \\ \frac{-\mu_1\varepsilon_1(t)}{2} & V_2(x) & \frac{-\mu_2\varepsilon_2(t)}{2} \\ 0 & \frac{-\mu_2\varepsilon_2(t)}{2} & V_3(x)-\hbar w_2(t) \end{pmatrix}\begin{pmatrix}\psi_1\\ \psi_2\\ \psi_3\end{pmatrix} \tag{4}$$

where I is the unit matrix. We will assume the Condon limit using the equilibrium configuration dipole moments: $\mu_1 = 4$ au, $\mu_2 = -2.5$ au [22]; $w_j(t) = w_{0,j} + \lambda\Delta t/2$ are the pulse frequencies, with carrier pulse $w_{0,j}$, which can be chosen so that the intermediate potential is off resonance (as in the AS and ADS scheme), and temporal chirp λ, which we use in the adiabatic passage between two electronic states. The frequency chirp crosses $w_{0,j}$ at half the pulse duration. The fields are given by $\varepsilon_j(t) = \varepsilon_j S_j(t)$ where the pulse shapes are parametrized as a three step function with switch on $\varepsilon_j(t) = \varepsilon_j \sin^2(\pi t/2\tau_{1j})$ between $0 \le t < \tau_{1j}$; plateau $\varepsilon_j(t) = \varepsilon_j$ between $\tau_{1j} \le t < \tau_{2j}$, and switch off $\varepsilon_j(t) = \varepsilon_j \cos^2(\pi(t-\tau_{2j})/2\tau_{3j})$ between $\tau_{2j} \le t < \tau_{3j}$. For the ultrashort pulses we make $\tau_{1j} = \tau_{3j}$ and $\tau_{2j} = 0$, so that the width of the pulse is given by $\tau = 2\tau_{1j}$. We solve the TDSE numerically with standard propagation routines. When required, results in the adiabatic representation are obtained by diagonalizing the Hamiltonian potential energy matrix and calculating the wave packet carried by this representation, from which the adiabatic wave packet's width $\langle\Delta x(t)\rangle$ can be measured.

In the paper, all the wave packet magnitudes are in scaled units: The width is scaled with respect to the initial (minimal width) vibrational wave function ($\langle\Delta x\rangle \rightarrow \langle\Delta x(t)\rangle/\langle\Delta x_0\rangle$), whereas the position is given as a displacement from the ground equilibrium configuration, x_0, scaled by the distance d between the initial position and the equilibrium configuration of the $A^1\Sigma_u$ potential [$\delta x = (\langle x(t)\rangle - x_0)/d$].

3. Results comparing the ISS and ADS schemes

In ISS the physical source of dynamical squeezing comes from the accumulated phase induced by the different force constants of the potentials between which the wave packet is transferred. The ISS recipe is clear [16]: switch the wave packet to a different potential, where it oscillates and breathes, wait for maximal stretching and then switch it back to the initial potential, where it oscillates and squeezes. One can wait for maximal squeezing and iterate again. Maximal squeezing

depends on previously having achieved maximal stretching. In the simplest harmonic oscillator model the degree of maximal squeezing achieved at the n iteration is given by

$$\frac{\langle \Delta x^{(n)} \rangle_{\min}}{\langle \Delta x_0 \rangle} = \gamma^{-n} \tag{5}$$

which depends on the ratio of harmonic frequencies, $\gamma = w_1/w_2$ (w_j are the vibrational quanta in V_1 and V_2, respectively, which should not be confused with the pulse carrier frequencies). The ISS scheme requires the use of ultrashort pump and dump pulses, to drive the sudden vertical transitions between the potentials at the appropriate Franck–Condon windows. It is known that the efficiency of these vertical transitions is very dependent on the energy spread of the wave packet (which can be observed in the absorption or emission spectra) and thus shorter pulses (larger bandwidth) will be needed in lighter molecules. For the initial pump pulse, for instance, the maximum duration of the pulse should be [16]

