

Simulating VAET + Bath

Mingyu Kang

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1 Quantized Model

The VAET model (spin-oscillator) without bath is the following.

$$\hat{H}_{SO} = \hat{H}_e + \hat{H}_O + \hat{H}_I \quad (1)$$

$$\hat{H}_e = \sum_{i=1}^n \varepsilon_i |i\rangle\langle i| + \sum_{i<j} V_{ij} (|i\rangle\langle j| + |j\rangle\langle i|) \quad (2)$$

$$\hat{H}_O = \sum_{k=1}^m \nu_k \hat{a}_k^\dagger \hat{a}_k \quad (3)$$

$$\hat{H}_I = \sum_{i=1}^n \sum_{k=1}^m \kappa_{ik} |i\rangle\langle i| (\hat{a}_k + \hat{a}_k^\dagger) \quad (4)$$

Here there are several (m) oscillators, which can be considered as modes of intra-molecular vibrations. This Hamiltonian can be coherently simulated using trapped ions.

However, this model has some limits. First, this model does not take into account that the vibrational modes are damped by the solvent environment. Next, since there are finite number of modes, this cannot be matched to the conventional chemistry simulations, which treat the (damped) oscillators as an environment with continuous spectral density.

Hence, we add a heat bath to the VAET model (spin-oscillator-bath).

$$\hat{H}_{SOB} = \hat{H}_{SO} + \hat{H}_B + \hat{H}_{BI} \quad (5)$$

$$\hat{H}_B = \sum_{\alpha} \omega_{\alpha} \hat{b}_{\alpha}^{\dagger} \hat{b}_{\alpha} \quad (6)$$

$$\hat{H}_{BI} = \sum_{k=1}^m \sum_{\alpha} \gamma_{k\alpha} (\hat{a}_k + \hat{a}_k^{\dagger}) (\hat{b}_{\alpha} + \hat{b}_{\alpha}^{\dagger}) \quad (7)$$

The k th vibrational mode is coupled to the heat bath with coupling strengths $\gamma_{k\alpha}$. The bath is assumed to have infinitely many degrees of freedom, so we can define a continuous spectral density function associated to k -th vibrational mode:

$$J_k(\omega) = 2\pi M_k \nu_k \sum_{\alpha} \gamma_{k\alpha}^2 \delta(\omega - \omega_{\alpha}) \rightarrow \eta_k \omega e^{-\omega/\Lambda} \quad (8)$$

Here, M_k is the mass of the k -th vibrational mode. The justification for the factor $2\pi M_k \nu_k$ will be clear in the next section. It is conventional to assume an Ohmic (proportional to ω) spectral

density for the heat bath, with damping strength η_k and some high-frequency cutoff Λ . I would like to emphasize that this spectral density is for the bath, and not oscillator + bath.

This VAET + bath model is more realistic than the VAET model, in a sense that the intramolecular vibrations are dissipative. Also, VAET + bath model allows matching to chemical simulations, as will be seen in the next section.

2 Matching Chemical Simulations

Here we treat the vibrational modes and the bath modes classically. For convenience, let's consider only one vibrational mode, thus omit index k . The coordinates for the vibrational mode is (y, P_y) and for the α -th bath mode is (x_α, p_α) . The oscillator-bath part of the Hamiltonian becomes the following:

$$H_{OB} = H_O + H_B + H_{BI} \quad (9)$$

$$H_O = \frac{P_y^2}{2M} + \frac{1}{2}M\nu^2 y^2 \quad (10)$$

$$H_B = \sum_{\alpha} \left(\frac{p_{\alpha}^2}{2m_{\alpha}} + \frac{1}{2}m_{\alpha}\omega_{\alpha}^2 x_{\alpha}^2 \right) \quad (11)$$

$$H_{BI} = \sum_{\alpha} \gamma_{\alpha}^2 \sqrt{2M\nu} \sqrt{2m_{\alpha}\omega_{\alpha}} y x_{\alpha} \equiv c_{\alpha} y x_{\alpha} \quad (12)$$

For H_{BI} we use $\hat{y} = \frac{1}{\sqrt{2M\nu}}(\hat{a} + \hat{a}^{\dagger}) \rightarrow y$, and defined

$$c_{\alpha} \equiv \gamma_{\alpha} \sqrt{2M\nu} \sqrt{2m_{\alpha}\omega_{\alpha}} \quad (13)$$

