In a recent publication [PRE 86, 04012 (2012)], Santos has presented a self-consistency condition that can be used to limit the possible forms of Fundamental Measure Theory. Here, the direct correlation function resulting from the Santos functional is derived and it is found to diverge for all densities.
In a recent contribution, Santos introduced a novel argument aimed at eliminating a source of ambiguity in the derivation of the Fundamental Measure Theory (FMT) approach to Density Functional Theory for hard spheres. The result is a new ansatz for improvement of FMT beyond the basic Rosenfeld functional. The proposal is quite interesting as the most accurate density functionals currently in use (such as the “White Bear” functional) are of exactly this type: heuristic improvements beyond functionals based on Rosenfeld’s original reasoning together with the additional requirement that the forms reproduce known, exact results in low-dimensional systems. The introduction of a new element that eliminates some of the arbitrariness of these extensions is therefore welcome. The purpose of this Comment is to examine one consequence of the proposed ansatz, namely the implied direct correlation function (DCF).

The direct correlation function is a fundamental element in DFT as it provides a connection between model free energy functionals and liquid-state properties, for which much is known. Given a (grand-canonical) free energy functional, \( \Omega [\rho] = F_{id} [\rho] + F_{ex} [\rho] - \mu \rho \), where \( \rho (r) \) is the ensemble-averaged local density, \( F_{id} \) is the ideal gas contribution, which is not relevant here, \( \mu \) is the chemical potential and \( F_{ex} [\rho] \) is the excess term, the (two-body) direct correlation function is given by taking two functional derivatives with respect to the density,

\[
c_2 (r_1, r_2) = -\frac{\delta^2 \beta F_{ex}[\rho]}{\delta \rho (r_1) \delta \rho (r_2)},
\]

where \( \beta = 1/k_B T \), \( k_B \) is Boltzmann’s constant and \( T \) is the temperature. This relation between the free energy functional and the DCF has always provided an important connection between free energy models and liquid-state properties: for example, one of the first indications of the utility of the White Bear functional was its improvement in the predicted DCF of hard spheres.

In DFT, the only unknown is the excess term and FMT is based on an ansatz of the form

\[
\beta F_{ex} = \int \Phi (n (r; [\rho])) \, dr
\]

where the weighted densities have the generic expressions

\[
n_i (r; [\rho]) = \int w_i (r - r') \rho (r') \, dr'
\]

Different models involve different collections of density-independent weight functions, \( w_i \), and of different forms for the function \( \Phi (n) \). The proposal of Santos makes use of the weight functions as were introduced by Rosenfeld \( (w_n (r_{12}) = \delta (\frac{\sigma}{2} - r_{12}), w_\eta (r_{12}) = \Theta (\frac{\pi \sigma^2}{2} - r_{12}) \) \), \( w_n (r_{12}) = f_{12} \delta \left( \frac{\pi \sigma^2}{2} - r_{12} \right) \) where \( \sigma \) is the hard-sphere diameter) and the \( \Phi \) function of the Rosenfeld, \( \Phi_B = s \Phi_1 (\eta) + \Phi_2 (\eta) (s^2 - v^2) + \Phi_3 (\eta) s (s^2 - 3v^2) \), where \( \eta (r; [\rho]) = n_\eta (r; [\rho]) \) is the weighted density formed from the weight function \( w_\eta (r_{12}) \), etc. The other terms are

\[
\Phi_1 (\eta) = -\frac{1}{2 \pi \sigma^2} \ln (1 - \eta), \quad \Phi_2 (\eta) = \frac{1}{2 \pi \sigma (1 - \eta)}, \quad \Phi_3 (\eta) = \frac{1}{2 \pi \sigma (1 - \eta)^2}
\]

