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High-frequency asymptotic behavior of self-energies in quantum impurity models

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We present explicit expressions for the high-frequency asymptotic behavior of electron self-energy of general quantum impurity models, which may be useful for improving the convergence of dynamical mean-field calculations and for the analytic continuation of the electron self-energy. We also give results, expressed in more physical terms, for the two-orbital and three-orbital rotationally invariant Slater-Kanamori interactions, in order to facilitate calculations of transition metal oxides.

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Quantum Monte Carlo solutions of quantum impurity models[1] play an important role in the dynamical mean-field theory (DMFT) of materials.[2,3] In these calculations, knowledge of the high-frequency behavior of the electron self-energy is useful, both to improve the convergence of the dynamical mean-field self-consistency process[4–6] and to provide the normalization needed for analytic continuation of the electron self-energy.[7] While the general procedure for deriving the high frequency behavior is known,[1,8–10] explicit expressions seem not to have appeared except for the single-orbital Anderson impurity model[9–11] and its cluster generalizations.[12,13]

In this Brief Report, we present these results for general quantum impurity models. We also present the specialization to the two and three orbital models with rotationally invariant (Slater-Kanamori)[14,15] on-site interactions, relevant to late and early transition metal oxides[16–19] where the physics is dominated by e_g and t_{2g} orbitals respectively. In supplementary material (available online) we present a Mathematica notebook which generates the needed terms and presents the resulting output.[20]

A quantum impurity model involves a set of "impurity" degrees of freedom coupled to a non-interacting bath. We begin by defining a general impurity model. We denote the impurity states by an index a representing spin and orbital degrees of freedom and define the operator that creates an electron into impurity state a as $\psi_a^\dagger$. We denote the bath states by an energy/momentum label k and a spin/orbital index α, with $c_{k\alpha}^\dagger$ the operator creating an electron into one of these state. The impurity model is the sum of a non-interacting ($H_{\rm nonint}$) and interacting ($H_{\rm int}$) terms

$$H = H_{\rm nonint} + H_{\rm int} \tag{1}$$

with

$$\begin{aligned} H_{\rm nonint} = &\sum_{k\alpha} \varepsilon_{k\alpha} c_{k\alpha}^\dagger c_{k\alpha} + \sum_{k\alpha b} \left(V_k^{\alpha b} c_{k\alpha}^\dagger \psi_b + H.c. \right) \\ &+ \sum_{ab} E^{ab} \psi_a^\dagger \psi_b, \end{aligned} \tag{2}$$

and

$$H_{\rm int} = \sum_{a_1 a_2 b_1 b_2} I^{a_1 a_2 b_1 b_2} \psi_{a_1}^\dagger \psi_{a_2}^\dagger \psi_{b_1} \psi_{b_2}. \tag{3}$$

The electron self-energy Σ is then a matrix in space of impurity model states a with components Σ^{ab}. An expression for the high-frequency behavior can be obtained from examination of the short-time behavior of the electron Green's function,[1,6,10] which in turn can be expressed in terms of the commutator of the Hamiltonian with electron operators. Evaluating Eqs. (215) and (216) of Ref. 1 we find:

$$\lim_{z\to\infty} \Sigma^{ab}(z) = \Sigma_\infty^{ab} + \frac{\Sigma_1^{ab}}{z}, \tag{4}$$

with

$$\Sigma_\infty^{ab} = 4 \sum_{a_1 b_1} I^{a a_1 b_1 b} \left\langle \psi_{a_1}^\dagger \psi_{b_1} \right\rangle, \tag{5}$$

and

$$\begin{aligned} \Sigma_1^{ab} = &\sum_{a_1 a_2 b_1 b_2} K_{a_1 a_2 b_1 b_2}^{ab} \left\langle \psi_{a_1}^\dagger \psi_{a_2}^\dagger \psi_{b_1} \psi_{b_2} \right\rangle \\ &+ \sum_{a_1 b_1} L_{a_1 b_1}^{ab} \left\langle \psi_{a_1}^\dagger \psi_{b_1} \right\rangle - \sum_\lambda \Sigma_\infty^{a\lambda} \Sigma_\infty^{\lambda b}, \end{aligned} \tag{6}$$

where

$$K_{a_1 a_2 b_1 b_2}^{ab} = \sum_\lambda \left(4 I^{a\lambda b_1 b_2} I^{a_1 a_2 \lambda b} - 16 I^{a a_1 b_1 \lambda} I^{\lambda a_2 b_2 b} \right), \tag{7}$$

and

$$L_{a_1 b_1}^{ab} = -8 \sum_\lambda I^{a a_1 \lambda_1 \lambda_2} I^{\lambda_1 \lambda_2 b_1 b}. \tag{8}$$

In these expressions the expectation values must in general be evaluated from the numerical solution of the quantum impurity model.

