A novel method based on spectral Green functions is presented for the simulation of driven open quantum dynamics that can be described by the Lindblad master equation in Liouville density operator space. The method extends the Hilbert space formalism and provides simple algebraic connections between the driven and non-driven dynamics in the spectral frequency domain. The formalism shows remarkable analogies to the use of Green functions in quantum field theory such as the elementary excitation energies and the Dyson self-energy equation. To demonstrate its potential, we apply the novel method to a coherently driven dissipative ensemble of 2-level systems comprising a single “active” subsystem interacting with N identical “passive” subsystems — a generic model with important applications in quantum optics and dynamic nuclear polarization. The novel method dramatically reduces computational cost compared with simulations based on solving the full master equation, thus making it possible to study and optimize many-body correlated states in the physically realistic limit where N is of several orders of magnitude.

**Introduction.** Open quantum dynamics takes into account the environment (outer degrees of freedom) and so more accurately describes real physical phenomena compared with closed quantum dynamics based entirely on the energy operator (inner Hamiltonian). The environment tends to quench coherent states and to purge quantum information by free thermal decay. On the other hand, it can cause irreversible dynamics that makes it possible to create and keep coherent states by continuous driving the system out of its thermal equilibrium \(\rho\). This makes driven open systems a fundamental object of quantum theory.

The mathematical description of the dynamics of open quantum systems is non-unitary and generally complex. This especially refers to correlated many-body systems that exhibit collective phenomena depending on their environment. In many cases the Lindblad master equation approach can be used that retains the positivity of the density operator and introduces the environmental effects through Markovian quantum jump operators that enter the dissipator in a simple algebraic way \[\rho\]. However, the Liouville space containing the trajectories of the density operator describing the evolution of a many-body open quantum system grows exponentially with the number of constituents and the dynamics is sensitive to a large number of physical parameters. Hence efficient mathematical tools are necessary to perform adequate approximations and state space restrictions in order to gain insight into the underlying physics.

For the description of collective phenomena in large-scale closed quantum systems at thermal equilibrium, a method involving the use of Green functions was developed that statistical physics adopted from quantum field theory \[\gamma\]. This method is based on the approximate calculation of correlations between dynamic operators that leads to self-consistent equations for the observables. Subsequently, this method was extended to non-equilibrium closed and non-Markovian open quantum systems describing various transport phenomena where, besides the standard time domain formalism, it was transformed to an inhomogeneous spectral problem in Hilbert space \[\gamma\].

Here we propose an extension of the non-equilibrium spectral approach to the important class of driven Markovian open quantum dynamics in the Liouville space of the density operator. To this end we show that the Green function for an inhomogeneous spectral problem can be formulated in terms of both Hamiltonian and dissipative parts of the Lindblad master equation. The steady state of the driven system is then obtained by a simple algebraic transform of the non-driven thermal equilibrium. Remarkably there is a close analogy between our proposed spectral formalism and the use of Green functions in quantum field theory, including the elementary excitation energies and the Dyson self-energy equation. For a demonstration, we apply the method to a coherently driven dissipative ensemble of correlated 2-level systems, a basic model used in quantum optics and dynamic nuclear polarization. We show that, because the computational cost of the method is significantly cheaper in comparison with the direct master equation simulation, it is possible to study and optimize the many-body correlated states in the realistically large-scale limit. This opens up new possibilities in simulations of many-body driven open quantum dynamics including the spectral response to the driving and fast search for the optimal parameter regions.

**Spectral Green functions for the Lindblad master equation.** The dynamics of open quantum systems is described in terms of the Lindblad master equation \[\gamma\]

\[
\dot{\rho} = \mathcal{M}\rho, \quad \mathcal{M} = -i[H, \cdot] + \mathcal{D} \quad (1)
\]

where \(\rho\) is the density operator, \(H\) is the Hamiltonian describing the (internal) energy of the system and \(\mathcal{D}\) is the dissipator that represents the effect of the (external) en-
verson. The latter uses Markovian jumps represented by (dimensionless) operators $X_j$ and system-environment exchange rates $\gamma_j$,  
\[ \mathcal{D} = \sum \gamma_j \mathcal{L}(X_j), \quad \mathcal{L}(X)\rho = X\rho X^\dagger - \frac{1}{2} \{X^\dagger X, \rho\}. \quad (2) \]

The dissipator tends to return the system to the state that is in thermal equilibrium with the environment while the Hamiltonian contains terms that drive the system out of thermal equilibrium, $\mathcal{D}\rho_{th} = 0$, $[H, \rho_{th}] \neq 0$. Eq. (1) describes then a driven open quantum dynamics in the Liouville space of the density operator.

