# Quantum Decoherence of the Particle on a Circle

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## I. INTRODUCTION

The quantum-mechanical nature of the world does not manifest itself easily in our day-to-day lives. The eigenstates of a free particle, for instance, are supposed to be plane waves delocalized over all of space. Yet we never see our pocketwatches in two places at once! Rather, macroscopic objects exist in *localized* position (psuedo)-eigenstates and obey classical laws of motion. If we believe that our world is fundamentally quantum mechanical, then there must be an explanation for why it behaves classically in warm, macroscopic conditions.

The theory of decoherence gives a compelling explanation of how classical mechanics emerges from quantum mechanics. It does not rely on arguments of consciousness or wavefunction collapse, but results from using the framework of well-established quantum theory to analyze quantum systems interacting with their environment. In essence, when a quantum system becomes *entangled* with its surrounding, its characteristic quantum property of *interference* no longer appears; its coherence becomes lost from a quantum superposition into a classical mixture of different states.

In many regimes, the (classical) mixture is comprised not of the energy eigenstates of the underlying quantum system, but rather of particular "pointer" states that are immune to decoherence [1]. These pointer states are determined by the interactions with the environment. So even though a pocketwatch has a free-particle Hamiltonian with plane-wave eigenstates, the interaction with its surroundings – scattering photons, emitting blackbody radiation, deflecting air molecules – causes it to localize in *position-space* rather than momentum-space. In fact, generally, for free particles coupled to a thermal bath, the environment induces a superselection of *minimal-uncertainty Gaussian states* [2]. As the theory explains, a (coherent) superposition of a macroscopic object in two positions rapidly decoheres into a mixture of narrow Gaussian wavepackets. Due to the interaction with the environment, the translational degrees of freedom of a macrosopic object behave classically.

In this paper, I wish to answer questions about how decoherence affects *rotational* degrees of freedom. As a motivational example, consider hydrogen chloride (HCl), a simple diatomic molecule which can be modeled as a rigid rotator (or equivalently, a particle on a sphere) whose rotational

eigenstates are the spherical harmonics. These states have distinctively quantum character: In the ground state ("s"), the hydrogen nucleus is delocalized uniformly over a sphere around the central chlorine atom; in higher-energy harmonics, the wavefunction of the hydrogen nucleus is similarly "smeared out" over many possible orientations. If we look at the rotational spectrum of HCl at room temperature, we see that the molecules occupy many such rotational states at room temperature. The rotational wavefunctions of HCl can in fact be selected and imaged [3]. Evidently, small objects can rotate *quantum mechanically* and exist comfortably in non-classical rotational eigenstates.

In contrast, larger objects rotate classically. The motion of a gyroscope, for instance, is described with the rigid-body-motion of classical mechanics. Even objects which are small by our day-to-day standards, such as proteins ($\sim 10^5 a.m.u.$), have rotational motion described by classical equations of motion. For these macroscopic objects, the rotational wavefunctions are well-localized in orientation, rather than spread out all over all possible orientations.

Somewhere between the length scale of a HCl molecule and a protein, the rotational behavior of a molecule must transition from quantum mechanical to classical. Decoherence theory says that this quantum-to-classical transition arises because the interactions with the environment becomes significant compared to the system's intrinsic dynamics. For rotations, the larger the rotating body, the more accurately its orientation is determined by collisions with particles in the environment, and the resulting entanglement causes the rotational wavefunction to localize in orientation and effectively behave classically.

Here I explore the quantum-to-classical transition of rotation. I expect the most classical-like rotational wavefunctions to be somewhat analogous to the coherent states of the harmonic oscillator; that is, I expect the environment to prefer states that are localized in (classical) phase space and minimize the Heisenberg uncertainty relation. For simple systems such as the harmonic oscillator, it is possible to show that indeed the environment-selected states are indeed coherent states by using arguments about minimizing the loss of purity [4]. I wish to explore whether rotational degrees of freedom also display similar behavior.

In brief, I aim to understand how the dynamics of a rigid rotor are altered by coupling to the environment.

*A comment.* Rather than considering the rotational states of a rigid rotor in three dimensions, I restrict myself to rotations in two dimensions. This is the problem of a planar rotor, or equivalently, of a free particle moving on a circle. I chose this system because it is much easier to work with than the particle moving on a sphere: the rotational states are labeled by one quantum number $|l\rangle$ rather

than two $|jm\rangle$, and the classical phase space has a topology much easier to reason about. Hopefully the planar rotor is sophisticated enough to capture any special features about the decoherence of rotations compared to translations.

This paper is organized as follows. First in Section II, I discuss the formalism of decoherence, explaining why a system loses its interference properties when it becomes entangled with its environment. Next in Section III, I introduce the quantum planar rotor and its behavior under free evolution. Finally in Section IV, I explore how the quantum planar rotor is affected by interactions with an external environment by deriving and analyzing the equation of motion of its reduced density matrix.

## II.   DECOHERENCE

The simple act of interacting with a surrounding environment causes a quantum system to lose much of its quantum behavior. In this section, I will illustrate why this is the case, and also introduce some of the formalism of reduced density matrices that will be used later on to describe the dynamics of the planar rotor. Much of this material is a review of the existing literature such as ref. [1], but I believe that one cannot claim to 'understand decoherence' without actually going through the calculations to show its effects. In any case, this background discussion is relevant for understanding the physics behind the later calculations in Section IV.