For the single-orbital Anderson impurity model,[21] the nonzero elements of the tensor $I^{a_1 a_2 b_1 b_2}$ may be chosen as $I^{\uparrow\downarrow\downarrow\uparrow} = I^{\downarrow\uparrow\uparrow\downarrow} = U/4$ and $I^{\uparrow\downarrow\uparrow\downarrow} = I^{\downarrow\uparrow\downarrow\uparrow} = -U/4$. One

can readily verify that Eqs. (5)-(8) yield the well-known results that[9–11]

$$\Sigma^{\uparrow\uparrow}_{\infty} = U\langle n_{\downarrow}\rangle, \tag{9}$$
$$\Sigma^{\uparrow\uparrow}_{1} = U^2\langle n_{\downarrow}\rangle(1-\langle n_{\downarrow}\rangle), \tag{10}$$

while $\Sigma^{\downarrow\downarrow}$ can be found by flipping all spin indices, and all "off-diagonal" self-energies (e.g. $\Sigma^{\uparrow\downarrow}$) are zero.

While these expressions [Eqs. (5)-(8)] may be straightforwardly programmed and evaluated for general impurity models and interactions, it is useful to present results for the important special case of single-site dynamical mean-field theory of d electrons subject to rotationally invariant Slater-Kanamori interactions,[14,15] because the results can be expressed in terms of physically relevant densities and the symmetries are automatically implemented, leading to fewer expressions to be evaluated.

The on-site interacting part of the Slater-Kanamori Hamiltonian is traditionally expressed in terms of three parameters, U, $U^{'}$ and J, as

$$\begin{aligned} H_{\rm int} = &\sum_{\alpha} U n_{\alpha\uparrow}n_{\alpha\downarrow} + \sum_{\alpha>\beta,\sigma} U' n_{\alpha\sigma}n_{\beta\overline{\sigma}} \\ &+ \sum_{\alpha>\beta,\sigma} (U'-J) n_{\alpha\sigma}n_{\beta\sigma} \\ &- \sum_{\alpha\neq\beta} J\left(\psi^{\dagger}_{\alpha\downarrow}\psi^{\dagger}_{\beta\uparrow}\psi_{\beta\downarrow}\psi_{\alpha\uparrow} + \psi^{\dagger}_{\beta\uparrow}\psi^{\dagger}_{\beta\downarrow}\psi_{\alpha\uparrow}\psi_{\alpha\downarrow}\right), \end{aligned} \tag{11}$$

where $n_{\alpha\sigma} = \psi^{\dagger}_{\alpha\sigma}\psi_{\alpha\sigma}$ and α labels the orbitals.

The five orbital states of the d-manifold are degenerate in free space, but in cubic symmetry are split into a doublet, transforming as the e_g representation of the cubic group, and a triplet, transforming as the t_{2g} representation. In many materials one may restrict attention to one of these two manifolds. Symmetry considerations then imply that the self-energy is diagonal in the orbital basis and, in paramagnetic states, diagonal in the spin states as well.

In the case of two-orbital model (e_g system), $\alpha,\beta \in \{1,2\}$. We can express the high-frequency behavior for the self-energy of a spin up electron on orbital 1 in terms of expectation values of orbital densities and spin-exchange operators as

$$\Sigma^{1\uparrow,1\uparrow}_{\infty} = U\langle n_{1\downarrow}\rangle + U'\langle n_{2\downarrow}\rangle + (U'-J)\langle n_{2\uparrow}\rangle, \tag{12}$$

$$\begin{aligned} \Sigma^{1\uparrow,1\uparrow}_{1} = &U^2\langle n_{1\downarrow}\rangle(1-\langle n_{1\downarrow}\rangle) + (U')^2\langle n_{2\downarrow}\rangle(1-\langle n_{2\downarrow}\rangle) \\ &+ (U'-J)^2\langle n_{2\uparrow}\rangle(1-\langle n_{2\uparrow}\rangle) \\ &+ 2UU'(\langle n_{1\downarrow}n_{2\downarrow}\rangle - \langle n_{1\downarrow}\rangle\langle n_{2\downarrow}\rangle) \\ &+ 2U(U'-J)(\langle n_{1\downarrow}n_{2\uparrow}\rangle - \langle n_{1\downarrow}\rangle\langle n_{2\uparrow}\rangle) \\ &+ 2U'(U'-J)(\langle n_{2\uparrow}n_{2\downarrow}\rangle - \langle n_{2\uparrow}\rangle\langle n_{2\downarrow}\rangle) \\ &+ J^2\left(\langle n_{1\downarrow}\rangle + \langle n_{2\downarrow}\rangle - 2\langle n_{1\downarrow}n_{2\downarrow}\rangle\right) \\ &+ 2J(J-U)\left\langle \psi^{\dagger}_{1\uparrow}\psi^{\dagger}_{2\downarrow}\psi_{1\downarrow}\psi_{2\uparrow}\right\rangle \\ &+ 2J(U+J-2U')\left\langle \psi^{\dagger}_{1\uparrow}\psi^{\dagger}_{1\downarrow}\psi_{2\downarrow}\psi_{2\uparrow}\right\rangle, \end{aligned} \tag{13}$$

and the forms for the other orbital/spin can be straightforwardly found by interchanging indices. Note that the off-diagonal elements are zero.

