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Preparation and alignment of highly vibrationally excited molecules by CARP – chirped adiabatic Raman passage

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Abstract

Three-dimensional time-dependent Schrödinger equation simulations of Raman excitation of the Cl_2 molecule are used to demonstrate the feasibility of efficient high vibrational excitation ($v \geq 20$) of symmetric non-polar bonds using short (ps) chirped frequency laser pulses at intensities below the ionization threshold ($I = 2 \times 10^{13} \text{ W/cm}^2$). The process called chirped adiabatic Raman passage (CARP) involves sequential Raman excitations $\Delta v = +1$ during the pulse accompanied by rotational transitions. It is shown that as a result of CARP, considerable laser alignment of the molecule is achieved in high vibrational levels, thus offering a new tool for the study of dynamics and reactivity of aligned excited molecules. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Laser control and manipulation of molecules is becoming an expanding area of research due to newly emerging laser technologies whereby one can control shapes, amplitudes and frequencies of laser pulses [1,2]. Intensity is becoming a new parameter, which one can use to control and orient molecules as it is now well understood that there exists ionization thresholds, usually $I = 5 \times 10^{13} \text{ W/cm}^2$, below which using short ($\tau \approx$ picosecond) pulses one can expose molecules to intense fields without having significant ionization in the non-resonant frequency range [3]. For highly polarizable molecules this leads to a new tool for manipulation,

alignment and focusing of molecules by intense short non-resonant laser pulses [4–7].

Another laser parameter which can easily be varied and controlled as a function of time is the frequency $\omega(t)$, leading to *chirped* pulses. We have shown earlier that such pulses can be used on a picosecond time scale to efficiently excite dipolar molecular vibrations in the mid-IR region [8–10], by adjusting the chirp rate ($d\omega/dt$) and frequency at each vibrational excitation step, $\Delta v = +1$, in order to account for inherent anharmonicities. Recently we have extended the scheme of IR chirped excitation to Raman processes, especially for the case of non-polar, symmetric molecular bonds such as those occurring in homonuclear diatomic molecules and metal-metal bonds. We call this chirped adiabatic Raman passage (CARP) [11–13]. Here, we show that the use of short, intense, chirped laser pulses in the near-IR region will lead generally to efficient population transfer

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to highly excited vibrational levels and the concomitant rotational excitation leaves the molecule considerably aligned with the direction of the laser polarization. We demonstrate this new result by accurate numerical solution of the three-dimensional time-dependent Schr  dinger equation, TDSE, for molecules in intense laser fields [14,15] using known molecular parameters for the Cl₂ molecule [16,17]. We follow the numerical procedure established in our previous work on the laser control of electron transfer in the photodissociation of Cl₂ [18,19].

Alignment of vibrationally excited molecules, as shown in the present work, should be of considerable use in rendering molecule–surface reactions more efficient [20]. It will allow to study the highly excited vibrational molecules, e.g., O₂, which play an important role in atmospheric chemistry [21,22]. Moreover, molecular alignment simplifies multiphoton molecular spectra [23] and even photoelectron spectra [24], by removing the need to average over random orientation of molecules in general.

2. Numerical method and TDSEs

A high degree of alignment can usually be achieved for molecular reactants by selective resonant photodissociation with a high-powered laser [25–27]. Non-destructive alignment is best achieved using non-resonant frequencies [4,28]. In the case of the Cl₂ molecule studied here, all parallel Σ – Σ transitions are far off-resonance, in particular the dominant ionic A¹ Σ_u^+ state [16], which allows to describe Raman processes via a static polarizability $\alpha_{\parallel}(R)$ [29], i.e., the laser–molecule interaction with the laser-induced dipole moment (Stark shift). Restricting the Stark shift to the quadratic term (in the quasi-static approximation), the motion of the molecule on the ground state surface (X¹ Σ_g^+) is described by the following Hamiltonian:

$$H_{\Sigma}^J = T^N(R) + V_{\Sigma}^J(R) - \frac{1}{2} \alpha_{\parallel}(R) \mathcal{E}(t)^2 \cos^2 \theta. \quad (1)$$

