

## Suppression of Decoherence in a Wave Packet via Nonlinear Resonance

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We propose a simple approach for suppressing decoherence of a wave packet excited in an anharmonic oscillator. We show that when a resonant external field forces the oscillator to follow the driving force, motion around the resonant trajectory inside a stable resonant island can be made almost completely immune to the environment. As an example, we study suppression of decoherence due to coupling to thermally populated rotations in vibrational wave packets in a Na<sub>2</sub> molecule.

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Wave packet motion in a multilevel quantum system eventually decoheres due to coupling to an environment. The coupling can be viewed as interaction between the degree of freedom of interest and other degrees of freedom of the same system. Diatomic molecules provide an example [1]: coherence of localized vibrational excitation is lost due to rovibrational coupling to the bath of many thermally populated rotational states. Thus, the coupling between different degrees of freedom reappears in the form of decoherence when using molecular wave packets for quantum information processing [2]. Since rovibrational coupling preserves  $J$  statistics, the rotational bath is not affected by the system (vibrations); only entropy flows between vibrations and rotations, leading to a purely dephasing interaction.

We propose a simple approach for suppressing decoherence in any weakly anharmonic oscillator. We present both analytical and numerical models, considering both an ideal bath and a realistic example of the dephasing of vibrational motion in rotationally hot Na<sub>2</sub> [1].

Methods developed to fight decoherence [3–5] in few-level systems are not readily applied to a molecule. For rovibrational coupling there are no decoherence free subspaces [1]; bang-bang methods are not entirely adequate [1] and are difficult to implement in a molecule [6]. Our brute-force approach complements methods which protect wave packets via carefully arranged interferences [8]; it requires neither detailed knowledge of the wave packet structure nor its coupling with the bath. Our approach is related to the suppression of dephasing in a resonantly driven two-level system [9] when a strong control field overwhelms fluctuations in the natural resonance frequency of the system.

Once dephasing is suppressed, several qubits can be encoded in phase, position, or symmetry of the wave packets [10]. This does not imply scalable molecular quantum computation, but rather a quantum computing-type approach to quantum control [10]. Here, we address a more general question of decoherence management in multilevel systems. When an anharmonic system is driven near reso-

nance, its trajectory is almost completely determined by the driving force. We propose to code information into oscillations *about* this trajectory. We show how in a driven anharmonic oscillator an island created in the phase space—the nonlinear resonance—not only stabilizes coherent wave packet oscillations around the resonant trajectory, but also suppresses coupling to other degrees of freedom. Thus, a protected subspace in the Hilbert space of an anharmonic oscillator can be created by rather simple means.

The basic idea can be understood using a very simple classical picture—that of several weakly anharmonic swings with slightly different (possibly fluctuating) quasi-harmonic frequencies, all driven by the same periodic force. The swings will oscillate with the drive frequency, and the trajectories of the swings will have a different phase lag with respect to the driving force. The excitation we are interested in is that of *deviations* about this dynamic equilibrium. The effective potential for the motion around the dynamic equilibrium is the same for all swings if the driving field is sufficiently strong. For this motion, different phase lags of different swings are irrelevant, and the response of different swings is the same.

Quantum mechanically, we illustrate the idea by considering the decoherence of vibrational wave packets due to rovibrational coupling, which can be formally mapped onto a standard model of decoherence [1]. The swings are replaced by Morse oscillators, describing the vibrational degree of freedom of a diatomic molecule. Different quasiharmonic frequencies of the swings arise from the rovibrational coupling: each Morse oscillator represents a different thermally populated rotational level  $|J, M\rangle$  with the corresponding centrifugal distortion  $-J(J+1)/2\mu R^2$  ( $\mu$  is the reduced mass and  $R$  is the internuclear distance.)