$$\sigma \sim \frac{\alpha \hbar}{m w_2^2 d \langle \Delta x_0 \rangle} \tag{6}$$

where m is the reduced mass of the molecule, d the spatial separation between the equilibrium configurations of V_1 and V_2, and α is an adimensional adjustable parameter that depends on the desired yield of population transfer (typically $\alpha \sim 2$–4 for excellent population transfer). For the $X^1\Sigma_g$ and $A^1\Sigma_u$ potentials in Na_2, Eq. (5) provides a minimum pulse timewidth of around 5 fs for very effective population transfer (better than 95%). Since the wave packet energy spread increases at successive iterations, the scheme cannot be extended beyond this first pump–dump sequence in Na_2. In fact, the problem is not only the wave packet vertical transitions (the source of linear phase accumulation) but the laser free "horizontal" displacement (the phase difference realization leading to squeezing) because the anharmonicity typically causes parabolic phase distortion which leads to wave packet spreading. However, during certain time the same anharmonicity can increase the squeezing beyond the harmonic limit.

Fig. 1(a) is a representation of the ISS dynamics showing the initial wave function, the maximally stretched wave packet in V_2 before the dump pulse, and the maximally squeezed wave packet in V_1 after the first (and final) iteration. The chosen fields have $\tau = 5$ fs with peak amplitude $\varepsilon_0 \approx 3.9 \times 10^{-3}$ au (implying peak intensities around 0.5 TW/cm^2) delayed by 200 fs. The dynamics of the process is shown in Fig. 2 where we calculate the average observables (average scaled displacement $\delta x(t)$ and width $\langle \Delta x(t) \rangle$), and the electronic populations and we show the laser pulses. The maximal squeezing is 39% [23], which improves the results assuming a harmonic model for the molecular potentials after the first iteration, $\langle \Delta x^{(1)} \rangle_{\min}/\langle \Delta x_0 \rangle = \gamma^{-1} \sim 0.74$. In fact, the wave packet stretches considerably more on V_2 than in the harmonic limit, and then squeezes also more in V_1. For the next iteration the wave packet could be pumped again using a 2.5 fs laser pulse, but the Franck–Condon window excites the population above the dissociation energy in V_2, so that no further squeezing can be achieved.

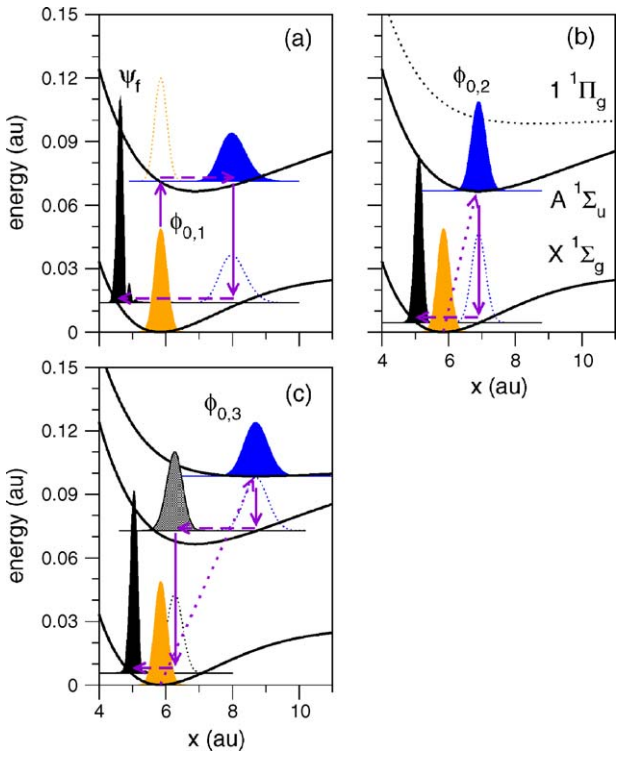

Fig. 1. Sketches of the ISS scheme (a) and of the ADS scheme via one photon excitation to V_2 (b) and via two-photon excitation to V_3 (c), showing snapshots of the wave packets.