The master equation preserves the unit trace $\text{Tr}\rho = 1$. The superoperator $\mathcal{M}$ transfers all operators to traceless operators and hence reduces the dimension of the Liouville space. As a consequence, $\mathcal{M}$ is degenerate with a non-trivial zero eigenspace. This eigenspace contains a non-thermal steady state that is eventually established in the driven system,  
\[ t \to +\infty, \quad e^{\mathcal{M}t} \rho_{th} \to \rho, \quad \mathcal{M}\rho = 0, \quad \text{Tr}\rho = 1. \quad (3) \]

In the fully dissipative case the zero eigenspace is 1-dimensional, Eq. (3) uniquely defines the steady-state for all initial conditions. In this case the only traceless solution to Eq. (3) is trivial,  
\[ \mathcal{M}\rho = 0, \quad \text{Tr}\rho = 0 \to \rho = 0. \quad (4) \]

Suppose the Hamiltonian can be represented in the form  
\[ H = P + H_0 + \zeta H_1, \quad [H_{0,1}, \rho_{th}] = 0, \quad [P, \rho_{th}] \neq 0 \quad (5) \]

where $\zeta$ is a real scalar parameter, the Hermitian operators $H_{0,1}$ are non-driving and the Hermitian operator $P$ contains the driving terms of the Hamiltonian. We will assume that the operator $H_1$ is dimensionless and $H_0, P$ and $\zeta$ are measured in frequency units. Extracting the thermal equilibrium part $\rho = \rho_{th} + \hat{\rho}$, $\text{Tr}\hat{\rho} = 0$ and introducing the superoperators $F_0 = -i[H_0, \cdot] + \mathcal{D}$, $P = i[P, \cdot]$, $H_1 = i[H_1, \cdot]$, the homogeneous Eq. (3) is rewritten as an inhomogeneous generalized spectral problem  
\[ (F_0 - P - \zeta H_1) \hat{\rho} = P\rho_{th} \quad (6) \]

where $\zeta$ plays the role of a spectral parameter and the solution $\hat{\rho}$ belongs to the subspace of traceless operators.

Assuming the validity of Eq. (4) for any real value of $\zeta$, the superoperator $F_0 - P - \zeta H_1$ is non-degenerate and hence invertible. Then the unique solution to Eq. (6) is $\hat{\rho} = \mathcal{G}(\zeta)\rho_{th}$, where the superoperator $\mathcal{G}(\zeta)$ must satisfy the equation  
\[ (F_0 - P - \zeta H_1) \mathcal{G}(\zeta) = 1 \quad (7) \]

with the unit superoperator in the right-hand side. The superoperator $\mathcal{G}(\zeta)$ is independent of the thermal equilibrium, acts in the subspace of traceless operators and plays the role of the Green function of the inhomogeneous spectral problem (6). We call the superoperator $\mathcal{G}(\zeta)$ the driven spectral Green function for Eqs. (1), (5). By virtue of the previous equations, the steady state is written as  
\[ \rho = [1 + \mathcal{X}(\zeta)]\rho_{th}, \quad \mathcal{X}(\zeta) = \mathcal{G}(\zeta)P. \quad (8) \]

Eq. (7) can be rewritten in the form  
\[ [1 - \mathcal{G}_0 P] \mathcal{G}(\zeta) = \mathcal{G}_0(\zeta) \quad (9) \]

where the superoperator $\mathcal{G}_0(\zeta)$ must satisfy the equation  
\[ (F_0 - \zeta H_1) \mathcal{G}_0(\zeta) = 1. \quad (10) \]

