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# Spin-dependent optimized effective potential formalism for open and closed systems

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Orbital-based exchange ($x$) correlation ($c$) energy functionals, leading to the optimized effective potential (OEP) formalism of density-functional theory (DFT), are gaining increasing importance in ground-state DFT, as applied to the calculation of the electronic structure of closed systems with a fixed number of particles, such as atoms and molecules. These types of functionals prove also to be extremely valuable for dealing with solid-state systems with reduced dimensionality, such as is the case of electrons trapped at the interface between two different semiconductors, or narrow metallic slabs. In both cases, electrons build a quasi-two-dimensional electron gas, or Q2DEG. We provide here a general DFT-OEP formal scheme valid both for Q2DEGs either isolated (closed) or in contact with a particle bath (open), and show that both possible representations are equivalent, being the choice of one or the other essentially a question of convenience. Based on this equivalence, a calculation scheme is proposed which avoids the noninvertibility problem of the density response function for closed systems. We also consider the case of spontaneously spin-polarized Q2DEGs, and find that far from the region where the Q2DEG is localized, the exact $x$-only exchange potential approaches two different, spin-dependent asymptotic limits. As an example, aside from these formal results, we also provide numerical results for a spin-polarized jellium slab, using the new OEP formalism for closed systems. The accuracy of the Krieger-Li-Iafrate approximation has been also tested for the same system, and found to be as good as it is for atoms and molecules.

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## I. INTRODUCTION

One of the main goals of current density-functional theory (DFT) [1,2] is the systematic improvement of the predictive power of the formalism. At zero temperature, which is the case discussed here, this predictive power is intrinsically related to the quality of the exchange ($x$) correlation ($c$) energy functional $E_{xc} = E_x + E_c$. As the exact exchange correlation functional is unknown, and DFT gives no clue on how to proceed to fulfill this goal, many alternatives have been suggested and crystallized in practical calculation schemes. Among them one can cite the local-density approximation (LDA), already introduced in the work by Kohn and Sham [2], taking as reference system the three-dimensional interacting homogeneous electron gas. Later, more sophisticated functionals along this line were developed, including dependencies on the gradients [3,4] (GGA's) and Laplacians [5,6] (Laplacian-level meta-GGA's) of the density. Most of the functionals used in calculations for bulk solid-state systems belong to this type. We will refer to them as density-based functionals.

This standard approach has some difficulties, however, in situations of reduced dimensionality, such as is the case for carriers (usually electrons) trapped at the interface between two different semiconductors, forming the so-called quasi-two-dimensional electron gases (Q2DEGs). Within this context, orbital-based functionals [7–9], which depend explicitly on Kohn-Sham (KS) orbitals and eigenvalues, but implicitly on the density, proved to be quite valuable. Among other advantages, these orbital-based functionals adapt automatically to situations of reduced dimensionality, through the spatial extension and the anisotropy of the orbitals (for example, extended in one plane, but localized along the direction perpendicular to the plane). Orbital-based functionals, for instance, are able to cover the full dimensionality crossover from the strict two-dimensional (2D) limit to the three-dimensional (3D) regime [10].

Compared with density-based functionals, the implicit dependence with density of orbital-based functionals has the consequence that the basic DFT evaluation of functional derivatives with respect to the density becomes more involved. The optimized effective potential (OEP) method [11,12] is especially suited for dealing with these implicit functionals within a KS-DFT framework. If the Hartree-Fock expression for the exchange energy functional $E_x$ is used, and the correlation functional $E_c$ is neglected, the OEP method is equivalent to the exact $x$-only implementation of KS theory. Several advantages have been reported from $x$-only OEP calculations for closed systems, either localized (atoms and molecules) or extended (solids). Among them, we can cite the cancellation of the spurious Hartree self-interacting energy [13], correct high-density limit [14], great improvement in the KS eigenvalue spectrum [15,16], semiconductor band structure and excitations [17–20], and nonlinear optical properties [21]. Concerning its application to Q2DEGs, we have developed and applied the formalism to the calculation of the electronic structure of $n$-doped semiconductor quantum wells, considered as open systems: In Ref. [22], an orbital-based correlation functional has been introduced, and the corresponding correlation potential generated through the OEP formalism. Closed Q2DEGs systems have been also considered using the same formalism, like the study of metallic slabs in the jellium approximation for the positive charge of the ions [23–25], and recent calculations for realistic semiconductor and insulator slabs [26,27]. The aim of this work is (a) to provide a general OEP formalism valid both for open and closed Q2DEGs, and determine the precise relationship between them; and (b) to generalize the formalism to the spin-dependent case, allowing for different spin-density polarizations.