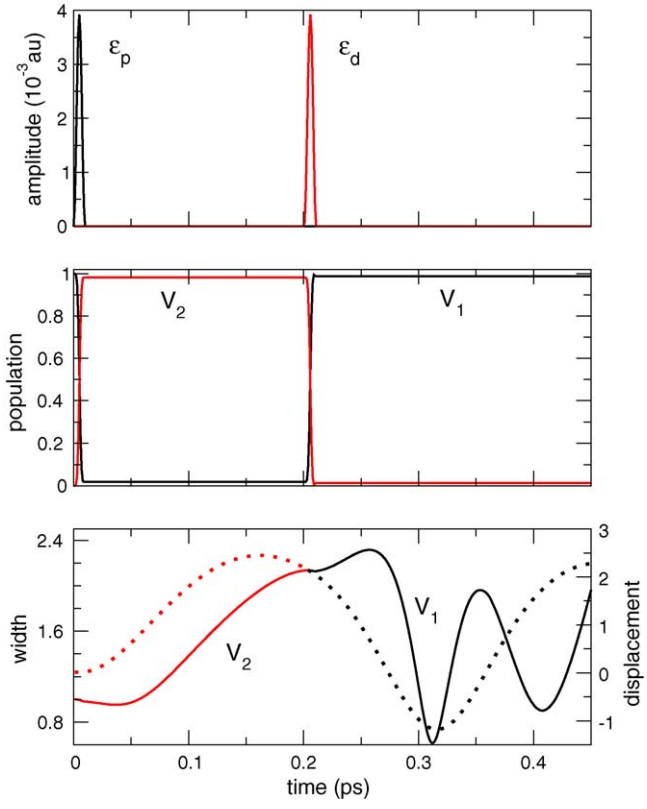

Fig. 2. Dynamics of the ISS scheme showing the pulse shapes, the electronic populations and the wave packet scaled width (solid line, left-side scale) and displacement (dotted line, right-side scale).

Dynamic squeezing can also be obtained via a slight modification of ISS, in which we first adiabatically transfer the initial wave function to the ground vibrational state of an excited potential (e.g. V_2) and then "vertically" dump it back to V_1, as Fig. 1(b) shows. According to the nomenclature of the introduction, this mechanism should be considered as the simplest realization of the ADS scheme. The adiabatic transition can be performed for instance by a linearly chirped pulse. In Fig. 3 we show the results of the dynamics. During the adiabatic transformation the width corresponds to that of the adiabatic wave packet. Firstly, perfect state-selective population transfer to $\phi_{0,2}$ is achieved using a pulse with $\tau = 3$ ps, peak amplitude of 0.85×10^{-3} au (peak intensity of 25 GW/cm^2) and linear chirp λ of 100 cm^{-1}/ps (which can be obtained via a 100 fs transform-limited pulse). Secondly, the diabatic transformation is performed by a 5 fs, $\varepsilon_0 = 3.9 \times 10^{-3}$ au, transform-limited pulse.

It can be easily proved that in the harmonic oscillator model, maximal squeezing is given by $\langle \Delta x \rangle_{\min} / \langle \Delta x_0 \rangle = 1/\sqrt{\gamma} \sim 0.86$, since the wave function dumped to V_1, $\phi_{0,2}$, is less stretched than the wave packet dumped in the ISS case. However, the anharmonicity of the dynamics again favors (at least at initial times) an increased compression of the wave function, so that the maximum squeezing achieved by this very simple scheme is 39%.

