Harmonic-Potential Theorem: Implications for Approximate Many-Body Theories

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We consider interacting particles in an external harmonic potential. We extend the B = 0 case of the generalized Kohn theorem, giving a "harmonic-potential theorem" (HPT), demonstrating rigid, arbitrary-amplitude, time-oscillatory Schrödinger transport of a many-body eigenfunction. We show *analytically* that the time-dependent local-density approximation (TDLDA) satisfies the HPT exactly. Other approximations, such as linearized TDLDA with frequency-dependent exchange correlation kernel and certain inhomogeneous hydrodynamic formalisms, do not. A simple modification permits such explicitly frequency-dependent local theories to satisfy the HPT, however.

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The Kohn theorem [1] concerns particles of mass m, and charge *e* with arbitrary momentum-conserving interactions plus a static external *B* field: It states that linear response to a uniform electric field $\mathbf{E}_0 \exp(-i\omega t)$ yields a sharp resonance at $\omega = \omega_c = eB/mc$, corresponding to cyclotron motion of the center of mass. Brey et al. [2] added an external scalar harmonic potential $V_{\text{ext}}(\mathbf{r}) = Kz^2/2$ extending throughout all space. Their result (the "generalized Kohn theorem" or "GKT") also predicts sharp resonances for a uniform exciting field. In the electron-gas context, the static external harmonic potential can be generated, via Poisson's equation, by a uniform positive charge background extending throughout space, with charge density $en_0 = \varepsilon K/4\pi e$, and this can be mimicked in GaAlAs quantum wells, wires, and dots [2-6]. (Here ε is the background medium's relative dielectric constant.) This paper concerns only the case B = 0, for which the single GKT resonance frequency $\omega_0 = (K/m^*)^{1/2}$ is also equal to the plasma frequency $\omega_p = (4\pi n_0 e^2/\varepsilon m^*)^{1/2}$ corresponding to the fictitious background density n_0 . The GKT resonance then corresponds to a "sloshing" or transverse plasmon motion of the electron gas.

Note that the GKT does *not* apply to a charge-neutral electron gas as in a neutral metal slab of finite thickness: There the electron gas samples the nonparabolic linear region of external potential lying just outside the jellium edge [4].

There is a fundamental difference between systems covered by the original Kohn theorem and those covered by the GKT: Unlike a *B* field, the harmonic scalar potential confines the density spatially so that, in the context of Coulomb-interacting systems, the GKT applies exactly to *bounded*, *non-neutral* electron gases with strong edge inhomogeneities [4]. The GKT is thus one of the few *exact* results known for the *dynamic* properties of an *inhomogeneous*, *interacting* many-particle system. We will show here that a slightly generalized GKT provides an interesting constraint on the general form of approximate theories of time-dependent phenomena in arbitrary inhomogeneous interacting systems. The GKT Hamiltonian for B = 0 with a spatially homogeneous time-dependent driving field $\mathbf{E} = -\mathbf{F}(t)/e$ is

$$H(\mathbf{r}_{1}, \mathbf{r}_{2}, \dots, \mathbf{r}_{N'}t) = H_{0} - \mathbf{F}(t) \cdot \sum_{j} \mathbf{r}_{j},$$

$$H_{0} = \sum_{j=1}^{N} \left[-\frac{\hbar^{2}}{2m} \left(\frac{\partial}{\partial \mathbf{r}_{j}}\right)^{2} + \frac{1}{2} \mathbf{r}_{j} \cdot \mathbf{K} \cdot \mathbf{r}_{j} \right] \qquad (1)$$

$$+ V(\{\mathbf{r}_{i} - \mathbf{r}_{j}\}).$$

Here V is an arbitrary interparticle potential which depends only on coordinate differences. By suitable choice of the spring-constant matrix **K** one can describe parabolic quantum wells [2-4], non-neutral quantum wires or dots [5,6], positively charged metal spheres, Hooke's atoms [7], or spherical nuclear models [8].