This work is divided in three parts. The first one (Secs. II and III) is dedicated to define the open and closed systems and the convenient representations (i.e., sets of variables) to describe them. With these elements precisely defined, the OEP equations for open and closed systems are derived. Based on them, the relation between the corresponding open and closed potentials is found: they differ by a spin-independent constant. The results of this part are general, i.e., not restricted to orbital-dependent functionals. The second part (Sec. IV) is dedicated to obtain explicit orbital-dependent expressions for the main elements of the formalism. First, explicit orbital expressions are derived for the potential functional derivatives and the derivatives with respect to orbital occupations, in the open- and closed-system representations. Using these basic results, explicit orbital-dependent expressions for the density response function and the magnetic susceptibility are obtained. For closed systems, it is found that the density response function is singular and nondiagonal in the spin variable, while for open systems it is diagonal in spin and nonsingular, thus invertible. These results, combined with those of the first part, have various important consequences, as for example the possibility of obtaining the closed-system potential (up to a spin-independent constant) by using the open-system density response function, which is invertible. Also, the magnetic susceptibility of the open system is insensitive to differences in spin channel filling, while that of the closed system is strongly dependent on it. Next, the OEP equations are recast in terms of the so-called orbital shifts. Written in this form, two exact conditions for the OEP potential are found. These are of great relevance, both from the formal and the practical points of view. As for example, these can be enforced during the self-consistency loop to improve convergence. They also determine both the asymptotic limit (open case) and the relative position of the spin-up and -down components of the potential (open and closed cases). For spin-polarized systems, it is found that both spin components of the exact $x$-only exchange potential tend to different asymptotic limits in the vacuum region. In the third part of the work (Sec. V) we provide numerical results for a spin-polarized metallic jellium slab. The main purpose is to provide an example on how to use the newly developed OEP formalism for closed systems. The OEP potential for the exchange-only case is calculated and compared with the LDA and Krieger-Li-Iafrate (KLI) approximations. It is found that, while LDA gives a fully ferromagnetic configuration, the OEP leads to a partial antiferromagnetic ground state. The KLI approximation gives results very close to OEP. The OEP potential is found to tend to different asymptotic values for different spin components, confirming the theoretical findings.

## II. QUASI-TWO-DIMENSIONAL ELECTRON GAS: OPEN AND CLOSED SYSTEMS

In the Q2DEG the electrons are confined in one spatial direction by a confinement potential (hereafter the quantum well), while they are free to move in the perpendicular direction. If translational invariance in the $x$-$y$ plane is assumed, and the confinement coordinate is $z$, the single-particle KS orbitals can be written as a product of a plane wave along $\boldsymbol{\rho} = (x,y)$ and a so-called subband orbital in $z$:

$$\psi_{i\sigma\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{A}} e^{i\mathbf{k}\boldsymbol{\rho}} \xi_{i\sigma}(z), \tag{1}$$

where $\mathbf{k} = (k_x,k_y)$ is the in-plane wave vector, $A$ is the area of the Q2DEG, $i$ is the subband index, and $\sigma$ the spin index, which can take the values $+1,-1$ (respectively $\uparrow,\downarrow$, depending on context). Some assumptions have been made already by writing Eq. (1). In the first place, the full factorization between the solution in the $x$-$y$ plane and the $z$ direction is only valid for a local potential [28]. However, since the OEP method lies within the framework of the KS implementation of DFT, the factorization implies no lack of generality, owing to the locality of the KS potential. On the other side, the assumption of translational invariance in the $x$-$y$ plane implies some lack of generality. According to the results in Ref. [29], however, the assumption is well justified for the relatively high 2D densities found typically in Q2DEGs. The energies $E_{i\sigma\mathbf{k}} = k^2/2 + \varepsilon_{i\sigma}$ of the single-particle KS orbitals in Eq. (1) are the sum of a continuous free-electron spectrum $k^2/2$ corresponding to the in-plane movement, and a discrete subband spectrum $\varepsilon_{i\sigma}$ related to the confined motion in the quantum well. The subband orbitals $\xi_{i\sigma}(z)$ are obtained from the KS equation for the confinement coordinate

$$\left[-\frac{1}{2}\frac{\partial^2}{\partial z^2} + V_{s\sigma}(z)\right]\xi_{i\sigma}(z) = \varepsilon_{i\sigma}\,\xi_{i\sigma}(z), \tag{2}$$