Additionally, one could use the same strategy on a different electronic potential, for instance V_3, by using a two-photon pump and dump excitation, as shown in Fig. 1(c). The state-selective population transfer to $\phi_{0,3}$ can be performed by many available adiabatic schemes [24]. Using the same electronic potentials in Na_2, we have shown for instance that with 1.5 ps pulses, an STIRAP counter-intuitive pulse sequence provides excellent yields [25].

There are now different choices regarding the two ultrashort dump pulses. Following the harmonic oscillator model, the highest efficiency $\langle \Delta x \rangle_{\min} / \langle \Delta x_0 \rangle = 1/\sqrt{\gamma} \sim 0.72$ (γ is now equal to w_1/w_3) can be achieved when the maximally stretched wave function is transferred to V_1, which in this case is $\phi_{0,3}$. After the adiabatic preparation, we can directly dump $\phi_{0,3}$ from V_3 to V_1 by two-photon emission, or we can dump it to V_2 and let it evolve for a quarter of the oscillator period, before dumping it to V_1. According to the harmonic oscillator model, maximal stretching would again occur at the repulsive potential wall of V_2, near $\delta x = 0$. In Fig. 4 we show the numerical results of the dynamics. As can be seen in Fig. 1(c) and Fig. 4(a), in the first case the Franck–Condon window is displaced to very far bond distances (in regions of high anharmonicity, $\delta x \sim 2.8$) and the breathing in V_1 never squeezes the wave packet beyond $\langle x_0 \rangle$. In the second case (Fig. 4(b)) the maximal stretching does not

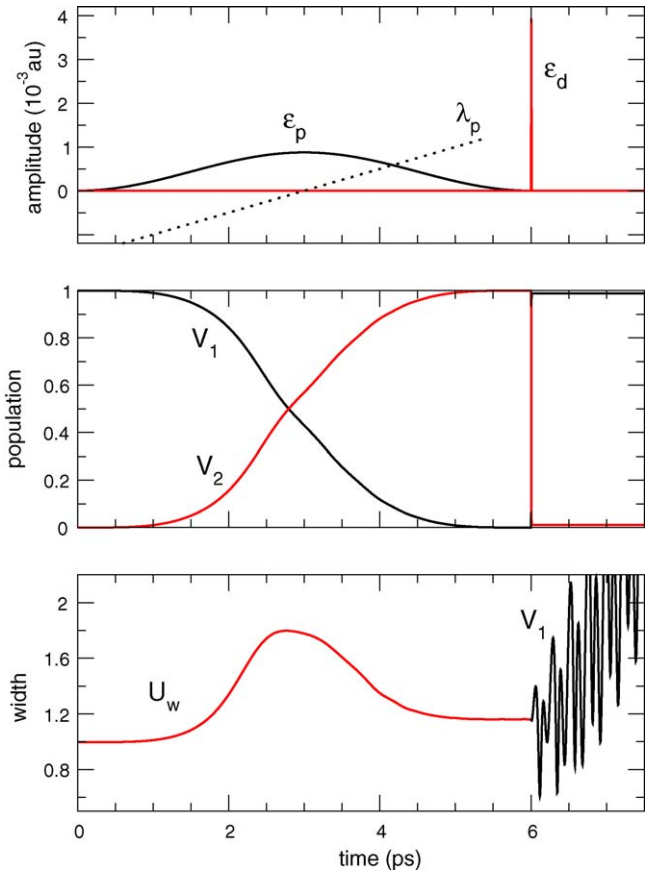

Fig. 3. Dynamics of the ADS scheme via one-photon excitation to V_2, showing the pulse shapes and chirp, the electronic populations and the wave packet scaled width.