Indeed, multiplying both sides of Eq. (9) by the invertible superoperator $F_0 - \zeta H_1$, we obtain to Eq. (7). Eq. (10) uniquely defines the superoperator $\mathcal{G}_0(\zeta)$ that is independent of the driving part $P$ of the Hamiltonian. We will call the superoperator $\mathcal{G}_0(\zeta)$ the non-driven spectral Green function. Using Eq. (9), the solution to Eq. (9) becomes $\hat{\rho} = \mathcal{X}(\zeta)\rho_{th} = [1 - \mathcal{G}_0(\zeta)P]^{-1}\mathcal{G}_0(\zeta)P\rho_{th}$. Applying the universal operator relation $1 + (1 - Y)^{-1}Y = (1 - Y)^{-1}$, we obtain then for the steady state  
\[ [1 - \mathcal{X}_0(\zeta)]\rho = \rho_{th}, \quad \mathcal{X}_0(\zeta) = \mathcal{G}_0(\zeta)P. \quad (11) \]

The dual Eqs. (9), (11) provide compact formulas for the steady state $\rho$ of the master Eq. (1) as a linear transformation of the thermal equilibrium $\rho_{th}$ defined by the product of the driving superoperator $P$ and the driven and non-driven spectral Green functions $\mathcal{G}$, $\mathcal{G}_0$ determined by Eqs. (7), (10) and connected via Eq. (9).

The superoperator $F_0 - P - \zeta H_1$ of Eq. (7) linearly depends on $\zeta$, hence the driven Green function $\mathcal{G}(\zeta)$ is rationally extendable into the complex plane of $\zeta$,  
\[ \mathcal{G}(\zeta) = \mathcal{G}(0) + \sum_{r=1}^{m} (\zeta - \zeta_r)^{-1}\mathcal{G}^{(r)}. \quad (12) \]

Here the poles $\zeta = \zeta_r$ and the residues $\mathcal{G}^{(r)}$ are given by (suitably normalized) solutions to the homogeneous driven spectral problem $(F_0 - P - \zeta H_1)\mathcal{G}^{(r)} = 0$. The superoperator $F_0 - P - \zeta H_1$ is real and non-degenerate for real $\zeta$, so the poles have nonzero imaginary parts and exist in complex conjugate pairs. Similarly, the non-driven Green function can be expanded  
\[ \mathcal{G}_0(\zeta) = \mathcal{G}_0(0) + \sum_{r=1}^{m_0} (\zeta - \zeta_{0r})^{-1}\mathcal{G}_0^{(r)} \quad (13) \]

with $(F_0 - \zeta_{0r} H_1)\mathcal{G}_0^{(r)} = 0$. In the case, where the superoperators $F_0$, $H_1$ commute, they have the same set of (traceless) eigenoperators $\{\rho_r\}$, $[F_0 - \lambda_r^{(0)}] \rho_r = \left[H_1 - \lambda_r^{(1)}\right] \rho_r = 0$. The non-degeneracy of $F_0$ provides that $\lambda_r^{(0)} \neq 0$. Then Eqs. (10), (13) yield that $\mathcal{G}_0(\zeta)$ is
operators $G_0(\zeta)$ play the roles of the elementary excitation energies. Indeed, the former requires only an operator inversion while the latter needs calculation by Eq. (3). In the same case $[G_0, H_1] = 0$ the non-driven spectral Green function $G_0(\zeta)$ can be written as a generalized Fourier transform

$$ G_0(\zeta) = -\int_{-\infty}^{+\infty} \tilde{G}_0(t)e^{-\zeta H_1 t} dt $$

(15)

where $\tilde{G}_0(t)$ is the Green function of the inhomogeneous non-driven dynamical problem for $\zeta = 0$

$$ \dot{\rho} = F_0\rho + f. $$

(16)

Indeed, for any bounded inhomogeneity $f$ the bounded solution to Eq. (16) is written as

$$ \rho(t) = \int_{-\infty}^{+\infty} \tilde{G}_0(t-t')f(t') dt', $$

$$ \tilde{G}_0(t) = e^{F_0 t}, \quad t \geq 0; \quad \tilde{G}_0(t) = 0, \quad t < 0. $$

We have then

$$ -\int_{-\infty}^{+\infty} \tilde{G}_0(t)e^{-\zeta H_1 t} dt = -\int_{0}^{+\infty} e^{(F_0 - \zeta H_1) t} dt = (F_0 - \zeta H_1)^{-1} G_0(\zeta). $$

The above equations imply that the magnitude $-G_0(\zeta)(\rho(0)$ describes the generalized spectrum of the free thermal decay of the traceless part of an initial state $\rho(0)$.

