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Dissipative quantum dynamics in terms of a reduced density matrix distributed over the environment energy

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Abstract. – A new perturbative quantum master equation is presented for the dissipative dynamics of a subsystem inside a nanosystem with a constant and finite total energy. The degrees of freedom besides those of the subsystem play the role of the environment. Our equation rules the time evolution of the distribution functions of the subsystem populations and coherences over the energy of the environment. Thanks to these distribution functions, we can take into account the effects of the conservation of the total energy. This equation is more general than the standard perturbative equations used for describing a subsystem interacting with an environment (such as the Redfield or Cohen-Tannoudji equations) because these latter equations can be deduced from it in the limit of an infinitely large environment. We numerically apply this equation to the spin-GORM model for the interaction of a two-level subsystem with an environment described by random matrices. We compare our equation with the exact von Neumann equation of the total system and show its superiority compared to the Redfield equation (in the Markovian and non-Markovian cases).

The study of the quantum dynamics of a spin or another subsystem interacting with its environment is an old but still very important problem in quantum mechanics and more specifically in non-equilibrium quantum statistical mechanics. The understanding of such problems is central for the study of very fundamental physical processes such as the relaxation to the thermodynamic equilibrium, the thermalization at the environment temperature, or the decoherence. A standard equation to describe the dynamics of a spin interacting with its environment is the non-Markovian Redfield equation (see, *e.g.*, ref. [1]). This equation describes the quantum dynamics in terms of the reduced density matrix of the subsystem which is coupled to its environment. Its non-Markovian character arises because of the dependence on time of the coefficients appearing in the equation. The other well-known equations previously used for the study of relaxation and decoherence (*i.e.*, the Markovian Redfield equation [1–5], the Cohen-Tannoudji equation [6] or the Lindblad equation [7]) can be derived from the non-Markovian Redfield equation. The Markovian form of this equation was first derived empirically for understanding NMR experiments [2]. It was later deduced formally by perturbation theory from the von Neumann equation of the total system (subsystem plus environment) and applied to wide areas of physics and chemistry [5]. In the Redfield theory, the fundamental assumption is that the environment is infinite and therefore not affected by the subsystem. However, recent advances in nanotechnology have focused the attention to isolated nanosystems which contain a few hundred or thousand degrees of freedom. In such nanosystems, one can encounter situations where a quantum degree of freedom such as a spin is interacting with the other degrees

of freedom of the isolated nanosystem. These other degrees of freedom may be sufficiently many that their energy levels form a dense quasi-continuous spectrum, allowing the relaxation and decoherence of the spin. The degrees of freedom other than the spin thus play the role of an environment or energy reservoir. In such isolated nanosystems, the fact is that the density of states of this nanoreservoir may vary on small energy scales of the order of the spin energy itself. In such circumstances, we may no longer expect the validity of the Redfield equation because the infinite environment hypothesis fails and the effects of the finiteness of the total energy of the nanosystem have to be taken into account.

The purpose of this letter is to show that this problem can be solved by distributing the reduced density matrix of the subsystem over the energy of the other degrees of freedom of the nanosystem. In this way, we obtain a description in terms of distribution functions of the populations and coherences of the subsystem or spin over the energy of the nanoreservoir or environment. These distribution functions evolve in time according to a new quantum master equation that takes into account the effect of the finiteness of the energy of the total system. This master equation is thus compatible with the conservation of the total energy of the nanosystem and it solves discrepancies observed when using the standard Redfield equation for nanosystems. In this letter, we present a general form of this master equation in terms of operators acting in the quantum state space of the subsystem or spin. Moreover, we apply this equation to the population dynamics of the spin-GORM model (a two-level subsystem interacting in a non-diagonal way with an environment described by Gaussian orthogonal random matrices) and we finally compare it to the exact von Neumann equation for the total system and to the Redfield equation. We will finally conclude presenting the perspectives created by this work.

