

Influence of a mesoscopic bath on quantum coherence

Onuttom Narayan¹ and Harsh Mathur²

¹*Department of Physics, University of California, Santa Cruz, California 95064, USA*

²*Physics Department, Case Western Reserve University, 10900 Euclid Avenue, Cleveland, Ohio 44106-7079, USA*

(Received 22 February 2005; published 15 July 2005)

For a quantum double well system interacting with a mesoscopic bath, it is shown that a “bath” consisting of a single particle is sufficient to substantially reduce tunneling between the two wells. This is demonstrated by considering an ammonia molecule in the center of a ring; in addition to halving the maser line frequency, there is an increase in intensity by four orders of magnitude. The tunneling varies nonmonotonically with the number N of electrons in the ring, reflecting the changing electronic correlations. Although the tunneling is reduced for small N , it turns around and grows to its free value for large N . This is shown to not violate Anderson’s orthogonality theorem. Experimental implementations are discussed.

DOI: [10.1103/PhysRevB.72.045338](https://doi.org/10.1103/PhysRevB.72.045338)

PACS number(s): 73.21.–b

The importance of decoherence for the transition from quantum to classical behavior is well known. Various authors have shown that when a system is coupled to a bath consisting of an infinite number of degrees of freedom, irreversibility—and therefore, decoherence—is a consequence; this has given rise to the field of dissipative quantum mechanics.^{1,2} Studies in this field have been further stimulated by the experimental realization of quantum coherent mesoscopic systems that are small on the macroscopic scale but large on the microscopic, atomic scale.³ Control of coupling to the environment is also critical to the success of quantum computing, a fast-expanding area of research.⁴

In this paper we consider a microscopic quantum system that is predominantly coupled to a *mesoscopic* bath. In dissipative quantum mechanics, it is generally presumed that the quantum system is coupled to a macroscopic bath with an essentially infinite number of degrees of freedom. The general belief is that “more is worse;” the greater the coupling to the environment, the more quantum effects are destroyed. Here we show that, for a mesoscopic bath, it is possible for the tunneling between the two wells of a quantum system in a double well potential to be highly nonmonotonic as a function of the number of particles in the bath. There is a substantial reduction in the tunneling with a *single* particle. As the number of particles is increased, the effect of the mesoscopic bath at first sharply decreases, then grows, and finally decreases again, essentially vanishing for sufficiently large particle number. We will show that the last assertion does not contradict Anderson’s orthogonality catastrophe.⁵

We demonstrate these results by considering an often studied double well system, the ammonia molecule,⁶ at the center of a mesoscopic ring. The mesoscopic bath is taken to be a one-dimensional ring; qualitatively similar results would be obtained for a multichannel ring or for a singly connected geometry such as a disk. If the ring has just one electron, the tunneling between the two configurations for the ammonia dipole is reduced by as much as 50%, reducing the frequency of the transition between the symmetric and antisymmetric configurations by the same amount. In addition, the coupling to the electron in the ring increases the dipole moment approximately 100-fold. This dramatically enhances the intensity of the spectral line for the transition, by $\sim 10^4$. As the

number of electrons on the ring is increased, a rich non-monotonic behavior unfolds: the transition frequency almost reverts to the free-molecule value for $N=2$, decreases again as N is increased, and then turns around to again approach the free-molecule value as $N \rightarrow \infty$. Possible experimental realizations are discussed towards the end of the paper.

Parenthetically we note that it is known the coupling of a molecule to light is enhanced by proximity to a metallic nanoparticle, an effect that is the basis of sensitive optical detection of molecules.⁷ The physical origin of this essentially classical effect is field enhancement due to the coupling of optical radiation to metallic plasmons. In contrast the enhancement in the spectral line identified here is quantum in origin.

