I. SPLINES AND FOURIER TRANSFORMATIONS

A. Direct Fourier Transformation

In our approach we do the direct Fourier transformation exactly, i.e. first we obtain coefficients of the cubic spline exploiting physical properties of GF transformed and then make analytical Fourier integration knowing the form of the splined curve. The cubic spline interpolation formula reads:

$$G(\tau) = a_i + b_i(\tau - \tau_i) + c_i(\tau - \tau_i)^2 + d_i(\tau - \tau_i)^3,\ \tau \in [\tau_i, \tau_{i+1}]$$

where coefficients a_i, b_i, c_i, d_i are equal to values of the function, it's first, second and third derivatives at knot i: $a_i = G(\tau_i)$, $b_i = G'(\tau_i)$, $c_i = G''(\tau_i)$, $d_i = G'''(\tau_i)$.

Or in terms of GF values, $G_i = G(\tau_i)$, and its second derivative, $M_i = G''(\tau_i)$, only:

$$\begin{aligned} a_i &= G_i, \\ b_i &= \frac{G_{i+1} - G_i}{h} - \frac{2M_i + M_{i+1}}{6}h, \\ c_i &= \frac{M_i}{2}, \\ d_i &= \frac{M_{i+1} - M_i}{6h}. \end{aligned} \tag{1}$$

From equations above we see that one needs to know the second derivatives, M_i, using tabulated values of GF, G_i, in order to get the cubic spline interpolation. To obtain M_i coefficients we use conditions of smoothness of the first derivative and continuity of the second one. As the result we have $L+1$ equations for $L+3$ unknowns:

$$\begin{bmatrix} 2 & \lambda_0 & & & & 0 \\ \mu_1 & 2 & \lambda_1 & & & \\ & \mu_2 & . & . & & \\ & & . & . & . & \\ & & . & . & 2 & \mu_{n-1} \\ 0 & & & & \mu_n & 2 \end{bmatrix} \begin{bmatrix} M_0 \\ M_1 \\ ... \\ ... \\ \\ M_n \end{bmatrix} = \begin{bmatrix} d_0 \\ d_1 \\ . \\ . \\ . \\ d_n \end{bmatrix} \tag{2}$$

where in addition to $L+1$ $M_0, ..., M_n$, $n = 0...L$, unknowns d_0 and d_n also should be provided. Last two unknowns entirely depend on the boundary conditions which we have to specify in order to have a unique solution of Eq. (2). If one knows the first derivatives at the end points then d_0 and d_n are known and in this case

$$\begin{aligned} \lambda_0 &= 1,\ d_0 = \frac{6}{h}\left(\frac{G_1 - G_0}{h} - G'_0\right) \\ \mu_0 &= 1,\ d_n = \frac{6}{h}\left(G'_n - \frac{G_n - G_{n-1}}{h}\right) \end{aligned}$$

and $d_i = \frac{3}{h}\left(\frac{G_{i+1}-G_i}{h} - \frac{G_i - G_{i-1}}{h}\right)$, $\lambda_i = \mu_i = \frac{1}{2}$, for $i \in [1, n-1]$. More detailed derivations of the above formulae one can find in[1].

We can reduce number of unknowns just putting M_0 and M_n to zero (it is so called natural spline boundary conditions). In this case

$$\lambda_0 = 0,\ d_0 = 0, \mu_n = 0, d_n = 0$$

and we have the number of unknowns matching the number of equations, $L+1$!

This boundary condition is good enough to compute FT of GF in the system at or close to half filling since the second derivative of the Green's function is small in absolute value in this regime. And using the natural spline boundary condition we do not impose a noticeable error. However, away from half filling when the asymmetry of the system grows, along with amplitude of one out of the two second derivatives, usage of the natural spline eventually leads to pathological behavior of the self energy. The signature of this pathology is in the "overshooting" effect when the self energy at some finite Matsubara frequency i.e. the imaginary part of the self energy becomes positive in some

frequency region on the positive Matsubara half-axis while it should be always negative. This, of course, amounts to having negative spectral weight for the self energy which is something that does not occur for fermionic response functions. The "overshooting" can get especially severe in the limiting cases of small temperatures, small particle densities or large interaction strength U.