where the KS potential $V_{s\sigma}(z) = V_e(z) + V_{\mathrm{H}}(z) + V_{xc\sigma}(z)$, with $V_e(z)$ the external potential, $V_{\mathrm{H}}(z)$ the Hartree potential, and $V_{xc\sigma}(z)$ the $xc$ potential. If not stated otherwise, Hartree atomic units are used throughout this work [30]. In a collinear spin situation as we are considering here, the subband wave functions in Eq. (2) can be taken to be real without loss of generality. The main quantity in spin-dependent DFT is the spin-resolved 3D electronic density [1], that is given by

$$n_\sigma(z) = \sum_i \theta_{i\sigma} n_{i\sigma} \xi_{i\sigma}(z)^2, \tag{3}$$

where the occupation factors $n_{i\sigma}$ are

$$n_{i\sigma} = \frac{\mu - \varepsilon_{i\sigma}}{2\pi}, \tag{4}$$

and $\theta_{i\sigma} \equiv \theta(n_{i\sigma})$, with $\theta(\ldots)$ the Heaviside step function: $\theta(x) = 1$ if $x > 0$ and $\theta(x) = 0$ if $x < 0$. $\mu$ denotes the chemical potential. The total number of electrons per unit area is

$$\eta = \sum_{i\sigma} \theta_{i\sigma} n_{i\sigma} = \frac{N_s}{2\pi}\mu - \sum_{i\sigma} \frac{\theta_{i\sigma}}{2\pi}\varepsilon_{i\sigma}, \tag{5}$$

where $N_s = \sum_{i\sigma} \theta_{i\sigma}$ is the total number of occupied subbands. Let us note that, as given above, $n_\sigma(z)$, $n_{i\sigma}$, and $\eta$ are dimensionless densities; to recover the corresponding dimensions, the right-hand side should be multiplied by $a_0^{-3}$, $a_0^{-2}$, and $a_0^{-2}$, respectively. Along the text we will use the term "areal density" to refer to $\eta$, which is a positive real number. Typically, this areal density is about $10^{14}$–$10^{15}$ electrons/cm$^2$ for the case of metallic jellium slabs, and about $10^{11}$ electrons/cm$^2$ for the case of Q2DEGs in semiconductor quantum wells.

We will consider two distinct physical situations. In one of them, the confined electron gas in the quantum well can exchange electrons with a particle reservoir that fixes the chemical potential $\mu$; we call this the *open system*. The second situation corresponds to an isolated quantum well with fixed areal density $\eta$; we will refer to this case as the *closed system*. Both cases impose different constraints on the density that can be expressed in terms of the possible density variations as follows:

$$\delta^{\mu} n_\sigma(z \in \text{reservoir}) = 0, \tag{6}$$

$$\int \delta^{\eta} n_\sigma(z)\, dz = 0, \tag{7}$$

where $\delta^r n_\sigma(z)$ denotes a spin-density variation that leaves $r$ unchanged. The superscript $r = \mu\,(\eta)$ is used to indicate the open (closed) case. The integrals on $z$ run from $z = -\infty$ to $\infty$ in the case $r = \eta$, while they exclude the region of the reservoir for $r = \mu$. The first condition [Eq. (6)] is sufficient for the reservoir to keep a fixed chemical potential $\mu$. Moreover, by definition the reservoir's density is not affected by arbitrary density variations in the region of the Q2DEG. This constraint implies that the energy optimization in the KS scheme applies only to the Q2DEG region and not to the reservoir, whose density is fixed, and its change of total energy per unit area is accounted simply by $-\mu\Delta\eta$ with $\Delta\eta = \sum_\sigma \int \delta^{\mu} n_\sigma(z)\, dz$. The second condition [Eq. (7)] expresses the fixed areal density constraint by restricting the density variations to only those which leave $\eta$ unchanged.

In all the following developments, $\eta$ and $\mu$ will be two fundamental variables of our Q2DEG. In some restricted sense, the areal density $\eta$ is somehow equivalent to the total number of electrons $N$ of a finite system, like an atom or molecule. For instance, we have shown in Ref. [24] that the $x$-only OEP potential and the associated work function of a jellium metallic slab suffers abrupt changes as $\eta$ changes, in particular every time the occupation factor $n_{i\sigma}$ corresponding to the highest slab discrete subband becomes infinitesimally occupied. This is qualitatively similar to the case of the exact exchange correlation potential and the chemical potential of finite systems, that have abrupt changes as $N$ passes through integer values [31,32].