### A.   Entanglement with the Environment Destroys Interference

Even weak interactions with the surrounding environment can profoundly affect the behavior of a quantum system. In essence, if a system becomes entangled with its environment, then the interference between states is no longer visible. As an illustration of why this is the case, let us consider how interactions with the environment affect the classic two-slit experiment.

Suppose a particle can hit a detector by passing through two possible slits, labeled $a$ and $b$. Say we block slit $b$ and force it to travel through slit $a$. Then the particle emerges in a state $|a\rangle$. When it hits the detector, its wavefunction is given by $\psi_a(x) = \langle x|a\rangle$, and according to the Born rule, the probability that the detector 'clicks' at location $x$ is given by $P_a(x) = |\psi_a(x)|^2$. If we instead block slit $a$ and force the particle through slot $b$, then the resulting probability distribution is given by $P_b(x) = |\psi_b(x)|^2$ where $\psi_b(x) = \langle x|b\rangle$.

If we let the particle travel through *both* slits, then it emerges in a superposition $|s\rangle = (|a\rangle +$

$|b\rangle)/\sqrt{2}$ with a corresponding wavefunction $\psi_{ab}(x) = \langle x|s\rangle = (\psi_a(x) + \psi_b(x))/\sqrt{2}$. The resulting probability distribution of clicks on the detector is given by

$$P_{ab}(x) = |\psi_{ab}|^2 \tag{1a}$$

$$= \frac{1}{2}|\psi_a(x) + \psi_b(x)|^2 \tag{1b}$$

$$= \frac{1}{2}\left[|\psi_a(x)|^2 + |\psi_b(x)|^2 + \psi_a^*(x)\psi_b(x) + \psi_a(x)\psi_b^*(x)\right] \tag{1c}$$

$$= \frac{1}{2}P_a(x) + \frac{1}{2}P_b(x) + \mathrm{Re}[\psi_a^*(x)\psi_b(x)]. \tag{1d}$$

Importantly, the probability is not a simple sum of the probability resulting from each slit. There is an extra term where the amplitudes of the two paths *interfere* with each other. This is the characteristic quantum effect that leads to the non-classical nature of the two-slit experiment.

In the preceding discussion, there was no mention of the environment around the quantum particle. As foreshadowed earlier, we expect that the environment destroys the extra interference term in Equation 1d. We will see that it is able to do so even without affecting the intrinsic dynamics of the particle!

To include the environment into the two-slit experiment, we have to expand our Hilbert space from just system states to a Hilbert space encompassing *product states* of the system and the environment. In other words, the kets now two parts, one of the system and one of the environment. The simplest environment is one that "smells out" which path the particle takes in the following sense. Say the environment is initially prepared in a state $|E_0\rangle$. If we block slit $b$ and force the particle to pass through slit $a$, then the system-environment interaction causes the environment to enter the state $|E_a\rangle$:

$$|a\rangle|E_0\rangle \longrightarrow |a\rangle|E_a\rangle.$$

If on the other hand the particle passes through slit b, then the system-environment state evolves as

$$|a\rangle|E_0\rangle \longrightarrow |b\rangle|E_b\rangle.$$

Because the Schrodinger Equation is linear, the superposition of states ("passing through both slits") must evolve as

$$\frac{1}{\sqrt{2}}\left(|a\rangle + |b\rangle\right)|E_0\rangle \longrightarrow \frac{1}{\sqrt{2}}\left(|a\rangle|E_a\rangle + |b\rangle|E_b\rangle\right). \tag{2}$$

The environment states $|E_a\rangle$ and $|E_b\rangle$ differ in subtle ways; perhaps the surrounding air molecules have been scattered in a slightly different way from the particle passing through; perhaps there has been a slight 'recoil' of the slit-board in the manner of Bohr and Einstein. The

main point is that the envrionment is in a different state depending on which slit the particle went through; if the system is in state $|a\rangle$, then the environment is in state $|E_a\rangle$, if the system is in state $b\rangle$ then the environment is in state $|E_b\rangle$. The environment has become *entangled* with the state of the system, and in a sense, it has 'measured' the state of the system.

To calculate the probability distribution of detector clicks in the presence of envrionmental entanglement, it is helpful to use the formalism of (reduced) density matrices. The full Hilbert space includes both the system and the environment, but if we wish to restrict our attention to just the system, we need to take a partial trace over the environmental degrees of freedom. The resulting reduced density matrix for the system captures all the observable behavior for the system, and "averages out" over the unimportant states of the environment.