To establish these results, first consider an ammonia molecule in the center of a ring of radius R . Assume for the moment that the dipole moment of the molecule is in the plane of the ring, and that there is only one electron in the ring. In the absence of the ring, for every orientation of the hydrogen plane, there are two possible positions of the nitrogen atom. The ground and first excited states are the symmetric and antisymmetric combinations of these. Transitions between the two states are used in the ammonia maser.

In the presence of the ring, the two possible positions of the nitrogen atom, which have opposite dipole moments, perturb the electron in the ring, changing its ground state to two different ground states. The possible states of the combined system of the ammonia molecule and the electron in the ring are then

$$|\uparrow\rangle \otimes |E_n\rangle, \quad |\downarrow\rangle \otimes |E'_n\rangle, \quad (1)$$

where $|E_n\rangle$ is the n th electronic eigenstate when the ammonia electric dipole moment is in one orientation, and $|E'_n\rangle$ is the corresponding electronic eigenstate when the dipole moment is in the other orientation. The fact that $|E_n\rangle$ and $|E'_n\rangle$ are different—partially orthogonal—states reduces the tunneling between the two dipole orientations, since the tunneling part of the Hamiltonian connects $|\uparrow\rangle \otimes |\psi\rangle$ to $|\downarrow\rangle \otimes |\psi\rangle$ with the same $|\psi\rangle$ for both states. Thus the splitting between the symmetric and antisymmetric state is reduced. The different ori-

entations of the hydrogen triangle do not affect this calculation, since there is no tunneling between the different orientations, only between the two different positions of the nitrogen atom for a fixed orientation of the hydrogen triangle.

The potential of the electron in the ring due to the ammonia dipole moment is $V(\theta) = ep \cos \theta / (4\pi\epsilon_0 R^2)$, causing matrix elements between the unperturbed electronic eigenstates. The unperturbed eigenstates of the electron are $\psi_0 = 1/\sqrt{2\pi}$ and $\psi_n(\theta) = \cos(n\theta)/\sqrt{\pi}$ for $n \neq 0$, with energies $\hbar^2 n^2 / (2mR^2)$. We neglect the $\sin(n\theta)$ wave functions, since they are not mixed into the ground state by $V(\theta)$. The Hamiltonian for the electron in the presence of the ammonia molecule is

$$H_{ij} = \frac{\hbar^2}{2mR^2} [j^2 \delta_{ij} + \rho(\delta_{i,j-1}\{1 + (\sqrt{2}-1)\delta_{i,0}\} + i \leftrightarrow j)], \quad (2)$$

where $\rho = mepl / (4\pi\epsilon_0 \hbar^2)$ is the dimensionless perturbation strength. Using $\rho \approx 0.58$ Debye for ammonia, H can be truncated to a $n \times n$ matrix, and its eigenvalues obtained numerically. Changing n from 4 to 5 changes the first four eigenvalues by less than 1%, so we use a 4×4 truncation. The eigenvalues are

$$[E_0, E_1, E_2, E_3] = \frac{\hbar^2}{2mR^2} [-0.48, 1.36, 4.05, 9.07]. \quad (3)$$

Including the configuration of the ammonia dipole, the Hamiltonian can be written in the basis $|\uparrow\rangle \otimes |E_n\rangle$, $|\downarrow\rangle \otimes |E'_n\rangle$, where $|E'_n\rangle$ are the electronic eigenstates with the dipole moment $p \rightarrow -p$ (with eigenvalues $E'_n = E_n$). This is an 8×8 matrix with our truncation. In the absence of tunneling between the dipole configurations, this would be a diagonal matrix, with doubly degenerate energy eigenvalues.

Without the electron in the ring, the splitting between the symmetric and antisymmetric levels of the ammonia molecule is 9×10^{-5} eV. In terms of the units used in Eq. (2), for a ring of $R = 10$ nm, the tunneling matrix element is thus

$$t = -0.13 \frac{\hbar^2}{2mR^2}, \quad (4)$$

with the level splitting equal to $2|t|$. Since the tunneling Hamiltonian does not act on the electron in the ring, the tunneling matrix element between $|\uparrow\rangle \otimes |E_n\rangle$ and $|\downarrow\rangle \otimes |E'_m\rangle$ is $t\langle E'_m | E_n \rangle$, which can be evaluated numerically.

