

Applications of $O(N)$ methods to linear response coefficients

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Polynomial expansion of the spectral measures are used to generate efficient algorithms for investigating coefficients of the linear response such as non-diagonal susceptibilities, or Kubo formula for diagonal conductivity and Hall conductivity in aperiodic systems. Applications for Chebyshev polynomials of first and second order are given.

Introduction

The purpose of this work is to present alternative approaches based on real space schemes to enable in particular to compute the so-called Rudermann Kittel Kasuya Yosida interaction (RKKY)^{1,2)} an effective coupling between two localized magnetic moments, mediated by conduction electrons in many different situations, as well as quantum transport coefficients. In high dimensional non periodic systems, one usually investigates linear coefficients by way of scaling analysis through diagonalization of periodic hamiltonian. But, if N is the number of states, exact diagonalization requires a CPU time scaling as $O(N^3)$, and memory scaling as $O(N^2)$. For sparse Hamiltonians, the use of Lanczos algorithm reduces memory and CPU time requirements to $O(N)$.^{1,3)} Here, we develop quantum transport coefficients by development of Kubo formula on orthogonal polynomials. The key point of the algorithm lies in the rescaling of the density of states after evaluating the upper and lower bounds on energy. In principle, any orthogonal polynomials could be used, but it turns out that manipulations of Chebyshev polynomials are particularly recommended as they are isomorphic to Fourier series.¹⁾

Calculation of the non-local susceptibility

Indirect RKKY interaction^{2,4)} stems from the coupling between localized magnetic moments and propagating electrons. If an electron in a state of energy $E < E_F$ undergoes a transition to a state of energy $E' > E_F$ because of the coupling with the localized moment in $|r_i\rangle$, then a hole-electron pair is created and propagates coherently during a certain time τ , with $|E' - E| \leq \hbar/\tau$, until the pair is destroyed by diffusion on another magnetic impurity located in $|r_j\rangle$. Accordingly the longer is the propagation time the smallest will be the vicinity around Fermi energy that account for RKKY. The generic form of the effective coupling between two magnetic impurities mediated by itinerant electrons reads $\mathcal{I}_{RKKY}(r_i, r_j, E) = J^2 \chi(r_i, r_j, E) \mathbf{S}_{r_i} \cdot \mathbf{S}_{r_j}$ with J is the interaction between the localized moment \mathbf{S}_{r_i} and the spin of the itinerant electrons, and $\chi(r_i, r_j, E_F)$ contains the sum of all the electron-hole propagation paths from $|r_i\rangle$ to $|r_j\rangle$. RKKY is then proportional to the electronic susceptibility $\chi(r_{ij})$ of itinerant electrons. When $J > 0$ (resp. $J < 0$), the configurations of parallel spins (resp. antiparal-

lel) will minimize the energy promoting ferromagnetic state (resp. antiferromagnetic state). The susceptibility as a contribution of all the scattering paths of the hole-electron pair can be written down as

$$\chi(r_i, r_j) = -\frac{1}{2\pi} \Im \int_{-\infty}^{+\infty} dE G_+(r_i, r_j, E) G_-(r_j, r_i, E)$$

introducing the retarded (G_-) and advanced Green's functions (G_+) which define the amplitude of propagation of the hole-electron pair. Note that there exists an exact sum rule between the susceptibility and the local density of states (LDoS) $\sum_j \chi(r_i, r_j) = \rho_i(E) = \frac{2}{\pi} \Im \langle r_i | G(z) | r_i \rangle$ which is used as a numerical test.

In metallic systems with space dimension D , the interaction is given by $\mathcal{I}_{RKKY}(r, E_F) \sim A(\mathbf{r}) \cos(2k_F r + \delta(\mathbf{r}))/r^D$ which manifests a long range oscillating behavior.²⁾ For a free electron gas, the A, δ are independent of \mathbf{r} , whereas for weak disorder limit, $A(\mathbf{r})$ becomes a random but smooth function of \mathbf{r} and $\delta(\mathbf{r})$ is the phase shift associated to the scattering of electrons on impurities. $\delta(\mathbf{r})$ becomes random for $r > l_{pm}$ (mean free path). To perform real space calculations of the non-local susceptibility, considering the Green operator $G(z) = (z - \mathcal{H})^{-1} = \int_{-\infty}^{+\infty} \frac{\delta(E - \mathcal{H})}{z - E} dE$, one starts from the general form of $\chi(r_i, r_j)$:

$$2\Re \int_{\substack{E > E_F \\ E' < E_F}} dE dE' \frac{\langle r_i | \delta(E - \mathcal{H}) | r_j \rangle \langle r_j | \delta(E' - \mathcal{H}) | r_i \rangle}{E - E'}$$

