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METHODOLOGICAL NOTES

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Relaxation of interacting open quantum systems

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Abstract. **We consider the transition from the description of a closed quantum system consisting of an open quantum system and a reservoir to the description of the open quantum system alone by eliminating the reservoir degrees of freedom by averaging over them. An approach based on the Lindblad master equation for the density matrix is used. A general scheme is developed for deriving the Lindblad superoperator that emerges after averaging the von Neumann equation over the reservoir degrees of freedom. This scheme is illustrated with the cases of radiation of a two-level atom into free space and the dynamics of the transition of a two-level atom from a pure state to a mixed state due to interaction with a dephasing reservoir. Special attention is paid to the open system consisting of several subsystems each of which independently interacts with the reservoir. In the case of noninteracting subsystems, the density matrix is a tensor product of the subsystem density matrices, and the Lindblad superoperator of the system is a sum of Lindblad superoperators of those subsystems. The interaction between the subsystems results not only in the emergence of the corresponding term in the Hamiltonian of the combined system but also in the nonadditivity of the Lindblad superoperators. This is often overlooked in modern literature, possibly leading, as is shown in this methodological note, to serious errors; for example, the second law of thermodynamics could be violated.**

Keywords: open quantum systems, Lindblad master equation, second law of thermodynamics

1. Introduction

Problems related to dissipation phenomena in the microworld have been attracting great attention recently. Physical quantities are known to be represented in quantum mechanics by Hermitian operators, which assumes the absence of dissipative processes in closed systems. However, we are usually dealing with open systems in contact with other systems, in particular, with a reservoir. To study separate phenomena, it is not necessary to extend the system under study to include the environment in order to finally obtain a closed system. Often, we do not need all of the exact information about the extended system and can restrict ourselves to the description of only a particular part of it.

Similarly, to find the distribution function of particles in Brownian motion, it is not necessary to solve the equations of motion for each particle; it is sufficient to solve only the Fokker–Planck equation. In quantum mechanics, the role of the distribution function is played by the density matrix, and, instead of the Fokker–Planck equation, the Lindblad equation obtained by eliminating the environment variables is to be solved. In other words, we can consider only the part of the system of interest to us, treating the remaining part as a reservoir. In this situation, the system under study is open, and it is necessary to correctly describe its non-Hermitian dynamics caused by interaction with the reservoir.

The first attempts at such a description used the Weisskopf–Wigner approximation [1], kinetic equations [2],

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or Maxwell–Bloch equations [3]. However, solutions of such equations often had no physical meaning, for example, negative levels of population inversion were obtained [2]. A breakthrough occurred after the publication of papers [4, 5]. It was shown there that an operator responsible for the energy exchange with a reservoir should have the form of a Lindblad superoperator. This guaranteed both the conservation of the trace of the density matrix (i.e., the total probability) and its positive definiteness.

The exact derivation of operators describing the interaction of the system with the reservoir proves to be rather complicated. Because of this, the Lindblad equation is often derived [6–19] using various approximations and phenomenological considerations. Below, we present the standard derivation of the Lindblad equation using controllable approximations and consider possible errors in the phenomenological approach. In particular, it is often assumed that Lindblad superoperators for interacting subsystems are given by sums of Lindblad superoperators for noninteracting subsystems. We show in this paper that such an approach does not guarantee the fulfilment of thermodynamic laws, and the mean values of operators can differ from the correct values by an order of magnitude.

2. Derivation of the Lindblad equation

We consider the quantum dynamics of some system S. If system S is closed, its dynamics is described by the Schrödinger equation. In many cases, we have to deal with a system interacting with some environment, a reservoir R. In this case, the ab initio approach for calculating the system dynamics involves the solution of the Schrödinger equation for system S and reservoir R, followed by averaging over the reservoir variables. However, such an approach cannot be realized in practice because the number of degrees of freedom of the reservoir is typically very large. Hence, the question arises as to whether it is possible to construct a closed equation that would describe the dynamics of only system S. This proved to be possible using some assumptions about the system dynamics, namely, in the Born–Markov approximation [4, 5, 20]. We derive the corresponding equation in this section following Davies's papers [21, 22].

We consider the dynamics of system S with a Hamiltonian $\hat{H}_{\rm S}$ and assume that this system interacts with reservoir R having the Hamiltonian $\hat{H}_{\rm R}$. We describe the interaction between the reservoir and the system by the Hamiltonian $\hat{H}_{\rm SR}$. The system and reservoir form a closed system and obey the Schrödinger equation for the common wave function $|\psi\rangle$. Because we are interested in the dynamics of an open system, it is convenient to consider not the wave function but the density matrix $\hat{\rho}$, which is related to the wave function for a closed system as $\hat{\rho} = |\psi\rangle\langle\psi|$ [20, 23]. Differentiating this expression over time and using the Schrödinger equation, we obtain the von Neumann equation for the density matrix

$$\frac{{\rm d}\hat{\rho}(t)}{{\rm d}t} = \frac{\rm i}{\hbar}\left[\hat{\rho}(t), \hat{H}_{\rm S} + \hat{H}_{\rm R} + \hat{H}_{\rm SR}\right]. \tag{1}$$

We assume that the interaction Hamiltonian $\hat{H}_{\rm SR}$ can be represented in the form $\hat{H}_{\rm SR} = \hbar\lambda\hat{S}\hat{R}$, where $\hat{S}$ and $\hat{R}$ are operators respectively depending on the dynamic variables of only the system and only the reservoir. In what follows, all the operators are dimensionless, and Planck's constant in the expression for $\hat{H}_{\rm SR}$ is isolated to make the coupling constant λ have the dimension of frequency.

We note that the Hamiltonian in Eqn (1) contains the eigenfrequencies $\omega_{\rm S}$ and $\omega_{\rm R}$ of the system and the reservoir, and also the coupling constant λ determining different time scales. In many physically important cases, the weak-coupling regime occurs when $\omega_{\rm S} \gg \lambda$. In the weak-coupling approximation, it is possible to obtain a time-local equation for the density matrix of the system [20]. For this, it is necessary to pass to slow amplitudes by averaging over times $\sim 1/\omega_{\rm S}$.

To separate slow amplitudes explicitly, it is convenient to pass to the interaction representation for the density matrix $\hat{\hat{\rho}}(t)$ in the interaction Hamiltonian $\hat{\hat{H}}_{\rm SR}(t)$:

$$\hat{\rho}(t) = \exp\left(-\frac{{\rm i}(\hat{H}_{\rm S} + \hat{H}_{\rm R})t}{\hbar}\right)\hat{\hat{\rho}}(t)\exp\left(\frac{{\rm i}(\hat{H}_{\rm S} + \hat{H}_{\rm R})t}{\hbar}\right),$$

$$\hat{H}_{\rm SR} = \exp\left(-\frac{{\rm i}(\hat{H}_{\rm S} + \hat{H}_{\rm R})t}{\hbar}\right)\hat{\hat{H}}_{\rm SR}(t)\exp\left(\frac{{\rm i}(\hat{H}_{\rm S} + \hat{H}_{\rm R})t}{\hbar}\right).$$

In this case, $\hat{\hat{\rho}}$ represents the 'slow' density matrix. Equation (1) in the interaction representation has the form

$$\frac{{\rm d}\hat{\hat{\rho}}(t)}{{\rm d}t} = \frac{\rm i}{\hbar}\left[\hat{\hat{\rho}}, \hat{\hat{H}}_{\rm SR}(t)\right]. \tag{2}$$

We note that in the absence of interaction, i.e., when $\lambda = 0$, the density matrix $\hat{\hat{\rho}}(t)$ does not change with time, which suggests that the characteristic variation time of $\hat{\hat{\rho}}$ is of the order of $1/\lambda$.

Time averaging in Eqn (2) as

$$\overline{\hat{F}(t_0)} = \frac{1}{\Delta t}\int_{t_0}^{t_0+\Delta t} \hat{F}(t)\,{\rm d}t$$

with $\Delta t \gg 1/\omega_{\rm S}$ gives

$$\frac{1}{\Delta t}\int_{t_0}^{t_0+\Delta t}\frac{{\rm d}\hat{\hat{\rho}}(t)}{{\rm d}t}\,{\rm d}t = \frac{1}{\Delta t}\int_{t_0}^{t_0+\Delta t}\frac{\rm i}{\hbar}\left[\hat{\hat{\rho}}, \hat{\hat{H}}_{\rm SR}(t)\right]{\rm d}t. \tag{3}$$

Because the operators $\hat{S}$ and $\hat{R}$ commute, the Hamiltonians $\hat{H}_{\rm S}$ and $\hat{H}_{\rm R}$ also commute, and the interaction operator $\hat{H}_{\rm SR} = \hbar\lambda\hat{S}\hat{R}$ in the interaction representation can be rewritten in the form

$$\hat{\hat{H}}_{\rm SR}(t) = \hbar\lambda\hat{\hat{S}}(t)\hat{\hat{R}}(t), \tag{4}$$

where

$$\hat{\hat{S}}(t) = \exp\left(\frac{{\rm i}\hat{H}_{\rm S}t}{\hbar}\right)\hat{S}\exp\left(-\frac{{\rm i}\hat{H}_{\rm S}t}{\hbar}\right),$$

$$\hat{\hat{R}}(t) = \exp\left(\frac{{\rm i}\hat{H}_{\rm R}t}{\hbar}\right)\hat{R}\exp\left(-\frac{{\rm i}\hat{H}_{\rm R}t}{\hbar}\right).$$

Using the evolution operator [20, 23] for Eqn (2)

$$\hat{U}(t, t_0) = {\rm T}\exp\left\{-\frac{\rm i}{\hbar}\int_{t_0}^{t}\hat{\hat{H}}_{\rm SR}(t')\,{\rm d}t'\right\},$$

where T is the chronological ordering operator, we can rewrite Eqn (3) as

$$\frac{\hat{\hat{\rho}}(t_0 + \Delta t) - \hat{\hat{\rho}}(t_0)}{\Delta t} = \frac{1}{\Delta t}\left(\hat{U}(t_0 + \Delta t, t_0)\hat{\hat{\rho}}(t_0)\hat{U}^{\dagger}(t_0 + \Delta t, t_0) - \hat{\hat{\rho}}(t_0)\right). \tag{5}$$

The expansion of $\hat{U}(t, t_0)$ in powers of the coupling constant λ through the second order has the form

$$\hat{U}(t, t_0) \approx 1 - \mathrm{i}\lambda \int_{t_0}^{t} \hat{\hat{S}}(t_1)\hat{\hat{R}}(t_1)\,\mathrm{d}t_1$$
$$- \lambda^2 \int_{t_0}^{t} \mathrm{d}t_1 \int_{t_0}^{t_1} \left(\hat{\hat{S}}(t_1)\hat{\hat{R}}(t_1)\right)\left(\hat{\hat{S}}(t_2)\hat{\hat{R}}(t_2)\right)\mathrm{d}t_2\,. \qquad (6)$$

We note that the criterion for the applicability of the perturbation theory developed here is a decrease of subsequent corrections to the evolution operator, i.e., $U_0 = 1 \gg \lambda U_1 \gg \lambda^2 U_2 \gg \ldots$. A dimensionless parameter providing the smallness of each next correction is $\lambda\Delta t$, where $\Delta t = t - t_0$ is the time over which expansion (6) is performed. Thus, expansion (6) is valid if $\lambda\Delta t \ll 1$.

We now substitute expansion (6) in (5). Keeping the terms of the first, $\hat{\hat{\rho}}_1$, and second, $\hat{\hat{\rho}}_2$, orders of smallness in powers of $\lambda\Delta t$, we then have

$$\frac{1}{\Delta t}\left(\hat{U}(t_0 + \Delta t, t_0)\hat{\hat{\rho}}(t_0)\hat{U}^{\dagger}(t_0 + \Delta t, t_0) - \hat{\hat{\rho}}(t_0)\right)$$
$$\approx \frac{1}{\Delta t}\left(\lambda\,\hat{\hat{\rho}}_1(t_0 + \Delta t) + \lambda^2\hat{\hat{\rho}}_2(t_0 + \Delta t)\right). \qquad (7)$$

We assume further that the reservoir is in thermal equilibrium *at all moments of time* and its density matrix does not change with time: $\hat{\rho}_{\mathrm{R}}(0) = \hat{\rho}_{\mathrm{R}}^{\mathrm{th}} = \exp(-\hat{H}_{\mathrm{R}}/T)/\operatorname{tr}\exp(-\hat{H}_{\mathrm{R}}/T)$. This is the first key approximation used to derive the master equation. This approximation, in which the reservoir state is assumed invariable at all moments of time, is called the Born approximation [20]. Physically, it corresponds to a reservoir being so large that the system cannot change its state.