Therefore, H_{OB} becomes

$$\begin{aligned} H_{OB} = & \frac{P_y^2}{2M} + \frac{1}{2}M \left(\nu^2 - \sum_{\alpha} \frac{c_{\alpha}^2}{Mm_{\alpha}\omega_{\alpha}^2} \right) y^2 \\ & + \sum_{\alpha} \left(\frac{p_{\alpha}^2}{2m_{\alpha}} + \frac{1}{2}m_{\alpha}\omega_{\alpha}^2 \left\{ x_{\alpha} + \frac{c_{\alpha}}{m_{\alpha}\omega_{\alpha}^2} y \right\}^2 \right) \end{aligned} \quad (14)$$

If we define the “classical frequency” $\Omega = \left(\nu^2 - \sum_{\alpha} \frac{c_{\alpha}^2}{Mm_{\alpha}\omega_{\alpha}^2} \right)^{1/2}$, then \hat{H}_{OB} becomes identical to the oscillator-bath part of Ref.[3] Eq. 1.3. Also, Ref. [3] defines the spectral density of the bath in Eq. 1.5 as

$$J(\omega) = \frac{\pi}{2} \sum_{\alpha} \frac{c_{\alpha}^2}{m_{\alpha}\omega_{\alpha}} \delta(\omega - \omega_{\alpha}) \rightarrow \eta \omega e^{-\omega/\Lambda} \quad (15)$$

By using Eq. 13, you can check that Eq. 15 agrees with the spectral density defined for the VAET + Bath model in Eq. 8.

Now, we can use the result of Ref.[3], which calculates the effective spectral density $J_{eff}(\Omega)$ of the oscillator + bath environment. Here Ref.[3] treats the electron occupation classically, by replacing $y_0(|D\rangle\langle D| - |A\rangle\langle A|)$ with a continuous variable q , where $y_0 = \frac{\kappa_D}{\Omega^2} \sqrt{\frac{2\nu}{M}} = -\frac{\kappa_A}{\Omega^2} \sqrt{\frac{2\nu}{M}}$ in the

donor-acceptor system. By using this approximation, Ref.[3] obtains Eq. 2.14:

$$J_{eff}(\omega) = \frac{\eta\omega\Omega^4}{(\omega^2 - \Omega^2)^2 + (\omega\eta/M)^2} \quad (16)$$

This is directly proportional to the imaginary part of the dynamic susceptibility of a damped harmonic oscillator with undamped frequency Ω and a friction coefficient η [3]. Therefore, this spectral density describes a *damped* oscillator.

Eq. 16 can be expressed as a sum of two Lorentzian curves [5]:

$$J_{eff}(\omega) = \frac{M\Omega^4}{2\omega_m} \left(\frac{\eta/2M}{(\omega - \omega_m)^2 + (\eta/2M)^2} - \frac{\eta/2M}{(\omega + \omega_m)^2 + (\eta/2M)^2} \right) \quad (17)$$

where the damped frequency $\omega_m = \sqrt{\Omega^2 - (\eta/2M)^2}$

In the weak damping regime ($\eta/M \ll \nu$), $\nu \approx \Omega \approx \omega_m$, so

$$J_{eff}(\omega) \approx \frac{M\nu^3}{2} \left(\frac{\eta/2M}{(\omega - \nu)^2 + (\eta/2M)^2} - \frac{\eta/2M}{(\omega + \nu)^2 + (\eta/2M)^2} \right) \quad (18)$$

$$\approx \frac{M\nu^3}{2} \frac{\eta/2M}{(\omega - \nu)^2 + (\eta/2M)^2} \quad (19)$$

where the last equation is valid at the proximity of ν , which is the region of interest when $\eta/M \ll \nu$ (Lorentzian peak is sharp). Note that the area under this Lorentzian curve is $\frac{\pi}{2}M\nu^3$.

