

Dynamical Correction to Linear Kohn-Sham Conductances from Static Density Functional Theory

S. Kurth^{1,2,3} and G. Stefanucci^{4,5,3}

¹*Nano-Bio Spectroscopy Group, Departamento de Física de Materiales, Universidad del País Vasco UPV/EHU, Centro Física de Materiales CSIC-UPV/EHU, Avenida Tolosa 72, E-20018 San Sebastián, Spain*

²*IKERBASQUE, Basque Foundation for Science, E-48011 Bilbao, Spain*

³*European Theoretical Spectroscopy Facility (ETSF)*

⁴*Dipartimento di Fisica, Università di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy*

⁵*INFN, Laboratori Nazionali di Frascati, Via E. Fermi 40, 00044 Frascati, Italy*

(Received 18 February 2013; revised manuscript received 30 May 2013; published 19 July 2013)

For molecules weakly coupled to leads the *exact* linear Kohn-Sham (KS) conductance can be orders of magnitude larger than the true linear conductance due to the lack of *dynamical* exchange-correlation (xc) corrections. In this work we show how to incorporate dynamical effects in KS transport calculations. The only quantity needed is the *static* xc potential in the molecular junction. Our scheme provides a comprehensive description of Coulomb blockade without breaking the spin symmetry. This is explicitly demonstrated in single-wall nanotubes where the corrected conductance is in good agreement with experimental data whereas the KS conductance fails dramatically.

DOI: [10.1103/PhysRevLett.111.030601](https://doi.org/10.1103/PhysRevLett.111.030601)

PACS numbers: 05.60.Gg, 31.15.ee, 71.15.Mb, 73.63.-b

The active field of molecular electronics [1] remains a challenge for *ab initio* methods. Density functional theory (DFT) is at present the only viable route for an atomistic description of complex molecular junctions. Nevertheless, DFT transport calculations still suffer from some practical difficulties. The fundamental sources of error in the linear conductance are the DFT exchange-correlation (xc) potential used to determine the Kohn-Sham (KS) conductance G_s and the dynamical xc correction [2–5] predicted by time-dependent (TD) DFT [6] [see Eq. (9) below]. Assessing their importance and mutual interplay is especially thorny in weakly coupled molecules where level alignment and charging effects play a prominent role. Toher *et al.* [7] and Koentopp *et al.* [4] showed that G_s evaluated with an accurate, and hence discontinuous [8], xc potential is suppressed, thus capturing the Coulomb blockade (CB) effect at even electron numbers N (closed shell). This may suggest the dynamical xc correction to be small. However, at odd N (open shell) G_s can be orders of magnitude larger than the true conductance G , even when the *exact* xc potential is employed [9]. A satisfactory “DFT explanation” of CB is, therefore, currently missing. In this Letter we provide a comprehensive picture of CB, valid for all N without breaking the spin symmetry. The key ingredient is the dynamical xc correction which, remarkably, can be expressed exclusively in terms of *static* DFT quantities. We propose a practical scheme to calculate G and demonstrate its validity by comparison with recent experiments on single-wall nanotubes.

At zero temperature and when transport is dominated by a single resonance (open shell) $G_s = G$ due to the Friedel sum rule [10]. In this regime G exhibits a Kondo plateau (no CB peaks) and the discontinuity is essential for G_s to

reproduce the plateau [9,11,12]. At temperatures higher than the Kondo temperature T_K , the exact discontinuous xc potential gives instead a $G_s \gg G$ [9]. To understand this discrepancy we model the resonance with a single level (HOMO or LUMO) of energy ν and Coulomb repulsion U coupled to left (L) and right (R) featureless leads contributing $\gamma = \gamma_L + \gamma_R$ to the broadening of the spectral peaks. Given the many-body (MB) spectral function $A(\omega)$ the number of electrons is

$$N = 2 \int f(\omega)A(\omega), \quad \int \equiv \int \frac{d\omega}{2\pi}, \quad (1)$$

