Linear and Non-Linear Optics of Lindbladian Systems

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Abstract

Non-equilibrium quantum systems can exhibit fundamentally different qualitative behavior than their equilibrium counterparts. Here we leverage Keldysh Green's functions to develop a linear and non-linear response formalism to probe the properties of open quantum systems subject to dissipative evolution by Lindbladian superoperators. We present results for fermionic systems and place a specific emphasis on optical conductivity and the example of Bernal bilayer graphene. Contrary to Hermitian systems:

1. The diamagnetic response may be real and exceptional points manifest themselves in the $\boldsymbol{k}$-resolved diamagnetic response
2. The paramagnetic response has a similar magnitude to the equilibrium system but does not manifest universal Dirac behavior
3. Asymmetric dissipation can enable a second order response in a centrosymmetric system

Our Previous Works

Lindbladians exhibit rich band structure phenomenology including a novel mechanism to realize flat bands as discussed in (PRB 106, L161109 (2022)), and large angle structures such as Bernal bilayer graphene and its generalizations exhibit extensively tunable low energy optical responses (PRB 107, L041408 (2023)) and unusual circular dichroic behavior (arXiv:2305.14472 (2023)).

Formalism

Lindbladian Systems

Lindbladians are the class of (non-Hermitian) time evolution super-operators, $\mathcal{L}[\rho] = i\dot{\rho}$, of the density matrix that are completely positive trace preserving (CPTP) and Markovian [1]. Explicitly,

$$\mathcal{L}[\rho] = [H,\rho] - \frac{i}{2}\sum_m \Gamma_m\left(\{J_m^\dagger J_m, \rho\} - 2J_m\rho J_m^\dagger\right)$$

which governs the evolution of a subsystem ρ with jump operators J_m that change the particle number and energy of the subsystem. J_m are normalized and Γ_m sets the rate of jumps. For linear jump operators

$$J_m(\boldsymbol{k}) = \sum_\alpha a_{m,\alpha}(\boldsymbol{k})c_{\boldsymbol{k},\alpha} + b_{m,\alpha}(\boldsymbol{k})c^\dagger_{-\boldsymbol{k},\alpha}$$

We can interpret the jump operators as measurement operators in the limit of continuous measurement by a memoryless observer. Physically this could correspond to (1) qubits in a quantum circuit subject to continuous measurements, (2) photons in a leaky cavity subject to jumps to and from free space modes, or (3) a 1D/2D material with jumps to and from a substrate.

Complex Fermion Representation

For Lindbladians that are quadratic in fermions, we can extend Prosen's "third-quantization" algebra [2] to place different parity sectors on equal footing. Doing so enables a concise matrix representation of the Lindbladian in terms of left- and right-contour super-fermions whose eigenfunctions are the normal modes of the full Lindbladian [3]. It reads

$$\hat{\mathcal{L}} = \boldsymbol{\Psi}^\dagger[L_{\text{coh}}(\boldsymbol{k}) + L_{\text{dis}}(\boldsymbol{k})]\boldsymbol{\Psi}$$

with the basis $\boldsymbol{\Psi} = (\boldsymbol{\ell}, \boldsymbol{r})$, where the left and right super-fermions are given by $\ell_{\boldsymbol{k},\alpha}\rho = c_{\boldsymbol{k},\alpha}\rho\mathcal{P}$ and $r_{\boldsymbol{k},\alpha}\rho = \rho c^\dagger_{\boldsymbol{k},\alpha}\mathcal{P}$ respectively for fermion parity $\mathcal{P}$.