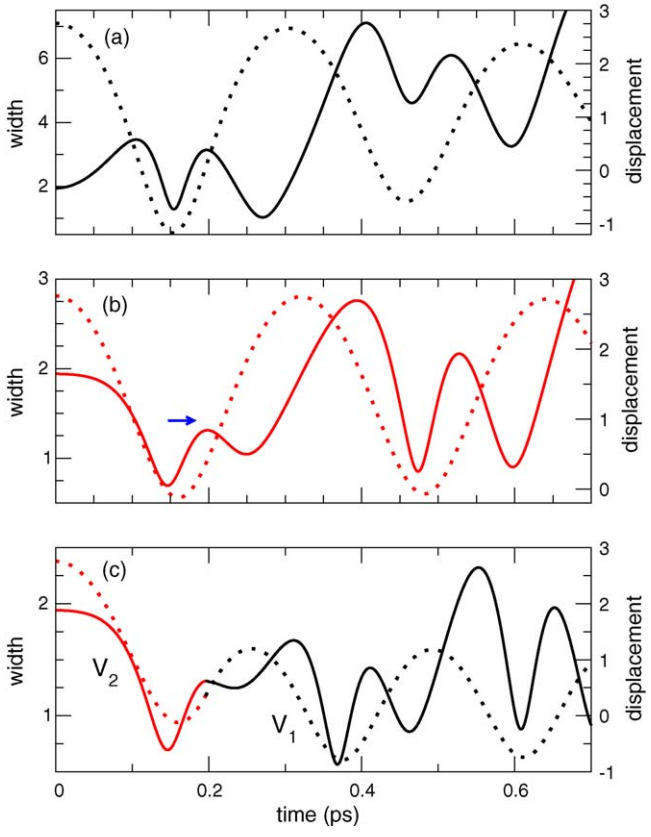

Fig. 4. Dynamics of the ADS scheme after the two-photon adiabatic excitation to V_3, showing the wave packet scaled width (solid line, left-side scale) and displacement (dotted line, right-side scale). In (a) the wave function $\phi_{0,3}$ is dumped to V_1 at initial times; in (b) $\phi_{0,3}$ is dumped to V_2 at initial times after which it evolves freely; in (c) we show the best implementation of the scheme, where the wave packet in V_2 is dumped to V_1 at the time marked by the arrow.

occur near $\delta x = 0$ but closer to the initial position of $\phi_{0,3}$ (at $\delta x \sim 1.7$). This is because the period of breathing is not very different to the period of oscillation, and not twice its value, as in the harmonic case. Then again if the wave packet is transferred at this time, no squeezing will be gained in V_1. The best results are achieved when the locally stretched wave packet (see the arrow in Fig. 4(b)) is dumped to V_1, where a maximal squeezing of 43% is obtained, as shown in Fig. 4(c). The gain, however, is small compared to the simpler ADS scheme using only one-photon pump–dump excitation via V_2, since the wave packet that is finally dumped from V_2 is not very different from $\phi_{0,2}$, which was the source of squeezing in the previous case.

Finally, one could also apply a two-photon pump and dump extension of the ISS scheme. However, the Franck–Condon region for the two-photon absorption lies above the dissociation energy in V_3.

4. Results comparing the AS and ADS schemes

In a previous contribution we have already applied the AS scheme to Na_2 [15], so that here we only outline the best results obtained. In the AS scheme the pulses are not switched off during the time that we want to keep the wave packet adiabatically squeezed [26]; thus, only the switch on sequence is important. The yield of the scheme depends on the final pulse parameters (pulse amplitudes or Rabi frequencies and detunings) which fix the shape of the LIP, whereas the efficiency depends on the dynamical parameters (peak amplitude or Rabi frequency, time duration of the switch on and initial time delay between the pulses) that allow or not the adiabatic excitation.

