Decoherence by a spin thermal bath: Role of spin-spin interactions and initial state of the bath

Shengjun Yuan,1 Mikhail I. Katsnelson,2 and Hans De Raedt1
1Department of Applied Physics, Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, NL-9747 AG Groningen, The Netherlands
2Institute of Molecules and Materials, Radboud University of Nijmegen, 6525 ED Nijmegen, The Netherlands

(Received 26 February 2008; published 7 May 2008)

We study the decoherence of two coupled spins that interact with a spin-bath environment. It is shown that the connectivity and the coupling strength between the spins in the environment are of crucial importance for the decoherence of the central system. For the anisotropic spin bath, changing the connectivity or coupling strength changes the decoherence of the central system from Gaussian to exponential decay law. The initial state of the environment is shown to affect the decoherence process in a qualitatively significant manner.

DOI: 10.1103/PhysRevB.77.184301 PACS number(s): 75.10.Jm, 03.65.Yz, 05.45.Pq, 75.10.Nr

I. INTRODUCTION

Understanding the decoherence in quantum spin systems is a subject of numerous works (for reviews, see Refs. 1 and 2). The issue seems to be very complicated and despite many efforts, even some basic questions about the character of the decoherence process are yet unsolved. Due to the interactions with and between the spin of the bath, an analytical treatment can be carried out in exceptional cases, even if the central systems contain one spin only. Recent works suggest that the internal dynamics of the environment can be crucial to the decoherence of the central system.3–15 In this paper, we present results of extensive simulation work of a two-spin system interacting with a spin-bath environment and show that the decoherence of the two-spin system can exhibit different behaviors, depending on the characteristics of the coupling with the environment, the internal dynamics, and the initial state of the latter. We also provide a simple physical picture to understand this behavior.

In general, the behavior of an open quantum system crucially depends on the ratio of typical energy differences of the central system and the energy of the environment. The $E\ll E_{se}$ case has been extensively studied in relation to the “Schrödinger cat” problem and the physics is quite clear:16,17 As a result of time evolution, the central system passes to one of the “pointer states”17 that, in this case, are the eigenstates of the interaction Hamiltonian $H_{se}$. In the opposite case, $E\gg E_{se}$ is less well understood. There is a conjecture that, in this case, the pointer states should be the eigenstates of the Hamiltonian $H_{se}$ of the central system, but this has been proven for a very simple model only.18 On the other hand, this case is of primary interest if, say, the central system consists of electron spins whereas the environment is nuclear spins, for instance, if one considers the possibility of quantum computation by using molecular magnets.19,20

II. MODEL

We consider a generic quantum spin model described by the Hamiltonian $H=H_{s}+H_{se}+H_{ee}$, where $H_{s}=-JS_{1}S_{2}$ is the Hamiltonian of the central system and the Hamiltonians of the environment and the interaction of the central system with the environment are given by

$$H_{s} = -\sum_{i=1}^{N-1} \sum_{j=i+1}^{N} \sum_{\alpha} \Omega_{ij}^{(\alpha)} I_{i}^{(\alpha)} I_{j}^{(\alpha)}$$

$$H_{se} = -\sum_{i=1}^{2} \sum_{j=1}^{N} \sum_{\alpha} \Delta_{ij}^{(\alpha)} S_{i}^{(\alpha)} I_{j}^{(\alpha)}$$

respectively. The exchange integrals $J$ and $\Omega_{ij}^{(\alpha)}$ determine the strength of the interaction between spins $S_{n}=(s_{n}^{x}, s_{n}^{y}, s_{n}^{z})$ of the central system and the spins $I_{n}=(I_{n}^{x}, I_{n}^{y}, I_{n}^{z})$ in the environment, respectively. The exchange integrals $\Delta_{ij}^{(\alpha)}$ control the interaction of the central system with its environment. In Eq. (1), the sum over $\alpha$ runs over the $x, y,$ and $z$ components of spin-1/2 operators $S$ and $I$. In the sequel, we will use the term “Heisenberg type” $H_{s}$ $(H_{se})$ to indicate that each $\Omega_{ij}^{(\alpha)}$ $(\Delta_{ij}^{(\alpha)})$ is a uniform random number in the range of $[-\Omega, \Omega]$ $([-\Delta, \Delta])$, with $\Omega$ and $\Delta$ being free parameters. In earlier work,14,15 we found that a Heisenberg type $H_{s}$ can induce close to perfect decoherence of the central system and therefore, we will focus on this case only.