whereas the linear (zero-bias) conductance reads

$$G = -2 \frac{\gamma_L \gamma_R}{\gamma} \int f'(\omega)A(\omega) \quad (2)$$

with the Fermi function $f(\omega) = 1/(e^{\beta(\omega-\mu)} + 1)$ at inverse temperature $\beta = 1/T$ and chemical potential μ . At $T \gg T_K$ the Abrikosov-Suhl (AS) resonance is suppressed and the spectral function is well represented by [13]

$$A(\omega) = nL_\gamma(\omega - \nu - U) + (1 - n)L_\gamma(\omega - \nu), \quad (3)$$

where $n = N/2$ and $L_\gamma(\omega) = \gamma/(\omega^2 + \gamma^2/4)$. For the KS system the spectral function is $A_s(\omega) = L_\gamma(\omega - \nu - \nu_{\text{Hxc}}[N])$. The Hartree-xc (Hxc) potential ν_{Hxc} is such that the number of electrons N which solves $N = 2 \int f(\omega)A_s(\omega)$ is the same as in Eq. (1). We obtain ν_{Hxc} by reverse engineering and find that, as function of N , it has the shape of a smeared step function (not shown). The smearing is due to the level broadening γ induced by contacting the level to the leads and develops into a true discontinuity (at $N = 1$) only in the limit $\gamma \rightarrow 0$, as it

should [11,12]. This v_{Hxc} is then used to calculate the KS conductance from

$$G_s = -2 \frac{\gamma_L \gamma_R}{\gamma} \int f'(\omega) A_s(\omega). \quad (4)$$

Despite the fact that the MB and DFT N - v curves are *identical*, see Fig. 1 top, the CB peaks present in G are completely absent in G_s ; see Fig. 1 bottom. The physical situation discussed here is distinct from that of Ref. [7] where the discontinuity keeps the HOMO doubly occupied and the LUMO empty when gating the molecule (closed shell). The discontinuity correctly suppresses G_s at even N but has the opposite effect at odd N .

Dynamical xc effects.—Open-shell molecules in the CB regime are probably the most striking example of the inadequacy of standard DFT transport calculations. Below we derive an exact formula for G in terms of TDDFT quantities. We take the leads as two jellia (the argument can be generalized to more realistic leads) and choose z as the longitudinal coordinate so that $z \rightarrow -\infty$ is in the left lead, $\alpha = L$, whereas $z \rightarrow \infty$ is in the right lead, $\alpha = R$. Let δV^α be the variation in the classical potential (external plus Hartree) of lead α . This perturbation generates a current [2,4]

$$\delta I = (\delta V^R - \delta V^L + \delta V_{\text{xc}}^R - \delta V_{\text{xc}}^L) G_s, \quad (5)$$

where $\delta V_{\text{xc}}^\alpha = \lim_{t \rightarrow \infty} \lim_{z \rightarrow s_\alpha \infty} \delta v_{\text{xc}}(\mathbf{r}, t)$, $s_{R/L} = \pm$ is the asymptotic value of the variation of the xc potential δv_{xc} . From linear-response TDDFT

$$\delta V_{\text{xc}}^\alpha = \int dt' d\mathbf{r}' \lim_{t \rightarrow \infty} \lim_{z \rightarrow s_\alpha \infty} f_{\text{xc}}(\mathbf{r}, \mathbf{r}'; t - t') \delta n(\mathbf{r}', t'), \quad (6)$$

where f_{xc} is the TDDFT kernel and $\delta n(\mathbf{r}, t)$ is the density variation. The assumption of a steady state implies that $f_{\text{xc}} \rightarrow 0$ for $|t - t'| \rightarrow \infty$ and $\delta n(\mathbf{r}, t \rightarrow \infty) = s_\alpha \delta n$

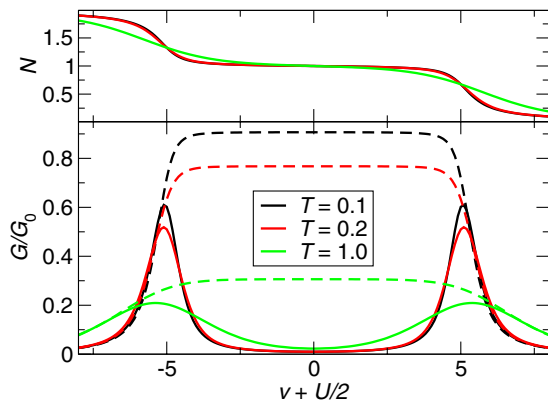


FIG. 1 (color online). Top: electron number N versus gate in MB and DFT (indistinguishable) for a single level coupled to featureless leads with $U = 10$, $\mu = 0$ at various temperatures T (all energies in units of γ). For these parameters $T_K = \sqrt{U\gamma} \exp(-(\pi U/8\gamma)) \approx 0.06$. Bottom: G from Eq. (2) (solid line) and G_s from Eq. (4) (dashed line) in units of $G_0 = 2e^2/h$.