The single particle matrix forms are (suppressing $\boldsymbol{k}$)

$$L_{\text{coh}} = \text{diag}(H, H)$$

$$L_{\text{dis}} = -\frac{i}{2}\sum_m \Gamma_m \begin{pmatrix} A_m - B_m & -2B_m \\ -2A_m & B_m - A_m \end{pmatrix}$$

Where the blocks are (outer product with α, β)

$$A_m = a^*_{m\alpha}a_{m\beta}, \quad B_m = b_{m\alpha}b^*_{m\beta}$$

Once formulated as a path integral, this description has a redundancy that can be removed using the Larkin-Ovchinnikov rotation to find $\widetilde{L}_{\text{coh}} = L_{\text{coh}}$ and

$$\widetilde{L}_{\text{dis}} = -\frac{i}{2}\sum_m \Gamma_m \begin{pmatrix} A_m + B_m & 2(A_m - B_m) \\ 0 & -(A_m + B_m) \end{pmatrix}$$

in terms of the modified fields $\widetilde{\boldsymbol{\Psi}} = (\boldsymbol{\psi}_1, \boldsymbol{\psi}_2)$.

Keldysh Green's Functions

We can obtain the generating functional $Z = e^{iS}$ as a path integral, where the Keldysh action is given by [4]

$$S = \int_{-\infty}^{\infty} dt \int_{BZ} \frac{d^D\boldsymbol{k}}{(2\pi)^D} \begin{pmatrix} \bar{\psi}_1 \\ \bar{\psi}_2 \end{pmatrix} \begin{pmatrix} G^R & G^K \\ 0 & G^A \end{pmatrix}^{-1} \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix}$$

Where the Green's function block is

$$G^{-1} = i\partial_t \mathbf{1} - (\widetilde{L}_{\text{coh}}(\boldsymbol{k}) + \widetilde{L}_{\text{dis}})$$

From which,

$$G^R = [i\partial_t\mathbf{1} - \Xi]^{-1} \qquad \Xi = H - \frac{i}{2}\sum_m \Gamma_m(A_m + B_m)$$

$$G^A = [i\partial_t\mathbf{1} - \Xi^\dagger]^{-1}$$

$$G^K = 2G^R\Sigma G^A \qquad \Sigma = -\frac{i}{2}\sum_m \Gamma_m(A_m - B_m)$$

Spectral Decomp. and Occupation

The right, $\Xi|u_m\rangle = \xi_m|u_m\rangle$, and left, $\langle\bar{u}_m|\Xi = \xi_m\langle\bar{u}_m|$ eigenvectors can decompose the Green's function as

$$G^R = \sum_m \frac{|u_m\rangle\langle\bar{u}_m|}{\omega - \xi_m}$$

and

$$G^K = 2\sum_{m,m'} \frac{\langle\bar{u}_m|\Sigma|\bar{u}_{m'}\rangle}{(\omega - \xi_m)(\omega - \xi^*_{m'})}|u_m\rangle\langle u_{m'}|$$

Where $\langle u| = (|u\rangle)^\dagger$ and $|\bar{u}\rangle = (\langle\bar{u}|)^\dagger$. Now, the equal-time inter-orbital correlation function (occupation) is

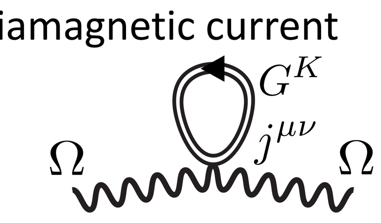
$$\langle c^\dagger_\alpha(t)c_\beta(t)\rangle = \frac{1}{2}\left[\delta_{\beta\alpha} - iG^K_{\beta\alpha}(t,t)\right]$$

Linear Optics

Diamagnetic Term

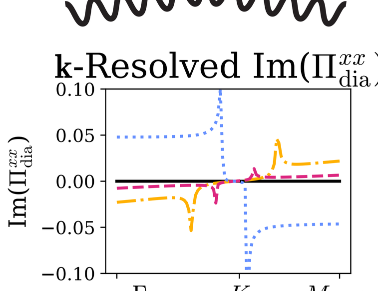
Let us consider a density response $\Pi^{\mu\nu} = -i\langle \mathsf{J}^{\mu\nu}\rangle$ corresponding to an instantaneous diamagnetic current

$$\mathsf{J}^{\mu\nu} = \sum_{\boldsymbol{k},\alpha,\beta} j^{\mu\nu}_{\alpha\beta}c^\dagger_\alpha c_\beta$$



The conductivity is a rescaling given by $\sigma^{\mu\nu} = (i/\Omega)\Pi^{\mu\nu}$.

