to the memory effects in the four-wave-mixing signal. In particular, we find that the time-integrated signal shows strong oscillations for negative delay times with period determined by the characteristic resonance energy.

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1:00 pm

Electronic Coherence Effects on Population Transfer by Intense Ultrashort Chirped Pulses in Molecules Coupled with a Dissipative Environment

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Chirped pulses are very efficient for achieving population transfer in atomic and molecular systems.¹ Total electronic population inversion of molecules can be achieved with intense positively chirped (PC) laser pulses, using coherent molecule-light interactions.² At the present time the following processes are recognized to be responsible for population transfer in a two-level electronic molecular systems: 1) intrapulse pump-dump process,^{3,4} 2) π pulses, and 3) adiabatic rapid passage (ARP).⁵ The first process in molecular systems coupled with a dissipative environment can be described in the framework of population wave packet picture.⁴ The last two processes are based on a coherent excitation of a molecule, and can, in principle, produce complete population transfer.

The effect of relaxation processes on a coherent excitation of a molecule coupled with a dissipative environment is an intriguing question. The purpose of this work is to answer the following question: "How does dissipative environment influence on a coherent excitation of a molecule?"

We have studied the influence of the chirp rate on the excited state population n_2 after the completion of pulse action. We used a model of Gaussian-Markovian modulation for description of relaxation processes. It can be characterized by the normalized correlation function $S(t) = \exp(-|t|/\tau_c)$. If $\tau_c \rightarrow \infty$, such a model describes a multilevel system without relaxation corresponding to a pure inhomogeneously broadened electronic transition.

We consider linear chirped pulses of the form

$$\frac{1}{2} \varepsilon(t) \exp \left[\frac{1}{2} i\mu(t-t_0)^2 - i\omega t \right] + c.c.$$

whose chirp rate $\mu = -4\Phi''(\omega) [\tau_p^4 + 4\Phi''(\omega)]^{-1/2}$, where $\Phi''(\omega) = \Phi''(\nu)/(4\pi^2)$ is the phase term.

QThG26 Fig. 1. Excited state population versus phase term Φ'' (v) for models with (solid lines) and without (dotted lines) relaxation. Frequency detuning $\Delta = (\omega - \omega_{21})/\omega_{21} = -1(a)$, $-0.5(b)$, $0(c)$, $0.5(d)$, $1(e)$.

Fig. 1 shows the calculation results of n_2 as a function of Φ'' (v) for different detunings of the exciting pulse carrier frequency ω with respect to the frequency of Franck-Condon transition ω_{21} , corresponding to models with and without relaxation. The saturation parameter $Q' \equiv \sigma_a(\omega_{21}) J_{\max} t_p = 3$. Here $\sigma_a(\omega_{21})$ is the cross section at the maximum of the absorption band, $J_{\max}$ is the power density of the exciting radiation in its maximum, t_p is the pulse duration.

One can see a strong dependence of n_2 on chirp sign induced by relaxation for positive detunings $\omega - \omega_{21}$, whereas n_2 does not depend on chirp sign for the model without relaxation. For large positive Φ'' (v) (PC) the excited state population n_2 tends to the constant value (that is about 1 for $Q' = 3$) which is very close to that given by the model without relaxation.

To understand these result, we will consider first the model without relaxation. One can obtain the following criterion for ARP in this model: $Q' \gg 1$. It conforms to the value of $Q' = 3$ used in our calculations. Thus, about a complete population transfer to the excited electronic state observed in our simulations for the model without relaxation, can be explained by ARP.

Furthermore, the model with relaxation gives the limit value of n_2 for large Φ'' (v) > 0 that is very close to that given by the model without relaxation. The last value is explained by ARP that is a coherent effect. Thus, a positive chirp is favorable to the reduction of relaxation effects on electronic coherence for detunings $\omega - \omega_{21} \approx 0$. This conclusion is supported by analytical considerations.

In conclusion, we have considered strong chirped pulse excitation of a molecule in solution taking into account electronic coherence effects.

The strong dependence of n_2 on chirp sign for positive detunings $\omega - \omega_{21}$ can be explained not only by pump-dump process for population wave packets based on incoherent consideration but the reduction of relaxation effects on electronic coherence for a positive chirp as well.

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Coherent LO Phonon-plasma Coupling in GaAs

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Coherent carrier and phonon dynamics in semiconductors and their nanostructures have been intensively studied during last decade.¹⁻³ It is generally believed that the transient field screening due to photoexcited electron-hole plasma can launch coherent phonon oscillation in bulk GaAs.⁴⁻⁶ At higher injected plasma density ($> 1 \times 10^{18}/\text{cm}^3$), the plasma oscillation resonates with the LO phonon and the LO-plasma coupling (LOPC) plays an important role in the dephasing of coherent LO phonons.⁴⁻⁷ It is therefore worth to understand this coupling process in more detail.

A two-pump time-resolved second-harmonic generation (TRSHG) technique was used to investigate this coupling dynamics. The two pumps and one probe were derived from a mode-locked Ti: sapphire oscillator, which delivers 35 fs, 1.56 eV and 82 MHz pulse train. In the experiment, the first pump at zero delay time injected low plasma density, $n_p \sim 6 \times 10^{17}/\text{cm}^3$, and initiated coherent LO phonons in the depletion region of n-type, $n_d \sim 2 \times 10^{17} \text{ Si}/\text{cm}^3$, GaAs (111) sample. A following intense second pump injected excess plasma, $n_p \sim 2 \times 10^{19}/\text{cm}^3$, into the same region, where the excess plasma could hybrid with the LO phonons. The reflected SH signal of the probe was then measured as a function of the time delay between the pump and probe. The reflected SH signal of the probe was detected in p_{in} - p_{out} geometry and obtained by processing the PMT output with a lock-in amplifier. It is necessary to note that only the initial pump was intensity-modu-