for \mathbf{r} in lead α . In Eq. (6) the contribution of the molecular region to the spatial integral is negligible in the thermodynamic limit. If we define

$$f_{\text{xc}}^{\alpha\beta} = \int dt' \int_{\text{lead } \beta} d\mathbf{r}' \lim_{z \rightarrow s_\alpha \infty} f_{\text{xc}}(\mathbf{r}, \mathbf{r}'; t'), \quad (7)$$

then $\delta V_{\text{xc}}^\alpha = \sum_{\beta=L,R} f_{\text{xc}}^{\alpha\beta} s_\beta \delta n$. We emphasize that $f_{\text{xc}}^{\alpha\beta}$ is not the static DFT kernel since the limit $t \rightarrow \infty$ is taken *after* the limit $|z| \rightarrow \infty$ and these two limits, in general, do not commute [14]. This implies that we cannot model $f_{\text{xc}}^{\alpha\beta}$ by performing DFT calculations on leads of finite length. Inserting the expression for $\delta V_{\text{xc}}^\alpha$ into Eq. (5) we find $\delta I = (\delta V^L - \delta V^R) G_s - \Phi G_s \delta n$ where

$$\Phi \equiv f_{\text{xc}}^{RL} + f_{\text{xc}}^{LR} - f_{\text{xc}}^{RR} - f_{\text{xc}}^{LL}. \quad (8)$$

The expression for δI is correctly gauge invariant. The kernel f_{xc} is defined up to the addition of an arbitrary function $q(\mathbf{r}) + q(\mathbf{r}')$ [15] and Φ is invariant under this transformation. In conclusion

$$G \equiv \frac{\delta I}{(\delta V^R - \delta V^L)} = \frac{G_s}{1 + \chi \Phi G_s}. \quad (9)$$

The quantity $\chi \equiv \delta n / \delta I \approx 1 / (v_F \sigma)$ with v_F the Fermi velocity and σ the cross section of the leads [16]. In the following we define $\chi \Phi G_s$ as the *dynamical xc correction* since Φ is expressed in terms of the TDDFT kernel.

Approximations to Φ .—To gain some insight into the density dependence of $\chi \Phi$ we consider again the single level model. For $N \neq 1$ and $T < \gamma$ the real and KS systems respond similarly and consequently $G \approx G_s$. On the other hand for $N = 1$ we have $G \approx 0$ whereas $G_s \approx G_0 = 2e^2/h$ the quantum of conductance. Therefore $\chi \Phi$ is small for $N \neq 1$ and large for $N = 1$. Interestingly the quantity $\partial v_{\text{Hxc}} / \partial N$ behaves similarly. Is there any relation between $\chi \Phi$ and $\partial v_{\text{Hxc}} / \partial N$? If so this relation would simplify enormously the problem of estimating the dynamical xc correction since $\partial v_{\text{Hxc}} / \partial N$ can be calculated from static DFT. In the following we show that in the CB regime this relation does actually exist.