## III. OEP EQUATIONS FOR THE Q2DEG

Framed in the KS implementation of the DFT formalism, the OEP method gives an integral equation for the spin-dependent $xc$ potential $V_{xc\sigma}(\mathbf{r}) \equiv \delta E_{xc}/\delta n_\sigma(\mathbf{r})$, where $E_{xc}$ is the exchange correlation energy. This integral equation is written in terms of the density response function $\chi_{\sigma\sigma'}(\mathbf{r},\mathbf{r}') = \delta n_\sigma(\mathbf{r})/\delta V_{s\sigma'}(\mathbf{r}')$ and $\delta E_{xc}/\delta V_{s\sigma}(\mathbf{r})$ [12]. In evaluating both these potential functional derivatives, the question arises as to how to properly account for the two different physical situations that we are considering: A given potential variation $\delta V_{s\sigma}(z)$ in the Q2DEG will in general lead to different density variations for open and closed systems, therefore, it is expected that the OEP equations for both cases will differ.

In the following, the $xc$ energy of the system (or any approximation to it) will be represented by a spin-density functional $\mathcal{F}[\{n_\sigma(z)\}] = E_{xc}$, where $\{n_\sigma(z)\}$ denotes the set of spin-up and -down densities $\{n_\sigma(z)\} \equiv \{n_\uparrow(z), n_\downarrow(z)\}$. In several indicated cases $\mathcal{F}$ will also represent an arbitrary functional of the density, therefore, we use this symbol so as to preserve the compactness of the derivations. In order to unambiguously define the potential functional derivatives appearing in the OEP formulation, it becomes necessary to work with a set of variables that determines $\{n_\sigma(z)\}$ unequivocally. We will call such a set of variables a representation. Suitable representations are, e.g., $\{V_{s\sigma}(z),\, \mu\}$ or $\{V_{s\sigma}(z),\, \eta\}$. In considering open Q2DEGs, it is convenient to work with the first representation, as $\delta\mu = 0$. In the closed case, where $\delta\eta = 0$, the second representation turns out to be more convenient. Accordingly, we will call the set $\{V_{s\sigma}(z),\, \mu\}$ the open-system representation, and the set $\{V_{s\sigma}(z),\, \eta\}$ the closed-system representation.

We will make use of the auxiliary noninteracting KS system, whose energy $E_s = T_s + A\sum_\sigma \int dz\, V_{s\sigma}(z) n_\sigma(z)$, with $T_s$ the noninteracting kinetic energy functional. The energy optimization of the KS system (plus the reservoir in the open case) for $r =$ constant leads to

$$\delta T_s = -A \sum_\sigma \int dz [V_{s\sigma}(z) - \mu] \delta^r n_\sigma(z). \tag{8}$$

The term proportional to $\mu$ in this equation is only relevant in the open case, as it vanishes in the closed system by virtue of Eq. (7). Additionally, we will make use of the restricted density response function, as defined by

$$\chi^r_{\sigma\sigma'}(z,z') := \left.\frac{\delta n_\sigma(z)}{\delta V_{s\sigma'}(z')}\right|_r. \tag{9}$$

The notation $\delta n_\sigma(z)/\delta V_{s\sigma'}(z')|_r$ means that the variation is made by keeping $r$ and $V_{s\underline{\sigma}'}(z')$ fixed (with $\underline{\sigma} = -\sigma$); for the sake of simplicity in notation, this is just denoted by the symbol $r$. From Eq. (9), we can express an $r$-conserving density variation by

$$\delta^r n_\sigma(z) = \sum_{\sigma'} \int dz' \chi^r_{\sigma\sigma'}(z,z') \delta V_{s\sigma'}(z'). \tag{10}$$

In Secs. III A and III B, we obtain the OEP equations for open and closed Q2DEGs, respectively. The relation between the corresponding potentials obtained from them is then analyzed in Sec. (III C).

### A. Open systems in the open-system representation

We consider an open system in contact with a particle reservoir which fixes the chemical potential $\mu$. The total energy of the interacting system plus the reservoir is

$$E = T_s + A \int dz [V_e(z) - \mu] n(z) + E_H + \mathcal{F}. \tag{11}$$

In terms of the open-system representation, the optimization of the total energy for fixed $\mu$ can be carried out with the help of Eqs. (8) and (10):

$$\frac{\delta^{\mu}E}{A} = -\sum_{\sigma'}\int dz'\Bigg\{\sum_{\sigma}\int dz[V_{s\sigma}(z) - V_H(z) - V_e(z)] \times \chi^{\mu}_{\sigma\sigma'}(z,z') - \frac{1}{A}\frac{\delta\mathcal{F}}{\delta V_{s\sigma'}(z')}\bigg|_{\mu}\Bigg\}\delta V_{s\sigma'}(z') = 0. \quad (12)$$