In an experiment [11], Na<sub>2</sub> vapor is created in a pipe oven at about 450 °C, thereby populating many rotational states. A short pump pulse excites vibrational wave packet onto the  $A$  surface from the ground  $X$  surface. To easier understand the rovibrational dynamics that follows, we

begin with the standard approximation for the rovibrational energies:

$$E_{v,J} = \omega_e(v + 1/2) - \omega_e x_e(v + 1/2)^2 + [B_e - \alpha_e(v + 1/2)]J(J + 1) - D_e J^2(J + 1)^2, \quad (1)$$

where  $\omega_e$ ,  $\omega_e x_e$ ,  $B_e$ ,  $\alpha_e$ ,  $D_e$ , are the standard spectroscopic constants [12] and  $v$ ,  $J$  are vibrational and rotational quantum numbers. For each state of the bath (i.e.,  $J$ ) the vibrational frequency is

$$\omega_{\text{vib}}(v, J) = \omega_e - 2(v + 1/2)\omega_e x_e - \alpha_e J(J + 1). \quad (2)$$

The  $v$  dependence of  $\omega_{\text{vib}}$  leads to coherent spreading and revivals of a wave packet [13]. The  $J$  dependence means that, for a thermal ensemble of  $J$ , there is an ensemble of vibrational oscillators with a vibrational wave packet excited in each oscillator. These wave packets oscillate with different  $J$ -dependent vibrational frequencies; see Eq. (2). As they spread over the vibrational orbit, the revivals are washed out. Figure 1 illustrates this effect. We use the full 3D rotational Boltzmann distribution for  $\text{Na}_2$  at  $T = 450^\circ\text{C}$ , which has maximum at  $J_0 = 48$  and half-width  $\Delta J \approx 35$ . Numerically, for this calculation we confine rotations to a plane: the rotational wave functions are  $\Phi_J(\theta) \propto \exp[iJ\theta]$  and the rovibrational energies are given by Eq. (2) with  $J(J + 1)$  replaced by  $J^2$ . An instantaneous excitation of the ground vibrational eigenstate of  $X^1\Sigma_g^+$  into the  $A^1\Sigma_u^+$  surface of  $\text{Na}_2$  [12] yields a coherent superposition of vibrational eigenstates centered near  $v_0 = 19$ , with a vibrational period  $T_{\text{vib}}(v_0; J_0) = 327$  fsec. The revivals clearly visible in Fig. 1(b) are washed out by rotational thermal averaging [Fig. 1(c)], with a rotational decoherence time of about  $30T_{\text{vib}}$ . The effect of a nonzero initial vibrational temperature is similar. The thermal vibrational state on the  $X$  surface is transferred into an

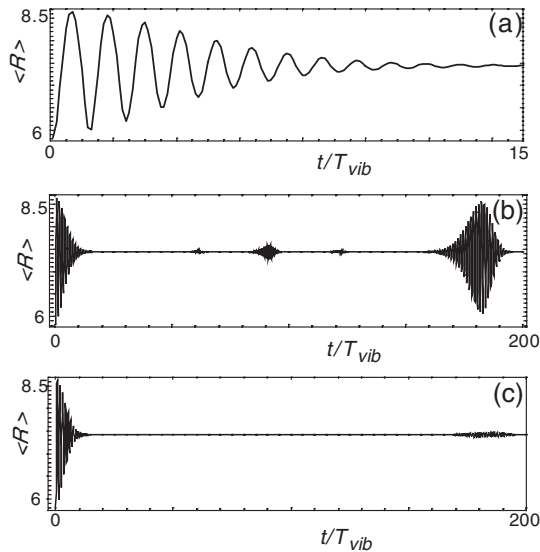


FIG. 1. Evolution of a vibrational wave packet with  $J = 48$  (a),(b), and weighted due to temperature (c).

incoherent distribution of coherent vibrational wave packets on the  $A$  surface, washing out revivals.

To illustrate our method of suppressing decoherence, we start with an analytical description of an idealized case in which the periodic force acts only on the vibrational degree of freedom. The rotational bath manifests itself via thermal ensemble of different  $J$ -dependent vibrational frequencies. The Hamiltonian is

$$\hat{H} = \hat{H}_0(R, \theta) + \hat{V}(R) \cos \Omega t, \quad (3)$$

where  $\hat{H}_0$  is the Hamiltonian of the field-free system with  $R$ ,  $\theta$  the coordinates of vibrational and rotational degrees of freedom, and  $\Omega$  is resonant with  $\omega_{\text{vib}}(v_0, J_0)$ . To show how resonant driving modifies the effective Hamiltonian, we follow Ref. [14] and look for the Floquet state that evolves from some  $|v_i, J\rangle$  as the field is turned on:

$$\chi(R, t) = e^{-iE_{v_i, J} t - i\gamma t} \sum_v C_v e^{-i\Omega(v - v_i)t} \psi_v(R). \quad (4)$$