The Rosenfeld functional reproduces the Percus-Yevik equation of state for a uniform fluid \( (\rho (r) = \rho \) with \( \rho \) being a position-independent constant) (see the Appendix for details). The idea of the extension discussed by Santos is that one would like to use knowledge of more accurate equations of state than Percus-Yevik to construct potentially more accurate approximations for \( \Phi \). Note that for a uniform system, Eq.(2) shows that the excess free energy of a uniform liquid is simply \( \beta F_{ex} = V \Phi (n (\rho)) \) where \( V \) is the total volume. Santos introduces a correction so that \( \Phi = \Phi_R + \Phi_S \) the form of which is fixed by the scaling relations to be

\[
\Phi_S (n) = (s^2 - v^2) \Phi_2 (\eta) \Delta \left( \frac{2 s (s^2 - 3v^2) \Phi_3 (\eta)}{(s^2 - v^2) \Phi_2 (\eta)} \right)
\]

Here, the function \( \Delta (u) \) is chosen so that the free energy in the uniform limit agrees with some chosen form (such as Carnahan-Starling). Calculation of the implied DCF in the uniform state based on Eq.(1) is straightforward (details are given in the Appendix). The result is
$\eta_1 = \pi/6 \rho \sigma^3$ and $c_{PY}(r; \rho)$ is the Percus-Yevik DCF that comes from $\Phi_R$. One feature that stands out is that this function diverges for $x = 0$ unless $\Delta'' \left( \frac{\eta}{1-\eta} \right) = 0$. This divergence is unphysical and, as shown in the Figure, spoils the otherwise reasonable agreement with the results of the White-Bear functional. Hence, if this undesirable behavior is to be avoided, the only possibility is the relatively restricted set of corrections given by $\Delta \left( \frac{\eta}{1-\eta} \right)$. However, Santos notes that in general one expects that $\Delta(y) \sim O(y^2)$ so this eliminates the possibility of a correction. Santos also offers a modified version of his proposal that appears to avoid the divergence in the DCF but at the cost of violating his self-consistency condition[1]. It is interesting to note that similar terms arise in deriving the DCF from the Rosenfeld part of the free energy, but they cancel thus leaving the (finite) Percus-Yevik result (for a demonstration, see, e.g., the Appendix).

In summary, the proposal of Santos produces a divergent DCF in the uniform liquid. Nevertheless, the reasoning behind it seems sound and it is to be hoped that there might still be a way to exploit it so as to eliminate some of the ambiguities of the usual extensions of FMT while retaining their advantages, one of which is an excellent description of the DCF for hard-spheres.

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CALCULATION OF THE DIRECT CORRELATION FUNCTION

1. The functional derivatives

In general,

\[
\frac{\delta \beta F_{ex}}{\delta \rho (\mathbf{r}_1)} = \frac{\delta}{\delta \rho (\mathbf{r}_1)} \int \Phi (n (\mathbf{r})) \, d\mathbf{r}
\]

\[
= \int \left( \frac{\partial}{\partial n_i} \Phi (n) \right) n_r (\mathbf{r}) \frac{\delta n_i (\mathbf{r})}{\delta \rho (\mathbf{r}_1)} \, d\mathbf{r}
\]

\[
\equiv \int \Phi_i (n (\mathbf{r})) \, w_i (\mathbf{r} - \mathbf{r}_1) \, d\mathbf{r}, \quad \Phi_i \equiv \frac{\partial}{\partial n_i} \Phi
\]

and

\[
\frac{\delta^2 \beta F_{ex}}{\delta \rho (\mathbf{r}_1) \delta \rho (\mathbf{r}_2)} = \int \Phi_{ij} (n (\mathbf{r})) \, w_i (\mathbf{r} - \mathbf{r}_1) \, w_j (\mathbf{r} - \mathbf{r}_2) \, d\mathbf{r}, \quad \Phi_{ij} \equiv \frac{\partial^2}{\partial n_i \partial n_j} \Phi
\]

so that the bulk limit is

\[
\lim_{\rho (\mathbf{r}) \to \rho} \frac{\delta^2 \beta F_{ex}}{\delta \rho (\mathbf{r}_1) \delta \rho (\mathbf{r}_2)} = \Phi_{ij} (n) \int w_i (\mathbf{r} - \mathbf{r}_1) \, w_j (\mathbf{r} - \mathbf{r}_2) \, d\mathbf{r}
\]

\[
\equiv \Phi_{ij} (n) \, w_i \ast w_j
\]

where the last line introduces a compact notation for the convolution in which the spatial arguments are suppressed and where \( n = \lim_{\rho (\mathbf{r}) \to \rho} n (\mathbf{r}) \).