We omit the term involving the permanent dipole moment, which is zero for symmetric homonuclear

diatomics. Note, that the Π surface in Cl₂ lies so low that for high vibrational excitations even one photon Σ – Π , perpendicular transitions may occur. Therefore, we do not describe them via a polarizability but rather include direct transitions (see below). In the present work, we use a chirped laser pulse defined as

$$\mathcal{E}(t) = \mathcal{E}^0(t) [\sin(\omega_p t) + \sin(\omega_s t + 0.5 \beta_s t^2)], \quad (2)$$

where $\mathcal{E}^0(t)$ is the pulse envelope, $\omega_{p(s)}$ the pump(p) and Stokes(s) frequencies in the range 9392 (1064 nm) $\leq \omega \leq$ 8736 cm^{–1} (1145 nm) and $\omega_p - \omega_s \approx \omega_v$, the vibrational frequency of the Cl₂ molecule. β_s is the chirping rate of the Stokes laser and is taken to be 72.5 cm^{–1}/ps for the intensity $I = c\mathcal{E}^2/8\pi = 2 \times 10^{13}$ W/cm² chosen here. The polarization of the total electric field is linear. This allows to sweep the vibrational spectrum of Cl₂ in a few picoseconds. A pulse rise and descent of 1 ps and a plateau of 2.5 ps are chosen for the numerical simulation described below, which involve propagating on a grid the numerical TDSE nuclear wave packets 3 ps after the pulse is terminated.

In order to include perpendicular transitions, we treat exactly the first Σ_g – Π_u transition, which is described via the dipole term coupling $\mu(R)\mathcal{E}(t)\sin\theta$ [16]. This dipolar moment in perturbation theory gives rise to the perpendicular polarizability $\alpha_{\perp}(R)$ in the non-resonant case. The total rotational–vibrational wave function is expanded using appropriate rotational functions $D'_{M_J, \Omega}(\theta, \phi, 0)$ [30] which results in the following one-dimensional coupled TDSEs for the nuclear functions $\Phi^J(R)$ (both bound and continuum):

$$\begin{aligned} i\hbar \frac{\partial \Phi_{\Sigma}^J(R, t)}{\partial t} = & \left[-\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} + V_{\Sigma}^J(R) \right. \\ & - \frac{1}{2} \mathcal{E}(t)^2 \alpha_{\parallel}(R) I_{\Sigma \rightarrow \Sigma}^{J \rightarrow J} \left. \right] \Phi_{\Sigma}^J(R, t) \\ & - \frac{1}{2} \mathcal{E}(t)^2 \alpha_{\parallel}(R) \left[I_{\Sigma \rightarrow \Sigma}^{J \rightarrow J+2} \Phi_{\Sigma}^{J+2}(R, t) \right. \\ & + I_{\Sigma \rightarrow \Sigma}^{J \rightarrow J-2} \Phi_{\Sigma}^{J-2}(R, t) \left. \right] \\ & + \sqrt{2} \mathcal{E}(t) \mu(R) \left[R(J) \Phi_{\Pi}^{J+1}(R, t) \right. \\ & + Q(J) \Phi_{\Pi}^J(R, t) + P(J) \Phi_{\Pi}^{J-1}(R, t) \left. \right], \end{aligned} \quad (3)$$

$$i\hbar \frac{\partial \Phi_{\Pi}^{J'}(R, t)}{\partial t} = \left[-\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} + V_{\Pi}^{J'}(R) \right] \Phi_{\Pi}^{J'}(R, t) \\ + \sqrt{2}\mathcal{E}(t)\mu(R) \left[R(J')\Phi_{\Sigma}^{J'+1}(R, t) \right. \\ \left. + P(J')\Phi_{\Sigma}^{J'-1}(R, t) \right], \quad (4)$$

$$i\hbar \frac{\partial \Phi_{\Pi}^{J''}(R, t)}{\partial t} = \left[-\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} + V_{\Pi}^{J''}(R) \right] \Phi_{\Pi}^{J''}(R, t) \\ + \sqrt{2}\mathcal{E}(t)\mu(R)Q(J'') \Phi_{\Pi}^{J''}(R, t). \quad (5)$$