In Fig. 5(a) we show the squeezed LIP $U_s(x)$ and the adiabatic wave packet prepared by the AS, which is the ground vibrational wave function of $U_s(x)$, $\phi_{0,s}$. In Fig. 6 we show the dynamics and pulses that are needed in order to achieve a best result of 8% squeezing. We use an intuitive, blue detuned sequence with 4 ps switch on time and 3 ps time delay [15]. The frequencies are shifted 0.014 au ($\sim$3100 cm^{-1}) above the $V_1 \rightarrow V_2$ resonance

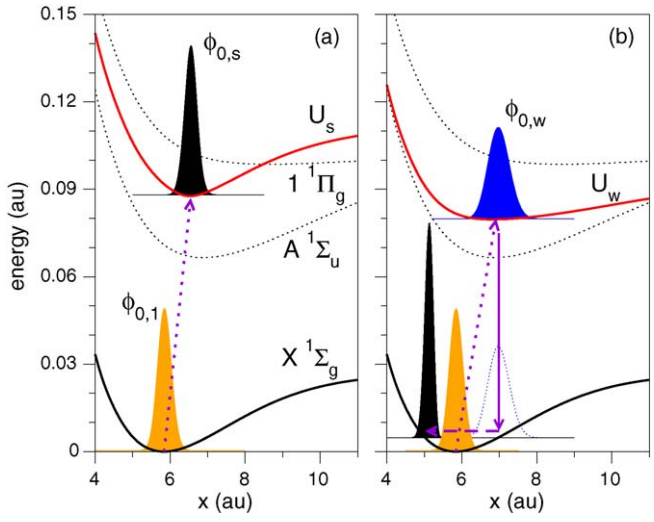

Fig. 5. Sketches of the AS scheme (a) and of the ADS scheme via LIP excitation (b), showing snapshots of the wave packets.

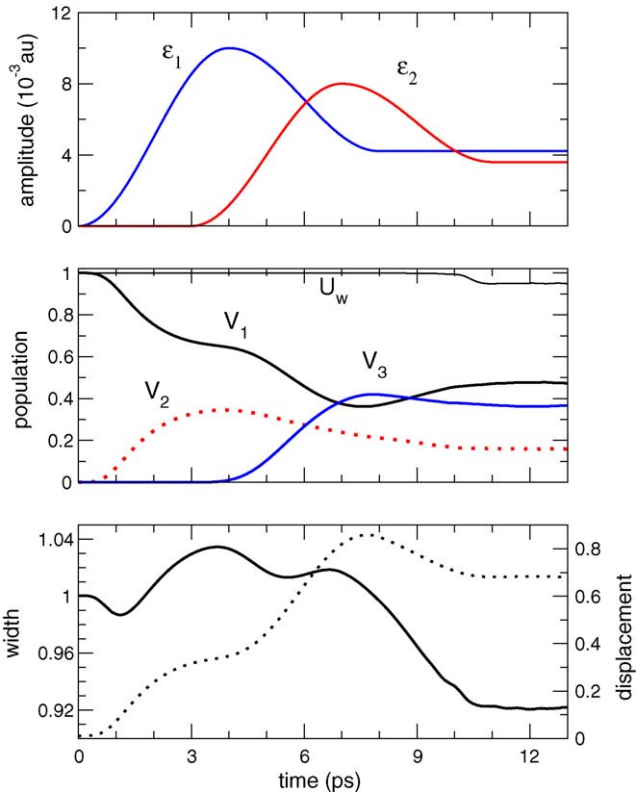

Fig. 6. Dynamics of the AS scheme showing the pulse shapes, the electronic populations and the wave packet scaled width (solid line, left-side scale) and displacement (dotted line, right-side scale).

and the final amplitudes that fix the shape of $U_s(x)$ are around 4×10^{-3} au (implying an intensity $\sim$0.5 TW/cm^2). The equilibrium configuration of the LIP is close to that of V_2 ($\delta x \sim 0.7$), but the overall squeezing is small. Slightly better results ($\sim$10%) can be obtained if the adiabatic demands are weakened.