If $(E_1 - E_0) \gg |t|$, the degenerate ground state in the 8×8 Hamiltonian is split by $2t\langle E_0 | E'_0 \rangle$. Numerically, for a ring of $R = 10$ nm, the splitting between the lowest two eigenvalues is 0.49 of the free-molecule splitting, and is to a good approximation equal to $2|t|\langle E_0 | E'_0 \rangle$. On the other hand, if R is sufficiently large so that $(E_1 - E_0) \ll |t|$, the radiation frequency is that of the free molecule. Numerically, this happens when $R \gg 30$ nm. If the temperature is large compared to $(E_1 - E_0)$, one has to consider transitions between excited states as well. For a 10 nm ring, the temperature T has to be much less than 8K to ignore the excited states.

For $R < 30$ nm and $T \ll 8$ K, to a good approximation the Hamiltonian can thus be truncated to a symmetric 2×2 matrix, for which the ground and first excited states are the symmetric and antisymmetric combinations $|I\rangle, |II\rangle = (1/\sqrt{2})[|\uparrow; E_0\rangle \pm |\downarrow; E'_0\rangle]$. In order to excite transitions between $|I\rangle$ and $|II\rangle$, one has to apply a time dependent electric field, which couples to the dipole moment of the states. With $P = P_{\text{amm}} + P_{\text{el}}$, one can verify that $\langle I|P|II\rangle = \langle \uparrow | P_{\text{amm}} | \uparrow \rangle + \langle E_0 | P_{\text{el}} | E_0 \rangle$. Since the electronic ground state has a dipole moment opposed to the direction of the ammonia moment, this changes the intensity of the spectral line. Numerically, one obtains

$$\langle I|P|II\rangle = 0.31e\text{\AA} - 0.65eR. \quad (5)$$

For $R = 10$ nm, $\langle I|P|II\rangle$ is changed by a factor of -210 , increasing the line intensity by about 4×10^4 .

We now re-examine the approximations made so far. For a general orientation of the ammonia molecule, only the component of its dipole moment in the plane of the ring couples to the electron in the ring, reducing the effects discussed above. For randomly oriented molecules, a broad band will be seen. Also, if the molecule is not in the center of the ring, the symmetry between the two orientations of its dipole moment is destroyed: the dipole moment prefers to align itself towards the closest point on the ring.

We now consider the effect of increasing the number of electrons in the ring. First, we consider the two electron case in detail. If θ_1 and $-\theta_2$ are the angular positions of the electrons with respect to the dipole moment of the ammonia, changing to $\theta_+ = (\theta_1 + \theta_2)/2$ and $\theta_- = (\theta_1 - \theta_2)$, the electronic Hamiltonian is

$$H = \frac{\hbar^2}{4mR^2} [\partial_+^2 + 4\partial_-^2] - 2\frac{ep}{R^2} \cos \theta_+ \cos\left(\frac{\theta_-}{2}\right) + \frac{e^2}{2R \cos \theta_-}. \quad (6)$$

In the absence of the ammonia dipole, the Hamiltonian is separable. Since the kinetic energy is small compared to the potential energy, $\theta_- \approx \pi$. Expanding θ_- as $\pi - 2\delta$, $H_\delta \approx (\hbar^2/mR^2)\partial^2/\partial\delta^2 + e^2\delta^2/4R$, so that

$$\langle \delta^2 \rangle = \hbar(mRe^2)^{-1/2}. \quad (7)$$

The wave function is uniform in the θ_+ coordinate.