To achieve the desired constraint condition on manybody theories we need the following slight extension of the GKT, which we shall term here the "harmonic potential theorem" (HPT). Let Ψ_0 be any many-body eigenstate of the Hamiltonian H_0 from (1), so that

$$H_0(\{\mathbf{r}_i\})\Psi_0(\{\mathbf{r}_i\}) = E_0\Psi_0(\{\mathbf{r}_i\}).$$
(2)

Consider applying a position-independent, time-dependent shift $\mathbf{x}(t)$, to all coordinates in Ψ_0 . Now construct the following state:

$$\Psi_{\text{HPT}}(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N, t) = \exp(-iE_0\hbar^{-1}t - iNS(t) + iN\hbar^{-1}m\frac{d\mathbf{x}}{dt} \cdot \mathbf{R}) \quad (3)$$
$$\Psi_0(\mathbf{\bar{r}}_1, \mathbf{\bar{r}}_2, \dots, \mathbf{\bar{r}}_N).$$

Here $\bar{\mathbf{r}}_j$ is a position operator relative to a moving origin and **R** is the center of mass operator:

$$\mathbf{\tilde{r}}_j = \mathbf{r}_j - \mathbf{x}(t), \qquad \mathbf{R} = \frac{1}{N} \sum_j \mathbf{r}_j.$$
 (4)

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The phase angle in (3) is

$$S(t) = \hbar^{-1} \int_{t_0}^t \left[\frac{m}{2} \dot{\mathbf{x}}(t')^2 - \frac{1}{2} \mathbf{x}(t') \cdot \mathbf{K} \cdot \mathbf{x}(t') \right] dt'.$$
(5)

By performing space and time differentiation of (3), canceling some equal and opposite terms which arise from differentiation of the phase factor and of the wave function Ψ_0 , and using (2) we obtain

$$\begin{bmatrix} i\hbar \frac{\partial}{\partial t} - H(\{\mathbf{r}_j\}, t) \end{bmatrix} \Psi_{\text{HPT}}(\{\mathbf{r}_j\}, t) = -[m\ddot{\mathbf{x}} + \mathbf{K} \cdot \mathbf{x} - \mathbf{F}(t)] \cdot \left[\sum_{j} \mathbf{r}_{j}\right] \Psi_{\text{HPT}}(\{\mathbf{r}_{i}\}, t). \quad (6)$$

Thus Ψ_{HPT} satisfies the time-dependent many-body Schrödinger equation provided that $\mathbf{x}(t)$ satisfies the classical driven harmonic-oscillator equation

$$m\ddot{\mathbf{x}} = -\mathbf{K} \cdot \mathbf{x} + \mathbf{F}(t). \tag{7}$$

The many-body density $|\Psi_{HPT}({\mathbf{r}_j}, t)|^2$ from (3) is $|\Psi_0({\mathbf{r}_j - \mathbf{x}(t)})|^2$, which is just the many-body density of the stationary state Ψ_0 , rigidity displaced by $\mathbf{x}(t)$. The same holds for the one-body density $n_{\rm HPT}({\bf r},t)$, and indeed for the entire many-body wave function, apart from the phase factor shown explicitly in (3). Note that the center of mass is tied to the accelerated $(\bar{\mathbf{r}})$ frame executing driven oscillator motion, and in this frame (3) is just the stationary state Ψ_0 : The phase factor in (3) transforms one back to the rest frame. Note also that a nontrivial free oscillator motion is demonstrated by (6) when $\mathbf{F}(t) = \mathbf{0}$. These results constitute what we term here the HPT. Much like the exchange (xc) hole normalization theorem in static xc theories, this theorem implies constraints on time-dependent many-body approximations.

We first prove *analytically* that the *unlinearized* version of the standard linearized adiabatic time dependent local density approximation (TDLDA) [9,10] satisfies the HPT. In this formalism the density is formed from time-dependent one-body wave functions satisfying

$$\left(-i\hbar\frac{\partial}{\partial t}-\frac{\hbar^{2}}{2m}\nabla^{2}+V_{\text{ext}}\left(\mathbf{r},t\right)+V_{\text{xc}}\left(\mathbf{r},t\right)\right)\phi_{j}\left(\mathbf{r},t\right)=0,$$
(8)

