Supplemental Material for:
Exact Generalized Kohn-Sham Theory for Hybrid Functionals

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I. Comparison of inverted KS potentials with previous results.

Figure S1: Comparison of KS potentials obtained from inversion in the present work with those obtained previously by Gould and Toulouse. [1]
II. Removal of noise from inverted potentials

In the main text, numerical noise arising from convergence difficulties at the empty focus of the prolate spheroidal grid was removed from $V_{R,xc}^\alpha$ (see Fig. 2). Here we provide the corresponding raw (noise-including) data for comparison.

Figure S2: Raw (noise-including) Exact remainder exchange-correlation potentials, $V_{R,xc}^\alpha$, found by inversion for GKS maps with various fractions $\alpha$ of Fock exchange, for: (a) Li$^+$, (b) Li$, (c)$ Be, and (d) F$^-$. 
III. Differences in exact GKS orbitals obtained from different fractions of Fock exchange.

Figure S3: $R^2 (\phi_\alpha^2 - \phi_{\alpha=0}^2)$, where $\phi_\alpha(r)$ is the exact GKS orbital obtained from inversion with $\alpha$ fraction of Fock exchange, for the two occupied orbitals of Be. Note small scale of differences, indicating a very weak dependence of the orbitals on the choice of $\alpha$. 
IV. Comparison of exact and approximate GKS potentials: \( \text{Li}^+, \text{Li}^-, \text{F}^- \)

Figure S4: Exact \( V_{R,xc}^\alpha \) of \( \text{Li}^+ \) obtained from inversion, compared with that obtained from PBE0, for various values of \( \alpha \).

- (a) \( \alpha = 0.00 \)
- (b) \( \alpha = 0.25 \)
- (c) \( \alpha = 0.50 \)
- (d) \( \alpha = 0.75 \)
- (e) \( \alpha = 1.00 \)
Figure S5: Exact $V_{R,xc}^\alpha$ of Li obtained from inversion, compared with that obtained from PBE0, for various values of $\alpha$.

Figure S6: Exact $V_{R,xc}^\alpha$ of F obtained from inversion, compared with that obtained from PBE0, for various values of $\alpha$. 

V. Decomposition of differences in $V_{Rxc}^\alpha$

By evaluating the PBE0 functional non-self-consistently on the exact density, we can decompose the total difference between the PBE0 and exact remainder exchange correlation potentials ($V_{\text{PBE0}[n_{\text{PBE0}}]} - V_{\text{exact}[n_{\text{exact}}]}$) into a functional-driven difference ($V_{\text{PBE0}[n_{\text{exact}}]} - V_{\text{exact}[n_{\text{exact}}]}$) and an exact density-driven difference ($V_{\text{PBE0}[n_{\text{PBE0}}]} - V_{\text{PBE0}[n_{\text{exact}}]}$). Figure S7 establishes that the overwhelming majority of the total difference in the systems studied is due to the functional, rather than the density.

Figure S7: Total difference ($V_{\text{PBE0}[n_{\text{PBE0}}]} - V_{\text{exact}[n_{\text{exact}}]}$), functional-driven difference ($V_{\text{PBE0}[n_{\text{exact}}]} - V_{\text{exact}[n_{\text{exact}}]}$), and density-driven difference ($V_{\text{PBE0}[n_{\text{PBE0}}]} - V_{\text{PBE0}[n_{\text{exact}}]}$) of remainder exchange-correlations potentials for Be.

REFERENCE:

[1] T. Gould and J. Toulouse, "Kohn-Sham potentials in exact density-functional theory at noninteger electron numbers," *Phys. Rev. A*, 90, 050502 (2014).