2. Useful formulae

For later use, I note that the convolutions are given by

\[
w_\eta \ast w_\eta = 2\pi \Theta (\sigma - r_{12}) \sigma^3 \frac{1}{24} (x - 1)^2 (x + 2)
\]

\[
w_\sigma \ast w_\eta = 2\pi \Theta (\sigma - r_{12}) \sigma^2 \frac{1}{4} (1 - x)
\]

\[
w_\sigma \ast w_\sigma = 2\pi \Theta (\sigma - r_{12}) \sigma \frac{1}{4x}
\]

\[
w_{\eta_1} \ast w_{\eta_1} = 2\pi \Theta (\sigma - r_{12}) \sigma \left( \frac{1}{4x} - \frac{1}{2} x \right)
\]

where \( x \equiv r_{12}/\sigma \). The weighted densities in the bulk limit are

\[
\eta (\mathbf{r}) \to 4\pi \frac{\sigma}{3} \left( \frac{\sigma}{2} \right)^3 \rho = \frac{\pi}{6} \rho \sigma^3
\]

\[
s (\mathbf{r}) \to 4\pi \left( \frac{\sigma}{2} \right)^2 \rho = \frac{6}{\sigma} \eta
\]

\[
v_i (\mathbf{r}) \to 0
\]

3. Rosenfeld functional

It is straightforward to see that the Rosenfeld functional gives

\[
\lim_{\rho (\mathbf{r}) \to \rho} \frac{\delta^2 \beta F_{ex}^{(R)}}{\delta \rho (\mathbf{r}_1) \delta \rho (\mathbf{r}_2)} = s \Phi_1'' (\eta) \, w_\eta \ast w_\eta + 2 \Phi_1' (\eta) \, w_\sigma \ast w_\eta
\]

\[
+ \Phi_2' (\eta) \, (s^2 - \nu^2) \, w_\eta \ast w_\eta + 2 \Phi_2 (\eta) \, (w_{\eta_1} \ast w_{\eta_2}) + 2 \Phi_2' (\eta) \, w_\eta \ast (2 w_{\eta_1} - 2 w_{\eta_2})
\]

\[
+ \Phi_3' (\eta) \, s \, w_\eta \ast w_\eta + 2 \Phi_3 (\eta) \, (s^2 - 3 \nu^2) \, w_\eta \ast w_\sigma + 2 \Phi_3' (\eta) \, s w_\eta \ast 2 \, (2 w_{\eta_1} - 2 w_{\eta_2})
\]

\[
+ 2 \Phi_3 (\eta) \, w_{\eta_1} \ast 2 \, (2 w_{\eta_1} - 2 w_{\eta_2}) + \Phi_3 (\eta) \, s2 \, (w_{\eta_1} \ast w_{\eta_2} - 3 w_{\eta_1} \ast w_{\eta_2})
\]
and since \( v_i = 0 \) in the bulk this becomes

\[
\lim_{\rho(r) \to \pi} \frac{\delta^2 \beta F_{ex}^{(R)}}{\delta \rho(r_1) \delta \rho(r_2)} = s\Phi''_1(\eta) w_\eta * w_\eta + 2\Phi'_1(\eta) w_s * w_\eta \\
+ \Phi''_2(\eta) s^2 w_\eta * w_\eta + 2\Phi_2(\eta) (w_s * w_\eta - w_{v_i} * w_{v_i}) + 4\Phi'_2(\eta) s w_\eta * w_s \\
+ \Phi''_3(\eta) s (s^2 - 3\nu^2) w_\eta * w_\eta + 2\Phi_3(\eta) (s^2 - 3\nu^2) w_\eta * w_s + 4\Phi'_3(\eta) s^2 w_\eta * w_s \\
+ 6\Phi_3(\eta) s (w_s * w_s - w_{v_i} * w_{v_i})
\]