Eqs. (7), (10) can be considered to be a Liouville space extension of the spectral Green function formalism in Hilbert space [10-12]. There are noteworthy analogies to quantum field theory. The real parts of the poles $\zeta = \zeta_\rho$, $\zeta = \zeta_0$ of the superoperators $G(\zeta)$, $G_0(\zeta)$ of Eqs. (12), (13) play the roles of the elementary excitation energies. Eq. (9) is a copy of the Dyson equation with the superoperators $G_0$, $G$ and $P$ playing the roles of the bare and dressed propagators and the self-energy [7,9]. The superoperator $G_0(t-t')$ of Eq. (15) plays the role of the retarded Green function that describes the free irreversible decay of correlations between the initial state and the thermal equilibrium.

The advantage of the method introduced in this section is that the use of Eqs. (7), (8) is generally much less computationally costly than calculating the steady state as the dynamic limit or an element of the zero eigenspace by Eq. (3). Indeed, the former requires only an operator inversion while the latter needs either calculation of an operator exponent or an operator diagonalization. Besides, the knowledge of the poles and residues of the rational structure [12] and Eqs. (3) can be used to evaluate the steady state once for all values of the spectral parameter, thus justifying the importance of the method for spectroscopy. At the poles $\zeta = \zeta_\rho$ the superoperator $X(\zeta)$ becomes infinite. Hence, the real values of the spectral parameter closest to the poles $\zeta \sim \text{Re}\zeta_\rho$ define the spectral peaks, i.e., the physical regions where the maximal response of the system to the driving should be expected. The imaginary parts $\text{Im}\zeta_\rho$ are responsible for the Lorentzian widths of the spectral peaks. Furthermore, Eq. (9) connects the driven and non-driven Green functions and their spectral expansions (12), (13). In fact, Eqs. (5), (11) imply that the superoperators $1 + X(\zeta)$, $1 - \lambda_0(\zeta)$ are inverse to each other and so $X(\zeta)$, $\lambda_0(\zeta)$ commute for all $\zeta$ and are diagonalized in the same basis. Eq. (11) admits the formal expansion (convergent for $\lambda_0(\zeta)$ close to nilpotent)

$$ \rho = [1 + \lambda_0(\zeta) + \lambda_0^2(\zeta) + \ldots] \rho_{\text{th}}, $$

(17)

featuring the zero, linear, quadratic, etc, responses of the steady state to the driving. Since the superoperator $X_0(\zeta)$ satisfies its own characteristic equation, $\pi_0(X_0(\zeta)) = 0$, Eq. (11) has the exact solution

$$ \rho = \bar{\pi}(X_0(\zeta)) \rho_{\text{th}}, \quad \bar{\pi}(x) = \frac{\pi_0(1) - \pi_0(x)}{\pi_0(1) - 1} $$

where $\pi_0(x)$ is the characteristic polynomial of $X_0(\zeta)$. The coefficients of $\pi_0(x)$ are functions of $\zeta$ and the equation $\pi_0(1) = 0$ defines the poles $\zeta = \zeta_\rho$ of $G(\zeta)$. The generalized Fourier transform (15) and Eq. (9) connect the non-driven dynamics at $\zeta = 0$ with the driven steady state for all values of $\zeta$.

**Application to ensemble of 2-level systems.** We now illustrate the method of spectral Green functions by its application to a driven dissipative ensemble of correlated 2-level quantum systems — the generic model system to study collective phenomena in quantum optics, magnetism and quantum information [13-17].

The model Hamiltonian of Eq. (1) that we will consider is built of $N + 1$ correlated 2-level quantum systems comprising one “active” subsystem described by the spin-1/2 angular momentum $S$ and $N$ “passive” subsystems characterized by spin-1/2 angular momenta $I^{(k)}$, $1 \leq k \leq N$, featuring the non-driven and driving parts of Eq. (5)

$$ H_0 = 0, \quad H_1 = S_z, \quad P = \Omega (I_+ S_- + I_- S_+). $$

(18)

