Supporting Online Material for

Coherent Control of Decoherence

Matthijs P. A. Branderhorst, Pablo Londero, Piotr Wasylczyk, Constantin Brif, Robert L. Kosut, Herschel Rabitz, Ian A. Walmsley*

*To whom correspondence should be addressed. E-mail: walmsley@physics.ox.ac.uk

DOI: 10.1126/science.1154576

This PDF file includes:

Materials and Methods
References
Materials and Methods

In this supporting material we provide details about the numerical simulations used to model the experiment and to understand the coherence preserving mechanism found by the closed-loop control scheme. In addition, we give further details about the experimental setup and genetic search algorithm.

Numerical simulations

We calculated the complete vibrational state and its dynamics using a model based on the known spectroscopy of the diatomic potassium molecule (S1, S2). The ro-vibrational level structure of diatomic potassium has been spectroscopically characterized in earlier studies (S3, S4, S5), so the experiment can be numerically simulated using these data. We take RKR potentials and Dunham terms up to third order. We take 40 vibrational energy levels and 200 rotational levels into account. The control pulse field that is used for the experiment is characterized with SPIDER (S6), at a position just before the gas cell. This field is used in the model to calculate the complete ro-vibrational density matrix by means of first order weak perturbation theory. Since the vibrational oscillation around the equilibrium internuclear separation is small, we assume that the transition dipole moment is constant.

In order to calculate the vibrational density matrix at a certain time after excitation, we trace the complete ro-vibrational density matrix over the rotational degree of freedom at that particular time. From the vibrational density matrix $\rho$ we can calculate the vibrational purity $\text{Tr}\rho^2$.

The detected fluorescence signal can be calculated as $p_{\alpha\gamma} = \text{Tr}O_{\alpha\gamma}\rho$, where $O_{\alpha\gamma}$ is a positive operator valued measure (POM) depending on the gate pulse shape, the spectrometer transfer function and the spectroscopic parameters of the electronic transition. The signal $p_{\alpha\gamma}$ is the probability to measure outcome $\alpha$ (either a photon count or no photon count) for the setting of the time- and frequency gates $\gamma$. The POMs are calculated using the derivation in (S7). The theoretical signal thus obtained is shown in figure 2 (c) of the main text and shows good correspondence with the data.

The second model in terms of a system-bath framework is based on (S8). In this article the following expression is derived for the evolution of the Wigner distribution

$$W(r, \varphi, t) = \int_0^{2\pi} d\alpha P(\varphi - \alpha, t)W(r, \alpha, 0), \quad (1)$$
where $P$ is the decoherence kernel

$$P(\varphi - \alpha, t) = \frac{e^{-(\varphi - \alpha)/\gamma t}}{\gamma t(1 - e^{-2\pi/\gamma t})}$$

(2)

and $W(r, \alpha, 0)$ is the initial distribution. The decoherence kernel acts only on the angular coordinate and spreads the distribution out over the phase space trajectory. In the $A$ state of $K_2$ the decoherence time $\gamma^{-1}$ is 3.26 ps for a temperature of 400°C. This model gives clear insight into the decoherence process, but is not as accurate as the first model because it depends on a number of approximations. The initial ro-vibrational state is assumed to be a product state of the vibrational system and the rotational bath; the rotational bath is approximated by a continuous distribution; all higher order coupling terms are neglected based on the energy difference between the vibrational- and rotational constants.

**Experimental setup**

An all-sapphire gas cell containing potassium vapour is kept at 400°C in a purpose-built oven ($S9$) providing a dimer density fraction of around 1% ($S10$). A train of 90 fs pulses centered at 840 nm with spectral width of 18 nm and power of 200 mW is generated in a CPA Ti:Sapphire system with a repetition rate of 2 kHz. The pulse train is split into two arms, forming the pump and gate pulses that can be delayed with respect to each other. The central output frequency of the CPA system is chosen to coincide with the transition that has maximum Franck-Condon overlap, from the ground vibrational level to the 14th vibrational level in the excited electronic state. The vibrational wavepacket is excited in $K_2$ by transferring population from the $X^1\Sigma_g^+$ ground state to the $A^1\Sigma_u^+$ state with a shaped resonant laser pulse of around 1 \(\mu\)J energy and the emitted fluorescence is collected by a pair of 30° off axis parabolic mirrors, with a diameter of 76.2 mm and a focal length of 326 mm, and imaged onto a nonlinear optical crystal (BBO). The energy of the excitation pulse is low relative to the output energy of the CPA system in order to avoid non-linear processes taking place in the molecules. Mixing the fluorescence with the gate pulse creates the sum frequency in the temporal window determined by the duration of the gate pulse. The monochromator acts as a frequency filter with a pass band of around 3 nm. The generated light is detected with a photomultiplier working in the photon-counting mode. The temporal scanning is done by varying the relative lengths of the two arms. The pump pulses are shaped in an acousto optical modulator pulse shaper with a resolution of 100 pixels.

The genetic algorithm searches the control space, which consists of 20 independent parameters. The field function of the acoustic wave is a superposition of 8 polynomial orders for the phase and 4 sine waves for the amplitude. Every ‘generation’ consists of 37 pulse shapes; the 5 pulse shapes with the highest fitness value are used to create 35 new pulse shapes by crossover, and the best pulse shape of each generation as well as the best over-all pulse shape are carried over to the next generation. The mutation rate is 35%, and
the initial choice of pulse shapes is random. The fitness value, which is optimized by the genetic algorithm, is the visibility of the optimized quantum beat. The signal is measured at the peak and at the valley of the beat, and the visibility $V$ is simply calculated from those two data points as $V = \frac{I_{\text{peak}} - I_{\text{valley}}}{I_{\text{peak}} + I_{\text{valley}}}$.

References


S9. D. G. Sarkisyan; Institute for Physical Research, NAS of Armenia-0203, Ashtarak, Armenia