Here the  $\psi_v(R)$  are field-free vibrational eigenfunctions and  $\gamma$  is the quasienergy measured from the field-free energy of the state  $|v_i, J\rangle$ . Substituting Eq. (4) into the Schrödinger equation, we apply the rotating wave approximation to obtain

$$[\omega_e x_e(v - v_i)^2 + \Delta\omega(v - v_i) + \gamma]C_v = \frac{V}{2}(C_{v+1} + C_{v-1}), \quad (5)$$

where  $\Delta\omega = \omega_{\text{vib}}(v_i, J) - \Omega$ . We have also made another standard approximation  $\langle v | \hat{V} | v \pm 1 \rangle \approx V$  for all  $v$ .

Let us introduce the function  $\Psi(\lambda) = \sum C_v \exp[i(v - v_i)\lambda]$ . The variable  $\lambda$  is conjugate to action and describes the phase of oscillations relative to the stable fixed point of the resonance. The stable point moves along a periodic classical phase-space trajectory with frequency  $\Omega$ .  $\Psi(\lambda)$  is  $2\pi$ -periodic and describes the *envelope* of the wave packet  $\chi(R, t)$  moving along the classical orbit [7]. The dynamics of  $\Psi(\lambda)$  derived from Eq. (5) is

$$\omega_e x_e \Psi''_{\lambda, \lambda} + i\Delta\omega \Psi'_{\lambda} + (V \cos \lambda - \gamma)\Psi = 0, \quad (6)$$

where primes indicate derivatives with respect to  $\lambda$ . Without the driving field ( $V = 0$ )  $\Psi(\lambda) = 1$  and  $\gamma = 0$ , as expected. Transformation

$$\bar{\Psi}(\lambda) = \exp\left[i\frac{\Delta\omega}{2\omega_e x_e}\lambda\right]\Psi(\lambda) \quad (7)$$

yields the well-known Mathieu equation, written as the stationary Schrödinger equation in the periodic  $-V \cos \lambda$  lattice potential,

$$-\omega_e x_e \bar{\Psi}'' - V \cos \lambda \bar{\Psi} = \left(\frac{\Delta\omega^2}{4\omega_e x_e} - \gamma\right)\bar{\Psi}. \quad (8)$$

For given  $\Delta\omega$  and  $V$ , the quasienergies and the widths of the corresponding Brillouin zones are given by characteristics and widths of stability zones on the Mathieu equation stability diagram [15].

Equation (8), which has appeared numerous times for various quantum systems driven near resonance (starting with Ref. [14]), allows us to see how resonant driving preserves coherent motion excited inside the effective “bucket” potential  $-V \cos \lambda$  formed by the nonlinear resonance. We note that eigenstates of such a potential form nonspreading wave packets moving with the bucket, which have been studied in various systems as “flotons” [16] or “Trojan” wave packets [17,18]. Their shape is not important for this work; rather, we show that the same nonlinear resonance that creates these states also stabilizes the system against decoherence.

Incoherent thermal distribution of rotational states means that we deal with an incoherent ensemble of detunings  $\Delta\omega = \omega_{\text{vib}}(v_i, J) - \Omega$ . Equations (7) and (8) show that  $\kappa = \Delta\omega/2\omega_e x_e$  is the quasimomentum of a particle with effective mass  $1/2\omega_e x_e$  moving in the effective lattice potential  $-V \cos \lambda$ ;  $\Delta\omega$  plays the role of a velocity and  $K = \Delta\omega^2/4\omega_e x_e$  is the effective kinetic energy in the absence of the driving field (i.e., at  $V = 0$ ). As the field is turned on, the initial energy  $K = \Delta\omega^2/4\omega_e x_e$  translates into a population of excited states in each well and different quasimomentum states in the lattice; see Fig. 2. Therefore, incoherent distribution of  $\Delta\omega$  translates into (i) incoherent population of excited states in each well and (ii) incoherent population of different wells, which is equivalent to incoherent population within a single Brillouin zone. Neither is a problem if incoherent excitations within each well lie near the bottom, where the well is almost harmonic. Then, if we excite large-amplitude coherent motion inside each well as a carrier of information (Fig. 2), its dynamics will not suffer from incoherent distribution over initial low-lying excitations. The condition under which initial excitations are squeezed to the bottom of the well is  $K \ll V$ , yielding

$$\overline{\Delta\omega} \ll 2\sqrt{V\omega_e x_e}, \quad (9)$$

where  $\overline{\Delta\omega}$  is the range of field-free detunings due to temperature (rotational or vibrational).