The bath is further characterized by the number of environment spins $K$ with which a spin in the environment interacts. If $K=0$, each spin in the environment interacts with the central system only. $K=2$, $K=4$, or $K=6$ corresponds to environments in which the spins are placed on a ring, square, or triangular lattice, respectively, and interact with the nearest neighbors only. If $K=N-1$, each spin in the environment interacts with all the other spins in the environment and, to give this case a name, we will refer to this case as “spin glass.”

If the Hamiltonian of the central system $H_{s}$ is a perturbation relative to the interaction Hamiltonian $H_{se}$, the pointer states are eigenstates of $H_{se}$.17 In the opposite case, that is the regime $|\Delta|\ll |J|$ that we explore in this paper, the pointer states are conjectured to be eigenstates of $H_{se}$.18 The latter is given by $|1\rangle = |T_{1}\rangle = |\uparrow\rangle$, $|2\rangle = |S\rangle = ((|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2}, |3\rangle = |T_{2}\rangle = (|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2}$, and $|4\rangle = |T_{4}\rangle = |\downarrow\rangle$, which satisfy $H_{se}|S\rangle = (3J/4)|S\rangle$ and $H_{se}|T_{i}\rangle = (-J/4)|T_{i}\rangle$ for $i=1,0,1$.

The simulation procedure is as follows. We generate a random superposition $|\phi\rangle$ of all the basis states of the environment. This state corresponds to the equilibrium density matrix of the environment at infinite temperature. Alterna-
tively, to study the effect of the thermal state of the environment on the decoherence processes, we take the two state of the environment to be its ground state. The spin-up–spin-down state (|↑⟩|↓⟩) is taken as the initial state of the central system. Thus, the initial state of the whole system reads |Ψ(t=0)⟩=|↑⟩|↓⟩⟨φ| and is a product state of the state of the central system and the initial state of the environment which, in general, is a (very complicated) linear combination of the 2^N basis states of the environment. In our simulations, we take N=16 which, from earlier work, is sufficiently large for the environment to behave as a “large” system.

For a given fixed set of model parameters, the time evolution of the whole system is obtained by solving the time-dependent Schrödinger equation for the many-body wave function |Ψ(t)⟩, which describes the central system plus the environment. It conserves the energy of the whole system to machine precision. We monitor the effects of the decoherence by computing the matrix elements of the reduced density matrix ρ(t) of the central system.

As explained earlier, in the regime of interest |Δ|≈|φ|, the pointer states are expected to be the eigenstates of the central systems. Hence, we compute the matrix elements of the density matrix in the basis of eigenvectors of the central system. We also compute the time dependence of quadratic entropy S(t)=−Tr[ρ(t)lnρ(t)], where ρ(t) is the density matrix for H_{ce}=0.

III. ISOTROPIC COUPLING TO THE BATH

If the interaction between the central system and environment is isotropic, we have [H_s,H_{ce}]=0. Then, as shown in Appendix, the expressions of the reduced density matrix ρ(t) and the Loschmidt echo L(t) simplify. Indeed, if Δ_i^φ=Δ_j^φ=Δ_{ij}^{(c)}=Δ for all i,j, then

\[ H_{ce} = -J \sum_{j=1}^{N} I_j \]  

commutes with H_s and it follows that the decoherence process of the central system is determined by H_{ce}, H_s, the initial state of whole system |Ψ(t_0)⟩, and the eigenstates of the central system [see Eqs. (A10) and (A11) in Appendix]. In other words, in this case, L(t) and |ρ(t)| are not dependent on the J, which is the interaction between the spins in the central system. Furthermore, if we take the interactions between the environment spins to be isotropic, that is, \[ Ω_{ij}^{(s)} = Ω_{ij}^{(c)} = Ω_{ij} = Ω_{i,j} \] for all i,j, then

\[ H_{ce} = -\sum_{i=1}^{N-1} \sum_{j=i+1}^{N} Ω_{ij} I_i I_j \]  

commutes with H_{ce}, and therefore H_s has no effect on the decoherence process [see Eq. (A13) in Appendix].

In Figs. 1 and 2, we show the time evolution of the elements of the reduced density matrix ρ(t) for different connectivities K and Ω for the case that H_{ce} is an isotropic Heisenberg model, i.e., Δ_i^φ=Δ_j^φ=Δ_{ij}^{(c)}=Δ for all i,j.