where the Hartree potential V_H and the xc potential $V_{xc} = \mu_{xc}(n(\mathbf{r}, t))$ are related adiabatically (i.e., at equal time) to the density. In the static case, nonlinear TDLDA reduces to local-density Kohn-Sham (KSLDA) theory, and $\phi_j(\mathbf{r}, t)$ is just $\exp(-i\omega_j t)\phi_j(\mathbf{r})$, where ϕ_j and $\hbar\omega_j$ are the Kohn-Sham eigenfunction and eigenvalue. Let the static KSLDA ground state for the HPT Hamiltonian H_0 have a particle number density $n_0(\mathbf{r})$, occupied wave functions $\phi_j^0(\mathbf{r})$ and eigenvalues $\hbar\omega_j^0$, a Hartree potential $V_{0H}(\mathbf{r})$ and exchange-correlation potential $V_{0xc}(\mathbf{r}) = \mu_{xc}(n_0(\mathbf{r}))$. Now consider the HPT case for which $V_{ext}(\mathbf{r}, t) = (1/2)\mathbf{r} \cdot \mathbf{K} \cdot \mathbf{r} - \mathbf{F}(t) \cdot \mathbf{r}$. The HP the-

orem requires that $n(\mathbf{r}, t) = n^0(\mathbf{r} - \mathbf{x}(t))$ also satisfies the equations, provided $\mathbf{x}(t)$ is an oscillator motion satisfying (7). As noted earlier, in the moving frame the corresponding HPT many-body state is just the ground state which corresponds to KSLDA wave functions $\phi_j^0(\mathbf{\bar{r}})$ with $\mathbf{\bar{r}} = \mathbf{r} - \mathbf{x}$. Transforming back to the rest frame via a phase factor, we try TDLDA wave functions

$$\phi_j(\mathbf{r}, t) = \exp[-iS(t) - i\omega_j t + im\hbar^{-1}\dot{\mathbf{x}} \cdot \mathbf{r}] \times \phi_j^0(\mathbf{r} - \mathbf{x}(t)), \qquad (9)$$

with S given by (5). The wave functions (9) square and sum to the required HPT density $n^{0}(\mathbf{r} - \mathbf{x}(t))$, and the corresponding Hartree potential is $V^0(\mathbf{r} - \mathbf{x}(t))$. In the adiabatic TDLDA for which, crucially, $V_{xc}(\mathbf{r}, t) =$ $\mu_{\rm xc}(n^0(\mathbf{r} - \mathbf{x}(t))) = V_{\rm xc}^0(\mathbf{r} - \mathbf{x}(t))$, one verifies by direct space and time differentiations that (9) satisfies (8) when (7) holds; the working is similar to that for Eq. (6). Thus we have an exact TDLDA solution for HPT motion. By choosing Ψ_0 to be the many-body ground state and then linearizing this solution, we deduce that the small-motion HPT is satisfied by the usual TDLDA [10] which is linearized about the KSLDA ground state. In terms of the bare KSLDA susceptibility (casual densitydensity response) χ_0 formed by perturbation theory [9– 11] from the static Kohn-Sham orbitals, we can express the linearized TDLDA as follows in the time domain:

$$\delta n (\mathbf{r}, t) = \int dt' d\mathbf{r}' \chi_0 (\mathbf{r}, \mathbf{r}', t - t') \times [\delta v^{\text{ext}} (\mathbf{r}, t') + \delta v^H (\mathbf{r}', t') + \delta v^{\text{xc}} (r', t')]. (10)$$

In (10), $\delta v^{\text{xc}}(r,t) = f_{\text{xc}}(n_0(\mathbf{r}))\delta n(\mathbf{r},t)$, where $f_{\text{xc}}(n) = d\mu_{\text{xc}}/dn$. The above rigidly moving HPT-TDLDA solution shows that (10) is satisfied, for a harmonically confines system with $\delta V_{\text{ext}} = -\mathbf{F}(t) \cdot \mathbf{r}$, by a density perturbation

$$\delta n(\mathbf{r},t) = -\mathbf{x}(t) \cdot \nabla n_0(\mathbf{r}), \qquad (11)$$

where $\mathbf{x}(t)$ is a classical motion obeying (7).