The quantum subsystem that we consider is described by the Hamiltonian \hat{H}_S , and its eigenvalues (respectively, its eigenvectors) are denoted by $\{E_s\}$ (respectively, by $\{|s\rangle\}$). The environment is another quantum system described by the Hamiltonian \hat{H}_B which has a dense spectrum of energy levels forming a quasi-continuum. Its eigenvalues (respectively, its eigenvectors) are denoted by $\{\epsilon\}$ (respectively, by $\{|\epsilon\rangle\}$). The interaction between the subsystem and its environment is generally taken to be the product of a subsystem operator \hat{S} and an environment operator \hat{B} . The coupling parameter λ measures the intensity of the interaction between the subsystem and the environment. Therefore, the Hamiltonian of the total system is given by $\hat{H}_{\text{tot}} = \hat{H}_S + \hat{H}_B + \lambda\hat{S}\hat{B}$. The exact time evolution of the total system is described by the von Neumann equation $\dot{\hat{\rho}}(t) = -i[\hat{H}_{\text{tot}}, \hat{\rho}(t)]$, where $\hat{\rho}(t)$ is the density matrix of the total system. The subsystem dynamics may be described by the reduced density matrix of the subsystem $\hat{\rho}_S(t) = \text{Tr}_B \hat{\rho}(t)$. The total system has a finite constant energy. At initial time, the environment is supposed to be in a microcanonical state at energy ϵ .

The ansatz at the basis of the Redfield derivation is to assume that the total density matrix evolves while keeping the following factorized form:

$$\hat{\rho}(t) = \hat{\rho}_S(t) \otimes \frac{\delta(\epsilon - \hat{H}_B)}{n(\hat{H}_B)}, \quad (1)$$

where $n(\hat{H}_B) = \text{Tr}_B \delta(\epsilon - \hat{H}_B)$ is the density of states (DOS) of the environment. We see that the environment part of the density matrix does not evolve, supposing that the environment is not affected by the dynamics. As we announced it, we want to include the effects of the finiteness of the total energy into the description. The main idea (the new ansatz) is to suppose that the total density matrix can be described at all times by a density matrix of the

following form:

$$\hat{\rho}(t) = \frac{\hat{P}(\hat{H}_B; t)}{n(\hat{H}_B)}, \quad (2)$$

where $\hat{P}(\epsilon; t)$ is an operator acting on the quantum state space of the subsystem and depending on the energy ϵ of the environment. The reduced density matrix of the subsystem is therefore given by $\hat{\rho}_S(t) = \text{Tr}_B \hat{\rho}(t) = \int d\epsilon \hat{P}(\epsilon; t)$. The only approximation made by the ansatz is that we neglect the contributions coming from the coherences of the environment. We see that the description can now depend on the environment energy. Taking the trace over the environment degrees of freedom of the von Neumann equation, performing a perturbative expansion up to the second order in λ and closing the equation using the ansatz (2), one gets our new equation:

$$\begin{aligned} \dot{\hat{P}}(\epsilon; t) = & -i[\hat{H}_S, \hat{P}(\epsilon; t)] + \lambda^2 \int_0^t d\tau \int d\epsilon' \left\{ \zeta(\epsilon', \epsilon, \tau) \hat{S}(-\tau) \hat{P}(\epsilon'; t) \hat{S} + \right. \\ & \left. + \zeta^*(\epsilon', \epsilon, \tau) \hat{S} \hat{P}(\epsilon'; t) \hat{S}(-\tau) - \zeta(\epsilon, \epsilon', \tau) \hat{S} \hat{S}(-\tau) \hat{P}(\epsilon; t) - \zeta^*(\epsilon, \epsilon', \tau) \hat{P}(\epsilon; t) \hat{S}(-\tau) \hat{S} \right\} \quad (3) \end{aligned}$$

(the details of the calculation leading to eq. (3) are given elsewhere [8]). The coupling to the environment is fully characterized by the function $\zeta(\epsilon, \epsilon', \tau) = F(\epsilon, \epsilon') n(\epsilon') e^{i(\epsilon - \epsilon')\tau}$, where $F(\epsilon, \epsilon')$ is given by averaging the matrix elements $|\langle \epsilon | \hat{B} | \epsilon' \rangle|^2$ over the dense spectrum of eigenvalues around ϵ and ϵ' . $\zeta(\epsilon, \epsilon', \tau)$ is proportional to the Fermi golden-rule transition probability rate for the environment to jump from an energy ϵ to an energy ϵ' , multiplied by an oscillating term at frequency $\epsilon - \epsilon'$. Performing the same derivation using the standard ansatz (1) instead of the new ansatz (2) would have led to the non-Markovian Redfield equation [1] which is given by replacing $\hat{P}(\epsilon; t)$ by $\hat{\rho}_S(t)$ in eq. (3). Doing this, one loses the dependence on the environment energy, and the non-Markovian Redfield equation can therefore be seen as the particular case of our equation when the DOS of the environment varies on a large energy scale compared to the typical energies of the subsystem. The Markovian approximation consists in taking the infinite-time limit of the time-dependent coefficients of (3) and therefore in replacing \int_0^t by \int_0^∞ in (3). This approximation is justified on long time scales when the dynamics of the environment is fast compared to the subsystem dynamics. Performing this approximation on our new equation (respectively, on the non-Markovian Redfield equation) gives the Markovian version of our new equation (respectively, the Redfield equation [1]).

