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CHARACTERISTICS OF QUANTUM OPEN SYSTEMS: FREE RANDOM VARIABLES APPROACH*

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Random Matrix Theory provides an interesting tool for modelling a number of phenomena where noises (fluctuations) play a prominent role. Various applications range from the theory of mesoscopic systems in nuclear and atomic physics to biophysical models, like Hopfield-type models of neural networks and protein folding. Random Matrix Theory is also used to study dissipative systems with broken time-reversal invariance providing a setup for analysis of dynamic processes in condensed, disordered media. In the paper we use the Random Matrix Theory (RMT) within the formalism of Free Random Variables (*alias* Blue's functions), which allows to characterize spectral properties of non-Hermitean "Hamiltonians". The relevance of using the Blue's function method is discussed in connection with application of non-Hermitean operators in various problems of physical chemistry.

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1. Introduction

Non-Hermitean operators are related to studies of damping or decay processes [1–5] although microscopic treatment of effectively dissipative systems involves always Hermitean Hamiltonians. It is often convenient, however, to

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give a "reduced description" of dissipative systems [1, 6, 7] by using generalized, non-Hermitean "Hamiltonians". Among possible applications of the procedure, the three are of special importance:

- Evolution of incomplete state vectors. A subset of basis vectors may have a special significance with a truncated part of the complete wave function being of special interest. The decomposition of the complete wave function, through say, Löwdin-Feshbach partition [8, 9] scheme leads to an effective non-Hermitean Hamiltonian and may be suggested by the structure of the original Hamiltonian. As an example, excited atoms, decaying to their ground states through spontaneous radiation can be described in this way. Similar approach is also valid to study long-lived resonances in the continuum of nuclei, atoms and molecules [4, 10]. The original unperturbed Hamiltonian in all aforementioned problems is assumed to have then a discrete and a continuum part in its spectrum and the discrete part furnishes the truncated Hilbert space of interest.

- Master Equation approach to dissipative quantum dynamics [1,2] with its natural applications to study irreversible chemical kinetics. Non-Hermitean operators in this approach are obtained by performing adiabatic elimination of "heat bath" from the underlying microscopic description. The models of that type cover *eg.* Bloch theory of spin relaxation [11] and Redfield theory [12] of molecular spectra.

- Scattering problems, for which the resonance structure in the energy dependence of scattering cross sections is related to poles of the scattering matrix in the complex energy plane [3, 5]. Feshbach optical model provides representation of the scattering matrix in which complex poles appear as the eigenvalues of some non-Hermitean Hamiltonian H.

2. Random matrix approach

Statistical properties of complex systems can be successfully investigated within the framework of the random-matrix theory [13] which turned out to be quite general and a powerful phenomenological approach to a description of various phenomena such as quantum chaos [14], complex nuclei [4], chaotic scattering [5] and mesoscopic physics [2]. Aspects of vastly different physical situations such as electron localization phenomena in disordered conductors and semiconductors [15], disordered quantum wires [16] and quantum Hall effect [17] can be described in the language of the random matrix theory. Variants of the Anderson model [18] are also used in neural networks models [19] and models of protein folding [20]. In all the realms mentioned above,

the Hamiltonian of the system is rather intricate to be handled or simply unknown. In such cases the integration of the exact equations is replaced by the study of the joint distribution function of the matrix elements of the Hamiltonian $P(\boldsymbol{H})$. If there is no preferential basis in the space of matrix elements, so that the system is as random as possible and equal weights are given to all kinds of interactions, one has to require the probability $P(\boldsymbol{H})d\boldsymbol{H}$ to be invariant under similarity transformations $\boldsymbol{H} \to R^{-1}\boldsymbol{H}R$ with R being orthogonal, unitary or a symplectic $N \times N$ matrix reflecting the fundamental symmetry of the underlying Hamiltonian.