The $\boldsymbol{k}$-space resolved diamagnetic response diverges at exceptional points and can be used to identify this non-Hermitian spectral feature.

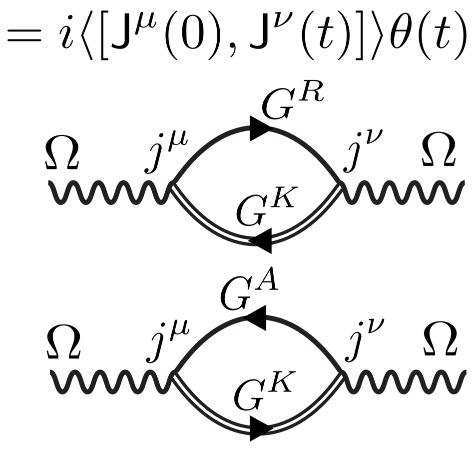


Paramagnetic Term

The paramagnetic current-current correlation function is the retarded response, $\Pi^{\mu\nu} = i\langle[\mathsf{J}^\mu(0), \mathsf{J}^\nu(t)]\rangle\theta(t)$ which can be evaluated by:

- Wick Expansion
- Reexpress as GFs
- Lehmann Representation
- Fourier Transform
- Residue Theorem

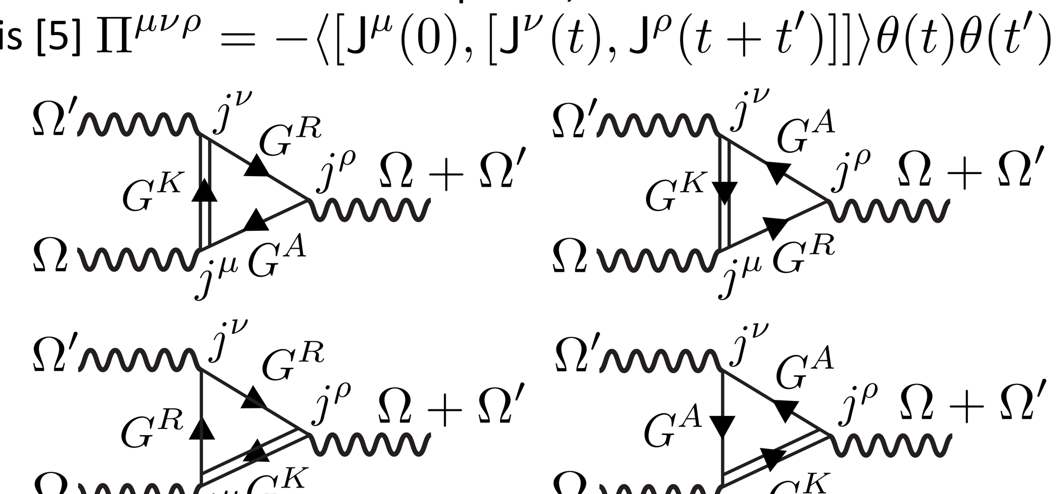
Note: $\langle\ldots\rangle = \text{Tr}[\ldots\rho_{ss}]$



Non-Linear Optics

Triangle Feynman Diagrams

For the second order response, the "new" contribution is [5] $\Pi^{\mu\nu\rho} = -\langle[\mathsf{J}^\mu(0), [\mathsf{J}^\nu(t), \mathsf{J}^\rho(t+t')]]\rangle\theta(t)\theta(t')$