$R(J)$, $Q(J)$ and $P(J)$ are the matrix elements of $\sin \theta$ for usual R , Q and P branches ($\Delta J = +1, 0, -1$). The P s are the $\cos^2 \theta$ matrix elements for the $\Delta J = 2, 0, -2$ transitions in Raman processes. These matrix elements depend on the value of M_J , the rotational sublevel quantum numbers. The M_J subscript is omitted, since due to the selection rules M_J does not change in the case of a linearly polarized laser field. Note, however, that the coefficients R , P and Q do depend on M_J . In Eqs. (1), (3)–(5), the symbols $V_{\Sigma}^J(R)$ and $V_{\Pi}^J(R)$ are molecular potentials (including the rotational energy) for the $X^1\Sigma_g^+$ and $^1\Pi_u$ states, correspondingly. In Eqs. (3)–(5), we consider that the $^1\Pi_u$ states are only coupled with the ground state. The $^1\Pi_u$ are coupled to other gerade states. The most possible other surface that can be coupled is the first $^1\Pi_g$ surface [17]. Because this state is repulsive, we can expect that the Franck–Condon factor between this state and the $^1\Pi_u$ states makes such transitions negligible.

A high-order split-operator method developed previously for coupled TDSEs [14,15] was used to solve the Eqs. (3)–(5) starting with the initial numerical wave function $v = 0$ in the ground $^1\Sigma_g^+$ for particular J 's and fixed M_J , $\Phi_{\Sigma}^J(R)$. The numerical integration involved propagating an exponential matrix spanning all J states used in the expansion of the total wave function, with a time step $dt = 0.01$ fs and a spatial grid $2 \text{ a.u.} \leq R \leq 13 \text{ a.u.}$, $dR = 0.025 \text{ a.u.}$ As an example, the total number of rotational functions Φ^J is 122 for $M_J = 0$ states, i.e., 61 for the ground state ($X^1\Sigma_g^+$) and 61 for the continuum nuclear state ($^1\Pi_u$). For $M_J \neq 0$, 60 additional states are added for the dissociative Π_u

states due to the Q branch ($\Delta J = 0$) which is allowed in this case [30]. Since $\Phi_{\Sigma}^J(R, t = 0) = \psi_{\Sigma}^{v=0,J}(R)$, propagation of the TDSEs (Eqs. (3)–(5)) allows us to calculate the total probabilities of excitation to particular ro-vibrational states $v \geq 0$ by projecting out $\psi_{\Sigma}^{v,J}(R)$'s that have been obtained by a numerical Numerov method,

$$P_v(t) = \sum_J \left| \int_{R_{\min}}^{R_{\max}} \psi_{\Sigma}^{v,J}(R) \Phi_{\Sigma}^J(R, t) dR \right|^2. \quad (6)$$

Eq. (6) shows that the final states are coherent superpositions of many rotational levels, J , from which one can numerically calculate the alignment at any time t [4,6,7],

$$\langle \cos^2 \theta \rangle = 2\pi \int_0^\pi \left[\int_{R_{\min}}^{R_{\max}} \left| \sum_J \Phi_{\Sigma}^J(R, t) Y_{M_J}^J(\theta, \phi) \right|^2 dR \right] \\ \times \cos^2 \theta \sin \theta d\theta. \quad (7)$$

Note, that since the $^1\Pi_u$ state surface is repulsive, once a high vibrational excitation is achieved, the molecule can dissociate via one photon or higher multiphoton processes. The coupled TDSEs allow to calculate the resulting continuum nuclear wave functions $\Phi_{\Pi}^J(R)$ exactly. Photodissociation probabilities are obtained by using an absorber at large internuclear distances ($R \geq 13 \text{ a.u.}$), which removes the wave packet from the grid and avoids reflections [31].

3. Results

Simulations have been performed for a laser chirped around the wavelength 1064 nm ($\omega = 9392 \text{ cm}^{-1}$). The chirping rate of the stimulated Raman or Stokes laser was set at $72.5 \text{ cm}^{-1}/\text{ps}$ in order to cover the vibrational frequency of Cl_2 , $\omega_v = 562 \text{ cm}^{-1}$. Fig. 1a shows the vibrational excitations obtained with different initial J 's = 0, 10, 20 for the $M_J = 0$ sublevels only. We note a Gaussian-like distribution around a maximum $v = 22$ level indicative of a diffusion-like process in the ladder climbing. Fig. 1b further shows the alignment calculated from $\langle \cos^2 \theta \rangle$, see Eq. (7), as a function of time. The $M_J = 0$ sublevels