When the particle passes through both slits, the full density matrix describing its entanglement with the surroundings can be written as

$$\rho_{SE} = |\Psi\rangle\langle\Psi| \tag{3a}$$

$$= \frac{1}{2}\Big[|a\rangle\langle a||E_a\rangle\langle E_a| + |b\rangle\langle b||E_b\rangle\langle E_b| + |a\rangle\langle b||E_a\rangle\langle E_b| + |b\rangle\langle a||E_b\rangle\langle E_a|\Big] \tag{3b}$$

Taking a partial trace of the system-and-environment density matrix yields the following reduced density matrix for just the system:

$$\rho_S = \mathrm{Tr}_E\big[\rho_{SE}\big] \tag{4a}$$

$$= \frac{1}{2}\Big\{|a\rangle\langle a|\,\mathrm{Tr}\big[|E_a\rangle\langle E_a|\big] + |b\rangle\langle b|\,\mathrm{Tr}\big[|E_b\rangle\langle E_b|\big] \tag{4b}$$

$$+ |a\rangle\langle b|\,\mathrm{Tr}\big[|E_a\rangle\langle E_b|\big] + |b\rangle\langle a|\,\mathrm{Tr}\big[|E_b\rangle\langle E_a|\big]\Big\} \tag{4c}$$

$$= \frac{1}{2}|a\rangle\langle a| + \frac{1}{2}|b\rangle\langle b| + \frac{1}{2}|a\rangle\langle b|\langle E_a|E_b\rangle + \frac{1}{2}|b\rangle\langle a|\langle E_b|E_a\rangle. \tag{4d}$$

Finally, the distribution of clicks on the detector is given by taking the trace against the position operator:

$$P_{ab}^{env} = \mathrm{Tr}\big[\hat{x}\rho_S\big] \tag{5a}$$

$$= \langle x|\rho_S|x\rangle \tag{5b}$$

$$= \frac{1}{2}\langle x|a\rangle\langle a|x\rangle + \frac{1}{2}\langle x|b\rangle\langle b|x\rangle + \frac{1}{2}\langle x|a\rangle\langle b|x\rangle\langle E_a|E_b\rangle + \frac{1}{2}\langle x|b\rangle\langle a|x\rangle\langle E_b|E_a\rangle \tag{5c}$$

$$= \frac{1}{2}|\psi_a(x)|^2 + \frac{1}{2}|\psi_b(x)|^2 + \mathrm{Re}\Big\{\langle E_a|E_b\rangle\psi_a^*(x)\psi_b(x)\Big\}. \tag{5d}$$

Notice that the strength of the interference term now depends on the *overlap* between the environmental states reading out the two possible paths. In particular, if $\langle E_a|E_b\rangle = 0$ and the

environmental states are completely distinguishable, then the probability distribution is

$$P(x) \to \frac{1}{2}|\psi_a(x)|^2 + \frac{1}{2}|\psi_b(x)|^2 \tag{6}$$

and the interference completely disappears! The probability becomes a classical sum of the probabilities of passing through individual slits.

If the environment *incompletely* measures the path of the particle $(0 < |\langle E_a|E_b \rangle|^2 < 1)$, then the interference pattern *partially* disappears. Remarkably, this result has been experimentally observed for the interference pattern of $C_{70}$ buckyballs – as the ambient gas pressure in the experimental chamber is increased, the visibility of the interference fringes goes down, indicating that collisions with surrounding gas particles cause the buckyballs to transition from quantum to classical behavior [5]. Philosophicallly, there is no definitive line between quantum and classical behavior, but rather, systems gradually transition from one regime to another as the amount of system-environment entanglement increases.

The above calculation is rather simple, but its implications are profound. Of particular note, the entanglement with the environment causes the system (that is, its reduced density matrix) to behave as a *classical mixture* of states $(\rho \sim |a\rangle\langle a| + |b\rangle\langle b|)$ rather than a *quantum superposition* $(\rho \sim (|a\rangle + |b\rangle)(\langle a| + \langle b|))$. Once it becomes a classical mixture, the characterstic coherence of the quantum state that gives rise to interference terms between different parts of a superposition – such as $\psi_a^*(x)\psi_b(x)$ – no longer appears in observable probabilities. This decoherence happens regardless of how exactly the system and the envrionment interact; in particular, it can happen without dissipation (energy loss) of the underlying quantum system. Furthermore, there is no need for an observing apparatus or a conscious observer; the decoherence happens as long as the system-environment interaction leaves a 'mark' on the environment about which of the two paths was traversed. The effective dynamics result simply from the entanglement with the environment.

## III.  THE PARTICLE ON A CIRCLE

Having paid our dues to decoherence, we turn now to the quantum mechanics of a particle on a circle. [Note that this problem is equivalent to a rotor restricted to a plane; the orientation of the rotor corresponds to the (angular) position on the circle, and its moment of inertia corresponds to the particle's mass.]

## A.  Position and Momentum Eigenstates

The position of the particle (or orientation of the rotator) is described by an angle $\phi$. We take the principal value of $\phi$ to lie in the range $-\pi \leq \phi < \pi$, where the points $\phi = -\pi$ and $\phi = \pi$ describe the same position. The eigenstates of this 'position' operator describe particles localized at one angle on the circle and are labeled as $|\phi\rangle$.

Since $\phi$ is inherently multi-valued with a sudden 'branch cut' somewhere on the unit circle, it is more convenient to work with the unitary operator

$$\hat{U} := e^{i\hat{\phi}} \tag{7}$$

with eigenbasis

$$\hat{U}|\phi\rangle = e^{i\phi}|\phi\rangle \tag{8}$$

representing the position on the (complex) unit cricle. We can also define corresponding Hermitian operators representing the $x$ and $y$ displacements on the circle, given by

$$\cos\hat{\phi} := \frac{\hat{U} + \hat{U}^\dagger}{2} \tag{9}$$

and

$$\sin\hat{\phi} := \frac{\hat{U} - \hat{U}^\dagger}{2i} \tag{10}$$

respectively.