With the dipole, the Hamiltonian of Eq. (6) is no longer separable, so we perform an approximate analysis. From classical arguments it is clear that $\langle \delta(\theta_+) \rangle$ is greatest when $\theta_+ = 0$, for which case by minimizing $-2(ep/R^2)\sin \delta + e^2/(2R \cos \delta)$ we find that $\langle \delta^2 \rangle \approx (4p/eR)^2$. By comparing with Eq. (7), we see that the change in the ground state wave function is negligible for the δ coordinate. For the θ_+ coordinate, with δ set at its optimal value ($\delta = 4p/eR$ for $\theta_+ = 0$, $\delta = 0$ for $\theta_+ = \pm \pi/2$), there is an effective potential energy $-(4p^2/R^3)\cos^2 \theta_+$. Since this is negligible compared to $\hbar^2/2mR^2$, the kinetic energy still forces the wave function to be essentially uniform in θ_+ . Thus the overlap in the ground state wave function of the two electrons with and without the

ammonia dipole potential is almost unity, as is therefore the overlap $\langle E'_0|E_0\rangle$. Hence, for $N=2$, the radiation frequency is essentially that of the free molecule.

If more electrons are added to the ring, $\langle E'_0|E_0\rangle$ decreases: for N electrons at low density the electronic state is essentially a Wigner crystal and the dipole potential shifts each electron from its lattice position by an angle $O(p/eRN)$, whereas the angular spread of each electron wave function is $O(1/N)$. Thus $\langle E'_0|E_0\rangle=[1-O(p/eR)^2]^N$, which tends to zero for large N . But in reality, as N increases, the Wigner crystal melts.⁸ The overlap $\langle E'_0|E_0\rangle$ then *reverses* its N dependence, approaching unity as $N\rightarrow\infty$. Physically this is because in the liquid state the electrons efficiently screen the dipole potential so that it does not significantly perturb the electronic ground state.

To make this plausible, let us consider the more tractable problem of N electrons in a spherical shell of inner radius R , perturbed by a point charge q placed at the center of the shell. Within the Thomas Fermi approximation, the point charge potential is screened within a distance $\sim d_s$ from R . If $d_s\ll R$, screening also changes $V(R)$ from q/R to qd_s/R^2 . The phase shift $\delta_l(k_F)$ due to this potential is approximately zero for $l\gg k_FR$, because of the centrifugal barrier, whereas for $l\ll k_FR$, it is approximately $\delta_0(k_F)$. Within the WKB approximation, $(\delta_0(k_F)\sim V(R)d_s/\hbar v_F)$. Following Anderson,⁵ the overlap between the ground state wave function with and without the point charge is bounded by $\exp[-\sum_l(2l+1)\sin^2\delta_l(k_F)\ln N]$. Combining our results so far, the prefactor to $\ln N$ is $\sim-(k_FR)^2(qd_s/R^2)^2(d_s/\hbar v_F)^2$. If N is increased at fixed R , since $1/d_s\sim k_F\sim N^{1/3}$, the overlap bound is $\exp[-\sim\ln N/N^{4/3}]$, which approaches unity as $N\rightarrow\infty$. This does not conflict with Anderson's result, which applies when N and the system size are both increased, with the particle density held constant.

Alternatively, we could consider N electrons in a spherical box of radius R perturbed by a point charge $-e$ placed at the center of the box. Anderson's bound on the overlap can also be expressed as $\exp[-k^2\sigma_{\text{tot}}\ln N/(12\pi^3)]$ where σ_{tot} is the cross section for an electron at the Fermi surface to scatter from the screened potential of the perturbing point charge. If we take the screened potential to be of the Thomas-Fermi form and use the Born approximation, both justified in the high density limit $k_F a_B\gg 1$ (where $a_B=4\pi\epsilon_0\hbar^2/me^2$ is the Bohr radius), we find

$$\langle E'_0|E_0\rangle\leq\exp\left[-\frac{1}{12\pi}\left(\frac{4}{9\pi}\right)^{1/3}\frac{1}{N^{1/3}}\frac{R}{a_B}\ln N\right] \quad (8)$$

which tends to 1 as $N\rightarrow\infty$ if R is held constant. Again, this does not conflict with Anderson's result.⁵