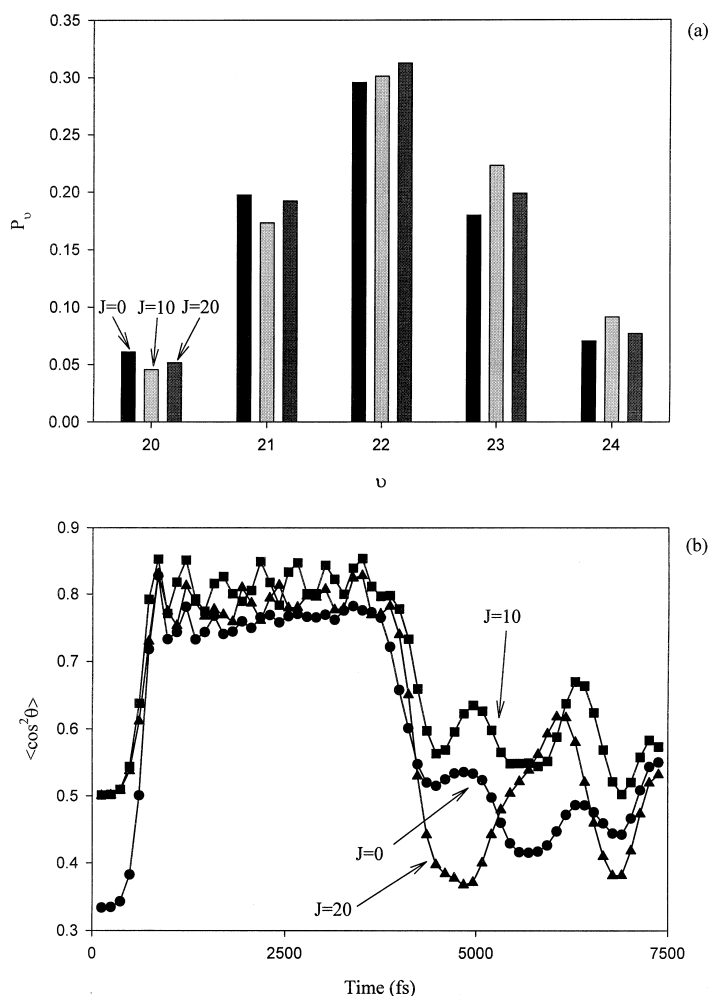


Fig. 1. (a) Distribution of vibrational populations ($P_v \geq 5\%$) after the pulse (4.5 ps) for the initial condition $v = 0$, $M_J = 0$ for $J = 0$ (black column), $J = 10$ (pale grey column) and $J = 20$ (dark grey column). (b) Alignment as a function of time for the same condition: $J = 0$ (circle), $J = 10$ (square) and $J = 20$ (triangle).

are the most strongly coupled to the field as they correspond to molecules *parallel* to the field, thus enhancing $\Sigma_g - \Sigma_u$ transitions and giving rise at high intensities to laser-induced molecular potentials, LIMP [32,33]. In general, starting with high J 's in $M_J = 0$ gives less alignment [4]. We have shown previously that such high J , $M_J = 0$ states in an intense field can be mapped onto one-dimensional lattice excitation models allowing for a qualitative interpretation of excitation migration in a ro-vibrational ladder [34]. An unusual behaviour is observed in that $J = 10$, $M_J = 0$ initial states show

somewhat better alignment than their $J = 0$ counterpart during the pulse. This can be correlated to the smaller dissociation of the $J = 10$ initial level (6.1%) compared to the $J = 0$ state (7.5%). This unusual behaviour is due to the fact that the rise of the pulse (1 ps) is not adiabatic with respect to rotational time scales. The oscillations after 4.5 ps are due to coherent superpositions of rotational states [6,7].

Different M_J 's have different couplings with the field, thus giving rise to different Rabi frequencies and can be responsible for the pulse breakup in

laser amplification at high intensities [35]. For Σ – Σ transitions, M_J 's that are close to zero are most strongly coupled and are responsible for the more efficient vibrational excitations as in Fig. 2a and give the best alignment (see Fig. 2b). In order to increase the efficiency of vibrational transfer for high M_J , we have the choice to increase the intensity or decrease the chirp rate according to the Landau–Zener theory [13]. Fig. 2 represents simulations for an initial $J = 10$ level and sublevels $M_J = 0, 5, 10$ with the same laser conditions as in Fig. 1. In order to obtain an even better alignment

than the 80–85% seen in Figs. 1 and 2, higher intensities might be contemplated. However, photodissociation will become a problem as illustrated in Fig. 3 for an initial $J = 10$ level. We see in Fig. 3 that for a fixed J , photodissociation increases with M_J because the alignment decreases due to less coupling via the parallel polarizability and the coupling increases via the perpendicular Σ – Π transitions for large M_J 's.