In the particular case where one further neglects the contributions of the coherences in eq. (3), one gets the following simplified Markovian equation for the population dynamics in the subsystem eigenbasis:

$$\begin{aligned} \dot{P}_{ss'}(\epsilon; t) = & +2\pi\lambda^2 \sum_{s' \neq s} |\langle s | \hat{S} | s' \rangle|^2 F(\epsilon, E_s - E_{s'} + \epsilon) n(\epsilon) P_{s's'}(E_s - E_{s'} + \epsilon; t) - \\ & -2\pi\lambda^2 \sum_{s' \neq s} |\langle s | \hat{S} | s' \rangle|^2 F(\epsilon, E_s - E_{s'} + \epsilon) n(E_s - E_{s'} + \epsilon) P_{ss}(\epsilon; t), \quad (4) \end{aligned}$$

where $P_{ss'}(\epsilon; t) = \langle s | \hat{P}(\epsilon; t) | s' \rangle$. Equation (4) satisfies detailed balance. Doing the same for the Redfield equation, one gets the well-known Cohen-Tannoudji equation [6]. Our simplified Markovian equations describe the evolution of the total system as a random walk between the states belonging to the same energy shell with transition probabilities given by the Fermi golden rule. This equation may look like the Pauli equation [9, 10] but it describes the time evolution of the distributions of populations over the energy of the environment which is the new feature of our equation. On the other hand, the difference between our simplified

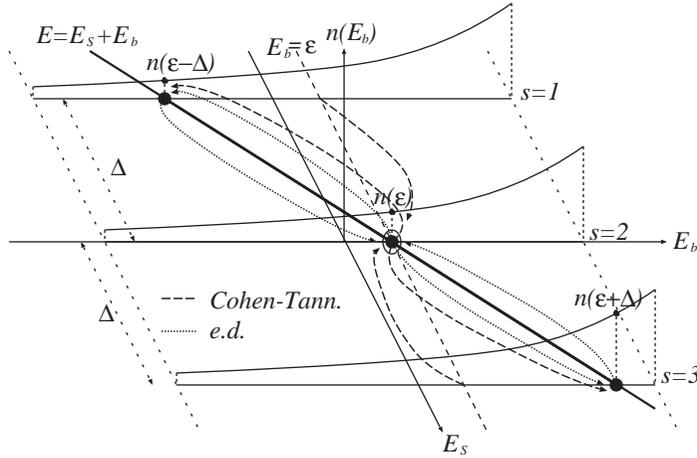


Fig. 1 – Representation of the energy exchanges described, respectively, by the Cohen-Tannoudji (Cohen-Tann.) and by the simplified Markovian version of our new energy-distributed equation (4) (e.d.) for a three-level system. The subsystem energy E_s increases along the x -axis, the environment energy E_b along the y -axis, and the DOS of the environment (supposed to be an increasing function of the environment energy) is depicted along the z -axis. The total energy of the system is given by E . The initial condition is denoted by the circled black dot. We see that transitions that preserve the energy of the total system have to occur along the total energy line $E = E_s + E_b$ (the thick full line) in the (x, y) -plane. Doing this, they satisfy the Fermi golden rule for the total system. One can see that only our energy-distributed equation (e.d.: dotted arrow) satisfies this condition. The Cohen-Tannoudji equation describes transitions (Cohen-Tann.: dashed arrow) that occur along a constant environment energy line and is therefore wrong when the subsystem energies are of the order of or larger than the typical energy scale of variation of the environment DOS.