3. Free random variables (FRM) and Blue's functions

Free random variables were introduced in a seminal paper by Voiculescu [21] in 1991. Since those days, they were promoted in physics with a pedagogical overview, recently published by Zee [22]. In this last approach [22,23] Zee has reformulated the Voiculescu idea in terms of the "Feynman diagrams" in the large N expansion, where N is the size of the matrix. The main idea of Voiculescu was to find an elegant way of "linearizing" the process of determining the average eigenvalue distribution for the convolution of non-commuting operators, hence finding an analogue of the logarithm of Fourier transformation for the usual convolutions. As an example let us consider two Hermitean operators in matrix representation, O_1 and O_2, populated from the statistical ensemble $P(O_1, O_2)$, provided that in the large N limit $P(O_1, O_2)$ factorizes into $P_1(O_1) \cdot P_2(O_2)$ (so-called *asymptotic freeness*[1]). Then resolvent

$$G(z) := \int dO_1 dO_2 P(O_1, O_2) \mathrm{Tr} \frac{1}{z - O_1 - O_2} \tag{1}$$

could be expressed (at least *a priori*) knowing only the individual resolvents

$$G_i(z) := \int dO_i \, P_i(O_i) \mathrm{Tr} \frac{1}{z - O_i} \quad \mathrm{i} = 1, 2 \,. \tag{2}$$

The working algorithm is:

- We construct *Blue's Functions* B_1 and B_2, as *functional inverses of the Green's functions* G_1 and G_2, *i.e.* $B_1[G_1(z)] = z$ and $B_2[G_2(z)] = z$.
- The Blue's $B(z)$ function for the sum comes from the *"additivity law"* $B(z) = B_1(z) + B_2(z) - \frac{1}{z}$.
- The desired resolvent (1) comes as a functional inverse of the Blue's function $B(z)$, and its average spectrum is given by the discontinuities in the analytical plane.

[1] For more precise definition of the "freeness" property, see [21]

3.1. Example

In the case of the Random Gaussian Ensembles, denoted by O_R, the resolvent (Green's function) is known as $G_R(z) = \frac{1}{2}(z - \sqrt{z^2-4})$, and it's imaginary part reproduces the seminal Wigner's semicircle law for the eigenvalues. The Blue's function for this ensemble is even simpler,

$$B_R(z) = z + 1/z\,. \tag{3}$$

It is easy to check, that "addition" of the deterministic operator O_D to the random one still fulfills the asymptotic freeness. Since the deterministic resolvent reads $G_O(z) = \sum_i \frac{1}{z-E_i}$, the addition law of the Blue's functions yields immediately the resolvent for the "*deterministic plus random Gaussian*", known also as a *Pastur equation*

$$G(z) = G_D(z - G(z))\,. \tag{4}$$

For example, in case when the deterministic Green's function corresponds to two-levels ($E_i = \pm E$), this equation is cubic

$$G^3(z) - 2zG^2(z) + (z^2 - E^2 + 1)G(z) - z = 0\,. \tag{5}$$

3.2. Other applications of Blue's functions

Differentiation of the Blue's function gives the positions of the end-points of the spectra and yields critical exponents [22,23]. *Integration* of the Blue's functions could show the *cusps* in "thermodynamic" quantities like pressure in Random Matrix Models (RMM)), thus allows to study singularities of the partition function and phase diagrams [24] of the RMM.

The generalization to the *non-Hermitean* case has been shown to require ([24, 25]) an introduction of 2×2 *matrix-valued generalized Blue's functions*. Solutions to this matrix-valued analogon of Blue's functions (with z replaced by matrix $\mathcal{Z} = \mathrm{diag}(z, \bar{z})$) belong generically to the two classes: (1) holomorphically separable (trivial) class, when we recover the holomorphic (analytical) Blue's function $B(z)$ accompanied by it's totally decoupled anti-holomorphic copy $\bar{B}(\bar{z})$; (2) non-holomorphic class, corresponding to the case when, despite the resolvent naively is the function of the z variable only, the support of the spectrum depends on both z and $\bar{z}$, forming therefore the two-dimensional islands, not one-dimensional cuts, on the complex plane (*cf.* also discussion in [2, 26]).

As a preliminary study of applications of the Blue's function formalism, we have solved the problem of the effective two-level system coupled to a noise reservoir [30].