In this approximation, if the system and reservoir states are uncorrelated at the initial instant, they remain uncorrelated at all the subsequent instants, i.e., $\hat{\hat{\rho}}(t) = \hat{\rho}_{\mathrm{S}}(t)\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}$. Then the operators $\hat{\hat{\rho}}_1(t_0 + \Delta t)$ and $\hat{\hat{\rho}}_2(t_0 + \Delta t)$ from (7) can be written in the form

$$\hat{\hat{\rho}}_1(t_0 + \Delta t) = \mathrm{i}\int_{t_0}^{t_0+\Delta t} \mathrm{d}t_1\,\hat{\rho}_{\mathrm{S}}(t_0)\hat{\hat{S}}(t_1)\left(\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\hat{\hat{R}}(t_1)\right)$$
$$- \mathrm{i}\int_{t_0}^{t_0+\Delta t} \mathrm{d}t_1\,\hat{\hat{S}}(t_1)\hat{\rho}_{\mathrm{S}}(t_0)\left(\hat{\hat{R}}(t_1)\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\right), \qquad (8)$$

$$\hat{\hat{\rho}}_2(t_0 + \Delta t) = \int_{0}^{t_0+\Delta t} \mathrm{d}t_1\left(\hat{\hat{S}}(t_1)\hat{\hat{R}}(t_1)\right)\left(\hat{\rho}_{\mathrm{S}}(t_0)\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\right)$$
$$\times \int_{0}^{t_0+\Delta t} \mathrm{d}t_1\left(\hat{\hat{S}}(t_1)\hat{\hat{R}}(t_1)\right) - \int_{t_0}^{t_0+\Delta t}\int_{t_0}^{t_1}\left(\hat{\hat{S}}(t_1)\hat{\hat{R}}(t_1)\right)$$
$$\times \left(\hat{\hat{S}}(t_2)\hat{\hat{R}}(t_2)\right)\mathrm{d}t_2\,\mathrm{d}t_1\left(\hat{\rho}_{\mathrm{S}}(t_0)\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\right) - \left(\hat{\rho}_{\mathrm{S}}(t_0)\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\right)$$
$$\times \int_{t_0}^{t_0+\Delta t}\int_{t_0}^{t_1}\left(\hat{\hat{S}}(t_2)\hat{\hat{R}}(t_2)\right)\left(\hat{\hat{S}}(t_1)\hat{\hat{R}}(t_1)\right)\mathrm{d}t_2\,\mathrm{d}t_1\,. \qquad (9)$$

To obtain the density matrix of the system only, we must take the trace $\hat{\hat{\rho}}_{(\mathrm{S})1,2}(t) = \operatorname{tr}_{\mathrm{R}}\hat{\hat{\rho}}_{1,2}(t)$ over the reservoir variables in expressions (8) and (9). Averages like $\operatorname{tr}_{\mathrm{R}}\left(\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\hat{R}\right)$ then appear in the integrand in (8). For many physically important reservoir types, such as the electromagnetic field in free space and various dephasing reservoirs, the condition $\operatorname{tr}_{\mathrm{R}}\left(\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\hat{\hat{R}}(t_1)\right) = 0$ is satisfied. As follows from (8), if $\operatorname{tr}_{\mathrm{R}}(\hat{\rho}_{\mathrm{R}}^{\mathrm{th}}\hat{\hat{R}}(t_1)) = 0$, then $\operatorname{tr}_{\mathrm{R}}\hat{\hat{\rho}}_1(t_0 + \Delta t) = 0$, and only the second-order term in λ, namely, $\hat{\hat{\rho}}_2(t)$, remains in (7).

We calculate the time integrals in $\hat{\hat{\rho}}_2(t)$ [see (9)] using the equality $\exp(\mathrm{i}\hat{H}_{\mathrm{S}}t)|k\rangle = \exp(\mathrm{i}\omega_k t)|k\rangle$, which is satisfied for the eigenstates $|k\rangle$ of the Hamiltonian $\hat{H}_{\mathrm{S}}$. This allows us to represent the operator $\hat{\hat{S}}(t)$ in the form

$$\hat{\hat{S}}(t) = \exp\left(\frac{\mathrm{i}\hat{H}_{\mathrm{S}}t}{\hbar}\right)\hat{S}\exp\left(-\frac{\mathrm{i}\hat{H}_{\mathrm{S}}t}{\hbar}\right) = \sum_{k'k''}\exp(\mathrm{i}\Delta\omega_{k'k''}t)\hat{S}_{k'k''}\,, \qquad (10)$$

where $\hat{S}_{k'k''} = \langle k'|\hat{S}|k''\rangle|k'\rangle\langle k''|$, $\hat{S}_{k'k''} = \hat{S}^{\dagger}_{k''k'}$ and summation is performed over frequency differences $\Delta\omega_{k'k''} = \omega_{k'} - \omega_{k''}$.

We note that the operators $\hat{S}_{k'k''}$ can be obtained explicitly if the eigenstates of the system Hamiltonian $\hat{H}_{\mathrm{S}}$ are known. If the eigenstates are not known, the Davies procedure does not work in general. Other methods for calculating $\hat{\hat{S}}(t)$, followed by averaging, should then be used.

The time dependence in (10) is contained only in the exponential factors. This allows us to easily average over a short time $\sim 1/\omega_{\mathrm{S}}$. This is a key moment in the procedure to eliminate the reservoir variables, allowing us to obtain the explicit time dependence $\hat{\hat{S}}(t)$.

Indeed, after the substitution of (10) in (9), integrals like

$$\frac{\lambda^2}{\Delta t}\sum_{k'k''k'''k''''}\int_{t_0}^{t_0+\Delta t}\int_{t_0}^{t_0+\Delta t}\mathrm{d}t_1\,\mathrm{d}t_2\,\hat{S}_{k'k''}\hat{\hat{\rho}}_{\mathrm{S}}(t_0)\hat{S}_{k'''k''''}$$
$$\times \exp\left(\mathrm{i}(\Delta\omega_{k'k''}t_1 + \Delta\omega_{k'''k''''}t_2)\right)\operatorname*{tr}_{\mathrm{R}}\left(\hat{\hat{R}}(t_2)\hat{\hat{R}}(t_1)\hat{\rho}_{\mathrm{R}}\right)$$
$$= \sum_{k'k''k'''k''''}\hat{S}_{k'k''}\hat{\hat{\rho}}_{\mathrm{S}}(t_0)\hat{S}_{k'''k''''}$$
$$\times \frac{1}{\Delta t}\int_{t_0}^{t_0+\Delta t}\mathrm{d}t_1\exp\left(\mathrm{i}(\Delta\omega_{k'k''} - \Delta\omega_{k'''k''''})t_1\right)$$
$$\times \int_{t_0-t_1}^{t_0+\Delta t-t_1}\mathrm{d}\tau\,\lambda^2\exp(\mathrm{i}\Delta\omega_{k'''k''''}\tau)\,F(\tau) \qquad (11)$$

remain, where $F(t_1 - t_2) = \operatorname{tr}_{\mathrm{R}}\left(\hat{\hat{R}}(t_2)\hat{\hat{R}}(t_1)\hat{\rho}_{\mathrm{R}}\right)$. To calculate time-averaged quantities, we recall that the averaging interval greatly exceeds all the characteristic times of the system. In other words, the integration limits in integrals in (11) can be set equal to infinity, which means considering the limit $\Delta t \gg 1/\omega_{\mathrm{S}}$. As a result, we obtain

$$\int_{t_0-t_1}^{t_0+\Delta t-t_1}\mathrm{d}\tau\,\lambda^2\exp(\mathrm{i}\Delta\omega_{k'''k''''}\tau)F(\tau)$$
$$\approx \int_{-\infty}^{\infty}\lambda^2\exp(\mathrm{i}\Delta\omega_{k'''k''''}\tau)F(\tau)\,\mathrm{d}\tau \equiv \lambda^2 G(\Delta\omega_{k'''k''''})\,, \quad (12)$$

$$\frac{1}{\Delta t}\int_{t_0}^{t_0+\Delta t}\mathrm{d}t_1\exp\left(\mathrm{i}(\Delta\omega_{k'k''} - \Delta\omega_{k'''k''''})t_1\right)$$
$$\approx \frac{\Delta t}{\Delta t}\,\delta_{\Delta\omega_{k'k''},\Delta\omega_{k'''k''''}} = \delta_{\Delta\omega_{k'k''},\Delta\omega_{k'''k''''}}\,, \qquad (13)$$

where $\delta_{\Delta\omega_{k'k''},\Delta\omega_{k'''k''''}}$ is the Kronecker delta.

An approximation like (13) leads to an important result: in sum (11), only terms with $\Delta\omega_{k'k''} = \Delta\omega_{k'''k''''}$ remain. To finally obtain the Lindblad equation, we use (12) and (13) to substitute (11) in (9) and then in (7). This gives

$$\frac{\hat{\hat{\rho}}(t_0+\Delta t)-\hat{\hat{\rho}}(t_0)}{\Delta t} = \lambda^2 \sum_{\Delta\omega} G(\Delta\omega)\Big(\Big[\hat{S}_{\Delta\omega}, \hat{\hat{\rho}}_{\mathrm{S}}(t_0)\hat{S}^{\dagger}_{\Delta\omega}\Big] + \Big[\hat{S}_{\Delta\omega}\hat{\hat{\rho}}_{\mathrm{S}}(t_0), \hat{S}^{\dagger}_{\Delta\omega}\Big]\Big), \tag{14}$$

where summation is performed over all possible frequency differences $\Delta\omega = \omega_{k'} - \omega_{k''}$. The difference equation is usually replaced with a differential equation [24] because the right-hand side is small, $\lambda^2 G(\Delta\omega)\Delta t \ll 1$. As a result, we obtain the Lindblad equation for the density matrix $\hat{\hat{\rho}}_{\mathrm{S}}$ of the system

$$\frac{\partial}{\partial t}\hat{\hat{\rho}}_{\mathrm{S}}(t) = \lambda^2 \sum_{\Delta\omega} G(\Delta\omega)\Big(\Big[\hat{S}_{\Delta\omega}, \hat{\hat{\rho}}_{\mathrm{S}}(t)\hat{S}^{\dagger}_{\Delta\omega}\Big] + \Big[\hat{S}_{\Delta\omega}\hat{\hat{\rho}}_{\mathrm{S}}(t), \hat{S}^{\dagger}_{\Delta\omega}\Big]\Big) \equiv \hat{L}\big[\hat{\hat{\rho}}_{\mathrm{S}}(t)\big]\,. \tag{15}$$

The right-hand side of (15) defines the Lindblad superoperator $\hat{L}\big[\hat{\hat{\rho}}_{\mathrm{S}}(t)\big]$. We can show that in the case of a reservoir in thermal equilibrium, the function $G(\Delta\omega)$ satisfies the Kubo–Martin–Schwinger relation [25, 26] $G(\Delta\omega)/G(-\Delta\omega) = \exp(\hbar\Delta\omega/T)$, where T is the reservoir temperature.

We recall that the averaging procedure in (12) and (13) requires that the averaging time Δt greatly exceed the characteristic inverse frequency of the system, $\omega_{\mathrm{S}}\Delta t \gg 1$. However, the averaging time Δt should not be too large, such that expansion (6) for the evolution operator hold. We can see from (14) and (15) that this requires the condition $\lambda^2 G(\Delta\omega)\Delta t \ll 1$. Thus, the criterion for the applicability of the Davies procedure for eliminating the reservoir variables is the condition $G(\Delta\omega)\lambda^2 \ll \omega_{\mathrm{S}}$.

Passing from the interaction representation to the Schrödinger representation, i.e., performing the inverse transformation $\hat{\rho}_{\mathrm{S}}(t) = \exp(-\mathrm{i}\hat{H}_{\mathrm{S}}t/\hbar)\hat{\hat{\rho}}_{\mathrm{S}}(t)\exp(\mathrm{i}\hat{H}_{\mathrm{S}}t/\hbar)$, we obtain

$$\frac{\partial}{\partial t}\hat{\rho}_{\mathrm{S}}(t) = -\frac{\mathrm{i}}{\hbar}\big[\hat{H}_{\mathrm{S}}, \hat{\rho}_{\mathrm{S}}\big] + \hat{L}\big[\hat{\rho}_{\mathrm{S}}(t)\big]\,. \tag{16}$$

3. Compatibility of the Lindblad equation with basic thermodynamic laws

It was shown previously (see, e.g., [27, 28]) that the structure of Lindblad equation (16) guarantees that the first and second, and under some conditions, the zeroth and third laws of thermodynamics are satisfied.

According to the zeroth law of thermodynamics, a system settles down to thermal equilibrium with a reservoir, i.e., their temperatures become equal, and the stationary state of the system is described by the Gibbs distribution $\hat{\rho}^{\mathrm{st}}_{\mathrm{S}} = \exp(-\hat{H}_{\mathrm{S}}/T)/\mathrm{tr}\exp(-\hat{H}_{\mathrm{S}}/T)$. For that state to be unique, there must be no operators $\hat{I}(\hat{S})$ that commute simultaneously with the Hamiltonian $\hat{H}_{\mathrm{S}}$ and with $\hat{L}\big[\hat{\rho}_{\mathrm{S}}(t)\big]$ [27, 29]. If such operators exists, their mean value determined by the initial state does not change with time and is an integral of motion.

The existence of an operator $\hat{I}(\hat{S})$ leads to the division of the total space of states of the system into subspaces consisting of vectors with the same eigenvalues of the integral of motion of $\hat{I}(\hat{S})$. Such subspaces are invariant in the sense that if the system is initially in this subspacee, it does not leave it in the course of evolution. Otherwise, the mean value of the integral of motion would change, but this mean is equal to one of the eigenvalues by definition. Depending on the initial conditions, the system finds itself with some probability in one such subspace. The interaction with the reservoir cannot take the system from one subspace to another. Each of these subspaces has its own set of eigenvalues of the system. In each subspace, the Gibbs distribution is established during evolution, with its own partition function for states of that subspace [29]. The final state is the sum of the obtained Gibbs states taken with the initial probabilities.

The first law of thermodynamics is the energy conservation law. In the case of an open system, the thermal flux J between the system and the reservoir is given by

$$J = \frac{\delta Q}{\delta t} \equiv \frac{E(t+\delta t)-E(t)}{\delta t} = \frac{1}{\delta t}\int_t^{t+\delta t}\frac{\mathrm{d}}{\mathrm{d}\tau}\,\mathrm{tr}_{\mathrm{S}}\big(\hat{H}_{\mathrm{S}}\hat{\rho}_{\mathrm{S}}(\tau)\big)\,\mathrm{d}\tau = \frac{1}{\delta t}\,\mathrm{tr}_{\mathrm{S}}\left(\hat{H}_{\mathrm{S}}\int_t^{t+\delta t}\frac{\mathrm{d}}{\mathrm{d}\tau}\,\hat{\rho}_{\mathrm{S}}(\tau)\,\mathrm{d}\tau\right). \tag{17}$$

Substituting the expression for $\mathrm{d}\hat{\rho}_{\mathrm{S}}(\tau)/\mathrm{d}\tau$ from Lindblad equation (16), we obtain

$$J = \mathrm{tr}_{\mathrm{S}}\left(\hat{H}_{\mathrm{S}}\hat{L}\big[\hat{\rho}_{\mathrm{S}}(t)\big]\right) \equiv \frac{\mathrm{d}\langle\hat{H}_{\mathrm{S}}\rangle}{\mathrm{d}t}\,, \tag{18}$$

which means that the heat flux is equal to the change in the system energy.