Markovian equation (respectively, the non-Markovian version of our equation) and the Cohen-Tannoudji equation [6] (respectively, the non-Markovian Redfield equation) holds in the fact that this last equation considers that the environment DOS is not affected by the system energy. This is represented in fig. 1. We will now apply our equation to the spin-GORM model. This model describes the evolution of a two-level system that interacts in a non-diagonal way with a complex environment. Here, the complexity is supposed to come from many-body interactions as in heavy nuclei or from a classically chaotic dynamics. Therefore, the environment operators are given by Gaussian orthogonal random matrices (GORM). Similar models have been studied in refs. [11–17]. The Hamiltonian of the total system is then $\hat{H}_{\text{tot}} = \frac{\Delta}{2}\hat{\sigma}_z + \frac{1}{\sqrt{8N}}\hat{X} + \lambda\hat{\sigma}_x \frac{1}{\sqrt{8N}}\hat{X}'$, where \hat{X} and \hat{X}' are Gaussian orthogonal random matrices of size N and probability density proportional to $\exp[-\frac{1}{4}\text{Tr}\hat{X}^2]$. The three fundamental parameters determining the model are Δ , λ , and N . The smoothed DOS of the environment is given by the Wigner semicircle $n(\epsilon) = \frac{4N}{\pi}\sqrt{\frac{1}{4} - \epsilon^2}$ with the convention that $\sqrt{x} = 0$ for $x < 0$. The smoothed DOS of the unperturbed total system ($\lambda = 0$) is the sum of two semicircles centered at the two energies $-\frac{\Delta}{2}$ and $\frac{\Delta}{2}$, as depicted in fig. 2.

In this paper, we restrict ourselves to the study of the small-coupling regimes $\lambda \ll 1$ because all the equations discussed in this paper are obtained perturbatively. Another restriction is related to the mean level spacing of the environment. The coupling between the levels (that is of order λ^2 because the first order in perturbation theory is zero due to the non-diagonal nature of the coupling) has to be of the order of or larger than the mean level spacing $\frac{1}{N}$ of the environment to induce a sufficient interaction between the levels belonging to the total

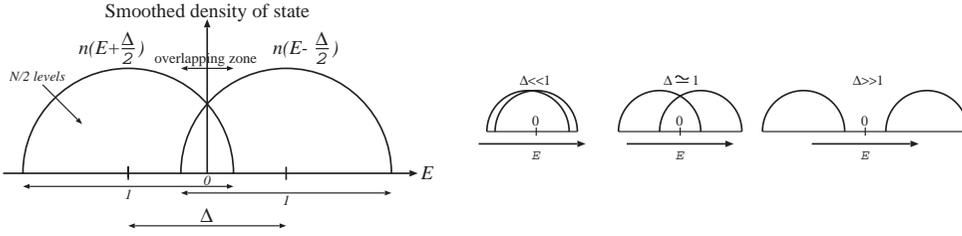


Fig. 2 – Schematic representation of the smoothed DOS of the unperturbed total system $n_{\text{tot}}(E) = \text{Tr} \delta(E - \hat{H}_S - \hat{H}_B) = n(E + \frac{\Delta}{2}) + n(E - \frac{\Delta}{2})$ for different values of Δ .

energy shell in order to reach a microcanonical distribution inside these shells. The criterion, in order that our master equation hold, is therefore $\lambda^2 \geq \frac{1}{N}$. Of course, the lower bound disappears in the continuum limit $N \rightarrow \infty$. The validity domain is shown in fig. 3. The detailed study of the lower bound of the coupling parameter has been done in ref. [17]. We apply our equation (3) to the spin-GORM model in order to study the time evolution of the subsystem populations in terms of $\hat{\sigma}_z$, which is the difference between the probability of being in the upper state of the subsystem minus the probability of being in the lower one:

$$\langle \hat{\sigma}_z \rangle^{NM}(t) = \int d\epsilon' [P_{++}(\epsilon'; t) - P_{--}(\epsilon' + \Delta; t)]. \quad (5)$$