4. Dissipative two-level system

We have considered the Hamiltonian with the Hilbert space spanned by N discrete states $|k\rangle$ and M continua $|\chi_n(E)\rangle$:

$$\begin{aligned}\mathcal{H} \;=\; & \sum_{k=1}^{N} E_k|\psi_k\rangle\langle\psi_k| + \sum_{n=1}^{M}\int dE E|\chi_n\rangle\langle\chi_n| \\ & +\gamma^{1/2}\sum_{n=1}^{M}\sum_{k=1}^{N}\int dE(V_k^n(E)|\psi_k\rangle\langle\chi_n| + h.c.)\,. \end{aligned} \tag{6}$$

The bound continuum coupling is characterized by the coupling constant γ and the energy dependent matrix $V_k^n(E)$. Note that in this model there is no direct coupling (interactions) between continuum channels n, n'. If V depends only weakly on the energy, the effective Hamiltonian H_{eff} takes the form

$$H_{\text{eff}} = \mathcal{H} - i\gamma VV^{\dagger}\,, \tag{7}$$

where $\mathcal{H}$ is $N \times N$ deterministic Hamiltonian with the bound-state energies $\varepsilon_1, ..., \varepsilon_N$ and the dissipative part is obtained via the usual Redfield equations [11, 12] in the so-called Markovian limit. Loosely understood, the Markovian property assumes exponential relaxation of response of the continuum when it acts on the test system and would correspond to the high temperature Debye relaxation bath. The operators $V_k^n = \langle\psi_k|V|\chi_n\rangle$ stand for the coupling of the bound-state eigenvectors ψ_k $(k = 1, \ldots, N)$ corresponding to eigenvalues $\varepsilon_1 = ... = \varepsilon_{N/2} \equiv \varepsilon$ and $\varepsilon_2 = ...+\varepsilon_{N/2} \equiv -\varepsilon$ with the continuum states χ_n $(n = 1, \ldots, M)$. Non-Hermiticity of the term $i\gamma VV^{\dagger}$ accounts for the depletion of the bound state subspace due to the leakage of the probability mass into the continua. The eigenvalues $\varepsilon = x - iy$ of the effective Hamiltonian H_{eff} are in general complex, their imaginary parts must be positive for damped systems and give the lifetimes of the corresponding eigenvalues as $1/|y|$.

The model Hamiltonian of that type (7) is widely used in quantum chaotic scattering problems [5] but has been also introduced to study unimolecular dissociation of selectively excited polyatomic molecules [28, 29]. For most dissociating molecules above the tunneling regime (when the excitation energy is well above the dissociation threshold) the coupling between the molecular complex and the continuum is strong and the density of strongly mixed molecular states is large, so that the unimolecular decay is characterized by overlapping resonance states.

We have solved the system in the regime when both M and N are large, but $m = M/N$ is fixed (which implies strongly overlapping resonances). To

simulate the effect of the "noise" represented by statistical properties of the operators $V, V^\dagger$ we have assumed that the N by M matrices V are random. For simplicity Gaussian randomness has been chosen. By use of the central limit theorem, Gaussianity is a legitimate choice for the infinite normalized sum of independent fluctuating contributions. It can be shown also that the distribution for normalized sum matrix, each sampled from a distribution follows the Gauss theorem of probability [22, 23].

To apply the addition law, we need only the functional form of the corresponding Blue's functions. For the deterministic part, the Green function is by definition

$$\mathcal{G}_D(\mathcal{Z}) = \frac{1}{2}(\frac{1}{\mathcal{Z}-\varepsilon} + \frac{1}{\mathcal{Z}+\varepsilon})\,, \tag{8}$$

therefore the Blue's function is given by the equation

$$\mathcal{Z} = \frac{1}{2}\left((\mathcal{B}_D(\mathcal{Z})-\varepsilon)^{-1} + (\mathcal{B}_D(\mathcal{Z})+\varepsilon)^{-1}\right)\,. \tag{9}$$

The Blue's function for the noise term could be determined by the projecting techniques [24] and reads

$$\mathcal{B}_N = m(1-\Gamma\mathcal{Z})^{-1}\Gamma + \frac{1}{\mathcal{Z}}\,, \tag{10}$$

here Γ is a matrix

$$\Gamma = \begin{pmatrix} -i\gamma & 0 \\ 0 & i\gamma \end{pmatrix}\,. \tag{11}$$

We could use the generalized addition formula for Blue's functions, resulting in

$$\mathcal{B}(\mathcal{Z}) = \mathcal{B}_D(\mathcal{Z}) + m(1-\Gamma\mathcal{Z})^{-1}\Gamma\,, \tag{12}$$

and by replacing in (12) $\mathcal{Z} \to \mathcal{G}$ and using the definition of the Blue's function we get

$$\mathcal{Z} = \mathcal{B}_D(\mathcal{G}) + m(1-\Gamma\mathcal{G})^{-1}\Gamma\,. \tag{13}$$