The case of a strictly one-dimensional ring deserves special consideration. For spinless electrons with short-range interactions at high density, the problem can be analyzed by bosonization, whereby the electron liquid is described in terms of non-interacting bosonic density-wave oscillators.⁹ In this description, the smooth ammonia dipole potential perturbs just one bosonic oscillator via a linear coupling. Thus, in contrast to the case of a sharp Kane-Fisher impurity,¹⁰ the problem remains trivially soluble. A simple calculation re-

veals that the overlap of the electronic ground states corresponding to the two configurations of the ammonia molecule is $\exp[-4\rho^2g/v^2N^2]$, essentially perfect as $N\rightarrow\infty$. Here v and g are Luttinger liquid interaction parameters.⁹ Physically, the reason for this dependence is that the single oscillator to which the ammonia molecule couples becomes stiffer as N (and therefore the Fermi velocity) increases.

In three dimensions, the single particle level spacing scales as $1/N^{1/3}$. Thus as N continues to grow, the bath will ultimately cross over to a macroscopic regime when the single particle level spacing falls well below the tunneling scale t . In this regime, the ammonia molecule can couple to an enormous number of possible particle-hole excitations of the bath, and its tunneling behavior should therefore be well-described in the conventional framework of the spin-boson model.^{2,11} In contrast to the mesoscopic bath, the imperfect overlap between the ground states of a macroscopic bath can renormalize the tunneling frequency of the two level system all the way down to zero, leading to a complete suppression of tunneling, or localization.² Also, there is damping for the macroscopic bath, but not the mesoscopic. Another distinction is the mesoscopic bath-induced amplification of the dipole coupling of ammonia to external radiation. This has, to our knowledge, no counterpart in the physical realizations of the spin-boson model studied so far.

We now briefly consider possible experimental realizations. Metallic rings are unsuitable because of the high density of electrons in metals: rings of the requisite size would contain far too many electrons. Conventional semiconductor devices such as silicon MOSFETs and GaAs MODFETs can have the required low electron density but suffer from the difficulty that the electron gas is buried deep inside the device below a dielectric layer of oxide or semiconductor. However it may be possible to circumvent this difficulty by fabricating devices in which the electron gas lies on the outer surface of the semiconductor and in which the vacuum plays the role of the dielectric layer. GaAs is unsuitable for such devices because of a high density of surface traps, but Kane and co-workers have recently succeeded in passivating the $\langle 111\rangle$ surface of silicon and making working devices of this type.¹² By suitably gating these devices, in principle, it should be possible to laterally confine the electrons to rings. Other systems in which the electron gas has low density and is not deeply buried include electrons deposited on liquid helium¹³ and newer semiconductor structures that are grown by bottom-up techniques and that may be injected with electrons by photoexcitation of a dye with which the structures have been coated.¹⁴ In the former system too electrons could be shepherded into rings by suitable gating;¹⁵ in the latter, the electrons would be confined to nanoscale semiconductor particles that are typically spherical in shape.

Apart from fabricating the rings, the other experimental task is to position ammonia molecules in their vicinity. A simple extension of the calculation above shows that a free ammonia molecule placed at the center of a ring experiences a force that tends to deflect it off-center where the coupling to the ring is much weaker. However this should not be an insurmountable problem at least in the solid state systems mentioned above. For example, with silicon devices, the ammonia molecules could be deposited at random on the silicon

surface, to which they would stick. Although ammonia sticks to a bare silicon surface by covalent bonding of the nitrogen atom¹⁶ that renders dipolar oscillations impossible, on a hydrogen passivated Si surface such as used in the devices of Ref. 12 it is expected that ammonia would stick without such covalent bonding.