The second law of thermodynamics can be written in the form [20, 29]

$$\frac{\mathrm{d}S}{\mathrm{d}t} - \frac{J}{T} \geqslant 0\,, \tag{19}$$

where $S = -\mathrm{tr}\big(\hat{\rho}_{\mathrm{S}}(t)\ln\hat{\rho}_{\mathrm{S}}(t)\big)$ is the von Neumann entropy and $J = \delta Q/\delta t$ is the energy flux between the system and the reservoir from (18). It can be rigorously proved that condition (19) for S and J introduced in this way is satisfied for finite-dimensional systems [27]. Thus, the second law of thermodynamics is a consequence of Lindblad equation (16).

The third law of thermodynamics states that the system entropy tends to zero if the reservoir temperature tends to zero. It can be shown that if the system does not have an integral of motion and the reservoir has a ground state, then the third law follows from (16) (see the details in [28]).

Thus, the form of Eqn (16) implies the first, second, and, under some conditions, the zeroth and third laws of thermodynamics. If we change (16), for example, pass to the tau-approximation or use a dissipative operator different from the Lindblad superoperator, the laws of thermodynamics can be violated. In other words, although the Lindblad equation approach is approximate, it correctly describes the thermodynamics of the system and provides the most reliable estimates.

4. Examples of Lindblad equation applications

4.1 Spontaneous emission of an atom in free space

In this case, a continuum of the electromagnetic modes in free space plays the role of a reservoir. We note that level populations can be obtained from the Weisskopf–Wigner theory [1]. However, this theory uses pure states of an atom with zero entropy. In other words, the Clausius inequality can be verified only using the Lindblad equation for the density matrix.

We consider an atom as a two-level system (TLS). The TLS Hamiltonian with a ground state $|g\rangle$ with zero energy and an excited state $|e\rangle$ with the energy $\hbar\omega_0$ can be written as

$$\hat{H}_{\rm S} = \hbar\omega_0 \hat{\sigma}^{\dagger}\hat{\sigma}\,, \tag{20}$$

where $\hat{\sigma}$ is a decreasing operator, $\hat{\sigma}|e\rangle = |g\rangle$, and $\hat{\sigma}^{\dagger}$ is an increasing operator, $\hat{\sigma}^{\dagger}|g\rangle = |e\rangle$. A quantized electromagnetic field is described by the Hamiltonian

$$\hat{H}_{\rm R} = \sum_{\alpha,\,\mathbf{k}} \hbar\omega_{\mathbf{k}} \hat{a}^{+}_{\alpha,\,\mathbf{k}} \hat{a}_{\alpha,\,\mathbf{k}}\,, \tag{21}$$

where $\mathbf{k}$ is the wave vector of a mode and α is its polarization. The dispersion of the electromagnetic field in free space determines the dependence of the field frequency on its wave vector, $\omega_{\mathbf{k}} = c|\mathbf{k}|$. The atom–free-space interaction Hamiltonian in the dipole approximation has the form

$$\begin{aligned} \hat{H}_{\rm SR} &= -\hat{\mathbf{d}}\hat{\mathbf{E}} \\ &= -\sum_{\alpha,\,\mathbf{k}} \sqrt{\frac{2\pi\hbar\omega_{\mathbf{k}}}{V}}\, \mathbf{e}_{\alpha,\,\mathbf{k}} \mathbf{d}_{eg} (\hat{\sigma} + \hat{\sigma}^{\dagger})(\hat{a}^{\dagger}_{\alpha,\,\mathbf{k}} + \hat{a}_{\alpha,\,\mathbf{k}}) = \hbar\lambda\hat{S}\hat{R}\,, \end{aligned} \tag{22}$$

where $\hat{S} = (\hat{\sigma} + \hat{\sigma}^{\dagger})$, $\hat{R} = \sum_{\lambda,\,\mathbf{k}} \sqrt{\omega_{\mathbf{k}}/\omega_0}\, \mathbf{e}_{\lambda,\,\mathbf{k}} \mathbf{n}_{eg} (\hat{a}^{\dagger}_{\lambda,\,\mathbf{k}} + \hat{a}_{\lambda,\,\mathbf{k}})$, and $\lambda = -\sqrt{2\pi\omega_0/\hbar V}\,|\mathbf{d}_{eg}|$. The TLS–electromagnetic-field coupling constant is determined by the transition dipole moment $\mathbf{d}_{eg}$, the unit vector $\mathbf{e}_{\lambda,\,\mathbf{k}}$ in the direction of polarization of the electromagnetic field, and the space volume V. Expansion (10) of the interaction operator $\hat{S} = \hat{\sigma} + \hat{\sigma}^{\dagger}$ in this case takes the form

$$\begin{aligned} \hat{\hat{S}}(t) &= \exp\,({\rm i}\omega_0 t)|e\rangle\langle g| + \exp\,(-{\rm i}\omega_0 t)|g\rangle\langle e| \\ &= \exp\,({\rm i}\omega_0 t)\hat{\sigma}^{\dagger} + \exp\,(-{\rm i}\omega_0 t)\hat{\sigma}\,. \end{aligned} \tag{23}$$

Following the Davies procedure described above, we obtain the Lindblad equation [20]

$$\begin{aligned} \frac{\partial}{\partial t}\,\hat{\rho}_{\rm S}(t) &= -{\rm i}\big[\hat{H}_{\rm S}, \hat{\rho}_{\rm S}\big] + \gamma_{\rm diss}\big(1 + N(\omega_0)\big) \\ &\quad\times \left(\hat{\sigma}\hat{\rho}_{\rm S}(t)\hat{\sigma}^{\dagger} - \frac{1}{2}\,\hat{\sigma}^{\dagger}\hat{\sigma}\hat{\rho}_{\rm S}(t) - \frac{1}{2}\,\hat{\rho}_{\rm S}(t)\hat{\sigma}^{\dagger}\hat{\sigma}\right) \\ &\quad + \gamma_{\rm diss}N(\omega_0)\left(\hat{\sigma}^{\dagger}\hat{\rho}_{\rm S}(t)\hat{\sigma} - \frac{1}{2}\hat{\sigma}\hat{\sigma}^{\dagger}\hat{\rho}_{\rm S}(t) - \frac{1}{2}\,\hat{\rho}_{\rm S}(t)\hat{\sigma}\hat{\sigma}^{\dagger}\right), \end{aligned} \tag{24}$$

where $\gamma_{\rm diss} = 4\omega_0^3|\mathbf{d}_{eg}|^2/3\hbar c^3$ and $N(\omega) = (\exp\,(\hbar\omega/kT) - 1)^{-1}$.

This equation looks especially simple in the eigenstate basis of Hamiltonian (20). Decomposing in this basis, $\hat{\rho} = \rho_{ee}|e\rangle\langle e| + \rho_{eg}|e\rangle\langle g| + \rho_{ge}|g\rangle\langle e| + \rho_{gg}|g\rangle\langle g|$, and substituting this decomposition in (24), we obtain diagonal elements of the density matrix

$$\begin{aligned} \dot{\rho}_{ee} &= -\gamma_{\rm diss}\big(N(\omega_0) + 1\big)\rho_{ee} + \gamma_{\rm diss}N(\omega_0)\rho_{gg}\,, \\ \dot{\rho}_{gg} &= -\gamma_{\rm diss}N(\omega_0)\rho_{gg} + \gamma_{\rm diss}\big(N(\omega_0) + 1\big)\rho_{ee}\,. \end{aligned} \tag{25}$$

The quantities $\gamma_{\rm diss}$ and $\gamma_{\rm diss}N(\omega_0)$ are respectively similar to the Einstein coefficients for spontaneous and induced transitions. We can see that relaxation is determined by both spontaneous and induced processes, whereas pumping is determined only by induced processes. Thus, the term in (24) proportional to $G(\omega) = \gamma_{\rm diss}(N(\omega) + 1)$ describes the energy relaxation into the reservoir due to spontaneous and induced processes, while $G(\omega) = \gamma_{\rm diss}N(|\omega|)$ describes the pumping of the system by the reservoir only due to induced processes.

For diagonal elements, we in fact obtained kinetic equations preserving the total probability $\rho_{ee} + \rho_{gg} = {\rm const}$; the density matrix trace is conserved at all time instants. The nondiagonal elements evolve as

$$\begin{aligned} \dot{\rho}_{eg} &= -\frac{\gamma_{\rm diss}\big(2N(\omega_0) + 1\big)\rho_{eg}}{2}\,, \\ \dot{\rho}_{ge} &= -\frac{\gamma_{\rm diss}\big(2N(\omega_0) + 1\big)\rho_{ge}}{2}\,, \end{aligned} \tag{26}$$

i.e., exponentially decay. Direct calculation shows that the positive definiteness of the density matrix $\rho_{ee}\rho_{gg} - |\rho_{eg}|^2 \geqslant 0$ holds for any instant of time and for any positive definite initial density matrix.

We note that the energy relaxation rate (relaxation of diagonal elements) is sometimes called the longitudinal relaxation rate, and the dipole moment decay rate (decay of nondiagonal elements) is called the transverse relaxation rate. The longitudinal relaxation rate for the reservoir under study is $\gamma_{\rm diss}$, and the transverse relaxation rate is $\gamma_{\rm diss}\big(2N(\omega_0) + 1\big)/2$.

We illustrate the fulfillment of thermodynamic laws. The system does not have an integral of motion, i.e., an operator commuting simultaneously with the Hamiltonian $\hat{H}$ and the system–reservoir interaction operator $\hat{S}$. Therefore, the zeroth law of thermodynamics must hold. Indeed, the stationary solution of system (25), (26) is the Gibbs distribution

$$\begin{aligned} &\rho_{ee} = \frac{N(\omega_0)}{2N(\omega_0) + 1}\,, \qquad \rho_{gg} = \frac{N(\omega_0) + 1}{2N(\omega_0) + 1}\,, \\ &\frac{\rho_{ee}}{\rho_{gg}} = \frac{N(\omega_0)}{N(\omega_0) + 1} = \exp\left(-\frac{\hbar\omega_0}{kT}\right), \end{aligned} \tag{27}$$

i.e., the zeroth law of thermodynamics holds.

Figure 1 shows the time dependences of the entropy derivative ${\rm d}S/{\rm d}t$ and the difference between the entropy derivative and the thermal flux ${\rm d}S/{\rm d}t - J/T$. We can see that the difference ${\rm d}S/{\rm d}t - J/T$ is always greater than zero, i.e., the second law of thermodynamics holds. We note that the quantity ${\rm d}S/{\rm d}t$ itself can change sign. For example, if the

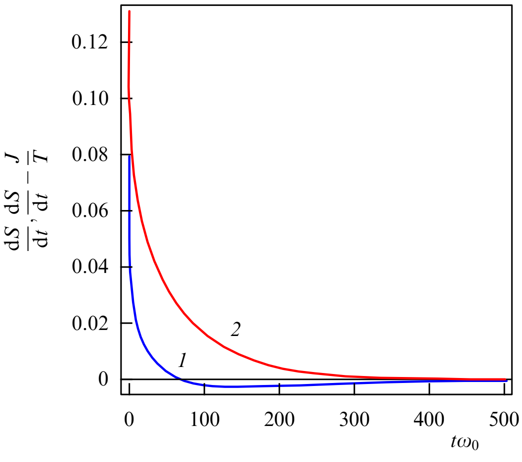

Figure 1. Time dependences of the entropy derivative ${\rm d}S/{\rm d}t$ (blue curve *1*) and the difference ${\rm d}S/{\rm d}t - J/T$ (red curve *2*). $kT/\hbar\omega_0 = 0.2$, $\gamma_{\rm diss}/\omega_0 = 0.01$.

system relaxes from the pure state $|e\rangle\langle e|$, then $\mathrm{d}S/\mathrm{d}t > 0$ during the transition to the mixed state $(|e\rangle\langle e| + |g\rangle\langle g|)/2$ with the maximum entropy. Then $\mathrm{d}S/\mathrm{d}t$ becomes negative, because the system undergoes a transition from the $(|e\rangle\langle e| + |g\rangle\langle g|)/2$ state with the maximum entropy to equilibrium state (27).

In the zero-temperature limit, $N(\omega_0) \to 0$, $\rho_{ee} \to 0$ and $\rho_{gg} \to 1$, i.e., the system undergoes a transition to the $|g\rangle\langle g|$ ground state. The entropy of the pure $|g\rangle\langle g|$ state is zero. Thus, the third law of thermodynamics also holds.

We note that Lindblad equation (24) for optical transitions at room temperature can be simplified. Because the photon energy greatly exceeds room temperature, the reservoir temperature can be assumed to be zero. Therefore, $N(\omega_0) = 0$, and only spontaneous processes responsible for relaxation occur. Lindblad equation (24) is simplified to

$$\frac{\partial}{\partial t}\,\hat{\rho}_{\mathrm{S}}(t) = -\mathrm{i}\big[\hat{H}_{\mathrm{S}}, \hat{\rho}_{\mathrm{S}}\big] + \gamma_{\mathrm{diss}}\left(\hat{\sigma}\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}^{\dagger} - \frac{1}{2}\,\hat{\sigma}^{\dagger}\hat{\sigma}\hat{\rho}_{\mathrm{S}}(t) - \frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}^{\dagger}\hat{\sigma}\right). \tag{28}$$

Thus, even in the absence of exciting photons in the reservoir, the relaxation of the excited system (in our case, a two-level atom) nevertheless occurs. This is related to the existence of zero-point oscillations of the reservoir energy, even at zero temperature. We also note that the ratio of the transverse and longitudinal relaxation rates in the optical range for the reservoir under study is 1:2.