The KS potential $V_{s\sigma}(z)$ that optimizes the total energy must fulfill this equation for an arbitrary variation $\delta V_{s\sigma'}(z')$; from these types of considerations the name *optimized effective potential* originates [33]. Therefore, the optimum $V_{s\sigma}(z)$ is the one that makes zero the term in curly braces. Hence, if we call $V^{\mu}_{s\sigma}(z)$ to the optimizing KS potential, it satisfies the integral equation

$$\sum_{\sigma}\int dz\big[V^{\mu}_{s\sigma}(z) - V_H(z) - V_e(z)\big]\chi^{\mu}_{\sigma\sigma'}(z,z') = \frac{1}{A}\frac{\delta\mathcal{F}}{\delta V_{s\sigma'}(z')}\bigg|_{\mu}.$$

By defining $V^{\mu}_{\mathcal{F}\sigma}(z) := V^{\mu}_{s\sigma}(z) - V_H(z) - V_e(z)$, we arrive at the OEP equation for the open system in the open-system representation:

$$\frac{1}{A}\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\mu} = \sum_{\sigma'}\int dz' V^{\mu}_{\mathcal{F}\sigma'}(z')\chi^{\mu}_{\sigma'\sigma}(z',z). \quad (13)$$

If $C$ is a spin-independent constant, $\{V_{s\sigma}(z),\, \mu\}$ and $\{V_{s\sigma}(z) + C,\, \mu + C\}$ define the same density $\{n_{\sigma}(z)\}$. Therefore, $\mathcal{F}$, being a functional of the density alone, must satisfy

$$\mathcal{F}[V_{s\sigma}(z) + C,\, \mu] = \mathcal{F}[V_{s\sigma}(z),\, \mu - C]. \quad (14)$$

This property can be recast in a different form: If $C$ is an infinitesimal, the left-hand side in Eq. (14) is, to first order in $C$,

$$\mathcal{F}[V_{s\sigma}(z) + C,\, \mu] = \mathcal{F}[V_{s\sigma}(z),\, \mu] + C\sum_{\sigma}\int dz\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\mu}.$$

Analogously, for the right-hand side of Eq. (14) we can write

$$\mathcal{F}[V_{s\sigma}(z),\, \mu - C] = \mathcal{F}[V_{s\sigma}(z),\, \mu] - C\frac{\partial\mathcal{F}}{\partial\mu}\bigg|_{V_{s\updownarrow}},$$

where $V_{s\updownarrow}$ means that the partial derivative with the chemical potential is done keeping fixed both spin components of the KS potential [34]. Equating the two equations above we arrive at the following identity:

$$\sum_{\sigma}\int dz\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\mu} = -\frac{\partial\mathcal{F}}{\partial\mu}\bigg|_{V_{s\updownarrow}}. \quad (15)$$

The only assumption in deriving this equation was that $\mathcal{F}$ is a functional of the spin density, therefore, its applicability is not limited to the *xc* energy functional. Summing on $\sigma$ and integrating on $z$ on both sides of Eq. (13), we can therefore apply Eq. (15) to the two potential functional derivatives in the resulting expression to obtain

$$\frac{1}{A}\frac{\partial\mathcal{F}}{\partial\mu}\bigg|_{V_{s\updownarrow}} = \sum_{\sigma}\int dz\, V^{\mu}_{\mathcal{F}\sigma}(z)\frac{\partial n_{\sigma}(z)}{\partial\mu}\bigg|_{V_{s\updownarrow}}. \quad (16)$$

This equation will be used in Sec. III C to obtain the relation between the OEP potentials for open and closed systems.

### B. Closed systems in the closed-system representation

The derivation for this case is completely analogous to Eqs. (11)–(13), but now in terms of the representation $\{V_{s\sigma}(z),\, \eta\}$ and without considering the reservoir. The OEP equation for closed systems is then

$$\frac{1}{A}\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\eta} = \sum_{\sigma'}\int dz' V^{\eta}_{\mathcal{F}\sigma'}(z')\chi^{\eta}_{\sigma'\sigma}(z',z). \quad (17)$$

If $C$ is a spin-independent constant, $\{V_{s\sigma}(z),\eta\}$ and $\{V_{s\sigma}(z) + C,\eta\}$ determine exactly the same density $\{n_{\sigma}(z)\}$. Therefore, the density functional $\mathcal{F}$ must satisfy

$$\mathcal{F}[V_{s\sigma}(z) + C,\eta] = \mathcal{F}[V_{s\sigma}(z),\eta]. \quad (18)$$