So, to avoid the problem with the self-energy and, hence, with the whole procedure of the self-consistency in DMFT-QMC program we need to use the proper boundary conditions. And in this case we have two possibilities to get unique solution for the system of Eq. (2) exploiting physical properties of studied GF: a) we can provide with the first derivatives at both ends separately (in the next section we show how to calculate those derivatives) or b) we can provide the sum of the first and the sum of second derivatives at the end points, so called the first and the second moments of the GF.

With the second choice of the boundary conditions b) the system of equations become three-diagonal one with two non-diagonal elements in the opposite corners of the matrix ($-M_{n-1}$ and $-\frac{1}{2}M_0$):

$$\begin{array}{cccccc}
4M_0 + M_1 & & & & -M_{n-1} & = d_0 \\
\frac{1}{2}M_0 + 2M_1 & + \frac{1}{2}M_2 & & & & = d_1 \\
\frac{1}{2}M_1 & + 2M_2 & + \frac{1}{2}M_3 & & & = d_2 \\
& \frac{1}{2}M_2 & + 2M_3 & + \frac{1}{2}M_4 & & = d_3 \\
& & \ddots & \ddots & \ddots & \vdots \\
& & \frac{1}{2}M_{n-3} & + 2M_{n-2} & \frac{1}{2}M_{n-1} & = d_{n-2} \\
-\frac{1}{2}M_0 & & & + \frac{1}{2}M_{n-2} & + 2M_{n-1} & = d_{n-1}
\end{array} \tag{3}$$

where $d_0 = \frac{6}{h}\left(\frac{G_1 - G_0}{h} + \frac{G_n - G_{n-1}}{h} - M^{(1)}\right) + 2M^{(2)}$, $d_{n-1} = \frac{6}{h}\left(\frac{G_n + G_{n-2} - 2G_{n-1}}{h}\right) - \frac{1}{2}B$, $G'_0 + G'_n = M^{(1)}$, $M_0 + M_n = M^{(2)}$.

Now having obtained the second derivatives M_i and coefficients a_i, b_i, c_i, d_i we can take Fourier integral analytically:

$$\begin{aligned}
&G_m(\omega_n) = \\
&\int_{\tau_{m-1}}^{\tau_m} d\tau[a + b(\tau - \tau_m) + c(\tau - \tau_m)^2 - d(\tau - \tau_m)^3)]\mathrm{e}^{(i\tau\omega_n)} = \\
&\frac{\mathrm{e}^{i\tau_m\omega_n}(-6d + 2ic\omega_n + b\omega^2 - ia\omega_n^3)}{\omega_n^4} - \\
&\frac{1}{\omega_n^4}(\mathrm{e}^{i\tau_{m-1}\omega_n}(-6d + 2ic\omega_n - 6i\Delta\tau d\omega_n + b\omega_n^2 - 2c\Delta\tau\omega_n^2 + \\
&3(\Delta\tau)^2 d\omega_n^2 - ia\omega_n^3 + ib\Delta\tau\omega_n^3 - ic(\Delta\tau)^2\omega_n^3 + i(\Delta\tau)^3 d\omega_n^3)).
\end{aligned} \tag{4}$$

Sum over m of $G_m(\omega_n)$

$$G(\omega_n) = \sum_{m=1}^{L} G_m(\omega_n)$$

will give us the Fourier integral in frequency space. This formula can be very easily identified in the corresponding subroutine of DMFT-QMC program.

The subroutine which does direct Fourier transformation requires three moments (real numbers) and a real L-dimension array of $G(\tau_i)$ as an input and produces complex N-dimension array of $G(\omega_n)$, where N is number of frequency points.

B. Inverse Fourier transformation

As it is well known Green function $G(\omega)$ falls off as $1/\omega$ when $\omega \to \infty$. In the program we deal with finite number of frequency points and cutting off $1/\omega$ tail one would make a rather crude approximation as the discontinuity of GF $G(\tau)$ (imaginary time domain!) has been removed. In such situation, the high-frequency tail has to be extracted from GF $G(\omega)$ and Fourier transformed analytically using the following Fourier relation

$$\frac{1}{i\omega_n - \epsilon} \leftrightarrow -[\Theta(\tau) + \zeta n(\epsilon)]\, e^{-\epsilon\tau}, \tag{5}$$

where $n(\epsilon) \equiv 1/[\exp\{\beta\epsilon\} - \zeta]$ and $\zeta = \pm 1$ depending on whether ω_n is bosonic or fermionic.