Making use of

\[
\Phi_1(\eta) = -\frac{1}{\pi \sigma^2} \ln (1 - \eta) \implies \Phi'_1(\eta) = \frac{1}{\pi \sigma^2 (1 - \eta)} \implies \Phi''_1(\eta) = \frac{1}{\pi \sigma^2 (1 - \eta)^2}
\]

\[
\Phi_2(\eta) = \frac{1}{2\pi (1 - \eta)} \implies \Phi'_2(\eta) = \frac{1}{2\pi (1 - \eta)^2} \implies \Phi''_2(\eta) = \frac{2}{2\pi (1 - \eta)^3}
\]

\[
\Phi_3(\eta) = \frac{1}{24\pi (1 - \eta)^2} \implies \Phi'_3(\eta) = \frac{2}{24\pi (1 - \eta)^3} \implies \Phi''_3(\eta) = \frac{6}{24\pi (1 - \eta)^4}
\]

gives

\[
\lim_{\rho(r) \to \pi} \frac{\delta^2 \beta F_{ex}^{(R)}}{\delta \rho(r_1) \delta \rho(r_2)} = \frac{1}{2} \frac{\eta}{(\eta - 1)^2} (x - 1)^2 (x + 2) - \frac{1}{\eta - 1} (1 - x)
\]

\[
+ \frac{9}{2} \frac{\eta^2}{(\eta - 1)^3} (x - 1)^2 (x + 2) + 6 \frac{\eta}{(\eta - 1)^2} (1 - x)
\]

\[
+ \frac{3}{2} \frac{\eta}{(\eta - 1)^2}
\]

or

\[
\lim_{\rho(r) \to \pi} \frac{\delta^2 \beta F_{ex}^{(R)}}{\delta \rho(r_1) \delta \rho(r_2)} = \frac{1}{2} \frac{\eta (2\eta + 1)^2}{(1 - \eta)^4} x^3 - \frac{3 \eta (\eta + 2)^2}{2 (1 - \eta)^4} x + \frac{(2\eta + 1)^2}{(1 - \eta)^4}
\]

which is the usual PY expression.

4. The new term

Next, we need the additional terms introduced by Santos. First note that

\[
\frac{\delta \beta \Phi_S}{\delta \rho(r_1)} = 2 (sw_s - v_i w_{v_i}) \Phi_2(\eta) \Delta
\]

\[
+ (s^2 - v^2) \Phi'_2(\eta) w_\eta \Delta
\]

\[
+ (s^2 - v^2) \Phi_2(\eta) (\Delta_s w_s + \Delta_{v_i} w_{v_i} + \Delta_{\eta} w_\eta)
\]

So, neglecting some non-contributing terms proportional to \( v_i \),

\[
\lim_{\rho(r) \to \pi} \frac{\delta^2 \beta \Delta F_{ex}^{(S)}}{\delta \rho(r_1) \delta \rho(r_2)} = 2 (w_s * w_s - w_{v_i} * w_{v_i}) \Phi_2(\eta) \Delta \\
+ 4s \Phi_2(\eta) (\Delta_s w_s * w_s + \Delta_{\eta} w_s * w_\eta) + s^2 \Phi'_2(\eta) w_\eta * w_\eta \Delta
\]

\[
+ 2s^2 \Phi'_2(\eta) (\Delta_s w_\eta * w_s + \Delta_{\eta} w_\eta * w_\eta)
\]

\[
+ s^2 \Phi_2(\eta) (\Delta_{ss} w_s * w_s + 2\Delta_{x\eta} w_s * w_\eta + 2\Delta_{s} w_s w_{v_i} + \Delta_{\eta \eta} w_\eta * w_\eta)
\]
Using

\[
\Delta = \Delta \left( 2 s \frac{(s^2 - 3v^2) \Phi_3(\eta)}{(s^2 - v^2) \Phi_2(\eta)} \right)
\]

\[
\Delta_s = \left( 2 s \frac{(s^4 + 3v^4) \Phi_3(\eta)}{(s^2 - v^2)^2 \Phi_2(\eta)} \right) \Delta' \left( 2 s \frac{(s^2 - 3v^2) \Phi_3(\eta)}{(s^2 - v^2) \Phi_2(\eta)} \right)
\]