The general state on the circle $|\alpha\rangle$ is described by a wavefunction $\psi_\alpha(\phi) = \langle\phi|\alpha\rangle$, where $\phi = \phi_0$ and $\phi = \phi_0 + 2\pi$ represent the same point on the circle. For the wavefunction to be single-valued, it must be periodic with a period of $2\pi$. As a result, the Fourier series of the wavefunction is composed of a *discrete* spectrum of momentum states.

To see how momentum is quantized, let us consider an (angular) momentum eigenstate $|\ell\rangle$. It is defined by the relation

$$\hat{L}|\ell\rangle = \ell|\ell\rangle, \tag{11}$$

where $\hat{L}$ is the angular momentum operator $-i\frac{\partial}{\partial\phi}$. As usual, the wavefunction of a momentum eigenstate in the position basis is given by

$$\psi_\ell(\phi) = \langle\phi|\ell\rangle = \frac{1}{\sqrt{2\pi}}e^{i\ell\phi}, \tag{12}$$

with the factor out front coming from normalization $1 = \int_{-\pi}^{+\pi} |\psi_\ell(\phi)|^2 d\phi$. For the wavefunction to be single-valued, $\ell$ must be an integer [6], and to complete the entire Hilbert space, $\ell$ must take on all positive and negative integer values: $\ell = 0, \pm 1, \pm 2, \ldots$.

Notice that $\hat{U}$ increases the angular momentum of a rotational eigenstate by one; i.e., it is a raising operator. To see this, we compute

$$\hat{U}|\ell\rangle = \hat{U} \int_{-\pi}^{+\pi} \frac{e^{i\ell\phi}}{\sqrt{2\pi}} |\phi\rangle d\phi \tag{13}$$

$$= \int_{-\pi}^{+\pi} \frac{e^{i\ell\phi}}{\sqrt{2\pi}} e^{i\phi} |\phi\rangle d\phi \tag{14}$$

$$= \int_{-\pi}^{+\pi} \frac{e^{i(\ell+1)\phi}}{\sqrt{2\pi}} |\phi\rangle d\phi \tag{15}$$

$$= |\ell + 1\rangle. \tag{16}$$

Similarly,

$$\hat{U}^\dagger = |\ell - 1\rangle. \tag{17}$$

In the language of Fourier Series, the momentum eigenstates form a basis of complex exponentials for the space of all functions with a period of $2\pi$. An arbitrary state $|\alpha\rangle$ can be written in the position representation as

$$|\alpha\rangle = \int_{-\pi}^{+\pi} \psi_\alpha(\phi) |\phi\rangle \, d\phi \tag{18}$$

or in the momentum representation as

$$|\alpha\rangle = \sum_{\ell=-\infty}^{+\infty} \hat{\psi}_\alpha[\ell] |\ell\rangle \tag{19}$$

where the momentum-space-coefficients $\hat{\psi}_\alpha[\ell]$ are the Fourier coefficients for the position-space wavefunction:

$$\psi_\alpha(\phi) = \sum_{\ell=-\infty}^{+\infty} \hat{\psi}_\alpha[\ell] \frac{e^{i\ell\phi}}{\sqrt{2\pi}}. \tag{20}$$

These statements can be summarized in the resolution of the identity:

$$I = \int_{-\pi}^{+\pi} |\phi\rangle\langle\phi| \, d\phi = \sum_{\ell=-\infty}^{+\infty} |\ell\rangle\langle\ell|. \tag{21}$$

### B.  Dynamics

The free particle Hamiltonian for the particle on a circle is analogous to the $p^2/2m$ for the praticle on a line, except the linear momentum is replaced with the angular momentum $L$, and the mass is replaced with the moment of inertia $I$. Hence the Hamiltonian is

$$H = \frac{L^2}{2I} \tag{22}$$

and the equation of motion is

$$\frac{d}{dt}|\Psi\rangle = -iH|\Psi\rangle = -i\frac{\hat{L}^2}{2I}|\Psi\rangle. \tag{23}$$

The energy eigenstates are the (angular) momentum eigenstates $|\ell\rangle$, which are delocalized over all the angles of the circle.

Rather than explicitly solving this equation, I will just describe the behavior qualitatively. If a particle begins in a momentum eigenstate, it stays in that eigenstate (but picks up a time-varying phase.) If it begins in a position eigenstate, it quickly spreads out, but unlike the like the free particle on a line, the probability density does not escape out to infinity but is trapped on the circle. The periodicity of $2\pi$ may cause some interesting beating effects, but in essence, the wavepacket slowly spreads over time. If the particle begins in a narrow superposition of position eigenstates in a wavepacket-like form, the time evolution still causes it to spread out and eventually spread all over the circle.

## IV.   DECOHERENCE OF THE PARTICLE ON A CIRCLE

Now we add environmental interactions to the dynamics of the free particle on a circle [7].