If |Δ|≫√K\Ω, which is in agreement with earlier work, we find that in the absence of interactions between the environment spins (\sqrt{K\Ω}=0) and after the initial decay, the central system exhibits long-time oscillations [see Fig. 1(a) (left)]. In this case and in the limit of a large environment, we have

\[ \text{Re} \, ρ_{23}(t) = \left[ \frac{1}{6} + \frac{1 - b t^2}{3} e^{-e^{r^2}} \right] \cos \omega t, \]  

where b=\:N\delta^2/4, c=b/2, and \:ω=J−Δ. Equation (4) clearly shows the two-step process, that is, after the initial Gaussian decay of the amplitude of the oscillations, the oscillations revive and their amplitude levels in Ref. 24. Due to conservation laws, this behavior does not change if the environment consists of an isotropic Heisenberg system (Ω_i^φ=Ω for all α, i, and j), which is independent of K. If, as in Ref. 23, we take Δ_0^φ=Δ_0^s=Δ_j^s∈[0,Δ] random instead of the identical, the amplitude of the long-living oscillations is no longer constant but very slowly decays (results not shown).

If |Δ|≪√K\Ω, the presence of Heisenberg-type interactions between the spins of the environment has little effect on the initial Gaussian decay of the central system, but it leads to a reduction and to a decay of the amplitude of the long-living oscillations. The larger K [see Figs. 1(b)–1(e), (left)] or Ω [see Figs. 2(a) and 2(c)], the faster the decay is. Note that for the sake of clarity, we have suppressed the fast oscillations by plotting instead of the real part, which is the absolute value of the matrix elements.

If |Δ|≪√K\Ω, keeping K fixed and increasing Ω smoothly changes the initial decay from Gaussian (fast) to exponential (slow), and the long-living oscillations are completely suppressed [see Figs. 2(b) and 2(d)]. For large Ω, the simulation data fit very well to

\[ |ρ_{23}(t)| = \frac{1}{2} e^{-\Delta K(t)}, \]  

with \:A_k(Ω) = Ω_1, Ω_2=9.13, and Ω_{N−2}=26.73. Note that, in principle, a closed quantum system cannot exhibit exponential decay. The fact that we observe a decay that is well described by a single exponential may be the result of tracing out the degrees of freedom of an environment that initially is in a state of random superposition of the basis states.

Physically, the observed behavior can be understood as follows. If |Δ|≪√K\Ω, a bath spin is affected by roughly the same amount by the motion of both the other bath spins and by the two central spins. Therefore, each bath spin has enough freedom to follow the original dynamics, much as if there were no coupling between bath spins. This explains why the initial Gaussian decay is insensitive to the values of K or Ω. After the initial decay, the whole system is expected to reach a stationary state, but because of the presence of Heisenberg-type interactions between the bath spins, a new stationary state of the bath is established, which suppresses the long-living oscillations.

For increasing K, the distance between two bath spins, which is defined as the minimum number of bonds connecting the two spins, becomes smaller. For instance, for K=2, this distance is (N−2)/2, and for K=N−1, it is zero. Therefore, for fixed Ω and increasing K, the fluctuations in the
spin bath can propagate faster and the evolution to the stationary state will be faster. Similarly, for fixed $K$, increasing the coupling strength between the bath spins will speed up the dynamics of the bath, that is, the larger the $K$ is, the faster the evolution will be to the stationary state.

In the opposite case $K$, $H_{ce}$ is a small perturbation relative to $H_e$ and the coupling between bath spins is the dominant factor in determining the dynamics of the bath spins. Therefore, by increasing $K$ or $\Omega$, the bath spin will have less freedom to follow the dynamics induced by the coupling to the two central spins, the influence of the bath on the central system will decrease, and the exponential decay will become slower.

According to the general picture of decoherence, for an environment with nontrivial internal dynamics that initially is in a random superposition of all its eigenstates, we expect that the central system will evolve to a stable mixture of its eigenstates. In other words, the decoherence will cause all the off-diagonal elements of the reduced density matrix to vanish with time. In the case of an isotropic Heisenberg coupling between the central system and the environment, $H_c$ commutes with the Hamiltonian $H$; hence, the energy of the central system is a conserved quantity. Therefore, the weight of the singlet $|S\rangle$ in the mixed state should be a constant (1/2), and the weights of the degenerate eigenstates $|T_0\rangle$, $|T_{-1}\rangle$, and $|T_1\rangle$ are expected to become the same (1/6).