By similar replacement in the equation (9) we get

$$\mathcal{G} = \frac{1}{2}\left((\mathcal{B}_D(\mathcal{G})-\varepsilon)^{-1} + (\mathcal{B}_D(\mathcal{G})+\varepsilon)^{-1}\right)\,. \tag{14}$$

Calculating $\mathcal{B}_D(\mathcal{G})$ from (13) and plugging it into (14) we arrive at the final equation for the Green function

$$\mathcal{G} = \frac{1}{2}\left((\mathcal{Z} - m(1-\Gamma\mathcal{G})^{-1}\Gamma - \varepsilon)^{-1} + (\mathcal{Z} - m(1-\Gamma\mathcal{G})^{-1}\Gamma + \varepsilon)^{-1}\right)\,. \tag{15}$$

We have solved this equation using the explicit parametrization of the matrix $\mathcal{G}$ with the unknown entries

$$\mathcal{G} = \begin{pmatrix} a & b \\ d & c \end{pmatrix} . \tag{16}$$

The relevant solution is the one when the off-diagonal element of the Green function is non-zero. In this case, the resolvent is nonanalytic, being the function of both variables $z, \overline{z}$. The support of the eigenvalues forms the two-dimensional "islands" of nonanalyticity, (in opposition to the hermitean case, when only cuts constitute the support if real eigenvalues) and the density distribution could be read-out from the "Gauss law" [26]

$$\mathrm{divE} = 4\pi\rho(z, \overline{z}) . \tag{17}$$

In this two-dimensional electrostatics divergence means differentiation with respect to $\overline{z}$, and electric field has two components, $E_x = 2\mathrm{Re}\,\mathcal{G}_{zz}$, $E_y = -2\mathrm{Im}\,\mathcal{G}_{zz}$.

The non-analytical solution is a consequence of the "spontaneous breakdown of analyticity". Naively, the resolvent

$$G(z) = \frac{1}{N}\ \mathrm{Tr}\left\langle \frac{1}{z - H} \right\rangle , \tag{18}$$

where

$$\langle \ldots \rangle = \int P(H) \ldots [dH] \tag{19}$$

is the function of z variable only. But the true ground state is not respecting holomorphic separability into z and $\bar{z}$ copies, resulting in the spontaneous breakdown of this "symmetry", by forming the mixed "condensates" $\mathcal{G}_{z\bar{z}}$ and therefore breaking the analyticity (non-analyticity) of the solution.

The system has been analyzed in terms of its spectral properties (see Fig. 1), investigating also occurrence of phase transition effects and partial trapping of the population in slow decay modes. The last phenomenon is of great importance for understanding the radiationless and multiphoton decay of polyatomic molecules.

Our calculations predict that distribution of eigenvalues of the effective Hamiltonian

$$\varrho = \frac{1}{4\pi}\left[\frac{2}{\gamma y}\left(1 + \frac{x^2 - \varepsilon^2}{y^2}\right) + \frac{m-1}{y^2} + \frac{x^2 + \varepsilon^2}{(x^2 - \varepsilon^2)^2}\right] \tag{20}$$

shows a gap (*cf.* Fig. 1) developing at some critical value of the coupling to continua. The formation of such a gap has been noticed in quantum and classical chaotic scattering systems studied by other authors [10, 31].