Generalizing TDLDA, Gross and Kohn [11] showed that driven time-dependent response starting from the ground state is described exactly by an equation of the form [10] with the same χ_0 , except that f_{xc} is *nonlocal* and *time delayed* (i.e., frequent dependency, in Fourier space). They further proposed keeping the time delay but using the spatial local density approximation, giving $f_{xc}(n_0(\mathbf{r}), \omega)$. This local, time-delayed approximation ("DLDA" for brevity) violates the HPT, as we now show. Consider a HPT system (with slab geometry, $\mathbf{r} = z$, for simplicity) in the ground state. A small steady z-independent force F is switched on at t = 0. The corresponding classical motion from (7) is of the form $\mathbf{x}(t) = \theta(t) a [1 - \cos(\omega_0 t)] \hat{\mathbf{z}}$, with a = const and $\omega_0 = (K/m)^{1/2}$: Equation (11) then gives the density perturbation $\delta n^{\text{HPT}}(\mathbf{r}, t)$, which has to satisfy both (10) and the DLDF version of (10). Subtracting these versions we find

$$0 = \int dt' dz' \chi_0(z, z', t - t') \Delta v^{\rm xc}(z', t'), \qquad (12)$$

where $\Delta v_{xc} = \delta v_{xc}^{DLDA} - \delta v_{xc}^{TDLDA}$. Now (12) represents the difference in linear density response of independent electrons to two different potentials, starting at t = 0with the common ground state. For (12) to be zero, the linear form of the Runge-Gross theorem [see Ref. [12], especially Eq. (6) which is explicitly linear] thus requires Δv_{xc} to be independent of z for t just greater than 0. We show that it is not. For the HPT density perturbation, denoting $f_{xc}^{DLDA} - f_{xc}^{TDLDA} = \Delta f_{xc}$, we have

$$\Delta \boldsymbol{v}_{xc}(z,t) = \int_{0}^{\infty} \Delta f_{xc}(\boldsymbol{n}_{0}(z), t - t') \times \left(a \frac{d\boldsymbol{n}_{0}}{dz} \left\{ \cos(\omega_{0}t') - 1 \right\} \right) dt'. \quad (13)$$

Now data in Ref. [11] imply that $\Delta f_{xc}(n,\tau) = \theta(\tau - 0^{-})[A(n)\delta(\tau) + B(\tau)]$, where $A(n) = f_{xc}(n, \omega \rightarrow \infty) - f_{xc}(n, \omega = 0)$ is positive for any n > 0, and $B(\tau)$ depends nonsingularly on time for $\tau > 0$. As $t \rightarrow 0$, (13) then gives

$$\Delta v_{\rm xc}(z,t) = -\frac{1}{2}\omega_0^2 t^2 a A(n_0(z)) \frac{dn_0}{dz} + O(t^3). \quad (14)$$

Equation (14) vanishes only at extrema of the (inhomogeneous) ground-state density where $dn_0/dz = 0$, so (14) *does* depend on z. Thus DLDA does not satisfy the HPT. (A minor modification allows it to do so, however; see below.)

Another case of HPT incompatibility occurs in inhomogeneous hydrodynamics, which can be regarded as a frequency-dependent LDA for the kinetic energy. The lowest-order inhomogeneous hydrodynamics theory [13–15] can be motivated starting from a Thomas-Fermi [13] condition $V_0(\mathbf{r}) + \varepsilon_F(n_0(\mathbf{r})) = \mu$ for the ground state. Taking a spatial gradient one obtains

$$F_{0\text{ext}}(\mathbf{r}) + \mathbf{F}_{0\text{int}}(\mathbf{r}) - \alpha \frac{\hbar^2}{2m} \nabla \left\{ \left[3\pi^2 n_0(\mathbf{r}) \right]^{2/3} \right\} = \mathbf{0}, \ (15)$$

where $\alpha = 1$ and, in the present case, $\mathbf{F}_{0\text{ ext}} = -\mathbf{K} \cdot \mathbf{r}$ is the Hooke's-law force arising from the static external potential in (1). $\mathbf{F}_{0\text{ int}}$ is the self-consistent Hartree force. One guesses that, in a dynamic situation, a "force" of the form of the left side of Eq. (15) equals the mass times the convective acceleration. Then linearizing in the small density perturbation $\delta n(\mathbf{r}, t)$ and fluid velocity \mathbf{u} one finds