Finally, we need to do some calibration. We consider the limit $\eta/2M \rightarrow 0$. Then, $J_{eff}(\omega) \rightarrow \frac{\pi}{2}M\nu^3\delta(\omega - \nu)$. This should match the VAET model (Eq.1-4), where the spectral density is conventionally defined as $J_{eff}(\omega) = \sum_k \kappa_k^2 \delta(\omega - \nu_k)$. Therefore, the final effective spectral density is

$$J_{eff}(\omega) = \frac{\kappa^2}{\pi} \frac{\eta/2M}{(\omega - \nu)^2 + (\eta/2M)^2} \quad (20)$$

This Lorentzian spectral density that describes the oscillator + bath environment can be classically simulated using chemical methods such as HEOM and path integral. However, it is worth emphasizing that to reach Eq. 20, not only the bath modes, but also the vibrational modes and the electron occupation, have been treated classically. Therefore, we expect the chemical methods to match the fully quantum simulation of VAET + bath model **only in the high-temperature regime**.

In practice, there should be some typical Lorentzian spectral density used in chemical simulations, with some FWHM and peak amplitude. Then we can find η and M from $\text{FWHM} = \eta/M$ and peak amplitude $M\nu^3/2 = \kappa^2/\pi$, and use these values to match quantum simulations. In particular, the spectral density of the bath coupled to k th vibrational mode (Eq. 8) becomes

$$J_k(\omega) = \left(\frac{2\kappa_k}{\nu_k} \right)^2 \sum_{\alpha} \gamma_{k\alpha}^2 \delta(\omega - \omega_{\alpha}) \rightarrow \eta_k \omega e^{-\omega/\Lambda} \quad (21)$$

and this will be used in quantum simulations, as described in the next sections.

3 Master Equation for a Damped Oscillator

Again for convenience, let's consider only one vibrational mode, thus omit index k . The quantum version of the oscillator-bath Hamiltonian is:

$$\hat{H}_{OB} = \hat{H}_O + \hat{H}_B + \hat{H}_{BI} \quad (22)$$

$$\hat{H}_O = \nu \hat{a}^\dagger \hat{a} \quad (23)$$

$$\hat{H}_B = \sum_{\alpha} \omega_{\alpha} \hat{b}_{\alpha}^\dagger \hat{b}_{\alpha} \quad (24)$$

$$\hat{H}_{BI} = (\hat{a} + \hat{a}^\dagger) \sum_{\alpha} \gamma_{\alpha} (\hat{b}_{\alpha} + \hat{b}_{\alpha}^\dagger) \quad (25)$$

We use the following approximations: 1. the bath is Markovian, 2. $\gamma_{\alpha} \ll \nu$ (weak damping), 3. secular approximation. Then, by standard procedure [1], we obtain the following master equation:

$$\begin{aligned} \partial_t \hat{\rho}_O(t) = & -i[\hat{H}_O + \hat{H}_{LS}, \hat{\rho}_O(t)] \\ & + \Gamma_+ \left(\hat{a}^\dagger \hat{\rho}_O(t) \hat{a} - \frac{1}{2} \{ \hat{a} \hat{a}^\dagger, \hat{\rho}_O(t) \} \right) \\ & + \Gamma_- \left(\hat{a} \hat{\rho}_O(t) \hat{a}^\dagger - \frac{1}{2} \{ \hat{a}^\dagger \hat{a}, \hat{\rho}_O(t) \} \right) \end{aligned} \quad (26)$$

where

$$\begin{aligned} \Gamma_+ & \equiv \Gamma_+(\nu) = 2\pi \sum_{\alpha} \delta(\nu - \omega_{\alpha}) \gamma_{\alpha}^2 \langle \hat{b}_{\alpha} \hat{b}_{\alpha} \rangle_B \\ & = \frac{\pi}{2} \left(\frac{\nu}{\kappa} \right)^2 J(\nu) \bar{n}_B(\nu) \end{aligned} \quad (27)$$

$$\Gamma_- = \frac{\pi}{2} \left(\frac{\nu}{\kappa} \right)^2 J(\nu) (\bar{n}_B(\nu) + 1) \quad (28)$$

Here we use Eq. 21. Also, $\bar{n}_B(\nu)$ is the average number of phonons with frequency ν in the bath. We assume Ohmic spectral density:

$$J(\omega) = \eta \omega e^{-\omega/\Lambda} \quad (29)$$

and thermal distribution:

$$\bar{n}_B(\omega) = 1/(e^{\beta\omega} - 1) \quad (30)$$

Therefore, as the cutoff frequency $\Lambda \rightarrow \infty$, we obtain the following:

$$\Gamma_+ = \frac{\pi}{2} \left(\frac{\nu}{\kappa} \right)^2 \frac{\eta \nu}{e^{\beta\nu} - 1}, \quad \Gamma_- = \frac{\pi}{2} \left(\frac{\nu}{\kappa} \right)^2 \frac{\eta \nu}{e^{\beta\nu} - 1} e^{\beta\nu} \quad (31)$$

At low temperature ($\beta\nu \gg 1$), $\Gamma_+ \rightarrow 0$ and $\Gamma_- \rightarrow \frac{\pi}{2} \left(\frac{\nu}{\kappa} \right)^2 \eta \nu$.