The aim of the method is to determine the coefficients $\langle r_i | \delta(E - \mathcal{H}) | r_j \rangle$ without exact diagonalization, usually limited to simple models and small finite size systems. Here we use a basis of orthogonal polynomials $\{\mathcal{P}_n(E)\}_{n \in N}$ associated to a normalized function $\rho(E)$, referred as a model density of states. If the spectral subset of $\rho(E)$ contains the one of the real hamiltonian, it can be shown that $\delta(E - \mathcal{H}) = \rho(E) \sum_n \mathcal{P}_n(E) \mathcal{P}_n(\mathcal{H})$ and the $\{\mathcal{P}_n(E)\}_{n \in N}$ satisfy to the orthogonality condition

$$\int_{-\infty}^{+\infty} \rho(E) \mathcal{P}_n(E) \mathcal{P}_m(E) dE = \delta_{nm}.$$

These relations enable to write $\langle r_i | \delta(E - \mathcal{H}) | r_j \rangle = \rho(E) \sum_{n \in N} \mathcal{P}_n(E) \alpha_{ij}^n$ with $\alpha_{ij}^n = \langle r_i | \mathcal{P}_n(\mathcal{H}) | r_j \rangle$. From these expressions, the susceptibility becomes

$$\chi(r_i, r_j) = \Re \sum_{nm} \mathcal{I}_{mn} \alpha_{ij}^n \alpha_{ji}^m$$

$$\mathcal{I}_{mn} = \int_{\substack{E > E_F \\ E' < E_F}} \rho(E)\rho(E') \frac{\mathcal{P}_m(E)\mathcal{P}_n(E')}{E - E'} dE dE'.$$

Accordingly, the calculation of $\chi(r_i, r_j)$ is splitted into two independent parts. The \mathcal{I}_{mn} have an analytical form, while recursive evaluation of coefficients α_{ij}^n is performed by means of the three term relations defining the orthogonal polynomials. Generically their definition equation reads $EP_n(E) = a_n\mathcal{P}_n(E) + b_n\mathcal{P}_{n+1}(E) + b_{n-1}\mathcal{P}_{n-1}(E)$ with $b_{-1} = 0, n \geq 0$, with a_n, b_n the associated recursion coefficients,³⁾ the main vectors to be evaluated recursively will follow from

$$|\varphi_i^n\rangle = \mathcal{P}_n(\mathcal{H}) |r_i\rangle = \sum_j \alpha_{ji}^n |r_j\rangle.$$

Pratically, we will consider the Chebyshev polynomials of second order, which are defined by $\mathcal{P}_n(\mathcal{H}) = \frac{1}{b}(\mathcal{H} - a)\mathcal{P}_n(\mathcal{H}) + \mathcal{P}_{n-1}(\mathcal{H})$ with $\mathcal{P}_{-1}(\mathcal{H}) = 0$ and $\mathcal{P}_0(\mathcal{H}) = \mathbb{1}$ and the corresponding weight is given by $\rho_{ab}(E) = \frac{1}{2\pi b^2} \sqrt{4b^2 - (E - a)^2}$ which is $\neq 0$ only for $E \in [a - 2b, a + 2b]$. The coefficients a and b are given by the calculated limits $a_{n \rightarrow \infty}, b_{n \rightarrow \infty}$ for the real densities of states. From the abovementioned relations, the $|\varphi_i^n\rangle$ will be given by $|\varphi_i^n\rangle = \frac{1}{b}(\mathcal{H} - a) |\varphi_i^{n-1}\rangle + |\varphi_i^{n-2}\rangle$ and $|\varphi_i^{-1}\rangle = 0, |\varphi_i^0\rangle = |r_i\rangle$. In the tight-binding scheme $\mathcal{H} = \sum_{pq} \gamma_{pq} |r_p\rangle \langle r_q|$, one shows that the α_{ij}^n coefficients have to be evaluated recursively through ($\alpha_{pj}^{-1} = 0, \alpha_{pj}^0 = \delta_{pj}, \forall p$)

$$\alpha_{ij}^{n+1} = \frac{1}{b} \left(\sum_p \alpha_{ip}^n \gamma_{pj} - a \alpha_{ij}^n \right) - \alpha_{ij}^{n-1}$$