\[
\Delta_\eta = \left( 2 s \frac{(s^2 - 3v^2) \Phi_3(\eta)}{(s^2 - v^2) \Phi_2(\eta)} \right) \Delta' \left( 2 s \frac{(s^2 - 3v^2) \Phi_3(\eta)}{(s^2 - v^2) \Phi_2(\eta)} \right)
\]

\[
\Delta_{v^2} = \left( -4 s \frac{s^3 \Phi_3(\eta)}{(s^2 - v^2)^2 \Phi_2(\eta)} \right) \Delta' \left( 2 s \frac{(s^2 - 3v^2) \Phi_3(\eta)}{(s^2 - v^2) \Phi_2(\eta)} \right)
\]

\[
\Delta_{ss} = \left( 2 s \frac{(s^4 + 3v^4) \Phi_3(\eta)}{(s^2 - v^2)^2 \Phi_2(\eta)} \right) \Delta'' \left( 2 s \frac{(s^2 - 3v^2) \Phi_3(\eta)}{(s^2 - v^2) \Phi_2(\eta)} \right)
\]

and the bulk limits

\[
\Delta \to \Delta \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right)
\]

\[
\Delta_s \to \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) \Delta' \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right)
\]

\[
\Delta_\eta \to 2s \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) \Delta' \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right)
\]

\[
\Delta_{v^2} \to \left( -4 s \frac{\Phi_3(\eta)}{s \Phi_2(\eta)} \right) \Delta' \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right)
\]

\[
\Delta_{ss} \to \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) ^2 \Delta'' \left( 2 s \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right)
\]

gives

\[
\lim_{\rho(\mathbf{r}) \to \rho \mathbf{r}} \frac{\delta^2 \beta \Delta F^{(S)}_{\text{ex}}}{\delta \rho (\mathbf{r}_1) \delta \rho (\mathbf{r}_2)} = 2 \left( w_s * w_s - w_{v_i} * w_{v_i} \right) \Phi_2(\eta) \Delta + 4s \left( w_s * w_\eta \right) \Phi_2(\eta) \Delta
\]

\[
+ 4s \Phi_2(\eta) \left( \Delta_s w_s * w_s + \Delta_\eta w_s * w_\eta \right) + s^2 \Phi_2''(\eta) w_\eta * w_\eta \Delta
\]

\[
+ 2s^2 \Phi_2'(\eta) \left( \Delta_s w_\eta * w_s + \Delta_\eta w_s * w_\eta \right)
\]

\[
+ s^2 \Phi_2(\eta) \left( 2 \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) ^2 \Delta'' w_s * w_s + 2\Delta_s w_\eta * w_\eta + \Delta_\eta w_\eta * w_\eta \right) + s^2 \Phi_2(\eta) \Delta' \left( -\frac{8 \Phi_3(\eta)}{s \Phi_2(\eta)} w_{v_i} w_{v_i} \right)
\]

or, rearranging a little,

\[
\lim_{\rho(\mathbf{r}) \to \rho \mathbf{r}} \frac{\delta^2 \beta \Delta F^{(S)}_{\text{ex}}}{\delta \rho (\mathbf{r}_1) \delta \rho (\mathbf{r}_2)} = \left( 2 \left( w_s * w_s - w_{v_i} * w_{v_i} \right) \Phi_2(\eta) + 4s \left( w_s * w_\eta \right) \Phi_2(\eta) + w_\eta * w_\eta s^2 \Phi_2''(\eta) \right) \Delta
\]

\[
+ 4s \Phi_2(\eta) \left( 2 \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) \left( w_s * w_s - w_{v_i} * w_{v_i} \right) + 3s \left( 2 \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) w_s * w_\eta + s^2 \left( 2 \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) w_\eta * w_\eta \Delta'
\]