Even better is to ensure that the spread in the effective kinetic energy  $K$  is below the zero-point energy of each well. This condition can be written as

$$\overline{\Delta\omega} \leq 32^{1/4} V^{1/4} \omega_e x_e^{3/4}. \quad (10)$$

For temperatures satisfying this condition, the initial state for excitation inside the well (Fig. 2) is well defined and the

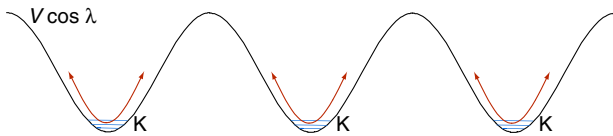


FIG. 2 (color online). In a strong driving field, detunings from resonance due to temperature are transferred into incoherent excitations at the bottom of the effective lattice. Information can be encoded in coherent oscillations inside each well.

coherence of the resulting motion is not compromised. Temperature is translated into incoherent population of different quasimomentum states within the lowest Brillouin zone, but the latter has vanishing width in the strong field limit  $V \gg \omega_e x_e$ .

In  $\text{Na}_2$ , one drives vibrations via the molecular polarizability. We use a linearly polarized bichromatic laser field of strength  $E$  and frequencies  $\omega_L - \Omega/2$  and  $\omega_L + \Omega/2$ , where  $\Omega$  is tuned in vibrational Raman resonance. Assuming that the field is detuned from electronic transitions, standard averaging over fast oscillations with  $\omega_L$  [19] yields the effective Hamiltonian

$$\hat{H} = \hat{H}_v(R) + \frac{\hat{J}^2}{2\mu R^2} - \frac{E^2}{2} \cos^2 \frac{\Omega t}{2} [\alpha_{\perp}(R) + \Delta\alpha(R) \cos^2 \theta], \quad (11)$$

where  $\hat{H}_v(R)$  is the vibrational Hamiltonian,  $R$  is the internuclear distance,  $\hat{J}$  is the angular momentum operator,  $\theta$  is the angle between molecular axis and laser polarization,  $\alpha_{\perp}(R)$ ,  $\alpha_{\parallel}(R)$  are perpendicular and parallel polarizabilities,  $\Delta\alpha = \alpha_{\parallel} - \alpha_{\perp}$ .

We first perform computationally much less demanding 2D numerical simulations with rotations confined to a plane,  $\hat{J}^2 = -\partial^2/\partial\theta^2$ , and then present limited 3D calculations. We use the split-operator fast-Fourier-transform algorithm method on spatial grid, not relying on Eq. (1). We assume that in the region of vibrational excitation  $\alpha(R) = \alpha(R_0)[1 + (R - R_0)/R_0]$ , where  $R_0 = 6.88$  a.u. is the equilibrium position (linear  $R$  dependence reflects the increasing volume available for electrons with increasing  $R$ ). For  $\alpha(R_0)$  we use static polarizabilities of the  $A^1\Sigma_u^+$  state of  $\text{Na}_2$ :  $\alpha_{\perp}(R_0) = 322$  a.u.,  $\Delta\alpha(R_0) = 236$  a.u. [20]. For this model and our rotational temperature, Eq. (9) requires that the sum intensity of both fields is  $I \gg 5 \times 10^{10}$  W/cm<sup>2</sup>. We use  $I = 2 \times 10^{11}$  W/cm<sup>2</sup>, so that almost all states within  $J_0 \pm \Delta J$  also meet the second criterion Eq. (10).