Consider the system in equilibrium. Using Eq. (1) the compressibility $\kappa = \partial N / \partial \mu$ can be written as $\kappa = \gamma / (\gamma_L \gamma_R) G + 2 \int f(\omega) \partial A(\omega) / \partial \mu$, where we identified the conductance G of Eq. (2). If we define the quantity $R \equiv -2 \int f(\omega) \partial A(\omega) / \partial N$ then $\kappa = \gamma / (\gamma_L \gamma_R) G / (1 + R)$. As the MB and DFT densities are the same, the MB and DFT compressibilities are the same too. Hence $\kappa = \gamma / (\gamma_L \gamma_R) G_s + 2 \int f(\omega) \partial A_s(\omega) / \partial \mu$, where we identified the KS conductance G_s of Eq. (4). The KS spectral function depends on μ through N , and the dependence on N is all contained in v_{Hxc} . Since $\partial A_s / \partial v_{\text{Hxc}} = -\partial A_s / \partial \omega$ [see definition of A_s below Eq. (3)] we have $\partial A_s / \partial \mu = -(\partial A_s / \partial \omega) (\partial v_{\text{Hxc}} / \partial N) (\partial N / \partial \mu)$. Using this result under the integral, solving for κ and equating the MB and DFT expressions one easily obtains

$$\frac{G}{G_s} = \frac{1 + R}{1 + \frac{\gamma}{\gamma_L \gamma_R} G_s \frac{\partial v_{\text{Hxc}}}{\partial N}}. \quad (10)$$

No approximations have been made so far. Let us study the dependence of R on temperature.

We first consider the low temperature case. For simplicity we take $\gamma_L = \gamma_R$ and set $v = -U/2$ at the particle-hole (ph) symmetric point (half-filling). At zero temperature $G = G_s = G_0$ and hence $R = R_0 \equiv (4G_0/\gamma)(\partial v_{\text{Hxc}}/\partial N)$ [17]. For temperatures $T > T_K$ the AS resonance broadens and its height decreases as $h(T/T_K)$ where h is a universal function which approaches zero at high T [21]. This means that $R \approx h(T/T_K)R_0$ remains large until the AS resonance disappears. No simple relation between Φ and $\partial v_{\text{Hxc}}/\partial N$ exists when Kondo correlations are present.

At temperatures $T \gg T_K$ thermal fluctuations destroy the Kondo effect and the MB spectral function is well approximated by Eq. (3). Therefore $R(v) = I(v) - I(v+U)$ where $I(E) \equiv \int f(\omega) L_\gamma(\omega - E)$. We can derive a more convenient expression for R by inserting Eq. (3) into Eq. (1) to find

$$N = \frac{2I(v)}{1 + I(v) - I(v+U)}, \quad (11)$$

and hence $1 + R = 2I(v)/N$. Unfortunately $I(v)$ is not an explicit function of N due to the implicit dependence of $v = v[N]$. However, for $v < \mu$, or equivalently for $N < 1$, we have $I(v+U) \ll 1$. Thus for $N < 1$ we can write $N \approx 2I(v)/(1 + I(v))$ from which we infer $I(v) \approx N/(2 - N)$. Using ph symmetry we therefore approximate R by the explicit function $1 + R = 2/(1 + |\delta N|)$ where $\delta N = N - 1$. Inserting this into Eq. (10) we deduce the main result of this Letter

$$\frac{G}{G_s} = \frac{2}{1 + |\delta N|} \frac{1}{1 + \frac{\gamma}{\gamma_L \gamma_R} G_s \frac{\partial v_{\text{Hxc}}}{\partial N}}. \quad (12)$$

Equation (12) provides a simple and implementable formula to correct the KS conductance. In fact, the dynamical xc correction of Eq. (9) is entirely expressed in terms of *static* DFT quantities. Moreover, whereas Φ involves the TDDFT kernel with coordinates in the *leads* the correction in Eq. (12) involves only the DFT v_{Hxc} in the *molecular junction*. The accuracy of Eq. (12) is examined in Fig. 2, and benchmarked against the MB conductance of Eq. (2). Even though the approximate R is not on top of the exact one, see inset, the agreement between the two conductances is extremely good. The position, width, and height of the peaks as well as the decay for large $|v|$ are all well reproduced. Most importantly the plateau of G_s , see Fig. 1, is completely gone.