Second Harmonic Generation and Shift

Second harmonic generation is given by the contraction

$$\Pi^{\mu\nu}_{\text{2HG}}(\Omega) = \Pi^{\mu\mu\nu}(\Omega, \Omega)$$

Meanwhile, shift is a non-linear DC response given by

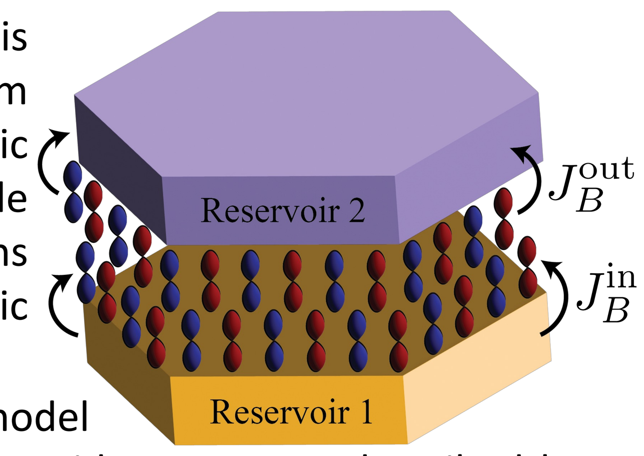
$$\Pi^{\mu\nu}_{\text{shift}}(\Omega) = [\Pi^{\mu\mu\nu}(\Omega, -\Omega) + \Pi^{\mu\mu\nu}(-\Omega, \Omega)]/2$$

For the full Π, diagrams like Π_{para} and Π_{dia} matter.

Example: Bernal Bilayer Graphene

System

Bernal bilayer graphene is a paradigmatic platform for realizing 2D electronic physics. Here we couple the bilayer to two baths and study the electronic response of the system.

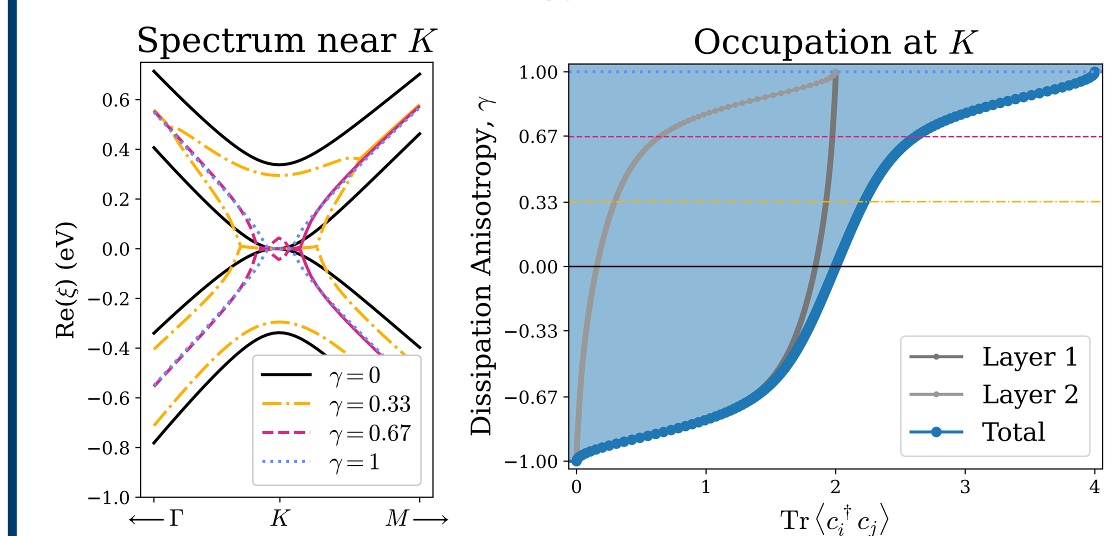


We use a tight-binding model for the bilayer [6] and consider a current described by single fermion jumps with anisotropic couplings to baths