As shown in Figs. 1(b)–1(e) (right), our simulations confirm that this picture is correct in all respects.

**IV. ANISOTROPIC COUPLING TO THE BATH**

In order to clarify the role of $K$ and $\Omega$, we change the coupling between the central system and the bath from Heisenberg to Heisenberg-type. From a comparison of the data in Figs. 2 and 3, it is clear that the roles of $K$ and $\Omega$ are the same in both cases, no matter whether the coupling to the bath is isotropic or anisotropic. However, there are some differences in the decoherence process. The most important parameter determining the decoherence process is the ratio of the typical interaction energy $\Delta$ to the mean-square energy of interactions in the thermal bath, $\sqrt{K\Omega}$.

If $|\Delta| > \sqrt{K\Omega}$, in the presence of anisotropic interactions between the central system and the environment spins, even in the absence of interactions between the bath spins, the second step of the oscillations decays and finally disappears as $K$ increases. This is because the anisotropic interactions...
break the rotational symmetry of the coupling between central system and environment, which is required for the long-living oscillations to persist.

If $|\Delta| \approx \sqrt{K\Omega}$, $|\rho_{23}(t)|$ can still be described by Eq. (5), but now $A_K(\Omega)$ is no longer a linear function of $\Omega$. For anisotropic $H_{ce}$, the energy of the central system is no longer a conserved quantity. Therefore, there will be energy transfer between the central system and the environment and the weight of each pointer state (eigenstate) in the final stable mixture needs not to be the same for all $K$ or $\Omega$.

For a change, we illustrate this point by considering the quadratic entropy $S_N(t)$ and the Loschmidt echo $L(t)$. We expect that these quantities will also be dependent on the symmetry of the coupling between the central system and the spin bath. In Fig. 4, we present results for large $K$ and $\Omega = 2$, which confirm this expectation. For isotropic (Heisenberg) $H_{ce}$ and perfect decoherence (zero off-diagonal terms in the reduced density matrix), we expect that max$S_N(t) = 1 - [(1/2)^2 + (1/6)^2] = 2/3$ in concert with the data of Fig. 4(a). For Heisenberg type $H_{ce}$, max$S_N(t)$ will depend on the coupling strengths, and as shown in Fig. 4(c), we find that max$S_N(t) = 1 - 4(1/4)^2 = 3/4$, corresponding to the case that all the diagonal elements in the reduced density matrix are the same (1/6) and all other elements are zero.

V. DISCUSSION AND CONCLUSIONS

In the foregoing, we have compared $\sqrt{K\Omega}$ to $|\Delta|$ to distinguish different regimes. As a matter of fact, $\sqrt{K\Omega}$ does not completely characterize the decoherence process, but it can be used to characterize its time scale. Indeed, as shown in Fig. 5, for different $\sqrt{K}$ and $\Omega$ but the same value of $\sqrt{K}\Omega$, the time evolution of $L(t)$ is very similar. Note that if $\sqrt{K}\Omega$ increases [compare Fig. 5(a) to Fig. 5(d)], the differences between the Loschmidt echoes increase. Additional simulations (results not shown) indicate that these differences are fluctuations that are due to the particular realization random parameters used in the simulation.

In conclusion, for a spin-bath environment that initially is in a random superposition of its basis states, we have shown how a pure quantum state of the central spin system evolves into a mixed state, and that if the interaction between the central system and environment is much smaller than the coupling between the spins in the central system, the pointer states are the eigenstates of the central system. Both these observations are in concert with the general picture of decoherence. Furthermore, we have demonstrated that, in the case that the environment is a spin system, the details of this spin system are important for the decoherence of the central system. In particular, we have shown that for the anisotropic spin bath, changing the internal dynamics of the environment (geometric structure or exchange couplings) may change the decoherence of the central spin system from Gaussian to exponential decay.

Finally, we would like to compare the present results with those of our earlier work in which we focused on the case in which the environment is initially in its ground state and demonstrated that, apart from the strength of different inter-
FIG. 3. (Color online) Same as Fig. 2 except that $H_{ce}$ is Heisenberg type and $\Delta=0.15$.