4.2 Relaxation of a two-level atom into a dephasing reservoir

In Section 4.1, we considered the system–reservoir interaction Hamiltonian (22), when energy relaxation in the system occurs. However, in some systems, for example, colloidal quantum dots, dye molecules, and superconducting qubits, processes can occur that induce only phase changes in the system without changing its energy. Such processes are called dephasing or decoherence. Two examples are the collisional broadening of emission lines in gases and the elastic scattering of phonons in a solid [30]. The dephasing rate can exceed the energy dissipation rate by several orders of magnitude [31–34]. Below, we present the simplest example of a reservoir producing dephasing of the system without changing its energy.

We consider a reservoir consisting of harmonic oscillators with the Hamiltonian

$$\hat{H}_{\mathrm{R}} = \hbar\sum_k \omega_k \hat{a}_k^{\dagger}\hat{a}_k\,. \tag{29}$$

We assume that the TLS Hamiltonian has form (20) and the interaction of TLSs with this reservoir is described by the Hamiltonian

$$\hat{H}_{\mathrm{SR}} = \hbar\sum_k \gamma_k\hat{\sigma}_z(\hat{a}_k^{\dagger} + \hat{a}_k) = \hbar\lambda\hat{S}\hat{R}\,, \tag{30}$$

where $\hat{\sigma}_z = |e\rangle\langle e| - |g\rangle\langle g|$, $\hat{S} = \hat{\sigma}_z$, and $\hat{R} = \sum_k(\gamma_k/\lambda)(\hat{a}_k^{\dagger} + \hat{a}_k)$ [30]. Hamiltonian (30) describes the process of absorption and emission of an exciting photon from the reservoir, in which the system energy does not change. In other words, it describes elastic scattering of photons from the reservoir by the system. In this case, expression (10) takes the form

$$\hat{\hat{S}}(t) = \exp(\mathrm{i}\hat{H}_{\mathrm{S}}t)\hat{S}\exp(-\mathrm{i}\hat{H}_{\mathrm{S}}t) = |e\rangle\langle e| - |g\rangle\langle g|\,. \tag{31}$$

We now substitute (31) in (9) and calculate integrals by using the second key approximation, equalities (12) and (13). After direct calculations, we obtain a Lindblad equation like (16),

$$\frac{\partial\hat{\rho}_{\mathrm{S}}(t)}{\partial t} = -\mathrm{i}\big[\hat{H}_{\mathrm{S}}, \hat{\rho}_{\mathrm{S}}\big] + \gamma_{\mathrm{deph}}\left(\hat{\sigma}_z\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}_z - \frac{1}{2}\,\hat{\sigma}_z\hat{\sigma}_z\hat{\rho}_{\mathrm{S}}(t) - \frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}_z\hat{\sigma}_z\right) = -\mathrm{i}\big[\hat{H}_{\mathrm{S}}, \hat{\rho}_{\mathrm{S}}\big] + \gamma_{\mathrm{deph}}\big(\hat{\sigma}_z\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}_z - \hat{\rho}_{\mathrm{S}}(t)\big)\,, \tag{32}$$

where $\gamma_{\mathrm{deph}} = \lim_{\omega\to 0} G(\omega) = G(0)$. We note that the relaxation constant γ_{deph} (dephasing rate) in this example is proportional to the number of photons in the reservoir at the zero frequency. This is explained by the fact that the system–reservoir interaction operator $\hat{H}_{\mathrm{SR}} = \hbar\sum_k \gamma_k\hat{\sigma}_z(\hat{a}_k^{\dagger} + \hat{a}_k)$ describes processes without changing the system energy, i.e., $\big[\hat{H}_{\mathrm{SR}}, \hat{H}_{\mathrm{S}}\big] = 0$. Therefore, the mean energy of the system does not change. Mathematically, this means that the system–reservoir interaction operator $\hat{S} = \hat{\sigma}_z$ is diagonal in the basis of the eigenstates of $\hat{H}_{\mathrm{S}}$ [see relation (31)]. To calculate the limit $\omega \to 0$ in the equality $\gamma_{\mathrm{deph}} = \lim_{\omega\to 0} G(\omega)$, the low-frequency properties of the reservoir must be used (see specific examples in [30, 35]). We note that in most optical systems, the dephasing rate greatly exceeds the energy relaxation rate (in real systems, their ratio can reach five orders of magnitude).

We note important properties of Eqn (32). Writing (32) in the eigenstate basis of $\hat{H}_{\mathrm{S}}$, we can easily obtain by direct calculation that

$$\frac{\mathrm{d}}{\mathrm{d}t}\,\rho_{ee} = \frac{\mathrm{d}}{\mathrm{d}t}\,\rho_{gg} = 0\,, \tag{33}$$

i.e., the diagonal elements of the density matrix are not related. This means that the system energy remains constant:

$$\langle\hat{\sigma}_z\rangle = \rho_{ee} - \rho_{gg} = \mathrm{const} = \big\langle\hat{\sigma}_z(0)\big\rangle\,. \tag{34}$$

For the nondiagonal elements of the density matrix, we have

$$\frac{\mathrm{d}}{\mathrm{d}t}\,\rho_{eg} = -\gamma_{\mathrm{deph}}\rho_{eg}\,. \tag{35}$$

This means that the TLS dipole moment $\hat{\mathbf{d}} = \mathbf{d}_{eg}(\hat{\sigma}^{\dagger} + \hat{\sigma}) \sim \mathrm{Re}\,\langle\hat{\sigma}\rangle$ decays exponentially:

$$\langle\hat{\sigma}\rangle = \rho_{eg} = \big\langle\hat{\sigma}(0)\big\rangle\exp(-\gamma_{\mathrm{deph}}t)\,. \tag{36}$$

The stationary density matrix takes the form

$$\hat{\rho}_{\mathrm{S}}^{\mathrm{st}} = \begin{pmatrix} \rho_{ee}(0) & 0 \\ 0 & \rho_{gg}(0) \end{pmatrix}. \tag{37}$$

Thus, the averaged dipole moment of the TLS tends to zero under the action of the dephasing reservoir [see (36)]; however, the mean square of the dipole moment and energy (34) do not change. That is why such a process is called dephasing.

We note that the dephasing reservoir considered here provides only the transverse relaxation, i.e., the relaxation of the dipole moment, but does not describe the longitudinal relaxation (energy relaxation). In real systems, both types of relaxation always exist, and therefore the consideration of only one dephasing reservoir is always an approximation. As shown above, interaction with the reservoir resulting in energy dissipation in the optical range gives the ratio of the longitudinal and transverse relaxation times equal to two. Because this ratio in a real system can reach 10^5, both the reservoir describing the energy relaxation and the dephasing reservoir must be used in the two-level approximation.

We discuss the fulfillment of thermodynamic laws. The system has a single integral of motion, the system Hamiltonian $\hat{H}_{\text{S}}$ itself. As noted above, in this case the system has invariant subspaces. The value $\langle \hat{H}_{\text{S}} \rangle$ of the integral of motion is the same in each subspace. In the situation considered here, there are two one-dimensional subspaces spanned by the eigenvectors $|e\rangle$ and $|g\rangle$. In this case, it is meaningless to speak about the energy distribution in one subspace. The stationary state of the system depends only on the initial state and is independent of the reservoir temperature [see (37)], i.e., the zeroth law of thermodynamics does not hold.

The first law does hold; however, it is interesting to point out that the heat flux to the reservoir is $J = 0$ in this example, and the second law of thermodynamics reduces to the inequality $\mathrm{d}S/\mathrm{d}t > 0$. Using expressions (33) and (35), the inequality $\mathrm{d}S/\mathrm{d}t > 0$ can be proved by direct calculation. Thus, this reservoir does not change the system energy and causes only an increase in its entropy.

5. Relaxation of composite systems. General consideration

So far, we have considered a system consisting of a single two-level atom. However, in reality, we have to deal with more complicated systems consisting of several subsystems, for example, systems of interacting atoms, logic qubit chains, or entangled photons. We call such systems composite systems. The Davies procedure for describing dissipation presented above is general. However, its application to composite systems involves considerable difficulties. This is explained by the fact that while the eigenfrequencies and eigenstates of the system Hamiltonian $\hat{H}_{\text{S}}$, used in decomposing the operator $\hat{S}$, were known in the examples considered in the preceding sections, the eigenstates are typically not known in the general case of the composite system Hamiltonian

$$\hat{H}_{\text{S}} = \hat{H}_{\text{S}}^{(1)} + \hat{H}_{\text{S}}^{(2)} + \hat{V}_{12} \,, \tag{38}$$

where $\hat{V}_{12}$ is the interaction between the first and second systems. To avoid this difficulty, the decomposition is often performed with respect to the eigenstates of noninteracting subsystems. If the subsystems do not interact, i.e., $\hat{V}_{12} = 0$, this approach is valid.

Indeed, from the standpoint of a rigorous theory, it is necessary to eliminate the reservoir variables for the total system. Because the subsystems do not interact, the density matrix of the total system is factored, $\hat{\rho}_{\text{S}}(t) = \hat{\rho}_{\text{S}}^{(1)}(t)\hat{\rho}_{\text{S}}^{(2)}(t)$, if $\rho_{\text{S}}(0) = \rho_{\text{S}}^{(1)}(0)\rho_{\text{S}}^{(2)}(0)$ at the initial instant. The general Lindblad equation can be formally obtained by using the Davies procedure with the eigenstate basis $|k^{(1)}\rangle|k^{(2)}\rangle$ with the eigenfrequencies $\omega_k^{(1)} + \omega_k^{(2)}$. Key assumption (10) remains valid because the equality

$$\exp\left(-\frac{\mathrm{i}\big(\hat{H}_{\text{S}}^{(1)} + \hat{H}_{\text{S}}^{(2)}\big)t}{\hbar}\right)|k^{(1)}\rangle|k^{(2)}\rangle = \exp\Big(-\mathrm{i}\big(\omega_k^{(1)} + \omega_k^{(2)}\big)t\Big)|k^{(1)}\rangle|k^{(2)}\rangle$$

holds, and we obtain the expressions

$$\frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}(t) = -\frac{\mathrm{i}}{\hbar}\left[\hat{H}_{\text{S}}^{(1)} + \hat{H}_{\text{S}}^{(2)}, \hat{\rho}_{\text{S}}(t)\right] + \hat{L}^{(1)}\big[\hat{\rho}_{\text{S}}(t)\big] + \hat{L}^{(2)}\big[\hat{\rho}_{\text{S}}(t)\big] \,, \tag{39}$$

where

$$\begin{aligned} \frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}^{(1)}(t) &= -\frac{\mathrm{i}}{\hbar}\left[\hat{H}_{\text{S}}^{(1)}, \hat{\rho}_{\text{S}}^{(1)}\right] + \hat{L}^{(1)}\big[\hat{\rho}_{\text{S}}^{(1)}(t)\big] \,, \\ \frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}^{(2)}(t) &= -\frac{\mathrm{i}}{\hbar}\left[\hat{H}_{\text{S}}^{(2)}, \hat{\rho}_{\text{S}}^{(2)}\right] + \hat{L}^{(2)}\big[\hat{\rho}_{\text{S}}^{(2)}(t)\big] \,. \end{aligned} \tag{40}$$

Here, $\hat{H}_{\text{S}}^{(1)}$ and $\hat{H}_{\text{S}}^{(2)}$ are Hamiltonians with the eigenfrequencies $\omega_k^{(1)}$ and $\omega_k^{(2)}$ and eigenstates $|k^{(1)}\rangle$ and $|k^{(2)}\rangle$. Each of these systems interacts with its reservoir, such that the Lindblad equations have form (40), where $\hat{\rho}_{\text{S}}^{(1)}$ and $\hat{\rho}_{\text{S}}^{(2)}$ are the density matrices of the first and second system. The validity of (39) can be confirmed by directly differentiating $\hat{\rho}_{\text{S}}(t) = \hat{\rho}_{\text{S}}^{(1)}(t)\hat{\rho}_{\text{S}}^{(2)}(t)$ over time:

$$\begin{aligned} \frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}(t) &= \left(\frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}^{(1)}(t)\right)\hat{\rho}_{\text{S}}^{(2)} + \hat{\rho}_{\text{S}}^{(1)}(t)\left(\frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}^{(2)}(t)\right) \\ &= -\frac{\mathrm{i}}{\hbar}\left[\hat{H}_{\text{S}}^{(1)}, \hat{\rho}_{\text{S}}^{(1)}(t)\right]\hat{\rho}_{\text{S}}^{(2)} + \hat{L}^{(1)}\big[\hat{\rho}_{\text{S}}^{(1)}(t)\big]\hat{\rho}_{\text{S}}^{(2)}(t) \\ &\quad -\frac{\mathrm{i}}{\hbar}\,\hat{\rho}_{\text{S}}^{(1)}(t)\left[\hat{H}_{\text{S}}^{(2)}, \hat{\rho}_{\text{S}}^{(2)}(t)\right] + \hat{\rho}_{\text{S}}^{(1)}(t)\hat{L}^{(2)}\big[\hat{\rho}_{\text{S}}^{(2)}(t)\big] \,, \end{aligned}$$

where the last expression is identically equal to the right-hand side of (39), because any operators of the first and second subsystems commute. Thus, as mentioned above, the Lindblad superoperators for noninteracting systems are additive.

It would seem that if after switching on the interaction between the subsystems, each of them continues to interact only with its reservoir, whose properties did not change after switching the interaction on, the Davies approach should not change the Lindblad superoperator associated with this subsystem. The general Lindblad superoperator remains the same as in the case of noninteracting subsystems, namely, is their sum. As a result, the Lindblad equation reduces to the form

$$\frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}(t) = -\frac{\mathrm{i}}{\hbar}\left[\hat{H}_{\text{S}}^{(1)} + \hat{H}_{\text{S}}^{(2)} + \hat{V}_{12}, \hat{\rho}_{\text{S}}\right] + \hat{L}^{(1)}\big[\hat{\rho}_{\text{S}}(t)\big] + \hat{L}^{(2)}\big[\hat{\rho}_{\text{S}}(t)\big] \,, \tag{41}$$

i.e., the interaction of systems is taken into account only in the Hermitian part of (41). Such an approach is often used in the literature and is called local or phenomenological [6–19, 28].