Application to physical systems.—In real molecules v_{Hxc} is an \mathbf{r} -dependent functional of the density. We write $v_{\text{Hxc}}(\mathbf{r}) = \delta v_{\text{Hxc}}(\mathbf{r}) + \bar{v}_{\text{Hxc}}$ as the sum of a functional δv_{Hxc} with a weak dependence on $N = \int_V d\mathbf{r}n(\mathbf{r})$ and

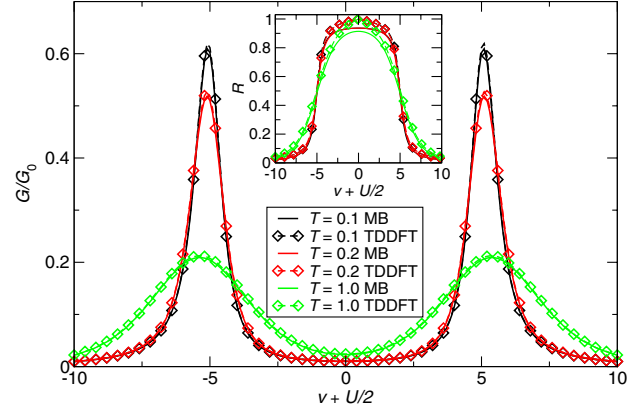


FIG. 2 (color online). Linear conductance from Eq. (2) using the spectral function Eq. (3) (MB, solid line) and from Eq. (12) (TDDFT, dashed line). The inset shows a comparison between the exact and the approximate R . Same parameters as in Fig. 1.

a spatially uniform part $\bar{v}_{\text{Hxc}} = 1/V \int_V d\mathbf{r}v_{\text{Hxc}}(\mathbf{r})$, where the integral is over the volume V of the molecular junction. For weakly coupled molecules \bar{v}_{Hxc} exhibits sharp steps as function of N when N crosses an integer. If we are at resonance and spin fluctuations are suppressed (no Kondo effect) then the KS conductance must be corrected according to Eq. (12) in which $\partial v_{\text{Hxc}}/\partial N \rightarrow \partial \bar{v}_{\text{Hxc}}/\partial N$. Below we argue that this correction applies out of resonance too. Let μ be in the HOMO-LUMO gap and consider a two-level system with Γ_α the 2×2 broadening matrix. For general Γ_α no simple analytic relation between G and N exists. However if $\Gamma_{\alpha,ml} = (\gamma_\alpha/2)\delta_{ml}$ then $N = 2 \int f(\omega) \text{Tr}[A(\omega)]$ and $G = -2(\gamma_L \gamma_R/\gamma) \times \int f'(\omega) \text{Tr}[A(\omega)]$. Discarding the dependence of δv_{Hxc} on μ (which is weak by definition) we can go through the same steps of the single-level derivation and find again Eq. (12). It is therefore reasonable to expect that the KS conductance should be corrected even out of resonance (closed shell) and that this correction should be proportional to $G_s \partial \bar{v}_{\text{Hxc}}/\partial N$.

We here propose a practical scheme to calculate G from DFT. Given the KS Hamiltonian matrix $h_{\text{KS},ml} = \delta_{ml}\epsilon_l$ and the broadening matrices $\Gamma_{\alpha,ml}$ we determine the density and G_s in the usual manner. G is then obtained from Eq. (12) where δN is the deviation of $(N - \text{Int}[N])$ from 1 whereas $\gamma_\alpha = \gamma_\alpha(N) = \Gamma_{\alpha,HH}$ if $\mu \approx \epsilon_H$ (resonance, open shell) and $\gamma_\alpha(N) = 1/2(\Gamma_{\alpha,HH} + \Gamma_{\alpha,LL})$ if $\mu \approx 1/2(\epsilon_L + \epsilon_H)$ is in the HOMO-LUMO gap (out of resonance, closed shell). One could improve the approximation to γ_α using different weights, but the qualitative features of the results are independent of these details.

To appreciate the decisive impact of the dynamical xc correction we consider two paradigmatic junctions in which δv_{Hxc} can be discarded. For \bar{v}_{Hxc} we choose a best fit of the zero-temperature limit of the single-level Hxc potential, but now sum over all possible charged states of the molecule [22], i.e.,