To evaluate \mathcal{I}_{mn} , we then first we rearrange the initial form

$$\mathcal{I}_{mn} = \int_{\substack{E > E_F \\ E' < E_F}} \rho(E)\rho(E') \frac{\mathcal{P}_m(E)\mathcal{P}_n(E')}{E - E'} dE dE'$$

by noticing that the factor $1/(E - E')$ can be written as

$$\oint_{\Gamma} \frac{dz}{(z - E)(z - E')} = \frac{2i\pi}{E - E'} (\Theta(E' - E_F)\Theta(E_F - E) - \Theta(E - E_F)\Theta(E_F - E'))$$

with the Heaviside function ($\Theta(x) = 0, x < 0$ and $\Theta(x) = 1, x > 0$) and the contour Γ in the complex plane is given for $\eta \rightarrow 0, R_{\Gamma} \rightarrow \infty$. It thus follows:

$$\Re(\mathcal{I}_{mn} + \mathcal{I}_{nm}) = -\frac{i}{2\pi} \oint_{\Gamma} dz \left(\int_{-\infty}^{\infty} dE \frac{\rho(E)\mathcal{P}_n(E)}{z - E} \right) \times \left(\int_{-\infty}^{\infty} dE' \frac{\rho(E')\mathcal{P}_m(E')}{z - E'} \right)$$

By application of Jordan Lemme, the integral on the contour Γ tends to zero when the radius goes to infinity, and only remains four integrals on the real axis respectively for $[-\infty, a - 2b], [a - 2b, E_F], [E_F, a + 2b], [a + 2b, +\infty]$. Using the relation between first and second order Chebyshev polynomials defined on $[-1, +1]$ and associated with $\rho(E) = \sqrt{1 - E^2}$

$$\lim_{\eta \rightarrow 0^{\pm}} \int_{-1}^{+1} \frac{\sqrt{1 - E^2} \mathcal{P}_n(a + 2bE)}{\omega + i\eta - E} dE = \pi \{ Q_{n+1}(\omega) \mp i\pi \sqrt{1 - \omega^2} \mathcal{P}_n(\omega) \}$$

it is easy to show that for $|\omega| \leq 1$, with $\omega = \cos \phi$ then $Q_n(\omega) = \cos n\phi$ and $\mathcal{P}_n(\omega) = \frac{\sin(n+1)\phi}{\sin \phi}$, and finally

$$\lim_{\eta \rightarrow 0^{\pm}} \int_{-1}^{+1} \frac{\sqrt{1 - E^2} \mathcal{P}_n(a + 2bE)}{\omega + i\eta - E} dE = \pi \exp(\mp i(n+1)\phi)$$

In conclusion, given that the integration outside $[-1, 1]$ leads to pure imaginary terms, the calculation of \mathcal{I}_{mn} reduces to $\Re(\mathcal{I}_{mn} + \mathcal{I}_{nm})$

$$\frac{1}{2\pi b} \left(\frac{\sin(m+n+3)\mathcal{A}_F}{m+n+3} - \frac{\sin(m+n+1)\mathcal{A}_F}{m+n+1} \right)$$

where $\mathcal{A}_F = \text{Arcos}(\frac{E_F - a}{2b})$. The final form of the electronic susceptibility for a general tight-binding hamiltonian will be defined by

$$\frac{1}{2\pi b} \sum_{nm} \alpha_{ij}^n \alpha_{ji}^m \left(\frac{\sin(m+n+3)\mathcal{A}_F}{m+n+3} - \frac{\sin(m+n+1)\mathcal{A}_F}{m+n+1} \right)$$

which is the final form of the algorithm. Test of the numerical convergence and illustrations on physical applications have been shown elsewhere.^{5,6)}

Kubo formula for diagonal conductivity

The real-space calculation of the Kubo formula of the electronic conductivity may be considered as an alternative of usual Landauer conductance calculations, or diagonalization methods. Indeed, the use of Landauer formula for investigating quantum zero temperature transport is usually associated with free escape boundary conditions. Direction perpendicular to the current density is assumed to be periodic whereas the other, of size L , is connected to metallic leads with different chemical potentials. Scaling analysis is performed through L . With recursion method, exact diagonalization of the Hamiltonian are avoided, so that in principle larger class of complex systems can be studied. To reduce the possible numerical instability at boundary conditions induced by the velocity operator (periodic boundary conditions will indeed generate a short-circuit across the sample), the following transformation the Kubo formula is very suitable ($\hat{\mathcal{X}}(t) = e^{i\mathcal{H}t/\hbar} \hat{\mathcal{X}} e^{-i\mathcal{H}t/\hbar}$ and $\hat{\mathcal{X}}$ is the component along direction x of the position operator, Ω the volume of the system):