The resulting equation is given by

$$\begin{aligned} \dot{P}_{\pm\pm}(\epsilon; t) = & +\frac{\lambda^2}{\pi} \sqrt{\frac{1}{4} - \epsilon^2} \int_{-\frac{1}{2}}^{+\frac{1}{2}} d\epsilon' P_{\mp\mp}(\epsilon'; t) \frac{\sin(\pm\Delta + \epsilon - \epsilon')t}{(\pm\Delta + \epsilon - \epsilon')} - \\ & -\frac{\lambda^2}{\pi} P_{\pm\pm}(\epsilon; t) \int_{-\frac{1}{2}}^{+\frac{1}{2}} d\epsilon' \sqrt{\frac{1}{4} - \epsilon'^2} \frac{\sin(\pm\Delta + \epsilon - \epsilon')t}{(\pm\Delta + \epsilon - \epsilon')}. \end{aligned} \quad (6)$$

(See ref. [8] for further details.) An important remark is that the Markovian version of our equation (3) and the simplified Markovian approximation (4) are the same for the spin-GORM model. Neglecting the coherence contributions, as done to get eq. (4), is here not necessary to obtain a decoupled population dynamics as eq. (6). We notice that, for the same reason, the Markovian Redfield and the Cohen-Tannoudji equations are identical for the spin-GORM model.

The Markovian version of our equation (6) admits analytical solutions of the form $P_{++}(\epsilon'; t) - P_{--}(\epsilon' + \Delta; t) = \langle \hat{\sigma}_z \rangle^M(t) \delta(\epsilon' - \epsilon)$, where

$$\langle \hat{\sigma}_z \rangle^M(t) = [\langle \hat{\sigma}_z \rangle^M(0) - \langle \hat{\sigma}_z \rangle_\infty^M] e^{-\gamma t} + \langle \hat{\sigma}_z \rangle_\infty^M, \quad (7)$$

with the relaxation rate

$$\gamma = \lambda^2 \left[\sqrt{\frac{1}{4} - \epsilon^2} + \sqrt{\frac{1}{4} - (\epsilon + \Delta)^2} \right],$$

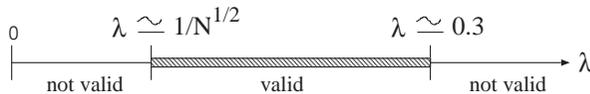


Fig. 3 – Schematic representation of the domain of validity of the master equation along the axis of the coupling parameter λ .