Figure 3(a) shows the temperature-weighted mean position  $\langle R \rangle(t)$ . The wave packet is excited vertically from the ground  $X$  state at the phase of the driving field  $\Omega t = 0$ , which places it at the bottom of the  $-V \cos \lambda$  bucket. After an initial transient regime, the wave packet mostly follows oscillations of the driving field.

Figure 3(b) shows the *difference* of temperature-weighted signal  $\langle R \rangle(t)$  for the vibrational wave packets excited at  $\Omega t_{\pm} = \pm\pi/3$ , so that they are positioned at  $\lambda_{\pm} = \pm\pi/3$  relative to the center of the resonance. One can see fast oscillations with vibrational frequency. They are of no interest to us. Additionally, there are slow vibrations in the effective bucket potential, which last well beyond  $200 T_{\text{vib}}$ . This is the motion which carries quantum information and is resistant to decoherence. As expected, it survives thermal averaging when the field meets the criterion Eq. (10). Additional slow modulation of the signal is mostly due to coherent wave packet spreading.

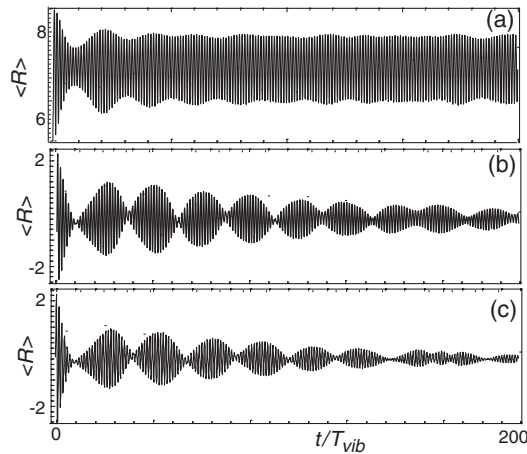


FIG. 3. (a) Temperature-weighted mean position for the wave packet caught in the center of the resonance; (b). Difference between the signals for  $\lambda_+ = +\pi/3$  and  $\lambda_- = -\pi/3$ ; (c). Same as (b) but in 3D.

Validity of our conclusions in 3D is supported by Fig. 3(c), which shows the same value as Fig. 3(b). The 3D calculations were done in the basis of rovibrational  $J$ ,  $M$ , and  $\nu$  states. Since linearly polarized field preserves  $M$ , each calculation is two-dimensional; yet averaging over rotational temperature involves a set of initial conditions for  $J$ ,  $M$ . To speed up averaging, we use only  $J = 0, 10, 20, 30, 40, 50, 60, 70, 80, 100, 120$ , and  $140$  for initial conditions. For  $J = 0 \dots 80$  we incremented  $M$  in steps of 10; for  $J = 100, 120, 140$  we incremented  $M$  in steps of 20. Accuracy of 3D calculations is lower than that of the 2D [Fig. 3(b)], but the results are in good agreement, validating our predictions.

The Hamiltonian Eq. (11) is different from the simplified case [Eq. (3)], as it includes additional coupling between vibrations and rotations via the term  $\Delta\alpha(R)\cos\theta$  in Eq. (11). The field also affects the rotational bath directly (by aligning the molecule). In our system, this leads to additional decoherence as different initial  $J$  states behave differently in the field: low  $J$  are aligned via  $\frac{1}{4}E^2\Delta\alpha\cos^2\theta$ , high  $J$  are not. However, numerical simulations clearly show that our approach still works. This is not surprising since we avoid significant alignment of characteristic  $J$  states: the deviation of  $\langle\cos^2\theta\rangle$  from the random orientation value of 0.5 (rotations are confined to a plane) was below 0.1 during most of the evolution, for all initial  $\bar{J} > 10$ .

In conclusion, resonant driving can decouple motion inside nonlinear resonant island from other degrees of freedom. Any initial thermal distribution is mapped onto excitations inside the resonance. Sufficiently strong driving restricts these excitations to the very bottom of a nearly harmonic bucket potential, where their effect is minimal. Information can then be written into coherent large-amplitude oscillations excited inside the resonant island. The effect is present in the ideal case when the bath is not driven by the external field. Even when the field interacts with the bath directly, separation of natural time scales

between the bath and the system preserves the effect. The relationship of our brute-force approach to the general method and recipes developed in [8] will be considered elsewhere.

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