$$\sigma_{xx}(E_F) = \frac{2\hbar e^2 \pi}{\Omega} \lim_{t \rightarrow \infty} \text{Tr}[\delta(E_F - H) \frac{(\hat{\mathcal{X}}(t) - \hat{\mathcal{X}}(0))^2}{t}]$$

where the asymptotic behavior of the quantum diffusion of the wave-packets can be easily kept under control.⁸⁾ The conductivity reads:

$$\frac{2\hbar e^2 \pi}{\Omega} \sum_{j_x, j_y} \mathcal{D}_j(t) \times \Im m(\langle \tilde{\Phi}_j(t) | G(E_F + i\eta) | \tilde{\Phi}_j(t) \rangle)$$

where $\mathcal{D}_j(t) = \frac{\langle \Psi_j(t) | \hat{\mathcal{X}}^2 | \Psi_j(t) \rangle}{t}$ and with $|\Psi_j(t)\rangle = e^{-i\mathcal{H}t/\hbar} |j_x, j_y\rangle, |\Phi_j(t)\rangle = \hat{\mathcal{X}} |\Psi_j(t)\rangle$ and $|\tilde{\Phi}_j(t)\rangle$ its normalized form. The summation should be done over the total basis of states $|j_x, j_y\rangle$, but it turns out that a limited number of initial sites is sufficient to achieve convergence of the calculation (given that there is an inherent averaging process).⁷⁾ The time-dependent evolution of a wave-packet initially localized at $|j_x, j_y\rangle$ is also evaluated by polynomial expansion of the evolution operator $e^{-i\mathcal{H}t/\hbar} = \sum_n (\int dE \mathcal{P}_n e^{-iEt/\hbar}) \mathcal{P}_n(\mathcal{H})$ where we choose Chebyshev polynomials of first kind. Here one has to notice that the dynamical properties given by the method are averaged over all the spectrum, which may be limited when properties are strongly dependent of the position of Fermi level. Improvements of the method to investigate energy-dependent wave packet diffusion have been made

recently.⁸⁾

Hall Kubo conductivity

For computing the non-dissipative conductivity σ_{xy} , one can also follow a real-space approach of Hall conductivity.^{9,10)} Starting from the general off-diagonal form of the Kubo conductivity, one shows that a possible algorithm is given by:

$$\sigma_{xy} = -\frac{ie^2\hbar}{2\Omega} \int_{\substack{E_2 > E_F \\ E_1 < E_F}} dE_1 dE_2 \frac{f(E_1) - f(E_2)}{E_1 - E_2} \langle \dot{Y}, \dot{X} \rangle$$

with $\langle \dot{Y}, \dot{X} \rangle = \text{Tr}[\delta(E_1 - \mathcal{H})\dot{Y}\delta(E_2 - \mathcal{H})\dot{X}]$. This formulation enables the expansion of the spectral measure on the basis of Chebyshev polynomials. Note that \dot{X} and \dot{Y} are the time dependent position operators in both respective directions of the plane. Some simple algebra yields two part to be evaluated separately:

$$\sigma_{xy} = -\frac{ie^2\hbar}{2\Omega} \sum_{m,n,i} I_{mn} \times \langle i_x, i_y | \mathcal{P}_n(\mathcal{H})\dot{Y}\mathcal{P}_m(\mathcal{H})\dot{X} | i_x, i_y \rangle$$

where I_{mn} is analytical and depends on the choice of the polynomial basis. In our case, it corresponds to ($A_F = \text{Arcos}(\frac{E_F - a}{2b})$), and a, b associated to the weight function of Chebyshev polynomials):

$$I_{mn} = \frac{1}{\pi b} \left\{ \frac{\sin(m+n+3)A_F}{(m+n+3)} - \frac{\sin(m+n+1)A_F}{(m+n+1)} \right\}$$

The other part implies the calculation of the coefficients $\langle j_x, j_y | \mathcal{P}_n(\mathcal{H})\dot{Y}\mathcal{P}_m(\mathcal{H})\dot{X} | j_x, j_y \rangle$. A reasonable number of initial sites $|j_x, j_y\rangle$ should be considered. The sum over m and n -indices is, given the form of the I_{mn} factors, limited by some appropriate cut-off.

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