### A.   Form of the Master Equation

As shown in section II, the interaction with the environment causes the system to no longer behave as a pure state but as a mixture. Thus, to describe how the system evolves in the presence of the environment, we cannot use an equation for a quantum state like Equation 23; we need to use an equation describing how the system's *density matrix* evolves.

Formally, the combined system-environment evolves unitarily according to the laws of quantum mechanics, but if we wish to study the dynamics of the system, we need to trace out over the environmental degrees of freedom and end up with the effective dynamics of the system's reduced

density matrix. Note that the tracing operation is non-unitary, so in general, the time evolution of the system will no longer be unitary. (This does not imply that quantum mechanics as a whole is non-unitary; it just means that if we only restrict our attention to the central system and ignore the environment, then the system effectively evolves non-unitarily.)

In the absence of the environment, the system's density matrix evolves as

$$\frac{\partial}{\partial t}\rho = \frac{d}{dt}\left(|\Psi\rangle\langle\Psi|\right) \tag{24}$$

$$= \left(\frac{d}{dt}|\Psi\rangle\right) + |\Psi\rangle\left(\frac{d}{dt}\langle\Psi|\right) \tag{25}$$

$$= \left(-iH|\Psi\rangle\right)\langle\Psi| + |\Psi\rangle\left(\langle\Psi|iH\right) \tag{26}$$

$$= -iH\rho + i\rho H \tag{27}$$

$$= -i[H,\rho]. \tag{28}$$

When we include the effects of the environment, there will be an extra non-unitary part:

$$\frac{\partial}{\partial t}\rho = -i\left[H,\rho\right] + \mathcal{D}\big[\rho\big] \tag{29}$$

The exact form of $\mathcal{D}[\rho]$ depends on the nature of the system-environment interaction in the particular system we are considering. If we wish to describe the rotational states of a molecule (gaseous HCl, say), then the extra term would contain contributions from the following processes:

- The molecule emits a photon and transitions from one rotational state to another (spontaneous emission).

- A pair of nearby molecules exchange a virtual photon, exciting one molecule and de-exciting another, entangling their rotational states.

- A molecule collides with another molecule, exchanging (translational) momentum and angular momentum.

The first two interactions involve the angular momentum operator of the rotating molecule, and hence do not localize the orientational state of the molecule. Collisions, on the other hand, depends on the exact orientation of the colliding molecules, and hence are capable of decohering any superpositions of different orientations of the molecule. More precisely, the molecule-molecule interaction depends on the *radial distance* between the nuclei – the position operator – and since the interaction depends on the positions of the nuclei, it is capable of 'measuring' the angular orientation of the molecule.

How effectively a collision decoheres superpositions of different orientations depends on the size of the molecule. For small molecules like HCl, a single collision is not able to measure with great precision the angle of the hydrogen atom. This is because the hydrogen atom is weakly interacting (compared to the Cl electron cloud), and because its distance from the center of mass is small (so that a small uncertainty in position means a large uncertainty in angle). Nevertheless, over the course of many collisions, the information about the orientation of the hydrogen atom will slowly become encoded in the environment (by entanglement with other molecules' orientations), and eventually, the hydrogen atom will be localized as a result of these collisions. For larger molecules such as proteins which consist of thousands of atoms locked in a (mostly) rigid body, the situation is different. In this case, a *single* collision with a surrounding solvent molecule is able to localize its overall rotational wavefunction to a particular orientation. This is because a single scattering event determines the location of an atom and hence the orientation of the molecule. In this case, the decoherence rate is maximal and simply equivalent to the total scattering rate of environmental particles off the rotating molecule.

In the langauge of the decoherence theory, the small-molecule and large-molecule cases differ by how much orientational information gets encoded in the environment as a result of a single collisional event. For small molecules, a collision provides *partial* which-path information, while for large molcules, it provides *full* which-path information. [Schematically, the overlap between the environmental states $\langle E_a | E_b \rangle$ is nonzero for small molecules, but is essentially zero for larger molecules.] In the full-which-path-information scenario, the decoherence rate is simply the rate of scattering events, but in the partial-which-path-information scenario, the dynamical processes can be more nuanced.

*An Analogy.* The theory of how collisions cause localization of *translational* degrees of freedom also involves two regimes. Here the question is about how well collisions can resolve a coherent superposition of two well-localized wavepackets located a distance $\Delta x$ apart ('positional resolution') [1]. If the (de Broglie) wavelength $\lambda_0$ of the scattered particle is much smaller than the coherent separation $\Delta x$, then one scattering event suffices to localize the particle. In the opposite regime where $\lambda_0 \gg \Delta x$, many scattering events are required to encode enough information about the location of the particle into the environmental degrees of freedom. These are known as the long-wavelength and short-wavelength limits, respectively. The discussion below happens in the long-wavelength limit where multiple scattering events are required in order to fully resolve the position.

## B.    The Caldeira-Leggett Model

Now I discuss the mathematical form of the non-unitary part $\mathcal{D}[\rho]$ in the master equation (Equation 29). I will use the Caldeira-Legget model [8], which treats the environment as a collection of harmonic oscillators bilinearly coupled to the position of the central degree of freedom. (This form of environment is quite general for many sorts of open quantum systems [8].)