Finally in Fig. 4, we give a detailed map of the J level distribution obtained by CARP excitation from a ground state $v = 0$, $J = 10$, $M_J = 0$ level

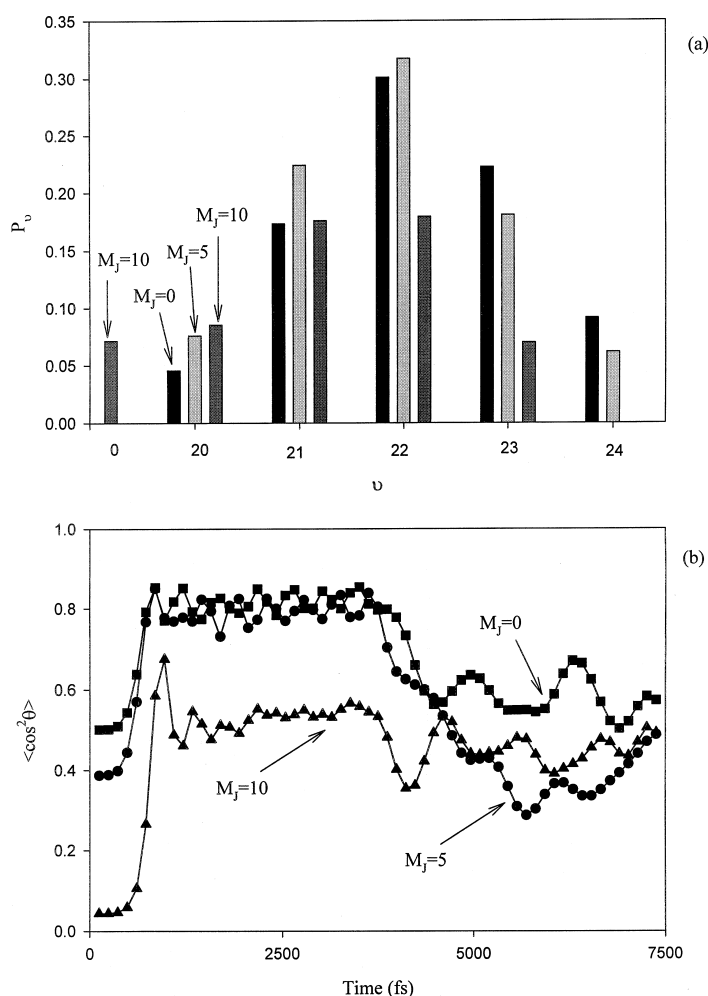


Fig. 2. (a) Distribution of vibrational populations ($P_v \geq 5\%$) after the pulse (4.5 ps) for the initial condition $v = 0$, $J = 10$ for $M_J = 0$ (black column), $M_J = 5$ (pale grey column) and $M_J = 10$ (dark grey column). (b) Alignment as a function of time for the same condition: $M_J = 0$ (square), $M_J = 5$ (circle) and $M_J = 10$ (triangle).

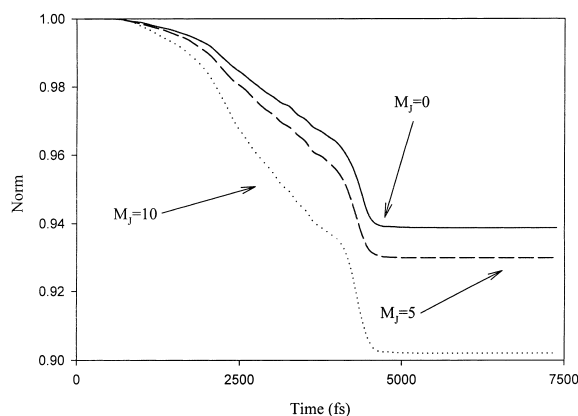


Fig. 3. Norm vs time for the initial condition $v = 0$, $J = 10$ for $M_J = 0$ (solid line), $M_J = 5$ (dashed line) and $M_J = 10$ (dotted line).