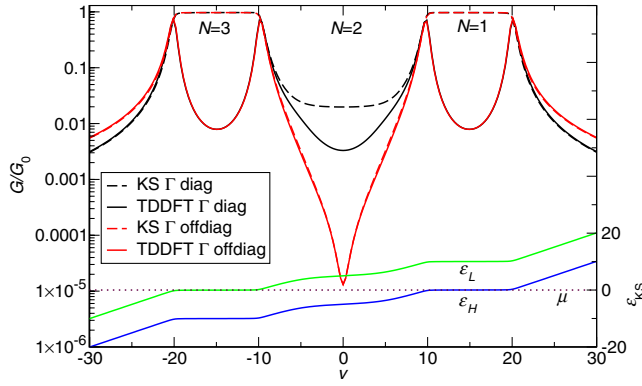


FIG. 3 (color online). Linear conductance from Eq. (4) (KS, dashed line) and from Eq. (12) (TDDFT, solid line) for the HOMO-LUMO model with diagonal and off-diagonal Γ matrices (left axis) and KS energies $\epsilon_{H/L} = \epsilon_{0H/L} + \bar{v}_{Hxc}$ (right axis). The electron number N for different ranges of v is also indicated.

$$\bar{v}_{Hxc} = \sum_K \frac{U(K)}{\pi} \arctan\left(\frac{N-K}{W(K)}\right). \quad (13)$$

The charging energies $U(N)$ are given by the xc part of the derivative discontinuity of the molecule with N electrons [8]. For the widths we take $W(N) = 0.16\gamma(N)/U(N)$ which is consistent with Ref. [19].

HOMO-LUMO model.—We study a two-level system with two electrons in the HOMO in the charge neutral state. Let $\epsilon_{0H} = -\epsilon_{0L} = -\epsilon_0 < 0$ be the noninteracting single-particle energies,

$$\Gamma_L = \Gamma_R = \gamma/2 \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}$$

and $U(N) = U$ independent of N . We solve the self-consistent equation for the density with $U = 10$, $\epsilon_0 = 5$, $\mu = 0$, and $\beta = 10$ (all energies in units of γ). G_s and G from Eq. (12) are shown in Fig. 3 (left axis). As expected the discontinuity of v_{Hxc} opens a gap in G_s for even N , in agreement with the results of Ref. [7]. Here the dynamical xc correction only weakly affects G_s since $\partial\bar{v}_{Hxc}/\partial N$ is multiplied by $G_s \ll 1$. For odd N the KS conductance exhibits a Kondo plateau due to the pinning of the KS level to μ ; see right axis. This is the regime previously discussed and no CB is observed. The dynamical xc correction remedies this serious deficiency by correctly suppressing the plateau. The results remain essentially unaltered if the off-diagonal matrix elements of Γ_α are discarded [23].

SWNT.—Experimental evidence of CB oscillations has recently been reported in metallic single-wall nanotubes (SWNT) quantum dots [24–26]. We now analyze the performance of Eq. (12) in these systems. The finite length of the SWNT causes a level quantization of the twofold degenerate bands. Since the wave vector is a good quantum number our approximation $\delta v_{Hxc} = 0$ is justified. For a SWNT quantum dot the constant interaction model [27]

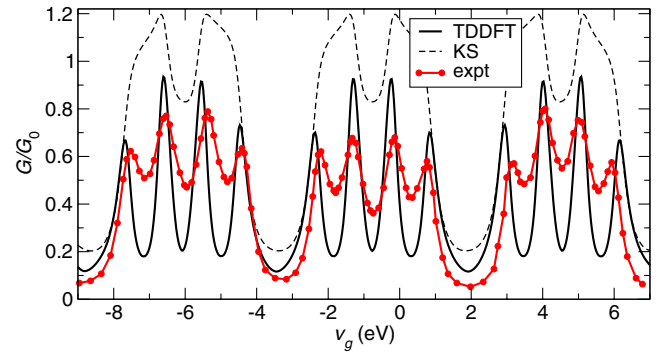


FIG. 4 (color online). Linear KS and TDDFT conductance [Eq. (12)] for a SWNT quantum dot in comparison to experimental conductance from Ref. [24], as function of gate voltage v_g .

has been refined by Oreg *et al.* [28] to account for the observed fourfold periodicity in the electron addition energy. We constructed the KS Hamiltonian corresponding to this model and approximated the broadening matrix as $\Gamma_{\alpha,ml} = (\gamma/2)\delta_{ml}$ [29] (no visible interference [23] from experiment). In Fig. 4 we compare the KS, TDDFT, and experimental conductance versus the gate voltage v_g . We clearly see that the conductance of Eq. (12) correctly exhibits the fourfold periodicity and represents a considerable improvement over G_s which, instead, shows two deformed Kondo plateaus per period. The qualitative behavior of G and G_s does not change by varying the parameters within a reasonable range around the average values reported in Ref. [24].