Here $I = \sum_k I^{(k)}$ is the total passive spin and $\Omega$ is the effective driving strength. The Hamiltonian is invariant to permutations of the passive subsystems, so the latter are identical and it is sufficient to represent the passive part by a single angular momentum $I$ with the spin quantum number $I = N/2$, similar to the Dicke
model [24, 28]. The occupation numbers are defined as $n = (n_+ - n_-) / 2 = -I, -I + 1, \ldots, I$ where $n_\pm$ are the numbers of passive subsystems in the excited/ground state. We have then

$$I_z = \sum_{n=-I}^{I} n|n\rangle\langle n|, \quad I_+ = \sum_{n=-I}^{I} \sqrt{\lambda_n} |n\rangle \langle n-1|, \quad I_- = I_+^\dagger, \quad \lambda_n = (I - n + 1)(I + n).$$

(19)

The Hamiltonian belongs to the subspace $\Lambda_0$ of zero-quantum coherences, $[I_z + S_z, H] = 0$, so the dynamics is closed in $\Lambda_0$. The density operator has the representation

$$\rho = \rho_0 + \rho_\perp S_z + \rho_+ S_+ + \rho_- S_-,$$

$$[I_z, \rho_{0,z}] = 0, \quad [I_z, \rho_{\pm}] = \pm \rho_{\pm}$$

with $\rho_{0,\pm}$ containing the passive spin components only. To describe the model dissipator of Eqs. (1), (2), we approximate the thermal equilibrium density operator as the zero-temperature Boltzmann distribution of the active subsystem $\rho_{0,z} = [2(N + 1)]^{-1} (1 - 2S_z)$, so that the active subsystem is in the ground state while the passive subsystems have all equally populated levels. The dissipation is built of separate active and passive parts $D = D_S + D_I$ satisfying the simplified conditions

$$D_S(1/2) = D_S S_z = -\Gamma_1 S_z, \quad D_S S_\pm = -\Gamma_2 S_\pm,$$

$$D_I(1) = 0, \quad D_I \rho_0 = -\gamma_1 \rho_0, \quad D_I \rho_\pm = -\gamma_2 \rho_\pm$$

for all traceless zero-quantum $\rho_0$ and single-quantum $\rho_\pm$ operators built of the passive spin components. Here $\Gamma_1, \Gamma_2, \gamma_1, \gamma_2 > 0$ are the effective active and passive longitudinal and transverse relaxation rates.

The chosen model has two important applications. In quantum optics, the active subsystem describes the pumped (solid atomic or molecular) gain medium while the passive ensemble plays the role of the population inverted amplifier [18, 19]. In high field solid state dynamic nuclear polarization, the active subsystem is formed by a microwave irradiated unpaired electron spin (of a free radical or paramagnetic ion) while the passive subsystems belong to hyperpolarized paramagnetic nuclear spins [20, 21]. Here initially the driving is caused by a time-periodic (optical or microwave) excitation and then the rotating wave approximation and an effective Hamiltonian must be applied by a suitable transformation [22, 23].

Multiplying both sides by $1 + \lambda_0(\zeta)$, Eq. (11) can be rewritten as $[1 - \lambda_0^2(\zeta)] \rho = [1 + \lambda_0(\zeta)] \rho_0$. The superoperator $\lambda_0^2(\zeta)$ is closed in the subspace $\Lambda_0^{(I)}$ of zero-quantum coherences of the passive ensemble, while $\lambda_0(\zeta)$ maps this subspace to the subspace of single-quantum passive coherences. Since $\rho_{th} \in \Lambda_0^{(I)}$, the projection $\rho^0$ of the steady state to $\Lambda_0^{(I)}$ satisfies the equation

$$[1 - \lambda_0^2(\zeta)] \rho^0 = \rho_{th}. \quad \text{By Eq. (14), the non-driven spectral Green function is diagonalized as}$$

$$G_0(\zeta) \rho_{th} = \frac{-\rho_\perp S_\perp}{\Gamma_2^2 + i\zeta}, \quad \Gamma_2 = \Gamma_2 + \gamma_2,$$

$$G_0(\zeta) S_z = \frac{-S_z}{\Gamma_1}, \quad G_0(\zeta) (\rho_{th} S_z) = \frac{-\rho_\perp S_\perp}{\Gamma_1 + \gamma_1},$$

$$G_0(\zeta) \rho_0 \left( \frac{1}{2} - S_z \right) = \frac{-\rho_0 (1/2 - S_z)}{\gamma_1}.$$ 