In summary, we have shown that a quantum two level system coupled to a mesoscopic bath responds to the bath in an extremely nonmonotonic manner as the number of particles in the bath is changed, and has a strong response even with one particle in the bath. Apart from its intrinsic interest, such an interaction could be relevant to a quantum computing architecture integrating mesoscopic solid state and flying atomic or ionic qubits,¹⁷ a potential application deserving further exploration. As a prototype, we have considered an ammonia molecule in the center of a ring with a small number of electrons. We have discussed possible experimental

techniques to make such devices with a single electron in a ring. The experimental signatures of the effects studied here are: (i) a shift in the ammonia spectral line due to the suppressed tunneling, (ii) a factor of 10^4 increase in the strength of the line due to the enhanced effective dipole moment, and (iii) strong inhomogeneous broadening of the line due to variations in the rings and the couplings between the rings and ammonia molecules. A key role in our analysis is played by the sensitivity of the electronic ground state to external perturbation. It is desirable to determine the dependence of this “wave function stiffness” on N and N/R more rigorously, e.g., numerically.

It is a pleasure to acknowledge helpful discussions with Mike Crommie, Arnie Dahm, Josh Deutsch, Carine Edder, Anupam Garg, Bruce Kane, Peter Littlewood, and Jie Shan.

¹A. O. Caldeira and A. J. Leggett, *Ann. Phys. (N.Y.)* **149**, 374 (1983).

²A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and W. Zwerger, *Rev. Mod. Phys.* **59**, 1 (1987).

³Y. Imry, *Introduction to Mesoscopic Physics* (Oxford University Press, New York, 2002).

⁴I. L. Chuang and M. A. Nielsen, *Quantum Computation and Quantum Information* (Cambridge University Press, New York, 2000).

⁵P. W. Anderson, *Phys. Rev. Lett.* **18**, 1049 (1967).

⁶R. P. Feynman, R. B. Leighton, and M. Sands, *The Feynman Lectures on Physics* (Addison-Wesley, Reading, 1965), Vol. III.

⁷A. J. Haes, D. McFarland, and R. P. Van Duyne, *Proc. SPIE* **5223**, 197 (2003); G. C. Schatz and R. P. Van Duyne, in *Handbook of Vibrational Spectroscopy*, edited by J. M. Chalmers and P. R. Griffiths (Wiley, New York, 2002).

⁸D. M. Ceperley and B. J. Alder, *Phys. Rev. Lett.* **45**, 566 (1980).

⁹For a pedagogical introduction see, e.g., J. von Delft and H. Schoeller, *Annalen der Physik* **7**, 225 (1998).

¹⁰C. L. Kane and M. P. A. Fisher, *Phys. Rev. Lett.* **68**, 1220 (1992).

¹¹If single particle spacings are treated as uniform over the energy range of interest, the particle-hole excitations of the electronic system may be considered an assembly of bosons via bosonization, as shown by R. Denton, B. Mühlischlegel, and D. J. Scalapino, *Phys. Rev. B* **7**, 3589 (1973). If the matrix element of the dipole potential between two single particle levels is also constant (or depends only on the difference in the energy) the ammonia-bath interaction is of the spin-boson form.

¹²K. Eng, R. McFarland, and B. Kane, *Bulletin, APS March Meeting* (2004); (private communication).

¹³M. W. Cole, *Rev. Mod. Phys.* **46**, 451 (1994).

¹⁴C. B. Murray, C. R. Kagan, and M. G. Bawendi, *Annu. Rev. Mater. Sci.* **30**, 545 (2000); G. M. Turner, M. C. Beard, and C. A. Schmittenmaer, *J. Phys. Chem. B* **106**, 11716 (2002).

¹⁵A. Dahm (private communication).

¹⁶R. J. Hamers, Ph. Avouris, and F. Bozso, *Phys. Rev. Lett.* **59**, 2071 (1987).

¹⁷A. Steane, *Nature (London)* **422**, 387 (2003), and references therein.