However, analyzing the derivation of the Lindblad superoperator based on the Davies procedure in detail, we can see that the key step in the derivation of (10),

$$\exp\left(-\frac{\mathrm{i}\big(\hat{H}_{\text{S}}^{(1)} + \hat{H}_{\text{S}}^{(2)} + \hat{V}\big)t}{\hbar}\right)|k^{(1)}\rangle|k^{(2)}\rangle = \exp\Big(-\mathrm{i}\big(\omega_k^{(1)} + \omega_k^{(2)}\big)t\Big)|k^{(1)}\rangle|k^{(2)}\rangle \,,$$

is no longer valid, and Eqn (41) cannot be obtained. The correct Lindblad equation should have the form

$$\frac{\partial}{\partial t}\,\hat{\rho}_{\text{S}}(t) = -\frac{\mathrm{i}}{\hbar}\left[\hat{H}_{\text{S}}^{(1)} + \hat{H}_{\text{S}}^{(2)} + \hat{V}_{12}, \hat{\rho}_{\text{S}}\right] + \hat{L}_V\big[\hat{\rho}_{\text{S}}(t)\big] \,, \tag{42}$$

where $\hat{L}_V\left[\hat{\rho}_\mathrm{S}(t)\right]$ describes the relaxation of the total system and does not reduce to the sum of $\hat{L}^{(1)}\left[\hat{\rho}_\mathrm{S}(t)\right]$ and $\hat{L}^{(2)}\left[\hat{\rho}_\mathrm{S}(t)\right]$. To obtain (42), it is necessary to follow the Davies procedure using the eigenfrequencies and eigenstates of the composite system.

We note that although Eqn (41) preserves the norm and positive definiteness of the density matrix, it is incorrect in the general case. Equation (41) can be used instead of (42) only if the difference between the solutions of (41) and (42) is small for small $\hat{V}_{12}$. However, in this case, intuition based of the perturbation theory gives a wrong answer. We show below that the stationary solutions of (41) and (42) can differ substantially (by an order of magnitude) even as $\hat{V}_{12} \to 0$. This is manifested especially significantly in the interaction with a dephasing reservoir.

6. Errors occurring in the phenomenological approach. Two coupled two-level systems interacting with a dephasing reservoir

We illustrate the incorrectness of the phenomenological approach with the example of dephasing of two coupled TLSs. We first consider a system of two noninteracting TLSs with the Hamiltonian

$$\hat{H}_\mathrm{S} = \hbar\omega_1\hat{\sigma}_1^\dagger\hat{\sigma}_1 + \hbar\omega_2\hat{\sigma}_2^\dagger\hat{\sigma}_2 = \hat{H}_\mathrm{S}^{(1)} + \hat{H}_\mathrm{S}^{(2)} . \tag{43}$$

The eigenstates of noninteracting TLSs are all possible combinations of the eigenstates of the first and second systems, $|e_1e_2\rangle$, $|e_1g_2\rangle$, $|g_1e_2\rangle$, and $|g_1g_2\rangle$. We assume that each TLS interacts with its dephasing reservoir, such that the interaction Hamiltonian has the form

$$\begin{aligned}\hat{H}_\mathrm{SR} &= \hbar\sum_k \gamma_k^{(1)}\hat{\sigma}_z^{(1)}\left(\hat{a}_k^{(1)\dagger} + \hat{a}_k^{(1)}\right) \\ &\quad + \hbar\sum_k \gamma_k^{(2)}\hat{\sigma}_z^{(2)}\left(\hat{a}_k^{(2)\dagger} + \hat{a}_k^{(2)}\right) = \hbar\lambda_1\hat{S}_1\hat{R}_1 + \hbar\lambda_2\hat{S}_2\hat{R}_2 ,\end{aligned} \tag{44}$$

$$\hat{S}_{1,2} = \hat{\sigma}_z^{(1,2)} , \qquad \hat{R}_{1,2} = \sum_k \frac{\gamma_k^{(1,2)}}{\lambda_{1,2}}\left(\hat{a}_k^{(1,2)\dagger} + \hat{a}_k^{(1,2)}\right) . \tag{45}$$

The operators $\hat{S}_1$ and $\hat{S}_2$ entering the system–reservoir interaction Hamiltonian can be decomposed with respect to the eigenstates of system (43) as

$$\begin{aligned}\hat{S}_1 &= \hat{\sigma}_z^{(1)} \\ &= |e_1e_2\rangle\langle e_1e_2| + |e_1g_2\rangle\langle e_1g_2| - |g_1e_2\rangle\langle g_1e_2| - |g_1g_2\rangle\langle g_1g_2| ,\end{aligned} \tag{46}$$

$$\begin{aligned}\hat{S}_2 &= \hat{\sigma}_z^{(2)} \\ &= |e_1e_2\rangle\langle e_1e_2| - |e_1g_2\rangle\langle e_1g_2| + |g_1e_2\rangle\langle g_1e_2| - |g_1g_2\rangle\langle g_1g_2|\end{aligned} \tag{47}$$

and are diagonal in the basis of eigenvectors of $\hat{H}_\mathrm{S}$. According to (39), the Lindblad equation for two noninteracting TLSs has the form

$$\begin{aligned}\frac{\partial\hat{\rho}_\mathrm{S}(t)}{\partial t} &= -\mathrm{i}\left[\hat{H}_\mathrm{S}, \hat{\rho}_\mathrm{S}\right] + \gamma_\mathrm{deph}^{(1)}\left(\hat{\sigma}_z^{(1)}\hat{\rho}_\mathrm{S}(t)\hat{\sigma}_z^{(1)} - \hat{\rho}_\mathrm{S}(t)\right) \\ &\quad + \gamma_\mathrm{deph}^{(2)}\left(\hat{\sigma}_z^{(2)}\hat{\rho}_\mathrm{S}(t)\hat{\sigma}_z^{(2)} - \hat{\rho}_\mathrm{S}(t)\right) \\ &= -\mathrm{i}\left[\hat{H}_\mathrm{S}^{(1)} + \hat{H}_\mathrm{S}^{(2)}, \hat{\rho}_\mathrm{S}\right] + \hat{L}_1\left[\hat{\rho}_\mathrm{S}(t)\right] + \hat{L}_2\left[\hat{\rho}_\mathrm{S}(t)\right] ,\end{aligned} \tag{48}$$

where $\hat{\rho}_\mathrm{S} = \hat{\rho}_\mathrm{S}^{(1)}\hat{\rho}_\mathrm{S}^{(2)}$. This corresponds to the case of two TLSs undergoing independent relaxation. As in the case of one TLS, the nondiagonal elements of each of the TLSs decay exponentially [see (36)],

$$\langle\hat{\sigma}_{1,2}\rangle = \langle\hat{\sigma}_{1,2}(0)\rangle\exp\left(-\gamma_\mathrm{deph}^{(1,2)}t\right) , \tag{49}$$

and the population inversion and energy of each TLS are conserved [see (34)]:

$$\langle\hat{\sigma}_z^{(1,2)}\rangle = \mathrm{const} = \langle\hat{\sigma}_z^{(1,2)}(0)\rangle . \tag{50}$$

Because the system Hamiltonian is an integral of motion, $\left[\hat{H}_\mathrm{S}, \hat{H}_\mathrm{S} + \hat{H}_\mathrm{SR} + \hat{H}_\mathrm{R}\right] = \left[\hat{H}_\mathrm{S}, \hat{H}_\mathrm{SR}\right] = 0$, as in the case of one TLS with a dephasing reservoir, one-dimensional invariant subspaces exist, each consisting of eigenvectors $|e_1e_2\rangle$, $|e_1g_2\rangle$, $|g_1e_2\rangle$, and $|g_1g_2\rangle$. The population probability of each eigenlevel does not change with time. The zeroth law of thermodynamics, as in the case of one TLS, does not hold.

We now assume that the TLSs interact with each other, and the Hamiltonian of the system has the form

$$\begin{aligned}\hat{H}_\mathrm{S} &= \hbar\omega_1\hat{\sigma}_1^\dagger\hat{\sigma}_1 + \hbar\omega_2\hat{\sigma}_2^\dagger\hat{\sigma}_2 + \hbar\Omega_\mathrm{R}\left(\hat{\sigma}_1^\dagger\hat{\sigma}_2 + \hat{\sigma}_2^\dagger\hat{\sigma}_1\right) \\ &= \hat{H}_\mathrm{S}^{(1)} + \hat{H}_\mathrm{S}^{(2)} + \hat{V}_{12} .\end{aligned} \tag{51}$$

We still assume that each TLS interacts with its dephasing reservoir and the interaction Hamiltonian has form (44).

As mentioned above, the phenomenological (or local) approach presents the Lindblad equation in the form

$$\begin{aligned}\frac{\partial\hat{\rho}_\mathrm{S}(t)}{\partial t} &= -\mathrm{i}\left[\hat{H}_\mathrm{S}^{(1)} + \hat{H}_\mathrm{S}^{(2)} + \hat{V}_{12}, \hat{\rho}_\mathrm{S}(t)\right] \\ &\quad + \hat{L}_1\left[\hat{\rho}_\mathrm{S}(t)\right] + \hat{L}_2\left[\hat{\rho}_\mathrm{S}(t)\right] .\end{aligned} \tag{52}$$

It can be shown formally that there is a single integral of motion for Eqn (52), namely, $\hat{I} = \hat{\sigma}_z^{(1)} + \hat{\sigma}_z^{(2)}$, for which $\mathrm{d}\langle\hat{I}_\mathrm{S}\rangle/\mathrm{d}t = \mathrm{tr}\,(\hat{I}_\mathrm{S}\dot{\hat{\rho}}_\mathrm{S}) = 0$. This operator has the eigenstates $|e_1g_2\rangle$ and $|g_1e_2\rangle$ with the eigenvalue 0, the state $|e_1e_2\rangle$ with the eigenvalue 2, and the state $|g_1g_2\rangle$ with the eigenvalue −2. As pointed out in Section 3, the phase space of the system must decompose into invariant subspaces, each of them having eigenstates with the same eigenvalues as the basis vectors. In this example, the number of such subspaces is three: two one-dimensional subspaces with respective basis vectors $|e_1e_2\rangle$ and $|g_1g_2\rangle$, and one two-dimensional subspace with basis vectors $|e_1g_2\rangle$ and $|g_1e_2\rangle$. Indeed, in matrix form Eqn (52) becomes

$$\begin{aligned}&\dot{\rho}^{(e_1e_2,e_1e_2)} = 0 , \qquad \dot{\rho}^{(g_1g_2,g_1g_2)} = 0 , \\ &\dot{\rho}^{(e_1g_2,e_1g_2)} + \dot{\rho}^{(g_1e_2,g_1e_2)} = 0 , \\ &\dot{\rho}^{(e_1g_2,e_1g_2)} - \dot{\rho}^{(g_1e_2,g_1e_2)} = -2\mathrm{i}\Omega_\mathrm{R}\left(\rho^{(e_1g_2,g_1e_2)} - \rho^{(g_1e_2,e_1g_2)}\right) , \\ &\dot{\rho}^{(e_1g_2,g_1e_2)} = \left(-\mathrm{i}(\omega_2 - \omega_1) - \gamma_\mathrm{deph}^{(1)} - \gamma_\mathrm{deph}^{(2)}\right)\rho^{(e_1g_2,g_1e_2)} \\ &\qquad - \mathrm{i}\Omega_\mathrm{R}\left(\rho^{(e_1g_2,e_1g_2)} - \rho^{(g_1e_2,g_1e_2)}\right) , \\ &\dot{\rho}^{(g_1e_2,e_1g_2)} = \left(\mathrm{i}(\omega_2 - \omega_1) - \gamma_\mathrm{deph}^{(1)} - \gamma_\mathrm{deph}^{(2)}\right)\rho^{(g_1e_2,e_1g_2)} \\ &\qquad + \mathrm{i}\Omega_\mathrm{R}\left(\rho^{(e_1g_2,e_1g_2)} - \rho^{(g_1e_2,g_1e_2)}\right) ,\end{aligned} \tag{53}$$

which shows that Eqn (52) decomposes into subsystems corresponding to invariant subspaces. In these subspaces, the Gibbs distribution over possible energy states should occur. However, direct substitution in phenomenological equation (52) shows that for the initial state $\hat{\rho}_\mathrm{S}(0) = |e_1g_2\rangle\langle e_1g_2|$ belonging to the subspace with basis vectors

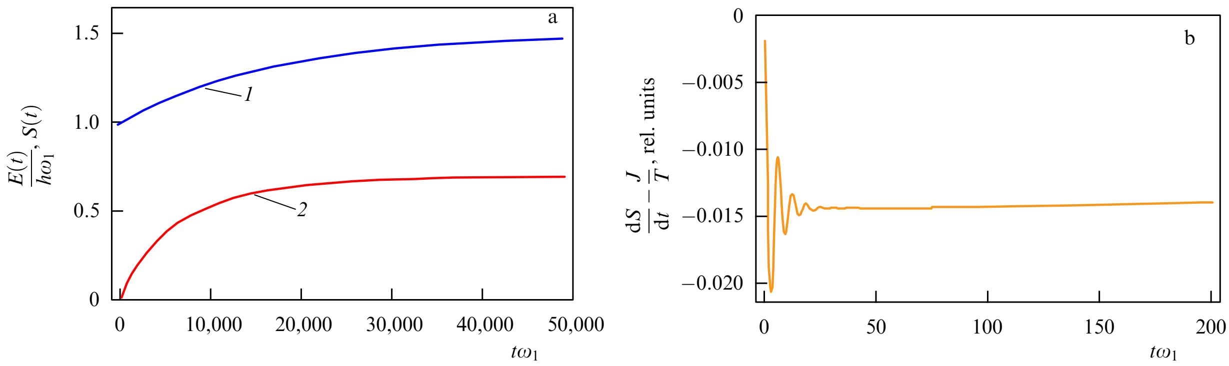

Figure 2. (a) Time dependences of the energy (blue curve *1*) and entropy (red curve *2*) and (b) time dependence of $\mathrm{d}S/\mathrm{d}t-\sum_i J_i/T_i$, predicted by Eqn (52) with the initial condition that the first TLS is in the excited state, the second TLS is in the ground state, and there are no correlations between the TLSs.