The inverse Fourier transformation for GF without the tail is made by straightforward summation over Matsubara frequencies. Once it is done we add the information about the tail using Eq. (5).

The subroutine for the inverse Fourier transformation requires complex N-dimension array of $G(\omega_n)$ and produces a real L-dimension array of $G(\tau_i)$ as an output.

II. MOMENTS

Moments, $M^{(k)}$, are nothing else as the expansion of GF in frequency domain:

$$G(\omega) = \sum_{k=0}^{N} \frac{M^{(k)}}{\omega^{k+1}}. \tag{6}$$

Another definition of k degree moment is the following:

$$M^{(k)} = \int_{-\infty}^{+\infty} d\omega \omega^k \rho(\omega), \tag{7}$$

where $\rho(\omega)$ is density of states (DOS).

Moments $M^{(k)}$ can be bind to sum of GFs and sum of it's derivatives in imaginary-time space as:

$$G^{(k)}(0) + G^{(k)}(\beta) = M^{(k)}, \tag{8}$$

where $k = 0, \dots N$.

To show this one needs to take Fourier integral in parts:

$$\begin{aligned} G(i\omega_n) &= \int_0^\beta \mathrm{e}^{i\omega_n \tau} G(\tau) d\tau \\ &= \sum_{k=0}^{N} \frac{(-1)^{k+1}(G^{(k)}(0) + G^{(k)}(\beta))}{(i\omega_n)^{k+1}} \\ &+ \frac{(-1)^{N+1}}{(i\omega_n)^{N+1}} \int_0^\beta \mathrm{e}^{i\omega_n \tau} \frac{\partial^{N+1} G(\tau)}{\partial \tau^{N+1}} d\tau. \end{aligned} \tag{9}$$

So, to solve the system of Eq.(3) we need to adhere to the proper boundary conditions which are expressed through the various moments of the Green's function. What we need finally it to provide the first three moments $M^{(0)}, M^{(1)}, M^{(2)}$. The first moment for Green's function is equal to one, the second moment proportional to the chemical potential in the system and the third one is a little bit more complicated and contains a density-density correlator. To show that we start with the Anderson model which reads

$$\begin{aligned} H_{SIAM} &= \sum_{k\alpha} \varepsilon_{k\alpha} c^\dagger_{k\alpha} c_{k\alpha} + \sum_{\alpha} (\varepsilon_\alpha + \frac{1}{2} \sum_{\alpha' \neq \alpha} U_{\alpha'\alpha}) f^\dagger_\alpha f_\alpha \\ &+ \sum_{k\alpha} V_{k\alpha} (f^\dagger_\alpha c_{k\alpha} + c^\dagger_{k\alpha} f_\alpha) \\ &+ \sum_{\alpha <} \sum_{\alpha'} (U_{\alpha\alpha'} n_\alpha n_{\alpha'} - \frac{1}{2}(n_\alpha + n_{\alpha'})), \end{aligned} \tag{10}$$

where $\tilde{\varepsilon}_\alpha = \varepsilon_\alpha + \frac{1}{2} \sum_{\alpha \neq \alpha'} U_{\alpha'\alpha}$, the first three moments moments be obtained from the following commutators

$$M^{(k)} = \left\langle \{\mathcal{L}^k f_\alpha; f^\dagger_\alpha\}_+ \right\rangle,$$

where $\mathcal{L}\mathcal{O} = [\mathcal{O},\mathcal{H}]$ denotes the commutator of operator $\mathcal{O}$ with the Hamiltonian, and $\{...\}_+$ is the anticommutator. After some algebra one finds the following expressions for the moments in the $SU(N)$ approximation:

$$M^{(0)} = \left\langle \left\{ f_\alpha, f_\alpha^\dagger \right\} \right\rangle = 1, \tag{11}$$

$$M^{(1)} = \left\langle \left\{ [f_\alpha, H], f_\alpha^\dagger \right\} \right\rangle = \tilde{\varepsilon}_\alpha + \sum_{\alpha' \neq \alpha} U_{\alpha\alpha'}(n_{\alpha'} - \frac{1}{2}), \tag{12}$$

$$\begin{aligned} M^{(2)} &= \left\langle \left\{ [[f_\alpha, H], H], f_\alpha^\dagger \right\} \right\rangle = \left\langle \left\{ [f_\alpha, H], [H, f_\alpha^\dagger] \right\} \right\rangle = \\ &\left\langle \tilde{\varepsilon}_\alpha^2 + 2\tilde{\varepsilon}_\alpha \sum_{\alpha' \neq \alpha} U_{\alpha\alpha'}(n_{\alpha'} - \frac{1}{2}) + \right. \\ &\left. \sum_{\alpha', \alpha'' \neq \alpha} \sum U_{\alpha\alpha'} U_{\alpha\alpha''}(n_{\alpha'} - \frac{1}{2})(n_{\alpha''} - \frac{1}{2}) + \sum_k V_{k\alpha}^2 \right\rangle. \end{aligned} \tag{13}$$