$$\delta \mathbf{F}(\mathbf{r},t) - \frac{2}{3} \alpha' \gamma \nabla \Big[n^0 (\mathbf{r})^{-1/3} \, \delta n \Big] = m \frac{\partial \mathbf{u}}{\partial t} \,, \qquad (16)$$

where $\gamma = \hbar^2 (2m)^{-1} (3\pi^2)^{2/3}$. In (16), the above motivation suggests the static value $\alpha' = \alpha = 1$. However, if (16) is used, with the Poisson and continuity equations,

for unforced motions at frequency ω in the *uniform* electron gas, it yields

$$\left[\beta^2\nabla^2+\omega^2-\omega_p^2\right]\delta n=0.$$

with $\beta^2 = \alpha' v_F^2/3$. But it is well known [14,15] from microscopic Lindhard response theory that high-frequency phenomena (e.g., plasmon dispersion) in a uniform gas are properly described by $\beta^2 = 3v_F^2/5$, corresponding to $\alpha' = 9/5$. Thus a frequency-dependent coefficient α is needed if both the static and high-frequency limits are to be described.

Within dispersive hydrodynamics, one commonly [14-16] described jellium via the uniform-gas dynamic equation (17) or similar, while description of edge inhomogeneities, both in the ground state and the dynamics, is avoided and replaced by boundary conditions at the edge. This approach is not of interest here, because we wish to see what can be learned by demanding that the HPT be satisfied, and this theorem concerns rigid displacement of a self-consistent inhomogeneous groundstate density. Thus the only suitable hydrodynamic theories are those which consistently describe both the inhomogeneous ground state [13,17] and the motions around it. We will find that the effective frequency dependent of α , while ensuring a correct uniform gas behavior, causes violation of the HPT for a class of systems including inhomogeneous non-neutral electron gases.

To see this, consider [13,17] the self-consistent inhomogeneous ground state density $n_0(\mathbf{r})$ satisfying the Thomas-Fermi equation for the HPT Hamiltonian H_0 . In free [$\mathbf{F}(t) = 0$] HPT motion, $n(\mathbf{r}, t) = n_0(\mathbf{r} - \mathbf{x}(t))$ yielding a Hartree force $\mathbf{F}_{int}(\mathbf{r}, t) = \mathbf{F}_{0int}(\mathbf{r} - \mathbf{x}(t))$ while \mathbf{F}_{ext} remains fixed at $\mathbf{F}_{0ext}(\mathbf{r})$. The fluid velocity is $\mathbf{u} = \partial \mathbf{x}/\partial t$ and the continuity equation is satisfied identically by the present $n(\mathbf{r}, t)$. Linearizing F_{int} and using (15), we have

$$\delta \mathbf{F} = -\mathbf{x}(t) \cdot \nabla \mathbf{F}_{0 \text{ int}}(\mathbf{r})$$

= $-\mathbf{x}(t) \cdot \nabla \Big[-\mathbf{F}_{0 \text{ ext}} + \alpha \gamma \nabla \Big(n_0^{2/3} \Big) \Big]$
= $-\mathbf{K} \cdot \mathbf{x}(t) - \alpha \gamma \mathbf{x}(t) \cdot \nabla \Big[\nabla \Big(n_0^{2/3} \Big) \Big].$ (18)

Using (18), (11), and (7) and noting $[\mathbf{x}, \nabla] = \mathbf{0}$ we find (16) is satisfied only if

$$(\boldsymbol{\alpha} - \boldsymbol{\alpha}') \mathbf{x} \cdot \nabla \Big[\nabla \Big(n_0 (\mathbf{r})^{2/3} \Big) \Big] = \mathbf{0}.$$
 (19)

For inhomogeneous ground states considered here, $\mathbf{x} \cdot \nabla[\nabla(n_0(\mathbf{r})^{2/3})]$ is nonzero for at least some values of \mathbf{r} , so that (19) gives $\alpha' = \alpha$ (frequency-independent pressure coefficient). But we have just seen that a correct description of the uniform gas requires $\alpha' = (9/5)\alpha$, so the inhomogeneous theory violates either the HPT of the uniform-gas limit. Hence, like the DLDA case studied above, our hydrodynamics violates the HPT because of

an explicit frequency dependence which was introduced [11,15] in order to model the uniform electron gas correctly.