$|e_1 g_2\rangle$ and $|g_1 e_2\rangle$, the state

$$\hat{\rho}_{\mathrm{st}}^{\,\mathrm{incor}}=\frac{|e_1 g_2\rangle\langle e_1 g_2|+|g_1 e_2\rangle\langle g_1 e_2|}{2} \tag{54}$$

is a stationary solution. Obviously, expression (54) is not the Gibbs distribution with the reservoir temperature, and therefore phenomenological approach (52) contradicts the zeroth law of thermodynamics, which should hold in this subspace.

In addition, despite the formal satisfaction of the first law of thermodynamics, the second law is also violated. Indeed, because stationary solution (54) obtained from the phenomenological approach is incorrect, the energy flux from the system to the reservoir is also calculated incorrectly. In particular, for $\omega_1<\omega_2$, Eqn (52) predicts the existence of a perpetuum mobile of the second kind: when only a low-frequency TLS is initially excited, the interaction with the reservoir leads to the excitation of a high-frequency TLS (Fig. 2a), irrespective of the relation between transition frequencies and the reservoir temperature.

The Clausius inequality $\mathrm{d}S/\mathrm{d}t-\sum_i J_i/T_i\geqslant 0$ is violated at all times (Fig. 2b). We note that the system entropy increases, $\mathrm{d}S/\mathrm{d}t>0$ (Fig. 2a). However, this increase in entropy is accompanied by the energy being 'pumped out' from the reservoir, $\sum_i J_i/T_i\geqslant 0$, the quantity $\mathrm{d}S/\mathrm{d}t-\sum_i J_i/T_i$ being negative. Thus, the phenomenological approach predicts the violation of the second law of thermodynamics.

Thermodynamic laws are violated because Lindblad superoperators $\hat{L}_1+\hat{L}_2$ do not correspond to any real Hamiltonian $\hat{H}_{\mathrm{SR}}$ of system–reservoir interaction.

We now show that the Lindblad equation obtained by correctly applying the Davies procedure leads to the satisfaction of all thermodynamic laws.

To correctly derive the Lindblad equation, we decompose the interaction operator with respect to the eigenstates of the system, which for Hamiltonian (51) can be found exactly and have the form

$$|\psi_1\rangle=|e_1,e_2\rangle\,,\qquad |\psi_2\rangle=|g_1,g_2\rangle\,, \tag{55}$$

$$|\psi_3\rangle=\sqrt{\frac{W-\Delta\omega}{2W}}\,|e_1,g_2\rangle+\sqrt{\frac{W+\Delta\omega}{2W}}\,|g_1,e_2\rangle\,, \tag{56}$$

$$|\psi_4\rangle=\sqrt{\frac{W+\Delta\omega}{2W}}\,|e_1,g_2\rangle-\sqrt{\frac{W-\Delta\omega}{2W}}\,|g_1,e_2\rangle \tag{57}$$

with the eigenenergies

$$E_1=\omega_1+\omega_2\,,\qquad E_2=0\,,\qquad E_{3,4}=\frac{\omega_1+\omega_2\pm W}{2}\,, \tag{58}$$

where we introduce the notation $W=\sqrt{\Delta\omega^2+4\Omega_{\mathrm{R}}^2}$ and $\Delta\omega=\omega_2-\omega_1$.

Decomposing the operators $\hat{S}_1$ and $\hat{S}_2$ with respect to eigenstates (55)–(57) and substituting the obtained expressions in Lindblad equation (1), we obtain

$$\frac{\partial\hat{\rho}_{\mathrm{S}}(t)}{\partial t}=-\mathrm{i}\left[\hat{H}_{\mathrm{S}}^{(1)}+\hat{H}_{\mathrm{S}}^{(2)}+\hat{V}_{12},\hat{\rho}_{\mathrm{S}}(t)\right]+\hat{L}_V\left[\hat{\rho}_{\mathrm{S}}(t)\right]\,, \tag{59}$$

where the collective Lindblad superoperator has the form

$$\begin{aligned}
\hat{L}_V\left[\hat{\rho}_{\mathrm{S}}(t)\right]&=\gamma_{\mathrm{deph}}^{(1)}\left(\hat{S}_{1d}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{1d}^{\dagger}-\frac{1}{2}\,\hat{S}_{1d}^{\dagger}\hat{S}_{1d}\hat{\rho}_{\mathrm{S}}(t)\right.\\
&\left.-\frac{1}{2}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{1d}^{\dagger}\hat{S}_{1d}\right)+\gamma_{\mathrm{deph}}^{(2)}\left(\hat{S}_{2d}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{2d}^{\dagger}-\frac{1}{2}\,\hat{S}_{2d}^{\dagger}\hat{S}_{2d}\hat{\rho}_{\mathrm{S}}(t)\right.\\
&\left.-\frac{1}{2}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{2d}^{\dagger}\hat{S}_{2d}\right)+\gamma_{\mathrm{deph}}^{(1)}\big(1+N(W)\big)\left(\hat{S}_{1nd}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{1nd}^{\dagger}\right.\\
&\left.-\frac{1}{2}\,\hat{S}_{1nd}^{\dagger}\hat{S}_{1nd}\hat{\rho}_{\mathrm{S}}(t)-\frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{1nd}^{\dagger}\hat{S}_{1nd}\right)+\gamma_{\mathrm{deph}}^{(1)}N(W)\\
&\times\left(\hat{S}_{1nd}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{1nd}-\frac{1}{2}\,\hat{S}_{1nd}\hat{S}_{1nd}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)-\frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{1nd}\hat{S}_{1nd}^{\dagger}\right)\\
&+\gamma_{\mathrm{deph}}^{(2)}\big(1+N(W)\big)\left(\hat{S}_{2nd}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{2nd}^{\dagger}-\frac{1}{2}\,\hat{S}_{2nd}^{\dagger}\hat{S}_{2nd}\hat{\rho}_{\mathrm{S}}(t)\right.\\
&\left.-\frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{2nd}^{\dagger}\hat{S}_{2nd}\right)+\gamma_{\mathrm{deph}}^{(2)}N(W)\left(\hat{S}_{2nd}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{2nd}\right.\\
&\left.-\frac{1}{2}\,\hat{S}_{2nd}\hat{S}_{2nd}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)-\frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{S}_{2nd}\hat{S}_{2nd}^{\dagger}\right)\,.
\end{aligned} \tag{60}$$

Here, operators $\hat{S}_{1d}$, $\hat{S}_{2d}$, $\hat{S}_{1nd}$, and $\hat{S}_{2nd}$ in the eigenstate basis (55)–(57) have the form

$$\hat{S}_{1d}=|\psi_1\rangle\langle\psi_1|-|\psi_2\rangle\langle\psi_2|+\frac{\Delta\omega}{W}\,|\psi_3\rangle\langle\psi_3|-\frac{\Delta\omega}{W}\,|\psi_4\rangle\langle\psi_4|\,, \tag{61}$$

$$\hat{S}_{2d}=|\psi_1\rangle\langle\psi_1|-|\psi_2\rangle\langle\psi_2|-\frac{\Delta\omega}{W}\,|\psi_3\rangle\langle\psi_3|+\frac{\Delta\omega}{W}\,|\psi_4\rangle\langle\psi_4|\,, \tag{62}$$

$$\hat{S}_{1nd}=-\frac{W^2-\Delta\omega^2}{W^2}\,|\psi_4\rangle\langle\psi_3|\,, \tag{63}$$

$$\hat{S}_{2nd}=\frac{W^2-\Delta\omega^2}{W^2}\,|\psi_4\rangle\langle\psi_3|\,. \tag{64}$$

We note that (60) cannot be represented as a sum of dissipative operators acting only on the first and second TLSs. Thus, for coupled subsystems, dissipative operators are not additive.

We study the satisfaction of the thermodynamic laws for the correct Lindblad equation (59) and compare its solution with that of phenomenological Lindblad equation (52).

As in the case of phenomenological equation (52), the correct Lindblad equation (59) has the integral of motion $\hat{I} = \hat{\sigma}_z^{(1)} + \hat{\sigma}_z^{(2)}$ commuting with Hamiltonian (51) of the system and the system–reservoir interaction Hamiltonian (44). According to the zeroth law of thermodynamics, the stationary distribution should be the Gibbs distribution in invariant subspaces determined by the integral of motion. It follows from Eqn (59) that if the state $\hat{\rho}_{\rm S}(0) = |e_1 g_2\rangle\langle e_1 g_2|$ lying in the invariant subspace with the basis $|e_1 g_2\rangle$ and $|g_1 e_2\rangle$ is taken as the initial state, then the stationary solution of Eqn (59) is given by the state

$$\hat{\rho}_{\rm st}^{\rm cor} = \left(|\psi_4\rangle\langle\psi_4| + \exp\left(-\frac{E_3 - E_4}{kT}\right)|\psi_3\rangle\langle\psi_3|\right) \times \left[\exp\left(-\frac{E_3 - E_4}{kT}\right) + 1\right]^{-1}, \qquad (65)$$

representing the Gibbs distribution over the eigenstates $|\psi_4\rangle$ and $|\psi_3\rangle$ belonging to the invariant subspace with basis vectors $|e_1 g_2\rangle$ and $|g_1 e_2\rangle$, which qualitatively differs from predictions of the phenomenological theory.

We now address the question about the satisfaction of the second law of thermodynamics, namely, of inequality (19). Figure 3 shows the time dependence of ${\rm d}S/{\rm d}t - \sum_i J_i/T_i$ for Lindblad equation (59). We can see that inequality (19) is always satisfied for Eqn (59). Thus, the correct representation of the Lindblad equation guarantees that the second law of thermodynamics (19) holds.

Dealing with a perpetuum mobile of the second kind, it is not sufficient to understand that it cannot be made. It is much more interesting to find where a logical error appears in its construction. We return to our example of the interaction of two TLSs with a dephasing reservoir. The cause of the logical error can be found based on the following simple considerations. For the Hamiltonians $\hat{H}_{\rm S}^{(1)} + \hat{H}_{\rm S}^{(2)}$ of two TLSs without interaction, the eigenstates are the states $|e_1 e_2\rangle$, $|e_1 g_2\rangle$, $|g_1 e_2\rangle$, and $|g_1 g_2\rangle$ of the first and second systems. Under interaction with dephasing reservoirs, the transition between the $|e_1 g_2\rangle$ and $|g_1 e_2\rangle$ states is forbidden because the matrix element $\langle e_1 g_2|\hat{H}_{\rm SR}|g_1 e_2\rangle$ vanishes. When the interaction $\hat{V}$ between TLSs is switched on, the eigenstates are linear combinations of the $|e_1 g_2\rangle$ and $|g_1 e_2\rangle$ states, namely, the $|\psi_3\rangle$ and $|\psi_4\rangle$ states specified by Eqns (56) and (57). The transition between $|\psi_3\rangle$ and $|\psi_4\rangle$ becomes possible because the matrix element $\langle\psi_3|\hat{H}_{\rm SR}|\psi_4\rangle$ is nonzero. Moreover, it is nonzero for an arbitrarily weak interaction $\hat{V}$. Thus, switching on the interaction $\hat{V}$ between TLSs removes the prohibition on transition between states (which was forbidden without the interaction), which means that an additional dissipation channel appears. It is clear that the stationary value of the density matrix can strongly change in this case. This example demonstrates that matrix elements of the stationary density matrix obtained from solutions of the correct and phenomenological Lindblad equations can be substantially different.

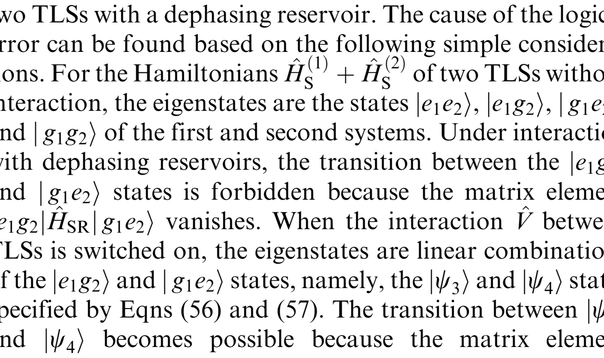

Figure 3. Time dependence of ${\rm d}S/{\rm d}t - \sum_i J_i/T_i$; $\omega_2/\omega_1 = 2$, $g/\omega_1 = 0.02$, $\gamma_{\rm deph}^{(1,2)}/\omega_1 = 0.05$, $kT_1/\hbar\omega_1 = 0.01$, $T_2 = T_1$, predicted by Eqn (59) with the same initial condition as in Fig. 2.

7. Creation of the positive population inversion of two-level systems by coherent pumping

We illustrate the impossibility of describing some phenomena within the phenomenological approach by considering the dynamics of a TLS interacting with a classical monochromatic electromagnetic wave.

It is generally accepted that coherent pumping cannot create a stationary positive inverse population of a TLS [36, 37]. This result is obtained in the phenomenological approximation when, at a positive temperature of the reservoir, energy is either transferred from the system to the reservoir if the reservoir is dissipative, or is not transferred at all if the reservoir is dephasing, because in that case the rates of direct and reverse processes are the same. It follows from the Kubo–Martin–Schwinger relations [see the discussion after (15)] that in the first case, the stationary state corresponds to the Gibbs distribution and the TLS population inversion is negative, while in the second case the inversion does not change at all. For one system and a reservoir, the correct description does not give fundamentally new results. However, if we add, for example, a time-dependent external field to the system, then the Kubo–Martin–Schwinger relation for initial energy levels is violated because the eigenstates of the new Hamiltonian have no fixed energy. This can be taken into account only using the correct approach. The equality of the rates of direct and reverse transitions for initial levels can be skewed in any direction, in particular, can lead to a stationary state with a positive population inversion $\langle\hat{\sigma}_z\rangle_{\rm st} > 0$ [28]. All the work applied to a TLS required to produce a positive population inversion is then performed by the external field.