The derivation of the Caldeira-Leggett equation involves many assumptions about the nature of the environment and of the interaction Hamiltonian. For the system discussed in this paper – the rotations of gas-phase HCl – most of these assumptions are met. These include the long-wavelength limit (i.e., many collisions are necessary to resolve orientation), a high-temperature environment compared to the energy spacings of the system, Markovian behavior (i.e., the environment forgets its state between collisions), and more [1]. Hence we can believe that the model discussed below adequately represents the physics of HCl.

In the Caldeira-Leggett model, the central system is a particle on a line. After a series of long manipulations and assumptions, Caldeira and Legget arrive at the following form of the master equation:

$$\frac{\partial}{\partial t}\rho = \underbrace{-i\left[H', \rho\right]}_{\text{unitary}} - \underbrace{i\gamma_0\left[\hat{x}, \hat{p}\rho + \rho\hat{p}\right]}_{\text{dissipation}} - \underbrace{2m\gamma_0 k_B T\left[\hat{x}, [\hat{x}, \rho]\right]}_{\text{decoherence}} \tag{30}$$

In this expression, $\hat{x}$ and $\hat{p}$ are the position and momentum operators for the particle on the line, $\gamma_0$ is a damping constant that depends on the coupling strengths of the central system to the surrounding harmonic oscillators, and $k_B T$ is the temperature. Each of the terms can be interpreted easily. The first term is the standard unitary evolution of a quantum system, slightly modified (lamb-shifted from $H$ to $H'$) due to the environment's effects. The second term is the classical momentum dissipation that takes energy out of the system and causes it to thermally equilibrate with the surrounding heat bath. The third term is distinctly quantum mechanical and results in decoherence [1].

Dissipation and decoherence occur on different timescales. We can figure out the ratio between these timescales by looking at the second and third terms in equation 30. The second term tells us that the dissipation rate is $\gamma_0$. The third term is a bit more complicated. The coefficient of $2mk_B T$ is related to the thermal de Broglie wavelength

$$\lambda_{dB} = 1/\sqrt{2mk_B T} \tag{31}$$

of the particle, and the double commutator in the dissipation term can be expanded out as

$$[\hat{x}, [\hat{x}, \rho]] = [\hat{x}, \hat{x}\rho - \rho\hat{x}] \tag{32}$$

$$= \hat{x}\hat{x}\rho + \rho\hat{x}\hat{x} - 2\hat{x}\rho\hat{x}. \tag{33}$$

If we write the density matrix out in its position state representation as

$$\langle x|\rho|x'\rangle = \rho(x, x'), \tag{34}$$

we see that the double commutator becomes

$$\langle x|[\hat{x}, [\hat{x}, \rho]]|x'\rangle = x^2\rho(x, x') + \rho(x, x')(x')^2 - 2x\rho(x, x')x' \tag{35}$$

$$= \left(x^2 + (x')^2 - 2xx'\right)\rho(x, x') \tag{36}$$

$$= (x - x')^2\rho(x, x'). \tag{37}$$

The size of the decoherence term is hence quadratic in the extent of coherent spatial spread $x - x'$ in the wavefunction. Putting these facts together, we see that decoherence occurs with a rate of

$$\Gamma = 2mk_BT\left(x - x'\right)^2\gamma_0 = \left(\frac{x - x'}{\lambda_{dB}}\right)^2\gamma_0. \tag{38}$$

It is helpful to plug in some numbers to compare the rates of dissipation and decoherence. For a HCl molecule, the wavefunction of the hydrogen nucleus is at most spread out over twice the length of the H-Cl bond, so $x - x' \approx 2.6$Å. At room temperature, its de Broglie wavelength is $\lambda_{dB} \sim 0.3$Å, so the decoherence occurs on a timescale roughly $(2.6/0.3)^2 \approx 75$ times more rapidly than dissipation. For larger molecules, the separation in timescales becomes even more pronounced, since the rotational eigenstate spans a larger spatial extent and the mass of the molecule increases.

Hence, if we just care about the behavior over decoherence timescales, we can ignore the dissipative term, simplifying the master equation to

$$\frac{\partial}{\partial t}\rho \approx -i\left[H', \rho\right] - 2m\gamma_0 k_BT\left[\hat{x}, [\hat{x}, \rho]\right] \tag{39}$$

$$= -i\left[H', \rho\right] - 2m\gamma_0 k_BT\left(\hat{x}\hat{x}\rho + \rho\hat{x}\hat{x} - 2\hat{x}\rho\hat{x}\right). \tag{40}$$

## C.  Caldiera-Legget for the circle

Now I extend the Caldeira-Legget model to describe the motion of a particle on a circle rather than on a line.

Say the particle of mass $m$ is constrained to move on a circle with radius $r_0$. The coordinates of the particle are described by $\hat{x} = r_0 \cos \hat{\phi}$ and $\hat{y} = r_0 \sin \hat{\phi}$, where $\phi$ is the angular coordinate along the circle. To model the environmental interaction, I suppose that each of the translational directions – $x$ and $y$ – are coupled to the surroundings in the same manner as the 1D Caldeira-Leggett model (Equation 39). This form of the interaction is justifiable if the the collisions in the two directions are identical and uncorrelated. In essense, in my model, I 'stitch on' two Caldeira-Leggett models in the $x$ and the $y$ directions, and constrain the particle to move on a circle of fixed radius.