FIG. 4. (Color online) The time evolution of the entropy $S(t)$ and the Loschmidt echo $L(t)$ of a central system (with $J=-5$), which interacts with a Heisenberg-type environment $H_e$ (with different $\Omega$) via a Heisenberg [(a) and (b)] $\Delta=-0.075$) or Heisenberg-type [(c) and (d)] $\Delta=0.15$) Hamiltonian $H_{ce}$ for the case $K=2$. The number next to each curve is the corresponding value of $\Omega$. 
actions, also their symmetry and the amount of entanglement of the ground state of the central system affect the decoherence.\textsuperscript{14,15} To facilitate the comparison, in Fig. 6, we present some data of the Loschmidt echoes for different $K$ but for fixed $\sqrt{K}\Omega$. Comparison of Fig. 5 with Fig. 6 indicates that if the environment is initially in its ground state, the decoherence process is qualitatively different from the one observed in the case that the initial state of the environment is a random superposition. Roughly speaking, it is more difficult for the central system to change from a pure quantum state to a classical, mixed state, which is of course consistent with the fact that the quantum effects become more prominent as the temperature decreases. In particular, from Fig. 6, it is clear that $\sqrt{K}\Omega$ is not enough to characterize the qualitative behavior of the Loschmidt echo for the cases shown.

FIG. 5. (Color online) The time evolution of the Loschmidt echo $L(t)$ of a central system (with $J=−5$), which interacts with a Heisenberg-type environment $H_e$ via a Heisenberg ($\Delta=−0.075$) Hamiltonian $H_{ce}$. In each panel, the values of $\sqrt{K}\Omega$ are the same: (a) $\sqrt{K}\Omega=0.1\sqrt{N−1}$, (b) $\sqrt{K}\Omega=0.15\sqrt{N−1}$, (c) $\sqrt{K}\Omega=0.25\sqrt{N−1}$, and (d) $\sqrt{K}\Omega=\sqrt{N−1}$. The different lines in each panel correspond to different $K$. Solid (black) line: $K=2$; dashed (red) line: $K=4$; dotted (green) line: $K=6$; and dash-dotted (blue) line: $K=N−1$.

FIG. 6. (Color online) The time evolution of the Loschmidt echo $L(t)$ of a central system (with $J=−5$), which interacts with a Heisenberg-type environment $H_e$ via a Heisenberg ($\Delta=−0.075$) Hamiltonian $H_{ce}$. The environment spins are initially prepared in the ground state. The different curves correspond to different $K$, but $\sqrt{K}\Omega=0.15\sqrt{N−1}$ is fixed. Solid (black) line: $K=2$; dashed (red) line: $K=4$; dotted (green) line: $K=6$; and dash-dotted (blue) line: $K=N−1$. 
The difference between the cases of an environment at a low-temperature\textsuperscript{14,15} and a high-temperature (chaotic) environment considered in the present paper is most important for the systems with very large connectivity. In the latter case, the ground state of the environment is a quantum spin glass that is a very effective source of decoherence.\textsuperscript{14,15} At the same time, for the case of infinite temperature of the bath considered in this paper, this case is not very special when compared to the case of short-range interactions within the environment (see Fig. 5). It would be of interest to see if, as the temperature decreases, the decoherence process changes as the environment goes into the spin-glass state (at $T \approx \sqrt{K}$), which is a problem that we will leave for future research.

\section*{ACKNOWLEDGMENT}

M.I.K. acknowledges support by the Stichting Fundamenteel Onderzoek der Materie (FOM).

\section*{APPENDIX}

Consider a generic quantum model described by the Hamiltonian $H=H_c+H_{ce}+H_e$, where $H_c$ and $H_e$ describe the central system and the bath ($[H_c,H_e]=0$), respectively, and $H_{ce}$ describes the coupling between them. If $[H_c,H_{ce}]=0$, then the time evolution operator of the whole system $e^{-iHt}$ can be represented as

$$
e^{-iHt} = e^{-iH_c t} e^{-i(H_{ce}+H_e)t}. \quad (A1)$$

Denote the eigenstates and corresponding eigenvalues of the central system by $|k\rangle$ and $\{E_k\}$, that is, $H_c|k\rangle = E_k|k\rangle$. The initial state $|\phi(t_0)\rangle$ of the central system can be represented as $|\phi(t_0)\rangle = \sum_k c_k|k\rangle$. For an isolated central system ($H_{ce}=0$), the time evolution of the density matrix of the central system is given by

$$\rho_c(t) = \sum_{k,l} e^{-i(E_k-E_l)t} a_k^* a_l |k\rangle \langle l|. \quad (A2)$$