The problem of the stationary state of a TLS in an external monochromatic field was first solved using phenomenological optical Bloch equations [30, 36, 38]

$$\frac{{\rm d}\langle\hat{\sigma}\rangle}{{\rm d}t} = -{\rm i}\omega_0\langle\hat{\sigma}\rangle - \frac{\rm i}{2}\,\Omega\exp\left(-{\rm i}\omega t\right)\langle\hat{\sigma}_z\rangle - \gamma_\perp\langle\hat{\sigma}\rangle\,, \qquad (66)$$

$$\frac{{\rm d}\langle\hat{\sigma}^\dagger\rangle}{{\rm d}t} = {\rm i}\omega_0\langle\hat{\sigma}^\dagger\rangle + \frac{\rm i}{2}\,\Omega\exp\left({\rm i}\omega t\right)\langle\hat{\sigma}_z\rangle - \gamma_\perp\langle\hat{\sigma}^\dagger\rangle\,, \qquad (67)$$

$$\frac{{\rm d}\langle\hat{\sigma}_z\rangle}{{\rm d}t} = {\rm i}\Omega\big(\langle\hat{\sigma}^\dagger\rangle\exp\left(-{\rm i}\omega t\right) - \langle\hat{\sigma}\rangle\exp\left({\rm i}\omega t\right)\big) - \gamma_\parallel\big(\langle\hat{\sigma}_z\rangle + 1\big)\,, \qquad (68)$$

where $\langle\ \rangle$ is the mean value of an operator, ω is the frequency of the incident monochromatic wave, $\Omega = -\mathbf{E}\mathbf{d}/\hbar$ is the Rabi frequency determining the interaction of the TLS with the external electric field $\mathbf{E}$, $\mathbf{d}$ is the matrix element of the dipole transition in the TLS [24, 30, 38], $\gamma_\parallel$ is the TLS longitudinal relaxation rate, and $\gamma_\perp$ is the TLS transverse relaxation rate introduced phenomenologically to describe relaxation.

It follows, in particular, from optical Bloch equations (66)–(68) that the TLS population tends to a negative stationary value $\langle\hat{\sigma}_z\rangle_{\rm st}$ at large times, even in the case of resonantly coinciding frequencies [30, 38]:

$$\langle\hat{\sigma}_z\rangle_{\mathrm{st}} = -\frac{1}{1+\Omega^2\gamma_{\parallel}\gamma_{\perp}/\left(\gamma_{\perp}^2+(\omega_0-\omega)^2\right)} < 0\,. \tag{69}$$

Thus, optical Bloch equations predict the fundamental impossibility of obtaining a stationary inverse population of TLSs $\langle\hat{\sigma}_z\rangle_{\mathrm{st}} > 0$ by coherent pumping.

It is known that the use of the Lindblad equation is more consistent. However, most papers and monographs on quantum optics [20, 30, 39] are based on the phenomenological approach, i.e., photon (22) and phonon (30) reservoirs are eliminated by using the eigenstates of an isolated TLS with the Hamiltonian $\hat{H}_{\mathrm{S}0} = \hbar\omega_0\hat{\sigma}^{\dagger}\hat{\sigma}$. The obtained Lindblad superoperators for the photon and phonon reservoirs have the respective forms (24) and (32). The presence of an external monochromatic wave is taken into account only by the addition of the term $\hbar\Omega(\hat{\sigma}^{\dagger}\exp(-\mathrm{i}\omega t)+\hat{\sigma}\exp(\mathrm{i}\omega t))$ to the Hamiltonian $\hat{H}_{\mathrm{S}0}$ describing the dipole interaction of the TLS with the external field in the rotating wave approximation. As a result, the Hamiltonian

$$\hat{H}_{\mathrm{S}}(t) = \hbar\omega_0\hat{\sigma}^{\dagger}\hat{\sigma} + \hbar\Omega\left(\hat{\sigma}^{\dagger}\exp(-\mathrm{i}\omega t)+\hat{\sigma}\exp(\mathrm{i}\omega t)\right) \tag{70}$$

of the system and the phenomenological Lindblad equation

$$\begin{aligned}\frac{\partial\hat{\rho}_{\mathrm{S}}(t)}{\partial t} &= -\mathrm{i}\left[\hat{H}_{\mathrm{S}}(t),\hat{\rho}_{\mathrm{S}}(t)\right] + \gamma_{\mathrm{diss}}\left(N(\omega_0)+1\right)\\ &\quad\times\left(\hat{\sigma}\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}^{\dagger} - \frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}^{\dagger}\hat{\sigma} - \frac{1}{2}\,\hat{\sigma}^{\dagger}\hat{\sigma}\hat{\rho}_{\mathrm{S}}(t)\right)\\ &\quad+\gamma_{\mathrm{diss}}N(\omega_0)\left(\hat{\sigma}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma} - \frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}\hat{\sigma}^{\dagger} - \frac{1}{2}\,\hat{\sigma}\hat{\sigma}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)\right)\\ &\quad+\frac{\gamma_{\mathrm{deph}}}{2}\left(\hat{\sigma}^{\dagger}\hat{\sigma}\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}^{\dagger}\hat{\sigma} - \hat{\rho}_{\mathrm{S}}(t)\right)\end{aligned} \tag{71}$$

are obtained [30, 38], where γ_{diss} and γ_{deph} are the dissipation and dephasing rates introduced in Sections 3 and 4. Lindblad equation (71) also predicts that a classical monochromatic electromagnetic wave cannot create a positive stationary inverse population in a TLS [20].

We note that the TLS is a successful idealized model of an atom. According to the existing concepts, incoherent pumping is required in order to obtain a positive population inversion in a TLS. However, the positive population inversion of the working level can also be created, for example, in a three-level system by coherent pumping at a frequency different from the working transition frequency. In passing from the three-level system to the TLS, the TLS Hamiltonian does not contain the term describing interaction with the coherent field, and the Lindblad equation contains the additional term [30]

$$\hat{L}_{\mathrm{pump}}(\hat{\rho}_{\mathrm{S}}) = \gamma_{\mathrm{pump}}\left(\hat{\sigma}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma} - \frac{1}{2}\,\hat{\rho}_{\mathrm{S}}(t)\hat{\sigma}\hat{\sigma}^{\dagger} - \frac{1}{2}\,\hat{\sigma}\hat{\sigma}^{\dagger}\hat{\rho}_{\mathrm{S}}(t)\right), \tag{72}$$

describing the TLS transition to the excited state, with the rate γ_{pump} called the incoherent TLS pumping rate. A similar result is obtained for the interaction of a TLS with a reservoir with a negative temperature. For a sufficiently high pumping rate $\gamma_{\mathrm{pump}} > \gamma_{\mathrm{diss}}$, the Lindblad superoperator $\hat{L}_{\mathrm{pump}}(\hat{\rho}_{\mathrm{S}})$ in Eqn (72) can produce a positive inverse population in the TLS. We note that it is assumed in this case that the system being pumped is physically different from the TLS [2, 40].

We consider the Lindblad equation obtained using the Davies algorithm considered in Section 2 [41]. Unlike the phenomenological approach (71), the correct Lindblad equation automatically contains a term like (72).

In contrast to the cases considered in Sections 3 and 4, Hamiltonian (70) of the system depends on time, and therefore, to find the correct Lindblad equation for the TLS density matrix, we perform the unitary transformation $\hat{U}_{\mathrm{ex}}(t) = \exp(-\mathrm{i}\omega\hat{\sigma}^{\dagger}\hat{\sigma}t)$ of Hamiltonian (70), Hamiltonians (21) and (29) of the reservoirs, and system–reservoir interaction Hamiltonians (22) and (30) [41]:

$$\begin{aligned}\hat{H}_{\mathrm{S}}' &= \hat{U}_{\mathrm{ex}}^{\dagger}(t)\hat{H}_{\mathrm{S}}(t)\hat{U}_{\mathrm{ex}}(t) - \mathrm{i}\hat{U}_{\mathrm{ex}}^{\dagger}(t)\,\frac{\partial\hat{U}_{\mathrm{ex}}(t)}{\partial t}\\ &= \hbar(\omega_0-\omega)\hat{\sigma}^{\dagger}\hat{\sigma} + \hbar\Omega(\hat{\sigma}^{\dagger}+\hat{\sigma})\,,\end{aligned} \tag{73}$$

$$\hat{H}_{\mathrm{R}}' = \hat{U}_{\mathrm{ex}}^{\dagger}(t)\hat{H}_{\mathrm{R}}\hat{U}_{\mathrm{ex}}(t) = \sum_{\nu}\hbar\nu\hat{a}_{1,\nu}^{\dagger}\hat{a}_{1,\nu} + \sum_{\nu}\hbar\nu\hat{a}_{2,\nu}^{\dagger}\hat{a}_{2,\nu}\,, \tag{74}$$

$$\begin{aligned}\hat{H}_{\mathrm{SR}}' &= \hat{U}_{\mathrm{ex}}^{\dagger}(t)\hat{H}_{\mathrm{SR}}\hat{U}_{\mathrm{ex}}(t)\\ &= \hbar\sum_{\nu}w_{1\nu}\left(\hat{\sigma}^{\dagger}\exp(\mathrm{i}\omega t)+\hat{\sigma}\exp(-\mathrm{i}\omega t)\right)\left(\hat{a}_{1,\nu}^{\dagger}+\hat{a}_{1,\nu}\right)\\ &\quad+\hbar\sum_{\nu}w_{2\nu}\hat{\sigma}^{\dagger}\hat{\sigma}(\hat{a}_{2,\nu}^{\dagger}+\hat{a}_{2,\nu})\,.\end{aligned} \tag{75}$$

After this transformation, Hamiltonian (70) of the system loses time dependence, and its eigenstates and eigenfrequencies have the form

$$\begin{aligned}&|\psi_+\rangle = \sqrt{1-\alpha^2}\,|e\rangle + \alpha|g\rangle\,,\\ &\lambda_+ = \frac{\omega_0-\omega}{2} + \sqrt{\frac{(\omega_0-\omega)^2}{4}+\Omega^2}\,,\end{aligned} \tag{76}$$

$$\begin{aligned}&|\psi_-\rangle = -\alpha|e\rangle + \sqrt{1-\alpha^2}\,|g\rangle\,,\\ &\lambda_- = \frac{\omega_0-\omega}{2} - \sqrt{\frac{(\omega_0-\omega)^2}{4}+\Omega^2}\,.\end{aligned} \tag{77}$$

The parameter α depends on the Rabi frequency Ω, the TLS transition frequency ω_0, and the incident field frequency ω as

$$\alpha = \frac{2\Omega}{\sqrt{4\Omega^2+\left(\omega_0-\omega+\sqrt{(\omega_0-\omega)^2+4\Omega^2}\right)^2}}\,. \tag{78}$$

It follows from (76) and (77) that $\lambda_+ > \lambda_-$ whenever the TLS is excited by a classical monochromatic electromagnetic wave. Below, these expressions are used to derive the Lindblad superoperators.

We note that the system–reservoir interaction Hamiltonian $\hat{H}_{\mathrm{SR}}'(t)$ acquires an explicit time dependence. However, this does not produce additional difficulties in the use of the Davies algorithm.

The eigenstates $|\psi_{\pm}\rangle$ of the Hamiltonian $\hat{H}_{\mathrm{S}}'$ in Eqn (73) are quantum superpositions of the ground $|g\rangle$ and excited $|e\rangle$ states of interacting TLSs (20). In addition, the ground state of the Hamiltonian $\hat{H}_{\mathrm{S}}'$ in (73) changes depending on the incident electromagnetic wave frequency ω. We can see from (76)–(78) that at incident field frequencies lower than the TLS transition frequency, $\omega < \omega_0$ ($\alpha < 0.5$), the $|g\rangle$ state makes a major contribution to the ground state $|\psi_-\rangle$, whereas in the opposite case $\omega > \omega_0$ ($\alpha > 0.5$), the major contribution to the ground $|\psi_-\rangle$ state comes from the $|e\rangle$ state.

The use of eigenstates (76) and (77) leads to the Lindblad equation containing six Lindblad superoperators related to the interaction of TLSs with the *photon* reservoir, instead of two superoperators obtained in the phenomenological approach [see (71)], and three Lindblad superoperators related to the interaction of TLSs with the *phonon* reservoir instead of one superoperator obtained in the phenomenological approach [see (71)] [41].

Explicit expressions for the new Lindblad superoperators are rather cumbersome [41] and we do not present them here. We note that the TLS relaxation dynamics related to new Lindblad superoperators appearing due to interaction with the *photon* reservoir do not differ from the dynamics predicted by phenomenological Lindblad equation (71).