The resulting decoherence term looks like

$$\mathcal{D}[\rho] = -2m\gamma_0 k_B T \Big\{ [\hat{x}, [\hat{x}, \rho]] + [\hat{y}, [\hat{y}, \rho]] \Big\}, \tag{41}$$

which can be expanded out as

$$\mathcal{D}[\rho] = -2m\gamma_0 k_B T \Big\{ \left( \hat{x}^2 \rho + \rho \hat{x}^2 - 2\hat{x}\rho \hat{x} \right) + \left( \hat{y}^2 \rho + \rho \hat{y}^2 - 2\hat{y}\rho \hat{y} \right) \Big\} \tag{42}$$

$$= -2m\gamma_0 k_B T \Big\{ r_0^2 \left( \sin^2 \hat{\phi} \rho + \rho \sin^2 \hat{\phi} - 2 \sin \hat{\phi} \rho \sin \hat{\phi} \right) + \tag{43}$$

$$r_0^2 \left( \cos^2 \hat{\phi} \rho + \rho \cos^2 \hat{\phi} - 2 \cos \hat{\phi} \rho \cos \hat{\phi} \right) \Big\} \tag{44}$$

$$= -2mr_0^2 \gamma_0 k_B T \Big\{ 2\rho - 2 \sin \hat{\phi} \rho \sin \hat{\phi} - 2 \cos \hat{\phi} \rho \cos \hat{\phi} \Big\}, \tag{45}$$

where between the last two lines I used the identity $\sin^2 \hat{\phi} + \cos^2 \hat{\phi} = I$. The coefficient can be simplified further: the $mr_0^2$ part is the moment of inertia $I$ of the particle on the ring, and so the $mr_0^2 k_B T \gamma_0$ can be identified as the rotational diffusion coefficient $D$ by using the fluctuation-dissipation relation for rotational diffusion. (Recall that when a classical angle variable $\phi$ is coupled to a heat bath, it diffuses around with the relation $\langle \phi^2 \rangle = 2(I k_B T \gamma_0) t$.)

Hence we have

$$\mathcal{D}[\rho] = 4D \left( \sin \hat{\phi} \, \rho \, \sin \hat{\phi} + \cos \hat{\phi} \, \rho \, \cos \hat{\phi} - \rho \right) \tag{46}$$

where the rotational diffusion coefficient $D$ is given by

$$D = mr_0^2 k_B T \gamma_0. \tag{47}$$

For further analysis it is helpful to rewrite the sines and cosines in terms of the raising and lowering operators $U = e^{i\hat{\phi}}$ and $U^\dagger = e^{-i\hat{\phi}}$. We find that

$$\cos \hat{\phi} \, \rho \, \cos \hat{\phi} + \sin \hat{\phi} \, \rho \, \sin \hat{\phi} = \left( \frac{U + U^\dagger}{2} \right) \rho \left( \frac{U + U^\dagger}{2} \right) + \left( \frac{U - U^\dagger}{2i} \right) \rho \left( \frac{U - U^\dagger}{2i} \right) \tag{48}$$

$$= \frac{1}{2} U^\dagger \rho U + \frac{1}{2} U \rho U^\dagger \tag{49}$$

so the overall master equation is

$$\frac{d}{dt}\rho(t) \approx -\frac{i}{2I}\left[\hat{L}^2, \rho(t)\right] + 4D\left(\frac{1}{2}U^\dagger \rho(t)\, U + \frac{1}{2}U\rho(t)\, U^\dagger - \rho(t)\right). \qquad (50)$$

### D.  Decoherence Dynamics

Now I analyze the dynamical behavior of Equation 50. There is no closed-form solution for this equation, so I will make some approximations and examine the behavior of limiting cases to try to gain some intuition about how the system behaves.

Let us consider what happens when the particle starts off in its pure ground state

$$|0\rangle = \frac{1}{\sqrt{2\pi}}\int_{-\pi}^{+\pi}|\phi\rangle\, d\phi \qquad (51)$$

where its wavefunction $\Psi(\phi, t = 0) = 1/\sqrt{2\pi}$ spread out uniformly all around the circle. This ground state is distinctly non-classical because it is a coherent superposition of different position eigenstates. We expect that the decoherence term will rapidly decohere it into a classical mixture rather than a quantum superposition.

To be more precise, we can look at the density matrix in the position representation, given by

$$\rho(t = 0) = |0\rangle\langle 0| = \frac{1}{\sqrt{2\pi}}\int |\phi\rangle\langle\psi|\, d\phi d\psi. \qquad (52)$$

The nonzero off-diagonal terms with $\phi \neq \psi$ indicate that there is quantum interference between the amplitudes of different positions. As the system evolves and decoheres, we expect that the off-diagonal terms (in the position basis) will decay with time. This is analogous to equation 4d, where the interaction with the environment causes a damping of the off-diagonal elements of the density matrix.