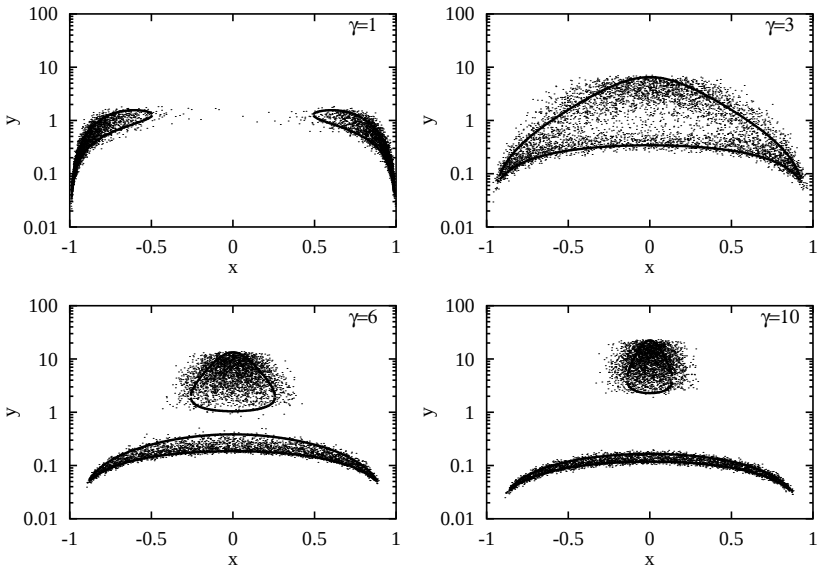

Fig. 1. The support of the eigenvalues of the two-state system for different coupling at fixed filling ratio $m = 0.25$. The dots correspond to the numerical simulation of the effective Hamiltonian, while the solid lines come from the analytical result [30]. Numerical simulations have been performed for 100 matrices of size $N = 100$ at coupling constants $\gamma = 1, \gamma = 3$ (the upper row of drawings), and for 30 matrices with $N = 400$ at $\gamma = 6$ and $\gamma = 10$ (the lower row). The density of the dots is related to the spectral density $\varrho(\varepsilon)$ evaluated in [30] by use of the Blue's function method (see the text). By functionally inverting the Blue's function relevant for that problem, we get the desired resolvent $\mathcal{G}$ from which the density of states $\varrho(x, y)$ follows according to Eq. (17).

The main achievement of using the Blue's function in that problem was fully analytical derivation of the formula for the generalized, nonholomorphic Green function that predicts spectral properties of the system through a non-perturbative approach. Our analytical results have been perfectly reproduced by numerical calculations (see Fig. 1) on samples of matrices. Our analysis has shown that:

- depending on the intensity of the coupling to the noise term, the system exhibits phase transitions which are observable through the structure of the spectral density;
- there are two different phase transitions, one of which is directly related to the imposition of randomness on otherwise deterministic structure of the effective 2-level Hamiltonian;

- due to inherent decay of the system through continua, the spectrum undergoes characteristic "collectivization" [10] induced by partial trapping of accessible states;
- for increasing values of the coupling constant, the system relaxation properties become distinctly non-exponential.

The last statement comes from the analysis of the integrated distribution $\varrho(y)$. By integrating $\rho(x,y)$ over the support of x we arrive at the formula representing distribution of "resonance widths". Using the symmetry of the spectra we arrive at

$$\varrho(y) = \int\limits_{\text{support}} dx\ \varrho(x,y) = R(x_u,y) - R(x_l,y)\,, \tag{21}$$

$$R(x,y) = \frac{1}{2\pi}\left\{\frac{2x}{\gamma y} + \frac{2x}{\gamma y^3}\left(\frac{x^2}{3} - \varepsilon^2\right) - \frac{(1-m)x}{y^2} + \frac{x}{\varepsilon^2 - x^2}\right\},$$

where x_u and x_l are the two positive end points of the support (for a continuous support $x_l = 0$). The resulting density can be used to determine average decay rate in the system. The bound state component $|\psi\rangle$ evolves in time according to

$$|\psi(t)\rangle = e^{-iH_{\text{eff}}t/\hbar}|\psi(0)\rangle\,, \tag{22}$$

and its effective decay rate can be defined as the logarithm derivative of its norm

$$k_{\text{eff}} = -\frac{d}{dt}\ln\langle\psi(t)|\psi(t)\rangle\,. \tag{23}$$

If the initial state has a nonzero overlap with only one eigenstate of H_{eff}, the above definition simplifies to $k_{\text{eff}} = 2y_i/\hbar$.

$$P(t) = \frac{1}{N}\langle \text{Tr}\{\exp(iH_{\text{eff}}^{\dagger}t)\exp(-iH_{\text{eff}}t)\}\rangle\,, \tag{24}$$

and *via* the obvious relation between the Green's function and the time-evolution operator, can be represented in the form of the Fourier integral [32]

$$P(t) = \frac{1}{4\pi^2 N}\int\limits_{-\infty}^{\infty} d\varepsilon e^{-i\varepsilon t}\int\limits_{-\infty}^{\infty} dz\langle \text{Tr}\mathcal{G}(z+\frac{1}{2}\varepsilon)\mathcal{G}^{\dagger}(z-\frac{1}{2}\varepsilon)\rangle\,. \tag{25}$$