The TLS relaxation dynamics associated with the new Lindblad superoperators appearing in the interaction with the *phonon* reservoir considerably differ in some cases from those predicted by the phenomenological approach, Eqn (71). This is explained primarily by the fact that in the presence of an external classical monochromatic wave, the TLS–phonon-reservoir interaction operator $\hat{\sigma}_z$ [see (30)] no longer commutes with TLS Hamiltonian (70):

$$\left[\hat{H}_{\mathrm{S}}, \hat{\sigma}_z\right] \neq 0\,. \tag{79}$$

This means that the phonon reservoir does not conserve the TLS energy and can facilitate the energy flow both into and out of the TLS. The presence of energy inflow and outflow due to the TLS–phonon-reservoir coupling is the main difference between the phenomenological approach and the Davies procedure approach. These processes correspond to the Lindblad operators

$$\begin{aligned}\hat{L}(\hat{\rho}'_{\mathrm{S}}) &= \gamma_{\mathrm{deph}}\,\frac{G(\Delta\lambda)}{G(0)}\left(\hat{S}\hat{\rho}'_{\mathrm{S}}\hat{S}^{\dagger} - \frac{1}{2}\,\hat{S}^{\dagger}\hat{S}\hat{\rho}'_{\mathrm{S}} - \frac{1}{2}\,\hat{\rho}'_{\mathrm{S}}\hat{S}^{\dagger}\hat{S}\right)\\ &\quad + \gamma_{\mathrm{deph}}\,\frac{G(-\Delta\lambda)}{G(0)}\left(\hat{S}^{\dagger}\hat{\rho}'_{\mathrm{S}}\hat{S} - \frac{1}{2}\,\hat{S}\hat{S}^{\dagger}\hat{\rho}'_{\mathrm{S}} - \frac{1}{2}\,\hat{\rho}'_{\mathrm{S}}\hat{S}\hat{S}^{\dagger}\right),\end{aligned} \tag{80}$$

where $\Delta\lambda = \lambda_+ - \lambda_-$,

$$\hat{S} = |\psi_-\rangle\langle\psi_-|\hat{\sigma}^{\dagger}\hat{\sigma}|\psi_+\rangle\langle\psi_+|\,, \tag{81}$$

and the prime at the TLS density matrix means that it is transformed by the unitary transformation $\hat{U}_{\mathrm{ex}}(t) = \exp\left(-\mathrm{i}\omega\hat{\sigma}^{\dagger}\hat{\sigma}t\right)$, similarly to (73)–(75).

Lindblad superoperator (80) contains two summands corresponding to the energy flow from and into the TLS. The relative rates of these processes are determined by temperature factors $G(\pm\Delta\lambda)$. At temperatures $kT \geqslant \hbar\Delta\lambda$, the energy outflow and inflow rates due to interaction with the phonon reservoir become equal. As a result, predictions of the phenomenological Lindblad equation (71) and the correct Lindblad equation do not differ considerably.

Considerable differences in the dynamics can be observed at low temperatures $kT < \hbar\Delta\lambda$. We consider the case $kT \ll \hbar\Delta\lambda$. Then only the first summand remains in the right-hand side of (80), because $\Delta\lambda = \lambda_+ - \lambda_- = \left[(\omega_0 - \omega)^2 + 4\Omega^2\right]^{1/2} > 0$. Thus, the Lindblad superoperator (80) takes the form

$$\hat{L}(\hat{\rho}'_{\mathrm{S}}) = \gamma_{\mathrm{deph}}\,\frac{G(\Delta\lambda)}{G(0)}\left(\hat{S}\hat{\rho}'_{\mathrm{S}}\hat{S}^{\dagger} - \frac{1}{2}\,\hat{S}^{\dagger}\hat{S}\hat{\rho}'_{\mathrm{S}} - \frac{1}{2}\,\hat{\rho}'_{\mathrm{S}}\hat{S}^{\dagger}\hat{S}\right). \tag{82}$$

For definiteness, we consider a weak monochromatic electromagnetic wave in the case where the Rabi frequency is considerably smaller than the detuning of the external field frequency for the TLS transition frequency, $|\Omega| \ll |\omega_0 - \omega|$. The operator $\hat{S}$ entering Lindblad superoperator (82) then strongly depends on which frequency, ω_0 or ω, is higher. From (76)–(78) and (81), in the leading order in $\Omega/|\omega_0 - \omega|$, we obtain

$$\hat{S} \approx \begin{cases} -\dfrac{\Omega}{|\omega_0 - \omega|}\,\hat{\sigma}\,, & \omega < \omega_0\,,\\[2mm] \dfrac{\Omega}{|\omega_0 - \omega|}\,\hat{\sigma}^{\dagger}\,, & \omega > \omega_0\,. \end{cases} \tag{83}$$

Such a behavior of the operator $\hat{S}$ is consistent with the rearrangement of the eigenstates of the Hamiltonian $\hat{H}'_{\mathrm{S}}$. Indeed, for $\omega > \omega_0$, the ground state is $|\psi_-\rangle \approx |e\rangle$ and $|\psi_+\rangle \approx |g\rangle$. Thus, it follows from (81) that $\hat{S} \propto |e\rangle\langle g| = \hat{\sigma}^{\dagger}$, which is consistent with (83). Using (82), we can see that for a small amplitude of the external field, $|\Omega| \ll |\omega_0 - \omega|$, positive detuning $\omega > \omega_0$, and low temperatures $kT \ll \hbar\Delta\lambda$, the Lindblad superoperator takes the form

$$\hat{L}(\hat{\rho}'_{\mathrm{S}}) = \frac{\Omega\gamma_{\mathrm{deph}}}{|\omega_0 - \omega|}\left(\hat{\sigma}^{\dagger}\hat{\rho}'_{\mathrm{S}}\hat{\sigma} - \frac{1}{2}\,\hat{\sigma}\hat{\sigma}^{\dagger}\hat{\rho}'_{\mathrm{S}} - \frac{1}{2}\,\hat{\rho}'_{\mathrm{S}}\hat{\sigma}\hat{\sigma}^{\dagger}\right). \tag{84}$$

This superoperator (84) corresponds in form to the Lindblad superoperator (72) for incoherent pumping. Thus, for certain parameters of the system, the phonon reservoirs coupled to the TLS in the presence of an external classical monochromatic electromagnetic wave can lead to incoherent pumping of the TLS.

Numerical simulations of the correct Lindblad equation for the density matrix show that the incoherent pumping caused by the phonon reservoir during excitation of the TLS by a classical monochromatic field leads in some cases to a positive inverse population of the TLS, even in the presence of a photon reservoir [40]. Figure 4 shows the dependence of the mean inverse population $\langle\hat{\sigma}_z\rangle$ on the Rabi frequency. At high temperatures $kT \gg \hbar\Delta\lambda$, the inverse populations obtained from the correct and phenomenological Lindblad equations do not differ considerably (Fig. 4b). At sufficiently low temperatures $kT \ll \hbar\Delta\lambda$, these two approaches give substantially different results: the correct Lindblad equation predicts the possibility of obtaining the inverse population in the TLS (Fig. 4a).

We address the question about energy fluxes and the satisfaction of the second law of thermodynamics. The phenomenological Lindblad equation can predict that at some instants, energy is transferred from the reservoir to the system (positive values of J in Fig. 5a). Such a behavior of the system contradicts the second law of thermodynamics: the value of $\mathrm{d}S/\mathrm{d}t - J/T$ becomes negative. The correct Lindblad equation gives a monotonic time dependence of the energy flux between the system and the reservoir (Fig. 5a). For the parameters chosen, the phonon reservoir makes the main contribution to the energy flux. The energy transferred from an external classical monochromatic electromagnetic wave is sufficient both for transfer to the reservoir (Fig. 5a) and for TLS pumping (Fig. 4a). Thus, we can see that according to the correct Lindblad equation, the system is pumped entirely due to the work produced by the external classical monochromatic electromagnetic wave interacting with the TLS.

We note in conclusion that the maximum difference between the phenomenological and correct Lindblad equa-

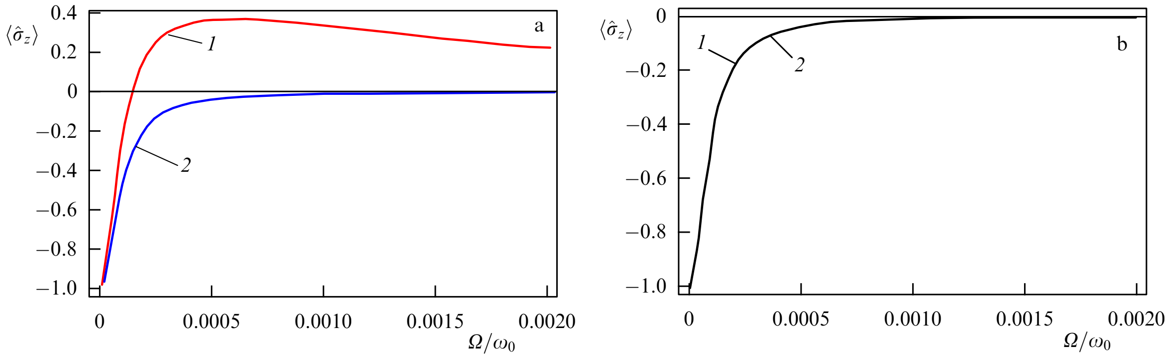

Figure 4. Dependence of the inverse population $\langle\hat{\sigma}_z\rangle$ on the TLS–laser-radiation coupling constant Ω for $|\omega-\omega_0| = 10^{-3}\omega_0$, $\gamma_{\text{diss}}/\omega_0 = 10^{-6}$, $\gamma_{\text{deph}}/\omega_0 = 10^{-4}$, (a) $T = 10^{-3}\omega_0$ and (b) $T = 10^{-1}\omega_0$. Red curve *1* is the solution of the correct Lindblad equation for the density matrix, blue curve *2* is the solution of the phenomenological Lindblad equation; at the initial instant, the TLS is in the ground state. Curves *1* and *2* in Fig. 4b virtually coincide.

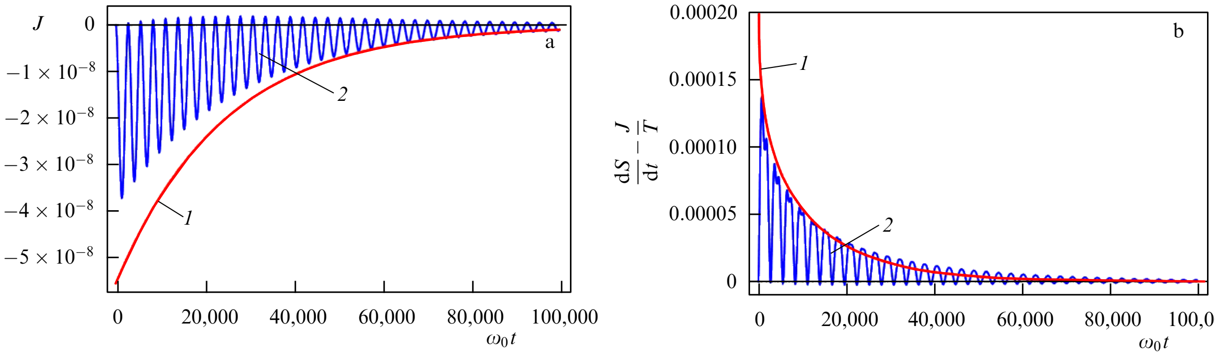

Figure 5. Time dependences of (a) the energy flux J between the reservoir and system and (b) $dS/dt - J/T$. At the zero instant, the TLS is in the ground state, $\Omega/\omega_0 = 10^{-3}$, $|\omega-\omega_0| = 10^{-3}\omega_0$, $\gamma_{\text{diss}}/\omega_0 = 10^{-6}$, $\gamma_{\text{deph}}/\omega_0 = 10^{-4}$, $T = 10^{-3}\omega_0$. Red curve *1* is the solution of the correct Lindblad equation for the density matrix, blue curve *2* is the solution of the phenomenological Lindblad equation; at the initial instant, the TLS is in the ground state.

tions appears in the limit $\max(|\omega-\omega_0|, \Omega) \gg \gamma_a, \gamma_{\text{deph}}$ (Fig. 4a). In the opposite limit case, when $\max(|\omega-\omega_0|, \Omega) \ll \gamma_a, \gamma_{\text{deph}}$, the difference is insignificant (Fig. 4b), and the phenomenological Lindblad equations for the density matrix can be used, as is accepted in the literature [20, 30, 39].

8. Conclusions

We have studied the relaxation of interacting open quantum systems. The Davies procedure for eliminating the reservoir variables and obtaining the Lindblad equation for the density matrix is presented. Solutions of this equation are positive definite density matrices with unit trace, ensuring that the first and second laws of thermodynamics hold.

Assuming that the Hamiltonians of the coupling of each of the subsystems to the reservoir are independent, the results obtained using the Davies and phenomenological approaches are compared. The latter are based on the assumption that the dissipative Lindblad superoperators appearing during the elimination of the reservoir variables remain for each of the subsystems, just as in the absence of coupling between the subsystems. In other words, the Lindblad superoperator of a complex system is assumed to be the sum of the Lindblad superoperators of individual subsystems. Taking into account that the subsystem–reservoir interaction Hamiltonians do not change after switching the interaction on, this assumption seems reasonable. We have shown, however, that this assumption, which substantially simplifies calculations, can lead to incorrect results contradicting the second law of thermodynamics. The reason is that the change in the eigenstates of the Hamiltonian of a composite system is neglected in the interaction between its parts. As a result, the dynamics of interaction of a composite system with the reservoir qualitatively changes. The interaction between subsystems leads not only to the appearance of the corresponding term in the Hamiltonian of the total system but also to the nonadditivity of Lindblad superoperators. In particular, for the same subsystem–reservoir interaction Hamiltonians, depending on the type of the coupling between subsystems, the Lindblad superoperator can cause pure dephasing or simultaneous dephasing and dissipation.

We note that the phenomenological approach is used in the literature in most cases because of its relative simplicity. For example, it is used for studying quantum dots [13], qubits [42], atoms interacting with resonator modes [43, 44], quantum transport in biological systems [9, 17], and plasmon structures [45]. In particular, it was stated in recent papers on quantum transport that the interaction with the reservoir can 'miraculously' improve the quantum transport properties [7, 12, 13, 16–18, 46, 47]. The main focus in these papers is on the density matrix dynamics, while the satisfaction of the second law of thermodynamics was almost never verified. In other words, the results obtained using the phenomenological approach have uncontrolled accuracy. In a number of cases, their incorrectness can be proved, while in other cases the difference in results is not qualitative but quantitative. Unfortunately, it is rather difficult to predict the type of specific situations in advance.

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