For the three-orbital case (t_{2g} system), α,β run from 1 to 3. The high-frequency behavior of the self-energy for a spin up electron on orbital 1 can be similarly expressed as

$$\Sigma^{1\uparrow,1\uparrow}_{\infty} = U\langle n_{1\downarrow}\rangle + U'(\langle n_{2\downarrow}\rangle + \langle n_{3\downarrow}\rangle) + (U'-J)(\langle n_{2\uparrow}\rangle + \langle n_{3\uparrow}\rangle), \tag{14}$$

$$\begin{aligned} \Sigma^{1\uparrow,1\uparrow}_{1} = &U^2\langle n_{1\downarrow}\rangle(1-\langle n_{1\downarrow}\rangle) + (U')^2\left[(\langle n_{2\downarrow}\rangle + \langle n_{3\downarrow}\rangle)(1-\langle n_{2\downarrow}\rangle - \langle n_{3\downarrow}\rangle) + 2\langle n_{2\downarrow}n_{3\downarrow}\rangle\right] \\ &+ (U'-J)^2\left[(\langle n_{2\uparrow}\rangle + \langle n_{3\uparrow}\rangle)(1-\langle n_{2\uparrow}\rangle - \langle n_{3\uparrow}\rangle) + 2\langle n_{2\uparrow}n_{3\uparrow}\rangle\right] + J^2(2\langle n_{1\downarrow}\rangle + \langle n_{2\downarrow}\rangle + \langle n_{3\downarrow}\rangle) \\ &+ 2(UU'-J^2)(\langle n_{1\downarrow}n_{2\downarrow}\rangle + \langle n_{1\downarrow}n_{3\downarrow}\rangle) - 2UU'\langle n_{1\downarrow}\rangle(\langle n_{2\downarrow}\rangle + \langle n_{3\downarrow}\rangle) \\ &+ 2U(U'-J)(\langle n_{1\downarrow}n_{2\uparrow}\rangle + \langle n_{1\downarrow}n_{3\uparrow}\rangle - \langle n_{1\downarrow}\rangle\langle n_{2\uparrow}\rangle - \langle n_{1\downarrow}\rangle\langle n_{3\uparrow}\rangle) \\ &+ 2U'(U'-J)\left[\langle n_{2\uparrow}n_{2\downarrow}\rangle + \langle n_{2\downarrow}n_{3\uparrow}\rangle + \langle n_{2\uparrow}n_{3\downarrow}\rangle + \langle n_{3\uparrow}n_{3\downarrow}\rangle - (\langle n_{2\uparrow}\rangle + \langle n_{3\uparrow}\rangle)(\langle n_{2\downarrow}\rangle + \langle n_{3\downarrow}\rangle)\right] \\ &+ 2J(J-U)\left(\left\langle \psi^{\dagger}_{1\uparrow}\psi^{\dagger}_{2\downarrow}\psi_{1\downarrow}\psi_{2\uparrow}\right\rangle + \left\langle \psi^{\dagger}_{1\uparrow}\psi^{\dagger}_{3\downarrow}\psi_{1\downarrow}\psi_{3\uparrow}\right\rangle\right) \\ &+ 2J(U+J-2U')\left(\left\langle \psi^{\dagger}_{1\uparrow}\psi^{\dagger}_{1\downarrow}\psi_{2\downarrow}\psi_{2\uparrow}\right\rangle + \left\langle \psi^{\dagger}_{1\uparrow}\psi^{\dagger}_{1\downarrow}\psi_{3\downarrow}\psi_{3\uparrow}\right\rangle\right) \\ &- J^2\left(\left\langle \psi^{\dagger}_{2\downarrow}\psi^{\dagger}_{2\uparrow}\psi_{3\downarrow}\psi_{3\uparrow}\right\rangle + \left\langle \psi^{\dagger}_{2\downarrow}\psi^{\dagger}_{3\uparrow}\psi_{2\uparrow}\psi_{3\downarrow}\right\rangle + \left\langle \psi^{\dagger}_{2\uparrow}\psi^{\dagger}_{3\downarrow}\psi_{2\downarrow}\psi_{3\uparrow}\right\rangle + \left\langle \psi^{\dagger}_{3\downarrow}\psi^{\dagger}_{3\uparrow}\psi_{2\downarrow}\psi_{2\uparrow}\right\rangle\right), \end{aligned} \tag{15}$$

while the forms for other orbitals/spins can be found by permuting indices. The off-diagonal elements are also zero. In this formulation only 27 correlators (with four fermionic operators) need to be measured, rather than the $6^4 = 1296$ correlators arising in a straightforward implementation of the general equations above.

As the number of orbitals increases, the number of terms proliferates, and for example in a treatment of the

full 5-fold degenerate d manifold more than 100 terms occur. The expressions are too lengthy to present in the published version, but we include as supplementary information a Mathematica notebook which generates the needed terms and presents the resulting output.[20] In the same file we have also presented the calculation for the two and three orbital cases. This code can also easily be manipulated to treat Hamiltonians with arbitrary interactions.

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