For small $C$, we can write to first order in $C$

$$\mathcal{F}[V_{s\sigma}(z) + C,\eta] = \mathcal{F}[V_{s\sigma}(z),\eta] + C\sum_{\sigma}\int dz\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\eta},$$

which we can equate to Eq. (18) with the result

$$\sum_{\sigma}\int dz\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\eta} = 0, \quad (19)$$

i.e., a constant spin-independent shift in the potential, at constant $\eta$, will not change $\mathcal{F}$. The same considerations made after Eq. (15), regarding its applicability to arbitrary density functionals, apply also to Eq. (19). Therefore, it can be used, for example, with $\mathcal{F} = n_{\sigma'}(z')$, to readily show the singularity of the density response function in the closed-system representation (see Sec. IV B 1).

Note that Eq. (17) is invariant upon the addition of a spin-independent constant to $V^{\eta}_{\mathcal{F}\sigma'}(z')$, as $\sum_{\sigma'}\int dz'\,\delta n_{\sigma'}(z') = \delta\eta = 0$.

### C. Relation between open- and closed-system potentials

In this section, we will find the relation between the potentials $V^{\mu}_{\mathcal{F}\sigma}(z)$ and $V^{\eta}_{\mathcal{F}\sigma}(z)$ defined by Eqs. (13) and (17), respectively. Implicit in the derivation is the assumption that the density functional $\mathcal{F}$ is well defined for both the open and closed systems, i.e., for arbitrary areal density and chemical potential. If we substitute the potential functional derivatives appearing in the OEP equation for open systems [Eq. (13)] by Eq. (A3) of Appendix A [note that for the replacement on the right-hand side of Eq. (13) we must consider the particular case $\mathcal{F} = n_{\sigma'}(z')$ in Eq. (A3)], we reobtain after simple algebra, and by virtue of Eq. (16), the OEP equation for closed systems [Eq. (17)] with $V^{\eta}_{\mathcal{F}\sigma}(z)$ replaced by $V^{\mu}_{\mathcal{F}\sigma}(z)$. This means that if $V^{\mu}_{\mathcal{F}\sigma}(z)$ is a solution of Eq. (13), then it will also be a solution of Eq. (17). On the contrary, if $V^{\eta}_{\mathcal{F}\sigma}(z)$ is a solution of the OEP equation for closed systems [Eq. (17)], it will not in general be a solution of the OEP equation for open systems [Eq. (13)]. The reason for this is that $V^{\eta}_{\mathcal{F}\sigma}(z)$ is defined within an additive constant, while $V^{\mu}_{\mathcal{F}\sigma}(z)$ is not. In order to find the precise relation between them, we begin by applying Eq. (A4) of Appendix A to the potential functional derivatives on both sides of Eq. (17) with the result

$$\frac{1}{A}\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\mu} = \sum_{\sigma'}\int dz'\, V^{\eta}_{\mathcal{F}\sigma'}(z')\frac{\delta n_{\sigma'}(z')}{\delta V_{s\sigma}(z)}\bigg|_{\mu} + \left(\frac{1}{A}\frac{\partial\mathcal{F}}{\partial\eta}\bigg|_{V_{s\updownarrow}} - \sum_{\sigma'}\int dz'\, V^{\eta}_{\mathcal{F}\sigma'}(z')\frac{\partial n_{\sigma'}(z')}{\partial\eta}\bigg|_{V_{s\updownarrow}}\right)\times\frac{\delta\eta}{\delta V_{s\sigma}(z)}\bigg|_{\mu}. \tag{20}$$

Taking into account that $\delta\eta = \sum_{\sigma'}\int dz'\,\delta n_{\sigma'}(z')$, Eq. (20) can be recast as

$$\frac{1}{A}\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\mu} = \sum_{\sigma'}\int dz'\,\left[V^{\eta}_{\mathcal{F}\sigma'}(z') + C\right]\frac{\delta n_{\sigma'}(z')}{\delta V_{s\sigma}(z)}\bigg|_{\mu},$$

where $C$ is identical to the parentheses on the right-hand side of Eq. (20). Comparing the equation above with the OEP equation for open systems [Eq. (13)], we get the precise relation between the open- and closed-system potentials, namely,

$$\begin{aligned} V^{\mu}_{\mathcal{F}\sigma}(z) = V^{\eta}_{\mathcal{F}\sigma}(z) + \frac{1}{A}\frac{\partial\mathcal{F}}{\partial\eta}\bigg|_{V_{s\updownarrow}} \\ - \sum_{\sigma'}\int dz'\, V^{\eta}_{\mathcal{F}\sigma'}(z')\frac{\partial n_{\sigma'}(z')}{\partial\eta}\bigg|_{V_{s\updownarrow}}. \end{aligned} \tag{21}$$

For $\mathcal{F} = E_{xc}$, this equation relates the two exchange correlation potentials $V^{\mu}_{xc\,\sigma}(z)$ and $V^{\eta}_{xc\,\sigma}(z)$ of our Q2DEG under the open-system (constant $\mu$) and closed-system (constant $\eta$) constraints.