A possible correction for this problem is as follows: In HPT motion the *relative* motion of the particles is as in the static equilibrium solution: thus the $\omega = 0$ coefficients α and f_{xc} are appropriate. In more general cases where the potential is not harmonic, or for non-HPT motions in a harmonic potential, one must separate out parts of the motion which merely translate the system from those which compress the system and/or rearrange the particle motions; only the latter should be described by finite- ω coefficients.

To achieve this, we first solve the relevant static equilibrium problem to obtain an inhomogeneous density $n_0(\mathbf{r})$. In an arbitrary perturbed state the linearized local fluid displacement $\mathbf{x}(\mathbf{r}, t)$ is defined by

$$\mathbf{x}(\mathbf{r},t) = \int_{t_0}^t \mathbf{u}(\mathbf{r},t') \, dt', \qquad \mathbf{u} = \frac{\partial \mathbf{x}}{\partial t}, \qquad (20)$$

where t_0 is the initial equilibrium time (e.g., $-\infty$). $\mathbf{u}(\mathbf{r}, t) = \mathbf{J}(\mathbf{r}, t)/n(\mathbf{r}, t)$ is the fluid velocity. In what follows we change to $\mathbf{x}(\mathbf{r}, t)$ as the basic variable. Time integration of the linearized continuity equation, with use of (20), shows that the density is recovered from \mathbf{x} by

$$\delta n(\mathbf{r},t) = -\nabla \cdot [n_0(\mathbf{r}) \mathbf{x}(\mathbf{r},t)]$$

= $\delta n_1(\mathbf{r},t) + \delta n_2(\mathbf{r},t),$ (21)

where

$$\delta n_2(\mathbf{r},t) = -\mathbf{x} (\mathbf{r},t) \cdot \nabla n_0(\mathbf{r})$$

$$\approx n_0(\mathbf{r} - \mathbf{x} (\mathbf{r},t)) - n_0(\mathbf{r}), \qquad (22)$$

$$\delta n_1(\mathbf{r},t) = -n_0(\mathbf{r}) \nabla \cdot \mathbf{x}(\mathbf{r},t)$$

$$\approx n(\mathbf{r},t) - n_0(\mathbf{r} - \mathbf{x}(\mathbf{r},t)), \qquad (22)$$

The idea is that δn_2 represents changes due to simple displacement of the local equilibrium density. Thus, for pressure terms in hydrodynamics or xc terms in the DLDF theory, δn_2 is associated with static coefficients $[\alpha = 1, f_{xc}(\omega = 0)]$ while δn_1 is associated with dynamic coefficients $[\alpha' = 9/5, f_{xc}(\omega)]$. Hence, in hydrodynamics, the Euler equation (16) is replaced by

$$\delta \mathbf{F}(\mathbf{r}) - \frac{2}{3} \gamma \nabla \left[n_0 \left(\mathbf{r} \right)^{-1/3} \left(\delta n_2 + \frac{9}{5} \delta n_1 \right) \right]$$
$$= m \frac{\partial \mathbf{u}}{\partial t}, \quad (24)$$

where γ is defined following Eq. (16). Similarly, the xc potential of DLDF theory [11] becomes, in Fourier space,

$$\delta V_{\rm xc}(\mathbf{r},\omega) = f_{\rm xc}(n_0(\mathbf{r}),\omega=0)\delta n_2 + f_{\rm xc}(n_0(\mathbf{r}),\omega)\delta n_1(\mathbf{r},\omega).$$
(25)

Note that δn_1 is zero for HPT motion, so that (25) reduces to TDLDA which uses the zero-frequency xc coefficient $f_{xc}(n, \omega = 0) = d\mu_{xc}/dn$. Similarly δn_2 is zero for the uniform gas, leading to a dynamic value of f_{xc} . Thus both these special cases are treated with the appropriate response formula. Equations (24) and (25) are not limited to these extreme cases, but they are merely suggestions and will require further investigation.

In summary, we have derived exact results for a harmonic external potential and used them to highlight a problem with local density formalisms containing frequency-dependent coefficients. The problem arises when one attempts to make the inhomogeneous groundstate calculation consistent with the dynamic response, while still preserving known results for the uniform-gas and harmonic-confinement cases. A simple remedy has been suggested for the difficulty, providing a basis for further investigation.

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