In conclusion we proposed a practical scheme to correct KS conductances. We highlighted the role of the discontinuity not only for an accurate G_s but also for an accurate dynamical xc correction to G . Approaches to generate discontinuous xc potentials are emerging both in the static [30] and dynamical [31] case. Our theory provides a coherent picture of CB within (TD)DFT without breaking the spin symmetry. By application to two different molecular junctions we further showed that the dynamical xc correction always reduces G_s , thus contributing to close the gap between theoretical predictions and experimental measurements.

S. K. acknowledges funding by the “Grupos Consolidados UPV/EHU del Gobierno Vasco” grant No. IT578-13. G. S. acknowledges funding by MIUR FIRB Grant No. RBFR12SW0J. We acknowledge support through travel grants [Psi-K2 4665 and 3962 (G. S.), and Psi-K2 5332 (S. K.)] of the European Science Foundation (ESF).

- [1] G. Cuniberti, G. Fagas, and K. Richter, *Introducing Molecular Electronics* (Springer, Heidelberg, 2005); J.C. Cuevas and E. Scheer, *Molecular Electronics: An Introduction to Theory and Experiment* (World Scientific, London, 2010).

- [2] G. Stefanucci and C.-O. Almbladh, *Phys. Rev. B* **69**, 195318 (2004); *Europhys. Lett.* **67**, 14 (2004).
- [3] N. Sai, M. Zwolak, G. Vignale, and M. Di Ventra, *Phys. Rev. Lett.* **94**, 186810 (2005).
- [4] M. Koentopp, K. Burke, and F. Evers, *Phys. Rev. B* **73**, 121403 (2006).
- [5] G. Vignale and M. Di Ventra, *Phys. Rev. B* **79**, 014201 (2009).
- [6] E. Runge and E. K. U. Gross, *Phys. Rev. Lett.* **52**, 997 (1984).
- [7] C. Toher, A. Filippetti, S. Sanvito, and K. Burke, *Phys. Rev. Lett.* **95**, 146402 (2005).
- [8] J. P. Perdew, R. G. Parr, M. Levy, and J. L. Balduz, *Phys. Rev. Lett.* **49**, 1691 (1982).
- [9] G. Stefanucci and S. Kurth, *Phys. Rev. Lett.* **107**, 216401 (2011).
- [10] H. Mera, K. Kaasbjerg, Y. M. Niquet, and G. Stefanucci, *Phys. Rev. B* **81**, 035110 (2010); H. Mera and Y. M. Niquet, *Phys. Rev. Lett.* **105**, 216408 (2010).
- [11] J. P. Bergfield, Z. F. Liu, K. Burke, and C. A. Stafford, *Phys. Rev. Lett.* **108**, 066801 (2012).
- [12] P. Tröster, P. Schmitteckert, and F. Evers, *Phys. Rev. B* **85**, 115409 (2012).
- [13] See, e.g., H. Haug and A.-P. Jauho, *Quantum Kinetics in Transport and Optics of Semiconductors* (Springer, New York, 2008).
- [14] The static DFT kernel is the response of the *equilibrium* v_{xc} to a density variation and, deep inside the leads, is determined by the condition of charge neutrality alone.
- [15] This follows from the fact that $\delta v_{xc}(\mathbf{r}, t \rightarrow \infty)$ is defined up to the addition of an arbitrary constant C ; see also M. Hellgren and E. K. U. Gross, *Phys. Rev. A* **85**, 022514 (2012).
- [16] A. M. Uimonen, E. Khosravi, A. Stan, G. Stefanucci, S. Kurth, R. van Leeuwen, and E. K. U. Gross, *Phys. Rev. B* **84**, 115103 (2011).