As discussed above, as $V^{\mu}_{\mathcal{F}\sigma}(z)$ is also a solution of Eq. (17), $V^{\eta}_{\mathcal{F}\sigma}(z)$ can be replaced by $V^{\mu}_{\mathcal{F}\sigma}(z)$ in Eq. (21) and the open-system potential then fulfills

$$\frac{1}{A}\frac{\partial\mathcal{F}}{\partial\eta}\bigg|_{V_{s\updownarrow}} = \sum_{\sigma}\int dz\, V^{\mu}_{\mathcal{F}\sigma}(z)\frac{\partial n_{\sigma}(z)}{\partial\eta}\bigg|_{V_{s\updownarrow}}. \tag{22}$$

This condition, toghether with Eq. (17) with $V^{\eta}_{\mathcal{F}\sigma}(z)$ replaced by $V^{\mu}_{\mathcal{F}\sigma}(z)$, provides a way of obtaining the open-system potential using the closed-system representation. Such a calculation scheme is fully analogous to that presented for finite systems in Ref. [35], as can be seen by comparing with Eqs. (36) and (13) of that work. However, while in the closed-system representation both Eqs. (17) and (22) are needed to find the open-system potential, in the open-system representation Eq. (13) (this work) alone suffices. The reason for that is that the condition of Eq. (22) [which is equivalent to Eq. (16), as will be shown in Sec. IV A 4] is already contained in the OEP equation in the open-system representation (see Sec. III A).

## IV. OEP FOR ORBITAL FUNCTIONALS IN THE Q2DEG

The OEP formalism is well suited for orbital-based $xc$ energy functionals, as in such a case the potential functional derivatives appearing in the formulation can be explicitly obtained from the chain rule for functional derivatives, with the subband orbitals and occupations as intermediate variables, and first-order perturbation theory. Accordingly, we develop in this section the OEP formalism for orbital-based functionals of the type

$$\mathcal{F} = \mathcal{F}[\{\xi_{i\sigma}, n_{i\sigma}\}], \tag{23}$$

where the set of orbital variables $\{\xi_{i\sigma}, n_{i\sigma}\}$ include, in general, both occupied and unoccupied subbands. We begin by finding explicit orbital-dependent expressions for the various restricted derivatives of an arbitrary orbital functional $\mathcal{F}$, namely, $\delta\mathcal{F}/\delta V_{s\sigma}(z)|_{r}$ and $\partial\mathcal{F}/\partial r|_{V s\updownarrow}$, $r = \mu, \eta$ (Sec. IV A). Next, we obtain explicit orbital expressions for the KS density response function (Sec. IV B 1) and the macroscopic magnetic susceptibility (Sec. IV B 2). In each case, the differences between the open and closed cases are analyzed. Using these results, the OEP equations are recast in terms of the so-called orbital shifts (Sec. IV C) and, in terms of them, explicit expressions for the OEP potential are found (Sec. IV E). Finally (Sec. IV F), the asymptotic limit of the $x$-only OEP potential $V^{r}_{x\,\sigma}(z \to \infty)$ is studied.

### A. Calculation of restricted derivatives of orbital-based functionals

In this section, we will use the chain rule for functional derivatives with $\{\xi_{i\sigma}, n_{i\sigma}\}$ as intermediate variables. We begin with the calculation of the two main derivatives appearing in the open-system representation, i.e., $\delta\mathcal{F}/\delta V_{s\sigma}(z)|_{\mu}$ and $\partial\mathcal{F}/\partial\mu|_{V_{s\updownarrow}}$.