At high temperature ($\beta\nu \ll 1$), $\Gamma_+ \approx \Gamma_- \approx \frac{\pi}{2} \left(\frac{\nu}{\kappa} \right)^2 \frac{\eta}{\beta}$.

To understand the physical meaning of the master equation, we calculate the evolution of the oscillator's average phonon number $\bar{n}(t) = \langle \hat{a}^\dagger \hat{a} \rangle_t$, and obtain the following:

$$\partial_t \bar{n}(t) = \Gamma_+ (\bar{n}(t) + 1) - \Gamma_- \bar{n}(t) \quad (32)$$

So Γ_+ and Γ_- can be interpreted as heating and cooling rate, respectively. Finally, the steady state phonon number is

$$\bar{n}_{SS} = \frac{\Gamma_+}{\Gamma_- - \Gamma_+} \quad (33)$$

4 Matching Trapped Ion Simulations

Now that we have the master equation for the damped oscillator, we should figure out how to make the ion chain's motional mode follow this master equation. We suggest using sideband cooling to simulate the phenomenon in low-temperature ($\beta\nu \gg 1$), and random stochastic fields in high-temperature ($\beta\nu \ll 1$).

4.1 Sideband Cooling

One can use sideband cooling with an auxiliary ion to cool the motional mode of the ion chain. Ideally this auxiliary ion is a different species than other ions that represent the electronic sites. Alternatively, if the optical pumping beam can address a single ion with very little optical crosstalk ($< 10^{-5}$) to the neighboring ions, sideband cooling can be performed with limited fidelity loss.

Sideband cooling is performed in two stages: 1. red sideband transition, 2. optical pumping that makes qubit state $|1\rangle$ decay to $|0\rangle$. First consider the optical pumping stage. Since $|1\rangle$ of Yb ions is inherently stable, we use Raman transition with the third excited state $|e\rangle$. Let's say $\Gamma_{0,1}$ is the linewidth of $|e\rangle$'s decay to $|0\rangle$ and $|1\rangle$, respectively. Also, let's use an optical pumping beam of Rabi frequency Ω_{aux} and detuning from $|1\rangle - |e\rangle$ transition Δ_{aux} . Then, the effective linewidth of $|1\rangle$'s decay to $|0\rangle$ is the following [6]:

$$\tilde{\Gamma} = \frac{\Omega_{aux}^2}{(\Gamma_0 + \Gamma_1)^2 + 4\Delta_{aux}^2} \Gamma_0 \quad (34)$$

Now consider the red sideband transition stage. We use laser with Rabi frequency Ω to cool the motional mode with frequency ω and Lamb-Dicke parameter η . Let's assume that we already pre-cooled to very low temperature, so that the phonon occupation n is either 0 or 1. Eq. 111 of Ref.[4] gives:

$$\dot{p}_0 = p_1 \frac{(\eta\Omega)^2}{\tilde{\Gamma}} - p_0 \left(\left(\frac{\tilde{\eta}\Omega}{2\omega} \right)^2 + \left(\frac{\eta\Omega}{4\omega} \right)^2 \right) \tilde{\Gamma} \quad (35)$$

$$\dot{p}_1 = -\dot{p}_0 \quad (36)$$

Here the two higher-order heating processes are: carrier excitation followed by decay on the blue sideband, and first blue sideband excitation followed by decay on the carrier. Note that $\tilde{\eta}$ is not necessarily equal to η - see Ref.[4].