\[
+ s^2 \Phi_2(\eta) \left( 2 \frac{\Phi_3(\eta)}{\Phi_2(\eta)} \right) ^2 w_s * w_s \Delta'' + 2\Delta_s w_\eta * w_\eta + \Delta_\eta w_\eta * w_\eta \right)
Next, using
\[
\Delta_{\eta s} = \left(2 \frac{s(s^2 - 3v^2)}{(s^2 - v^2)} \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right)\right) \left(2 \frac{(s^4 + 3v^4)}{s^2 - v^2} \Phi_3(\eta)\right) \Delta'' \left(2 \frac{s(s^2 - 3v^2)}{(s^2 - v^2)} \Phi_3(\eta)\right)
\]
\[
+ \left(2 \frac{(s^4 + 3v^4)}{(s^2 - v^2)} \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right)\right) \Delta' \left(2 \frac{s(s^2 - 3v^2)}{(s^2 - v^2)} \Phi_3(\eta)\right)
\]
\[
\rightarrow 4s \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) \Delta'' + 2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) \Delta'
\]
\[
\Delta_{\eta \eta} = \left(2 \frac{s(s^2 - 3v^2)}{(s^2 - v^2)} \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right)\right)^2 \Delta'' \left(2 \frac{s(s^2 - 3v^2)}{(s^2 - v^2)} \Phi_3(\eta)\right)
\]
\[
+ \left(2 \frac{s(s^2 - 3v^2)}{(s^2 - v^2)} \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right)\right) \Delta' \left(2 \frac{s(s^2 - 3v^2)}{(s^2 - v^2)} \Phi_3(\eta)\right)
\]
\[
\rightarrow 4s^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right)^2 \Delta'' + 2s \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) \Delta'
\]
results in
\[
\lim_{\rho(r) \to \rho \rho(r_1) \rho(r_2)} \frac{\delta^2 \beta \Delta F_e^{(S)}}{\delta \rho(r_1) \delta \rho(r_2)} = (2 (w_s \ast w_s - w_{v_s} \ast w_{v_s}) \Phi_2(\eta) + 4s (w_s \ast w_\eta) \Phi_2(\eta) + w_\eta \ast w_\eta s^2 \Phi_2'') \Delta
\]
\[
+ 4s \Phi_2(\eta) \left(2 \frac{\Phi_3(\eta)}{\Phi_2(\eta)} (w_s \ast w_s - w_{v_s} w_{v_s}) + 4s \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) w_\eta + s^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right)^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) w_\eta \ast w_\eta\right) \Delta'
\]
\[
+ s^2 \Phi_2(\eta) \left(2 \frac{\Phi_3(\eta)}{\Phi_2(\eta)} w_s \ast w_s + 8s \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) w_\eta + 4s^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right)^2 w_\eta \ast w_\eta\right) \Delta''
\]
Inserting the convolutions and the bulk value of s gives
\[
\lim_{\rho(r) \to \rho \rho(r_1) \rho(r_2)} \frac{\delta^2 \beta \Delta F_e^{(S)}}{\delta \rho(r_1) \delta \rho(r_2)} = \Theta(\sigma - r_{12}) \Theta(r_{12}/\sigma)
\]
\[
K(x) = 2\pi\sigma \left(x \Phi_2(\eta) + 6\eta (1 - x) \Phi_2'(\eta) + \frac{3}{2} \eta^2 (x - 1)^2 (x + 2) \Phi_2''(\eta)\right) \Delta
\]
\[
+ 48\pi\eta \Phi_2(\eta) \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)} x + 6\eta \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) (1 - x) + \frac{3}{2} \eta^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) w_\eta + \frac{3}{2} \eta^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) w_\eta \ast w_\eta\right) \Delta'
\]
\[
+ 72 \pi\eta^2 \Phi_2(\eta) \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)} \frac{1}{x} + 12\eta \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) (1 - x) + 6\eta^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) w_\eta + 6\eta^2 \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) \left(\frac{\Phi_3(\eta)}{\Phi_2(\eta)}\right) w_\eta \ast w_\eta\right) \Delta''
\]
Finally, using the explicit forms for \(\Phi_2(\eta)\) and \(\Phi_3(\eta)\) and noting that
\[
\frac{\Phi_4(\eta)}{\Phi_2(\eta)} = \frac{1}{12 \frac{1}{\sigma} - \eta}
\]
\[
\frac{\Phi_3(\eta)}{\Phi_2(\eta)} = \frac{\eta}{1 - \eta}
\]
results in the form given in the main text.

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