After fiddling around with the equation for a bit, one realizes that the dynamics do not cause the different angular momentum eigenstates to mix with each other. This means that we can look for an ansatz of the form

$$\rho(t) = \sum_{\ell=-\infty}^{+\infty} c_\ell(t)|\ell\rangle\langle\ell|, \qquad (53)$$

where the coefficients $c_\ell(t)$ are the dynamical variables to solve for. Our initial condition of $\rho(0) = |0\rangle\langle 0|$ corresponds to $c_\ell(0) = \delta_{0,\ell}$. (Note that the density matrix is diagonal in the momentum basis but not the position basis.)

The first term in Equation 50 (describing unitary evolution) is zero because both the density matrix (Equation 53) and the system Hamiltonian are diagonal in the momentum basis. To find the

second term, we plug in the ansatz and sandwich both sides of the master equation with $\langle \ell | \dots | \ell \rangle$. This gives us

$$\dot{c}_\ell(t) = 4D \left( -c_\ell(t) + \frac{1}{2} \langle \ell | U^\dagger \left[ \sum_m c_m(t) |m\rangle\langle m| \right] U |\ell\rangle + \text{``h.c.''} \right) \tag{54}$$

$$= 4D \left( -c_\ell(t) + \frac{1}{2} \sum_m c_m(t) \left\{ \langle \ell | U^\dagger |m\rangle\langle m| U |\ell\rangle + \langle \ell | U |m\rangle\langle m| U^\dagger |\ell\rangle \right\} \right) \tag{55}$$

$$= 4D \left( -c_\ell(t) + \frac{1}{2} \sum_m c_m(t) \Big\{ \underbrace{\langle \ell+1 |m\rangle\langle m| \ell+1 \rangle}_{\delta_{\ell+1,m}} + \underbrace{\langle \ell-1 |m\rangle\langle m| \ell-1 \rangle}_{\delta_{\ell-1,m}} \Big\} \right) \tag{56}$$

$$= 4D \left( -c_\ell(t) + \frac{1}{2} c_{\ell+1}(t) + \frac{1}{2} c_{\ell-1}(t) \right). \tag{57}$$

Remarkably, the coefficients $c_\ell(t)$ evolve as the probabilities of a continuous-time, discrete-space random walk. We can interpret the $\ell$ values as discrete sites spaced out along a line, and the coefficients $c_\ell(t)$ as the probability that a random walker occupies the site $\ell$ after time $t$. Every $1/4D$ unit of time, there is a probability of $\frac{1}{2}$ that the walker can jump to the left or to the right. The resulting fluxes of probability into and out of each location yields Equation 57.

The exact probability distribution resulting from this stochastic process is complicated, so for our purposes, it is best to invoke the central limit theorem and work in a regime where $\ell \gg 1$ but $\ell$ is not too many standard deviations away from the mean. Under this approximation, the distribution over $\ell$s is a Gaussian whose variance grows linearly with time (as expected for a random walk), and so the density matrix evolves roughly as

$$\boxed{\rho(t) \approx \frac{1}{\sqrt{8\pi Dt}} \sum_\ell e^{-\ell^2/8Dt} |\ell\rangle\langle\ell|.} \tag{58}$$

Note that this expression is not to be taken too literally. For small values of $\ell$, the discrete nature of angular momentum means that the Gaussian approximation is not very good. For large values of $\ell$ more than a few standard deviations $\sqrt{8Dt}$ away from $\ell = 0$, the tails of the probability distribution are exponential rather than Gaussian, so the approximation fails there as well. Additionally, this spreading-out-in-angular-momentum cannot continue indefinitely, because the larger $\ell$ states have greater energy and the environment cannot put energy into the rotor forever. Eventually as time goes on, the system reaches thermal equilibrium with its environment, meaning that the density matrix becomes

$$\rho_{eq} = e^{H/kT} = \sum_\ell e^{\ell^2/2IkT} |\ell\rangle\langle\ell|. \tag{59}$$

The reason that the spread in $\ell$ eventually stops growing is that the dissipative term (Eq. 30) starts to matter once $t$ approaches the dissipative timescale $t \sim 1/\gamma_0$. We expect that the dissipative term, instead of putting energy into the rotor, will take energy *out* of the rotor. The balancing between the decoherence and dissipative effects will result in thermal equilibrium. As a sanity check, if we set the thermal density matrix (Eq. 59) equal to the approximation from the central limit theorem neglecting dissipation (Eq. 58), we find that they look like each other once

$$2IkT \sim 8Dt \qquad \Longrightarrow \qquad t \sim 1/4\gamma_0 \tag{60}$$

(keeping in mind the fluctuation-dissipation relation $D = IkT\gamma_0$) [9].

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[1] M. A. Schlosshauer, *Decoherence: and the quantum-to-classical transition* (Springer Science & Business Media, 2007).

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[6] This condition is true if we wish to describe rotations of a rotator. For an actual particle travelling on a circle, bosons have integer $\ell$, and fermions have half-integer $\ell$. In general, if there is magnetic flux through the circle that couples to a charged particle, then the offset of $\ell$ from an integer is determeined by the amount of flux. In all cases, though, the adjacent values of $\ell$ are exactly one unit apart.

[7] This section (Section IV) consists primarily of my own analysis rather than literature review.

[8] A. Caldeira and A. J. Leggett, Quantum tunnelling in a dissipative system, Annals of physics **149**, 374 (1983).

[9] There is a bit more analysis that I wish to include still; I will add it soon in an updated version.