in order to illustrate an important observation that maximum rotational excitation extends to $J_{\max} \approx 60$. This was also observed for other initial J states such as $J = 0$ and 20, with all the probabilities calculated 3 ps after the end of the pulse, i.e., a total propagation time of 7.5 ps for the TDSEs equations, see Eqs. (3)–(5). The J_{\max} value is consistent with our previous model of high-intensity molecular rotational coherences [34]. Using an excitation dressed state representation it was

shown that ro-vibrational multiphoton transitions at high intensity and high J can be mapped for $M_J = 0$ exactly onto a one-dimensional discrete lattice diffusion model with a cut-off at a J_{\max} due to quantum localization [36]. From this model, the maximum change of rotational quantum number $m = J_{\max} - J_0$ where J_0 is the initial J can be estimated from the energy exchange between the molecule and the field: $2Bm(m+1) \approx \Omega_R$ where Ω_R is the Rabi frequency. In the present Cl_2 case, $B = 0.244 \text{ cm}^{-1}$, $\Omega_R \approx \mathcal{E}^2 \alpha_{\parallel} / 4 = 8700 \text{ cm}^{-1}$ ($\alpha_{\parallel} = 40 \text{ a.u.}$, $\mathcal{E} = 0.02 \text{ a.u.}$), then the above relation gives $m = \Delta J \approx 42$, in agreement with $J_{\max} \approx 60$ observed in Fig. 4.

4. Conclusion

We have shown from numerical solutions of the TDSE that using short intense chirped near-IR pulses we can achieve efficient excitation of high vibrational levels of symmetric molecular bonds. Thus in the case of Cl_2 , chirping the Stokes pulse by a rate of $72.5 \text{ cm}^{-1}/\text{ps}$ during 4.5 ps resulted in efficient transfer ($P_v \geq 30\%$) into the $v = 22$ level of the ground state of the molecule by successive adiabatic climbing of the vibrational ladder of Cl_2 and 80% between $v = 20$ and 24. This is achieved

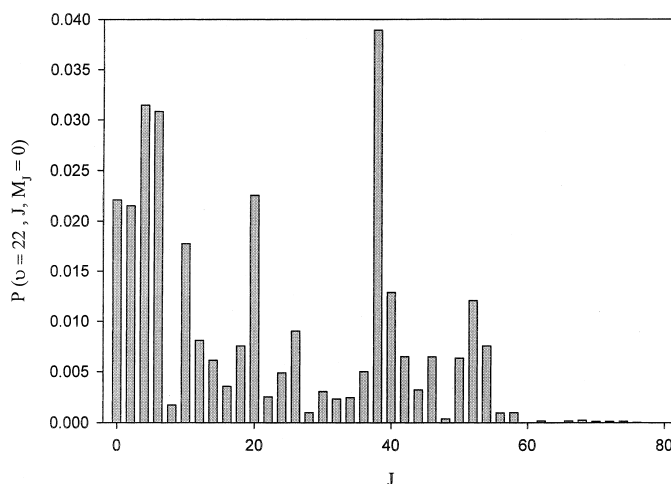


Fig. 4. Distribution of rotational population for $v = 22$ after the pulse (4.5 ps) for the initial condition $v = 0$, $J = 10$ and $M_J = 0$.

with a laser intensity $I = 2 \times 10^{13} \text{ W/cm}^2$, below the ionization threshold. Higher v states cannot be excited because of photodissociation. The wavelength chosen for the pump laser determines the highest v which can be efficiently populated. The lower the wavelength, the higher the v 's that can be populated up to the one photon resonance between the ground and $^1\Pi_u$ states. Note, that this limitation in vibrational excitation is specific to Cl_2 (and to all molecules having similar low repulsive surfaces) and one may expect that higher vibrational energy excitation can be achieved for other molecules, e.g., O_2 , H_2 , where resonant transitions with repulsive states occur at higher energies or are not available. Since, each vibrational transition can be accompanied by a $\Delta J = +2$ rotational excitation, once obtains simultaneously high J states with small M_J 's due to $\Delta M_J = 0$ selection rule. This results in considerable alignment of the molecule at the end of the pulse. The CARP excitation scheme proposed here achieves the preparation and alignment of highly vibrationally excited molecules on picosecond time scales at intensities below ionization. In the case of symmetric molecules, this means that such highly vibrationally excited states will have long lifetimes and could be used for further laser manipulation. In particular, the enhanced reactivity of such states with surfaces [20] and in atmospheric chemistry [21,22] should make CARP a new useful tool for the preparation of such molecules.

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