Summing up similar terms $SU(N)$ approximation we get

$$M^{(1)} = \varepsilon_\alpha + (2N-1)Un, \tag{14}$$

$$\begin{aligned} M^{(2)} &= \varepsilon_\alpha^2 + 2\varepsilon_\alpha(2N-1)Un + \\ &U^2\left[(2N-1)n + \langle nn \rangle\right] + \sum_k V_{k1}^2, \end{aligned} \tag{15}$$

where n is filling per band and per spin, $n = \frac{1}{2N}\sum_\alpha n_\alpha$, and double occupancy is defined as $\langle nn \rangle = \sum_{\alpha \neq \alpha'} \langle n_\alpha n_{\alpha'} \rangle$.

The second way to make the correct cubic spline as we mentioned before is to provide with local first derivatives at both ends as the boundary conditions. To find the first derivatives at the ends one can use the following definition of the first derivative of finite-temperature GF:

$$-\frac{\partial}{\partial\tau}\left\langle T_\tau f_\alpha(\tau) f_\alpha^\dagger(0) \right\rangle = -\left\langle T[H, f_\alpha] f_\alpha^\dagger \right\rangle = G'_\alpha(\beta).$$

Using as the Hamiltonian $H = H_{SIAM}$ we can easily obtain the derivatives at the ends:

$$\begin{aligned} G'_\alpha(\beta) &= \varepsilon_\alpha(1 - n_\alpha) - \left\langle \sum_k V_{k\alpha} c_{k\alpha} f_\alpha^\dagger \right\rangle + \\ &\sum_{\alpha' \neq \alpha} U_{\alpha\alpha'}\left(n_{\alpha'} - \langle n_{\alpha'} n_\alpha \rangle\right), \\ G'_\alpha(0) &= \varepsilon_\alpha n_\alpha + \left\langle \sum_k V_{k\alpha} c_{k\alpha}^\dagger f_\alpha \right\rangle + \\ &\sum_{\alpha' \neq \alpha} U_{\alpha\alpha'} \langle n_{\alpha'} n_\alpha \rangle, \end{aligned} \tag{16}$$

where averages e.g. $\left\langle \sum_k V_{k\alpha} c_{k\alpha} f_\alpha^\dagger \right\rangle$ can be calculated from the following expression:

$$\left\langle \sum_k V_{k\alpha} c_{k\alpha} f_\alpha^\dagger \right\rangle = T \sum_n \Delta_\alpha(i\omega_n) G_\alpha(i\omega_n) e^{i0^+}.$$

In below obtained formulae (Eqs. (11)-(15)) we should know filling, n_α, for each band and spin as well as we should know density-density correlator $\langle n_\alpha n_{\alpha'} \rangle$. The filling we can extract from GF itself. Calculation of the correlator in QMC highlights one of the advantages of the method: the correlator is provided by the QMC itself and one does not need to rely on additional approximations to obtain it as e.g. in the case of multiband IPT method where coherent potential approximation is used to get the correlator. At each time slice the density-density correlator is also computed from GF but in imaginary time domain where it is simply a product of two Green's functions in (τ, τ') space. We should note here that we compute the correlator along with other parameters in the system at each iteration step and once the self-consistency is reached we have correctly obtained all the components and parameters in the system. And

finally, with small enough imaginary time step $\Delta\tau$ one can completely avoid the "overshooting" problem. Keeping in mind the main limitation in QMC program $U\Delta\tau/2 < 1$. In the present computations we choose $\Delta\tau = 1/4$ which is good enough for the range of parameters we use in the current paper.

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[1] J. Stoer and R. Bulirsch, *Introduction to Numerical Analysis* (Springer-Verlag, New York Berlin Heidelberg, 1980).