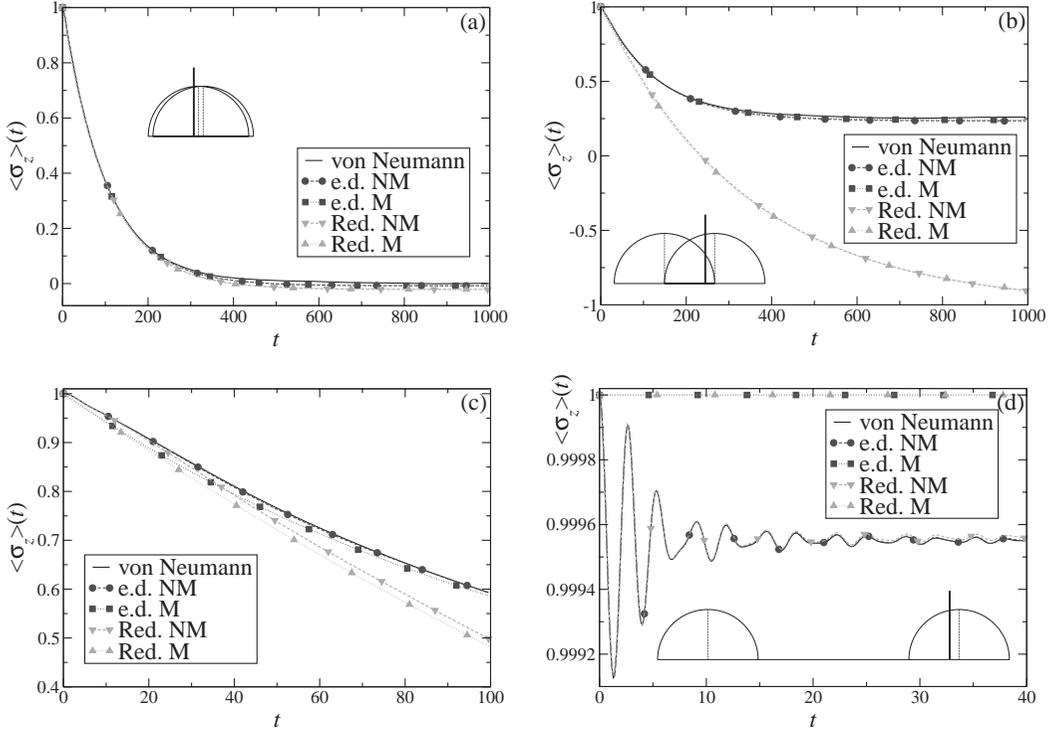


Fig. 4 – Dynamics of the two-level subsystem for the spin-GORM model. “NM” stands for non-Markovian, “M” for Markovian. “e.d.” is our new energy-distributed equation and “Red” is the Redfield equation. The energy splitting between the two levels are, respectively, (a) $\Delta = 0.05$, (b) and (c) $\Delta = 0.5$, and (d) $\Delta = 2.5$.

and the equilibrium population

$$\langle \hat{\sigma}_z \rangle_{\infty}^M = \frac{\sqrt{\frac{1}{4} - \epsilon^2} - \sqrt{\frac{1}{4} - (\epsilon + \Delta)^2}}{\sqrt{\frac{1}{4} - \epsilon^2} + \sqrt{\frac{1}{4} - (\epsilon + \Delta)^2}}.$$

Notice that the relaxation rate and the equilibrium population are independent of the initial condition on the spin itself.

We now start the discussion of the comparison between the different equations based on the numerical simulations of fig. 4. We always take the initial condition $\langle \hat{\sigma}_z \rangle^M(0) = 1$ for the spin. The initial state of the environment is characterized by the energy of the microcanonical distribution $\epsilon = -0.1$. The width of the energy shell is always $\delta\epsilon = 0.05$. The coupling term is taken to be small, $\lambda = 0.1$. The curves are averages over $\chi = 10$ realizations of GORM of size $N = 2000$ and over the different eigenstates in the environment energy shell. If one considers the long-time relaxation of the system, we notice, as we announced it, that our new equation is required when the subsystem energy Δ is of the order of magnitude of the energy scale of variation of the environment smoothed DOS: $n(\epsilon + \Delta) \neq n(\epsilon)$ (see fig. 4b). On the other hand, if Δ is small enough and therefore $n(\epsilon + \Delta) \approx n(\epsilon)$, our equation reduces to the Redfield equation (see fig. 4a). Notice that, on this time scales, the Markovian (M) and the non-Markovian (NM) equations coincide. At short time, because $\frac{\sin(\pm\Delta + \epsilon - \epsilon')t}{(\pm\Delta + \epsilon - \epsilon')}$ is not yet a delta distribution, the non-Markovian equation describes a spread of the probability

in energy around the total unperturbed energy shell. On time scales longer than the typical environment time ($\sim 2\pi$), this spreading becomes negligible and almost only the central part of the function contributes. The Markovian approximation neglects the spreading because it restricts at all times the dynamics inside the microcanonical energy shell of the unperturbed spectrum according to the Fermi golden rule. Therefore, the Markovian approximation is generally valid on long time scales, as one can see in fig. 4a, b and especially in fig. 4c. Because the microcanonical energy shell is not inside the overlapping zone in fig. 4d (see also fig. 2 in the case $\Delta \gg 1$), the subsystem dynamics is purely non-Markovian and the Markovian curves completely miss the dynamics. Notice that when $n(\epsilon + \Delta) \neq n(\epsilon)$, even if the non-Markovian Redfield equation predicts a completely wrong relaxation on long time scales, it gives correct results for very short non-Markovian time scales ($\sim 2\pi$), as one can see in fig. 4c and d.

We can conclude that our new equation is always valid in the small-coupling limit independently of the energy ratio between the system and the environment. It describes the non-Markovian short-time dynamics of the subsystem as well as the long-time one. It is therefore an important equation for the study of nanosystems. This equation reduces to the Redfield equation in case the subsystem energies differences $E_s - E_{s'}$ are very small compared to the typical energy scale of variations of the environment DOS. It can be shown that it is also in this limit that the equilibrium values of the system populations thermalize to a canonical distribution corresponding to the microcanonical temperature of the environment [17]. The dynamics of the quantum coherences will be investigated in future work.

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