Under the condition $\Gamma_1 \gg \gamma_1$, we obtain then

$$\lambda^2_0(\zeta) \rho_0 \left( \frac{1}{2} - S_z \right) = \frac{2\Omega^2 \Gamma_2 L_-(\xi) \rho_0}{\gamma_1 (\Gamma_2^2 + \zeta^2)} \left( \frac{1}{2} - S_z \right).$$

The form of $\rho_{th}$ implies $\rho^0 = \rho_0 (1/2 - S_z)$ where $\rho_0$ satisfies the equation

$$[1 - \chi(\zeta) L(\zeta)] \rho_0 = \frac{1}{N + 1}, \quad \chi(\zeta) = \frac{2\Omega^2 \Gamma_2}{\gamma_1 (\Gamma_2^2 + \zeta^2)}, \quad \zeta = \frac{1}{\gamma_1} (N + 1), \quad \chi_n(\zeta) = \lambda_n \chi(\zeta), \quad n = I, I - 1, \ldots, -I + 1.$$ 

Using the fact that $\rho_0$ is diagonal in the basis of the occupation numbers, $\rho_0 = \sum_n c_n |n\rangle \langle n|$, based on Eqs. (2), (21), we come to the following recurrency for the coefficients $c_n$ that can easily be resolved numerically

$$c_I = [1 + \chi(\zeta)]^{-1} (N + 1)^{-1},$$

$$c_{n-1} = [1 + \chi_{n-1}(\zeta)]^{-1} \left( [(N + 1)^{-1} + \chi_n(\zeta) c_n] \right), \quad \chi_n(\zeta) = \lambda_n \chi(\zeta), \quad n = I, I - 1, \ldots, -I + 1.$$ 

Eq. (21) shows that the poles of the driven Green function $G(\zeta)$ are given by the equations $1 + \lambda_n \chi(\zeta) = 0$ leading to the expressions

$$\zeta_n = \pm i \Gamma_2 \sqrt{1 + \frac{2\Omega^2 \lambda_n}{\gamma_1 \Gamma_2}}, \quad n = -I + 1, \ldots, I.$$ 

(22)
featuring a combination of Lorentzian peaks at the same location \( \text{Re} \, \zeta = 0 \) with different half-widths \( |\text{Im} \, \zeta_n| \).

Eqs. [21] can be used to calculate two important steady-state characteristics of the driven open quantum system, the polarization and self-correlation of the total \( z \)-component of the passive spin \( \langle I_z \rangle = \sum_n n c_n, \langle I_z^2 \rangle = \sum_n n^2 c_n \). Numerical results for a set of system parameters are plotted in FIG. 1b. It is evident that at \( \zeta \sim 0 \) the passive ensemble is almost fully polarized (population inverted) with \( \langle I_z \rangle \sim -N/2 \) that is accompanied with creation of correlations between the passive subsystems. For the parameters independent of the number of passive subsystems, the width of the “absorption line” grows with \( N \) in accordance with Eq. \( \Box \). Proceeding now to an ensemble of many active subsystems, each “serving” \( N \) passive subsystems, we can assume that the transverse relaxation rate of the active ensemble is proportional to the square of its relative concentration, \( \Gamma_2 = \Gamma_{2c}^2 \). For \( \Gamma_2 \gg \gamma_2 \), the simulation by Eqs. [21] implies then that the total peak polarization of the passive ensemble \( \xi \langle I_z \rangle \) at \( \zeta = 0 \) has an active concentration optimum whose both location and peak value increase with \( N \), FIG. 1b. Note finally that the method of spectral Green functions is exact (within the approximation \( \gamma_1/T_1 \ll 1 \)) and computationally efficient up to \( N \sim 10^9 \). The direct simulation in terms of the initial master equation is either impossible due to the lack of computational memory or takes a dramatically longer computational time.

**Conclusion and acknowledgement.** We have proposed a novel method of simulation of driven Markovian open quantum dynamics based on Green functions in spectral frequency domain. We demonstrated that the method is computationally highly efficient and opens up new ways in simulation, spectroscopy and optimization of many-body quantum dynamics in realistically large-scale limit. This work was funded by the British Engineering and Physical Science Research Council (EPSRC) through grant EP/N03404X/1 to WK.

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