$\bar{n} = p_1$, so to match Eq. 32 and 33, following should hold:

$$\Gamma_- - \Gamma_+ = \frac{(\eta\Omega)^2}{\tilde{\Gamma}} + \left(\frac{\tilde{\eta}\Omega}{2\omega} \right)^2 + \left(\frac{\eta\Omega}{4\omega} \right)^2 \approx \frac{(\eta\Omega)^2}{\tilde{\Gamma}} \quad (37)$$

$$\frac{\Gamma_+}{\Gamma_- - \Gamma_+} = \left(\frac{\tilde{\Gamma}}{2\omega} \right)^2 \left(\left(\frac{\tilde{\eta}}{\eta} \right)^2 + \frac{1}{4} \right) \quad (38)$$

where the approximation uses $\Omega \ll \omega$. Therefore, given Γ_- and Γ_+ that we want to simulate, we can find the corresponding Ω and $\tilde{\Gamma}$ ($\Rightarrow \Omega_{aux}$), and perform sideband cooling accordingly.

The results above are limited to low temperature, such that $n = 0$ or 1 . In principle, there should be a way to simulate higher temperature, but the math and the sideband cooling scheme will be somewhat complicated, because the pulses for cooling $n = 1, 2, 3, \dots$ states are all different. Also,

both multi-species cooling and individually addressed optical pumping beam are not experimentally prepared at the current stage.

4.2 Random stochastic fields

Here we simulate a damped oscillator in high temperature by applying random stochastic fields on the ion chain's collective mode, and then averaging over many instances. We should apply lasers with the mode's blue and red sideband frequencies on an auxiliary ion. The Hamiltonian becomes the following:

$$H(t) = \frac{\eta\Omega}{2} \left(\hat{a}e^{i(\delta t + \phi_m(t))} + \hat{a}^\dagger e^{-i(\delta t + \phi_m(t))} \right) \left(\hat{\sigma}_x \sin \phi_s + \hat{\sigma}_y \cos \phi_s \right) \quad (39)$$

If we set $\delta = 0$, $\phi_s = \pi/2$,

$$H(t) = \frac{\eta\Omega}{2} \left(\hat{a}e^{i\phi_m(t)} + \hat{a}^\dagger e^{-i\phi_m(t)} \right) \hat{\sigma}_x \quad (40)$$

We can fix the qubit state of an auxiliary ion to $|+\rangle$. Then, we have a simple Hamiltonian applied to the motional mode

$$H(t) = z(t)\hat{F} + z^*(t)\hat{F}^\dagger \quad (41)$$

where $z(t) = \frac{\eta\Omega}{2}e^{i\phi_m(t)}$ and $\hat{F} = \hat{a}$.

We consider a stepwise constant $\phi_m(t)$, where each step is the Trotterization step. At each i th step, $\phi_m^i = \phi_m(t_i)$ is randomly drawn from a uniform distribution $U([0, 2\pi])$. This makes $z(t)$ satisfy the following:

$$\overline{z(t)} = 0, \quad \overline{z(t)z(t')} = 0, \quad \overline{z(t)z^*(t')} = C_\tau(t - t') \quad (42)$$

Then, in the limit where Trotterization time step τ is much shorter than the relaxation time scale, the two-point correlation $C_\tau(t - t')$ approximately becomes a single square-pulse-shaped peak of width τ and height $(\eta\Omega/2)^2$. In other words,

$$\lim_{\tau \rightarrow 0} \int_0^\infty ds C_\tau(s) = \left(\frac{\eta\Omega}{2}\right)^2 \tau \equiv \frac{\gamma}{2} \quad (43)$$

It is known that under such random stochastic Hamiltonian, the quantum state averaged over random ensemble evolves under the following master equation [2]:

$$\partial_t \bar{\rho}(t) = \gamma \left(\hat{F} \bar{\rho}(t) \hat{F}^\dagger - \frac{1}{2} \{ \hat{F}^\dagger \hat{F}, \bar{\rho}(t) \} + \hat{F}^\dagger \bar{\rho}(t) \hat{F} - \frac{1}{2} \{ \hat{F} \hat{F}^\dagger, \bar{\rho}(t) \} \right) \quad (44)$$

Therefore, by averaging over multiple instances, we can simulate the master equation in Eq. 26, where

$$\Gamma_+ = \Gamma_- = \gamma = (\eta\Omega)^2 \tau / 2 \quad (45)$$

$\Gamma_+ \approx \Gamma_-$ only at high temperature ($\beta\nu \ll 1$), so this method can only simulate high-temperature bath.

The interaction in Eq. 39 is exactly same as the interaction used to simulate spin-motion coupling and run MS gates. Therefore, this method can be performed with the current experimental setup, with the cost of running the circuit many ($\sim 100?$ 1000?) times to find the average.

References

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