If the central system is coupled to the bath ($|\phi(t_0)\rangle$), the initial state of the whole system can be represented as

$$|\Psi(t_0)\rangle = \sum_k a_k |k\rangle |\phi(t_0)\rangle, \quad (A3)$$

and the state at later time $t$ is

$$|\Psi(t)\rangle = e^{-iHt} |\Psi(t_0)\rangle = \sum_k e^{-iE_k t} a_k e^{-i(H_{ce}+H_e)t} |k\rangle |\phi(t_0)\rangle. \quad (A4)$$

As $[H_c,H_{ce}]=0$, we have $H_{ce}|k\rangle |\phi(t_0)\rangle = |k\rangle M_k |\phi(t_0)\rangle$, therefore,

$$e^{-i(H_{ce}+H_e)t} |k\rangle |\phi(t_0)\rangle = \sum_m \frac{(-i)^m M_k^m |k\rangle |\phi(t_0)\rangle}{m!} = \sum_m |k\rangle \langle k| e^{-iM_k t} \langle k| |\phi(t_0)\rangle = |k\rangle \langle k| e^{-iE_k t} |\phi(t_0)\rangle = |k\rangle |\phi_k(t)\rangle, \quad (A5)$$

where we introduced

$$|\phi_k(t)\rangle = e^{-iM_k t} |\phi(t_0)\rangle. \quad (A6)$$

Hence, the state at time $t$ becomes

$$|\Psi(t)\rangle = \sum_k a_k e^{-iE_k t} |k\rangle |\phi_k(t)\rangle. \quad (A7)$$

The density matrix $\rho(t)$ of the whole system is

$$\rho(t) = \langle \Psi(t)|\langle \Psi(t) | = \sum_{k,j} e^{-i(E_k-E_j)t} a_k^* a_j |k\rangle |\phi_k(t)\rangle \langle \phi_j(t) | = \sum_{k,j} e^{-i(E_k-E_j)t} a_k^* a_j |\phi_k(t)\rangle \langle \phi_j(t) |. \quad (A8)$$

and the reduced density matrix $\rho_c(t)$ of the central system is

$$\rho_c(t) = \text{Tr}_e \rho(t) = \sum_{k,l} e^{-i(E_k-E_l)t} a_k^* a_l |\phi_k(t)\rangle \langle \phi_l(t) |. \quad (A9)$$

The Loschmidt echo $L(t)$ of the central system can be calculated as

$$L(t) = \text{Tr} \rho_c(t) \rho_0(t) = \sum_{k,l} e^{-i(E_k-E_l)t} a_k^* a_l |\phi_k(t)\rangle \langle \phi_l(t) | = \sum_{k,l} e^{-i(E_k-E_l)t} a_k^* a_l |\phi_k(t)\rangle \langle \phi_l(t) |. \quad (A10)$$

It is clear that if $[H_c,H_{ce}]=0$, the decoherence process is determined by the initial state of the central system $|\phi(t_0)\rangle$ and the time evolution of the $|\phi(t)\rangle$. As shown in Eq. (A6), the $|\phi_k(t)\rangle$ are determined by the initial state of the bath $|\phi(t_0)\rangle$, the eigenstates $|k\rangle$ of the central system, and the Hamiltonian $H_{ce}$ and $H_c$. The eigenvalues $E_k$ have no effect on the decoherence process. Thus, multiplying $H_c$ by a constant does not change the $L(t)$ and the diagonal elements of the reduced density matrix $\rho_c(t)$. The time evolution of the absolute value of the off-diagonal elements

$$|\rho_{c,l}(t)| = |a_l^* a_k |\langle \phi_l(t) | \phi_k(t) \rangle | \quad (A11)$$

is independent of $H_c$.  

184301-7
Finally, we consider the case that not only $[H_c, H_{cc}] = 0$ but also $[H_{cc}, H_e] = 0$. Then, Eq. (A6) becomes

$$|\psi(t)\rangle = e^{-i(M_e t + H_c t)} \langle \psi(0) | + e^{-iM_e t} e^{-iH_c t} |\psi(0)\rangle,$$

therefore we have

$$\langle \phi(t) | \phi(t) \rangle = \langle \phi(0) | e^{iH_c t} e^{iM_e t} e^{-iM_e t} e^{-iH_c t} | \phi(0) \rangle$$

$$= \langle \phi(0) | e^{-i(M_e t - M_l t)} | \phi(0) \rangle,$$

which implies that $|\rho_e(t)_{kl}|$ and $L(t)$ are not dependent on $H_e$.