Instead of adiabatically squeezing the wave packet in a narrower LIP, one can more easily stretch it on a wider LIP, and then transfer it back to V_1 where it will dynamically squeeze. Under this general principle one can optimize the ADS scheme, that is, find the pulse parameters such that U_w is maximally stretched. Using bound excited potentials as V_2 and V_3 to conform the LIP, it is unlikely that the width of the ground vibrational wave function of U_w, $\phi_{0,w}(x)$, will be larger than that of $\phi_{0,3}(x)$. Additionally, one has to balance the need of maximal width with that of small displacement of the equilibrium configuration (the Franck–Condon window for the diabatic transformation), such that the dynamics will not proceed through regions of high anharmonicity. However, it is not sufficient to minimize the displacement, since the effects of the anharmonicity over the width are highly non-linear. For instance, we have observed in the results of Section 3 that the anharmonicity increases the effect of squeezing at least if the wave packet does not depart from very far regions of the potential, $\delta x \sim 1$. Finally, it is also advisable to constrain the allowed pulse amplitudes. Thus, it is not easy to know "a priori" which are the most important features of the LIP that will induce the maximal squeezing, and which can be incorporated on the optimization functional [1].

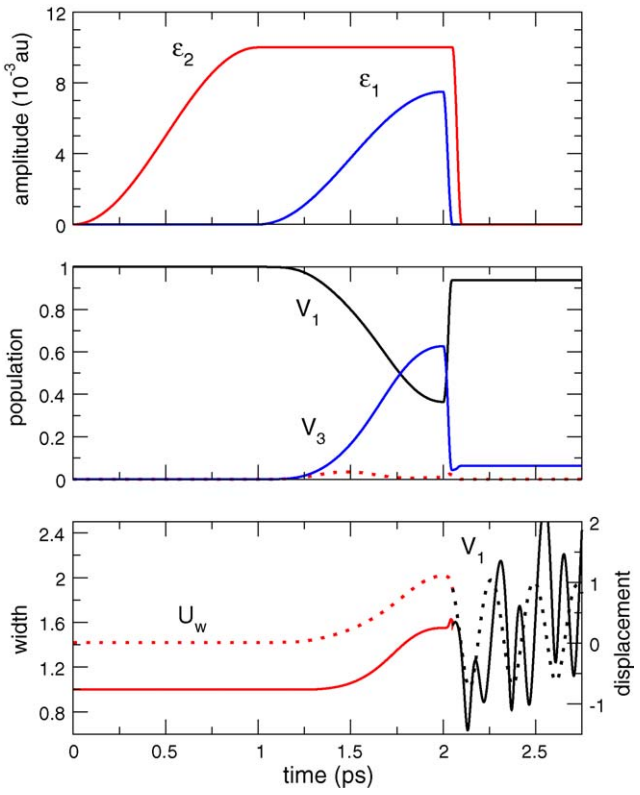

Fig. 7. Dynamics of the ADS scheme via a wide LIP showing the pulse shapes, the electronic populations and the wave packet scaled width (solid line, left-side scale) and displacement (dotted line, right-side scale).

In Fig. 5(b) we show the sketch of the method with snapshots of the wave packets and in Fig. 7 we show the dynamics. The detunings were fixed and the pulse amplitudes were chosen to maximize the width of U_w such that $\delta x \in [0.8,1.2]$ with $\varepsilon_1,\varepsilon_2 < 0.01$. The "optimal" adiabatic transformation is driven by a counterintuitive sequence with a first (instead of pump) pulse of $\tau_1 = 1$ ps and $\varepsilon_1 = 7.5 \times 10^{-3}$ au (peak intensity of 2 TW/cm^2) time-delayed 1 ps from a second (instead of dump) pulse with $\tau_1 = \tau_2 = 1$ ps and $\varepsilon_0 = 0.01$ au (peak intensity of 3.5 TW/cm^2). The sudden or diabatic transformation can be performed by a fast switching off of the pulses ($\tau_3 = 50$ fs for both pulses) reverting the sequence, that is, the first pulse is switched off before the second pulse. For this particular case, however, the maximal dynamical squeezing in V_1 is only 37%, that is, it does not improve the results of the simpler implementations of the ADS scheme.