- [17] The compressibility is weakly dependent on temperature up to $T \approx \gamma$ [18]. Therefore the v_{Hxc} obtained by reverse engineering is a good approximation even for $T = 0$. At the ph symmetric point ($N = 1$) this approximation gives $\partial v_{Hxc}/\partial N \sim U^2/\gamma$ [19]. At $T = 0$ the MB spectral function for $\omega \approx \mu$ is dominated by the AS resonance $A(\omega \approx \mu) \approx (4T_K/\gamma)L_{T_K}(\omega - \mu)$ and thus $\int f(\omega) \times (\partial A(\omega)/\partial \mu) \approx -2/(\pi\gamma)$. For the compressibility we have from the Bethe-Ansatz [20] $\kappa = (8\gamma)/(\pi U^2) \times [1 + \mathcal{O}(\gamma/U)]$ from which it follows that also R goes like $(U/\gamma)^2$.
- [18] I. Maruyama, N. Shibata, and K. N. Ueda, *J. Phys. Soc. Jpn.* **73**, 434 (2004).
- [19] F. Evers and P. Schmitteckert, *Phys. Chem. Chem. Phys.* **13**, 14 417 (2011).
- [20] P. B. Wiegmann and A. M. Tselick, *J. Phys. C* **16**, 2281 (1983).
- [21] T. A. Costi, *Phys. Rev. Lett.* **85**, 1504 (2000).
- [22] E. Perfetto and G. Stefanucci, *Phys. Rev. B* **86**, 081409 (2012).
- [23] This amounts to neglecting interference effects; see G. Stefanucci, E. Perfetto, S. Bellucci, and M. Cini, *Phys. Rev. B* **79**, 073406 (2009); T. Markussen, R. Stadler, and K. S. Thygesen, *Nano Lett.* **10**, 4260 (2010); J. P. Bergfield, G. C. Solomon, C. A. Stafford, and M. A. Ratner, *Nano Lett.* **11**, 2759 (2011).
- [24] W. Liang, M. Bockrath, and H. Park, *Phys. Rev. Lett.* **88**, 126801 (2002).
- [25] M. R. Buitelaar, A. Bachtold, T. Nussbaumer, M. Iqbal, and C. Schonenberger, *Phys. Rev. Lett.* **88**, 156801 (2002).
- [26] S. Sapmaz, P. Jarillo-Herrero, J. Kong, C. Dekker, L. P. Kouwenhoven, and H. S. J. van der Zant, *Phys. Rev. B* **71**, 153402 (2005).
- [27] L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, *Rep. Prog. Phys.* **64**, 701 (2001).
- [28] Y. Oreg, K. Byczuk, and B. I. Halperin, *Phys. Rev. Lett.* **85**, 365 (2000).
- [29] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.111.030601> for details on the implementation.
- [30] N. A. Lima, L. N. Oliveira, and K. Capelle, *Europhys. Lett.* **60**, 601 (2002); N. A. Lima, M. F. Silva, L. N. Oliveira, and K. Capelle, *Phys. Rev. Lett.* **90**, 146402 (2003); P. Mori-Sanchez, A. J. Cohen, and W. T. Yang, *Phys. Rev. Lett.* **102**, 066403 (2009); F. Malet and P. Gori-Giorgi, *Phys. Rev. Lett.* **109**, 246402 (2012); X. Gao, A.-H. Chen, I. V. Tokatly, and S. Kurth, *Phys. Rev. B* **86**, 235139 (2012); J. Lorenzana, Z.-J. Ying, and V. Brosco, *Phys. Rev. B* **86**, 075131 (2012); E. Krausler and L. Kronik, *Phys. Rev. Lett.* **110**, 126403 (2013).
- [31] C. Verdozzi, *Phys. Rev. Lett.* **101**, 166401 (2008); S. Kurth, G. Stefanucci, E. Khosravi, C. Verdozzi, and E. K. U. Gross, *Phys. Rev. Lett.* **104**, 236801 (2010); D. Hofmann and S. Kümmel, *Phys. Rev. B* **86**, 201109 (2012); P. Elliott, J. I. Fuks, A. Rubio, and N. T. Maitra, *Phys. Rev. Lett.* **109**, 266404 (2012); S. E. B. Nielsen, M. Ruggenthaler, and R. van Leeuwen, *Europhys. Lett.* **101**, 33 001 (2013).