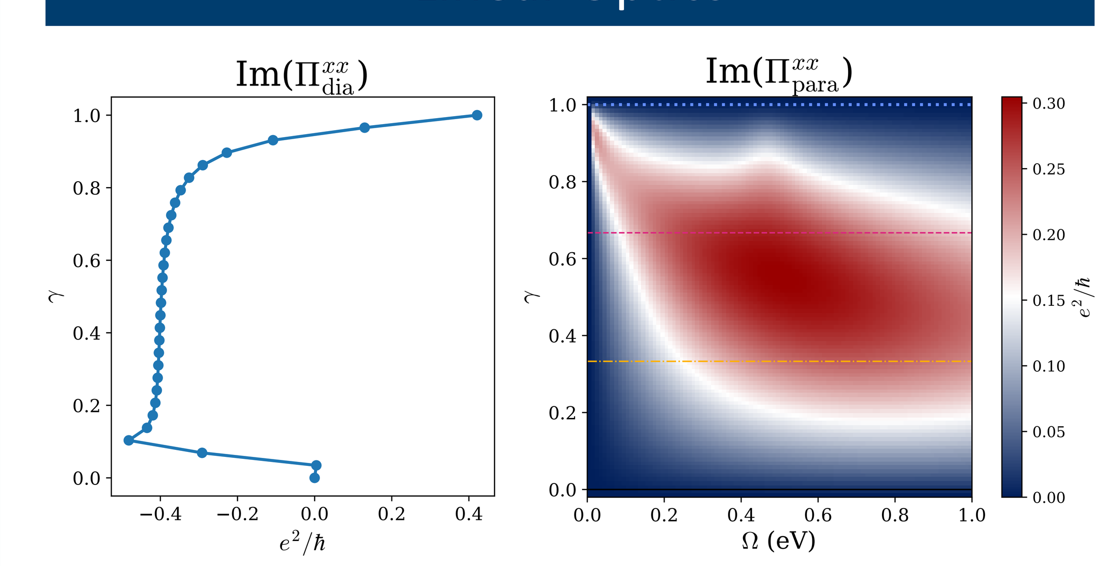
$$J^{\text{in}}_{A/B} = \Gamma(1+\gamma)c^\dagger_{1A/B}, \quad J^{\text{out}}_{A/B} = \Gamma(1-\gamma)c_{2A/B}$$

Spectrum and Occupation

We consider the low energy behavior (near K) for $\Gamma = 1$

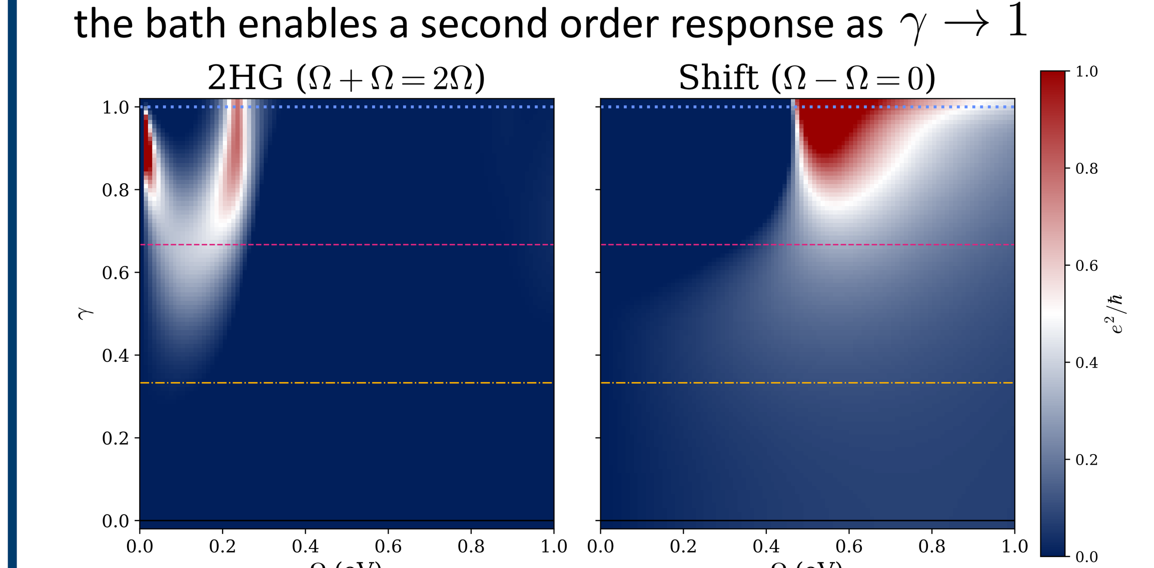


Linear Optics



Non-Linear Optics

Inversion symmetry breaking induced in the system by the bath enables a second order response as $\gamma \to 1$



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