In general case when resonances overlap, the average survival probability over the ensemble can be extracted from the diagonal part of $P(t)$

$$P_d(t) = \int\limits_0^{\infty}\varrho(y)\exp(-yt)dy\,, \tag{26}$$

and is related to the distribution of kinetic rates $\varrho(y)$ by the Laplace transform.

$$\varrho(y) = (2\pi)^{-1} \int_S P_d(t) \exp(yt) dt \,, \tag{27}$$

where S represents the path of integration in the complex plane. The effective kinetic rate constant reads

$$k_{\text{eff}} = -\frac{d[lnP_d(t)]}{dt} \,. \tag{28}$$

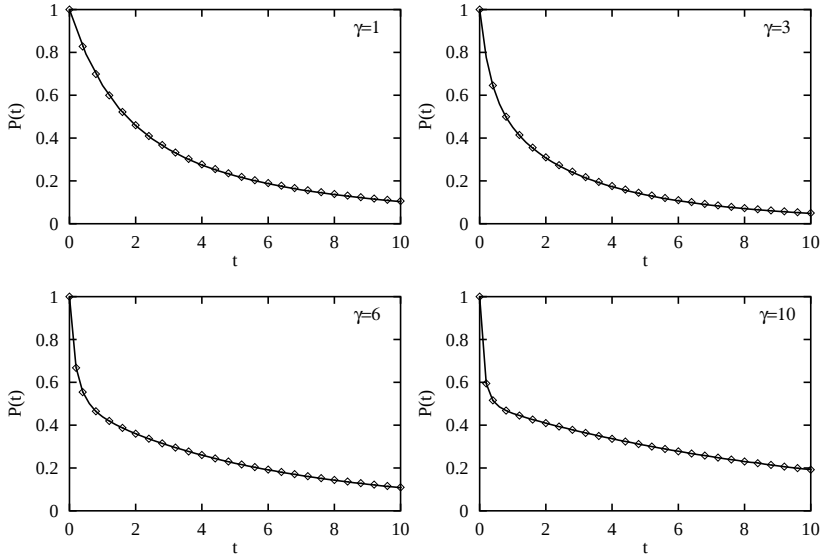

Fig. 2. The decay of the survival probability (26) for various values of the coupling constant γ. Nonexponential decay becomes transparent for increasing values of γ. The solid line is the Laplace transform of the analytical width distribution (22). The dots correspond to the Laplace transform of the density obtained numerically with a set of 30 random matrices of size $N = 400$.

The result is displayed in Fig. 2 showing the decay with some average decay rate for low couplings and the appearance of roughly two distinct decay rates for large couplings. The time scale is of the order of a femtosecond for $\varepsilon = 1eV$, and scales linearly with energy. (Note, that within the paper the mean-square dispersion of elements of V matrix has been put equal 1.) A similar analysis of the rate constant for the unimolecular reaction in the regime of overlapping resonances has been proposed [28] indicating possible use od Feshbach optical model in analysis of state-selected spectra of

molecular dissociation. It has been argued that in the overlapping regime, the observed linewidths are not related in a simple fashion either to the unimolecular decay rate or to the resonance widths.

5. Conclusions

Non-hermitean operators appear in a broad class of problems in statistical physics describing diffusion and growth analyzed in terms of the stochastic Master equation approach [33]. The statistical properties of random asymmetric matrices may be also important in the understanding of the behaviour of certain dynamical systems far from equilibrium. One possible example is the dynamics o f neural networks [34], where the coupling between the neurons given by a synaptic matrix J_{ij} is, in general, asymmetric and influenced by a substantial degree of disorder. In the paper we have presented analysis of a two-level system coupled to a random source. The models of similar type serve as a paradigm in the theory of transport properties in condensed matter [35]. Instead of using the standard language of the Green's function formalism, we have chosen to work with the functional inverse of the Green's function ("Blue's function"). The method stems from the powerful mathematical concept of free random variables [21] which has been shown to be an elegant tool in various applications of the random matrix theory [22–25]. The simplicity of the method makes it worth pursuing in studies of the vast family of nonhermitean models applied in spectroscopy, theory of condensed phase chemical reaction or biological physics.

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