5. Conclusions

We have shown that the minimum width stationary wave function of Na_2 can be squeezed using basically two laser pulses, via simple physical processes. The different schemes were labeled and organized according to the transformations (adiabatic, diabatic or a combination of both) involved. From an experiment-oriented point of view, the methods could be divided between those using pump–dump control (in Section 3) and those using two-pulse sequences (in Section 4). The first ones rely on controlling the time-delay between the pulses, and typically require full population transfer between electronic states via resonant excitation; the second ones rely on controlling the pulse sequence (the time delay at the switch on and off of two pulses) and require off-resonant partial population transfer to excited states.

Except for the adiabatic squeezing method, which creates stationary laser-molecule wave packets, in all other schemes the wave packet is dynamically squeezed in the ground electronic potential, with approximately the same yield of squeezing, ~40%. This is perhaps surprising, given that we use two excited electronic potentials, $A^1\Sigma_u$ and $1^1\prod_g$, with very distinct structure, to generate the necessary phase difference that gives raise to the dynamical squeezing. Probably, the coincidence comes from the need of high harmonic frequency ratios (which favor the use of $1^1\prod_g$) and small displacements (which favor the use of $A^1\Sigma_u$) so that, in all cases, the wide wave function that is finally dumped to V_1 is not very different than $\phi_{0,2}$, except perhaps in the ISS scheme. In any case, from the theoretical point of view, it is somehow disappointing that the results of the simplest scheme cannot be greatly improved by the more sophisticated ones. The results of this paper should be compared with those of optimal control theory, applied to the Na_2 as well. Averbukh et al. [14] obtained nearly 50% squeezing on an excited potential using phase-controlled pulses. The mechanism of the method was observed to be caused by interference between two wave packets promoted at different times. The control over the motion of the wave packet should be more efficient using phase-modulated pulses, also in the strong-pulse regime [27]. We believe this should prove more important on highly anharmonic motions. For Na_2 however, the optimal yield and in fact the optimal mechanism (although time-ordered in a different way) look relatively similar to those of the ISS scheme [23].

Finally, we would like to comment on the experimental feasibility of implementing these schemes. Obviously, the first problem is assessing the degree of squeezing, that is, designing the appropriate probes [5–7]. A second problem comes from the need of orienting the molecules. In our model we have assumed a fixed oriented sample. Essentially the same results would have been obtained if the molecules were initially pre-oriented, either by adiabatic or sudden alignment [21], since the rotational periods of Na_2 are typically larger than the time-scales of the pulses that we use for squeezing. Therefore, the essential problem consists on whether the initial vibrational state is or not too much disturbed by the laser-alignment. A third problem that we did not address in the paper is the study of the influence of the temperature on the yield of squeezing. Finally, the laser transformations in our schemes require very strong pulses, which may violate the RWA assumptions of the numerical model. For the ultrashort resonant diabatic steps the effects of the counter-rotating terms in the Hamiltonian can be neglected. However, this is not the case for the non-resonant adiabatic steps. In both results of Section 4 the competition between the two pathways of two-photon excitation hinder the yield of squeezing. Additionally in the resonant transitions the $2^1\prod_g$ state, which we did not introduce in the model, will likely be excited by two-photon absorption to an important extent [28]. Finally, although the intensity of the pulses is not exceedingly large, some ionization may further reduce the yield of the squeezing. More accurate molecular

models are therefore required in order to quantify the yield of squeezing that can be achieved using the proposed schemes with current technology in Na_2 or other molecules.

Acknowledgments

This work was partially supported by the Korea Research Foundation Grant funded by the Korean Government (MDEHRD) (KRF-2005-041-C00115), and by the Direccion General de Investigacion of Spain under Project no. CTQ2005-04430.

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