### *1. Calculation of $\delta\mathcal{F}/\delta V_{s\sigma}(z)|_{\mu}$*

Using the chain rule we get

$$\begin{aligned} \frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\mu} &= \sum_{i\sigma'}\int dz'\frac{\delta\mathcal{F}}{\delta\xi_{i\sigma'}(z')}\frac{\delta\xi_{i\sigma'}(z')}{\delta V_{s\sigma}(z)}\bigg|_{\mu} \\ &\quad + \sum_{i\sigma'}\frac{\partial\mathcal{F}}{\partial n_{i\sigma'}}\frac{\delta n_{i\sigma'}}{\delta V_{s\sigma}(z)}\bigg|_{\mu}. \end{aligned} \tag{24}$$

From first-order perturbation theory, the potential functional derivative of the KS orbitals is

$$\frac{\delta\xi_{i\sigma'}(z')}{\delta V_{s\sigma}(z)} = \delta_{\sigma\sigma'}\xi_{i\sigma}(z)G_{i\sigma}(z',z), \tag{25}$$

$$G_{i\sigma}(z',z) = \sum_{j\neq i}\frac{\xi_{j\sigma}(z')\xi_{j\sigma}(z)}{\varepsilon_{i\sigma} - \varepsilon_{j\sigma}}, \tag{26}$$

independently of $\mu$. For the potential functional derivative of the occupations in the right-hand side of Eq. (24), we have from Eq. (4) and first-order perturbation theory

$$\frac{\delta n_{i\sigma'}}{\delta V_{s\sigma}(z)}\bigg|_{\mu} = -\frac{\delta_{\sigma\sigma'}}{2\pi}\,\xi_{i\sigma}(z)^2. \tag{27}$$

Replacing Eqs. (25) and (27) in (24) we obtain

$$\begin{aligned} \frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\bigg|_{\mu} &= \sum_{i}\int dz'\frac{\delta\mathcal{F}}{\delta\xi_{i\sigma}(z')}\xi_{i\sigma}(z)G_{i\sigma}(z',z) \\ &\quad - \frac{1}{2\pi}\sum_{i}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}\,\xi_{i\sigma}(z)^2. \end{aligned} \tag{28}$$

### 2. *Calculation of $\partial\mathcal{F}/\partial\mu|_{V_{s\updownarrow}}$*

The electronic structure remains unchanged as long as $V_{s\sigma}$ remains fixed, therefore, it is not necessary to consider changes in the shape of the KS orbitals when evaluating $\partial\mathcal{F}/\partial\mu|_{V_{s\updownarrow}}$. Using Eq. (4) we get

$$\left.\frac{\partial\mathcal{F}}{\partial\mu}\right|_{V_{s\updownarrow}} = \sum_{i\sigma}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}\left.\frac{\partial n_{i\sigma}}{\partial\mu}\right|_{V_{s\updownarrow}} = \frac{1}{2\pi}\sum_{i\sigma}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}. \tag{29}$$

Note that this result is also obtained if we integrate on $z$ both sides of Eq. (28), taking into account that $\int dz\,\xi_{i\sigma}(z)G_{i\sigma}(z',z) = 0$ due to the orthogonality of the KS orbitals and also the property of Eq. (15).

We continue in the following with the remaining derivatives related to the closed-system representation.

### 3. *Calculation of $\delta\mathcal{F}/\delta V_{s\sigma}(z)|_{\eta}$*

Applying the chain rule, we have

$$\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\eta} = \sum_{i\sigma'}\int dz'\frac{\delta\mathcal{F}}{\delta\xi_{i\sigma'}(z')}\left.\frac{\delta\xi_{i\sigma'}(z')}{\delta V_{s\sigma}(z)}\right|_{\eta} + \sum_{i\sigma'}\frac{\partial\mathcal{F}}{\partial n_{i\sigma'}}\left.\frac{\delta n_{i\sigma'}}{\delta V_{s\sigma}(z)}\right|_{\eta}. \tag{30}$$

The result for the derivative $\delta\xi_{i\sigma'}(z')/\delta V_{s\sigma}(z)|_{\eta}$ is identical to Eq. (25) because the subband wave functions are fully determined by $V_{s\sigma}(z)$ [this can also be seen from Eq. (A4) with $\mathcal{F} = \xi_{i\sigma}(z)$ and the fact that $\partial\xi_{i\sigma}/\partial\eta|_{V_{s\updownarrow}} = 0$]. For the potential functional derivative of the occupation factors we have, taking into account Eqs. (A3) and (29) with $\mathcal{F} = n_{i\sigma'}$ and Eq. (27),

$$\left.\frac{\delta n_{i\sigma'}}{\delta V_{s\sigma}(z)}\right|_{\eta} = \frac{1}{2\pi}\left[\left.\frac{\delta\mu}{\delta V_{s\sigma}(z)}\right|_{\eta} - \delta_{\sigma\sigma'}\xi_{i\sigma}(z)^2\right]. \tag{31}$$

The functional derivative of the chemical potential is obtained from Eq. (5) together with the first-order perturbation theory result $\delta\varepsilon_{i\sigma}/\delta V_{s\sigma'}(z) = \delta_{\sigma\sigma'}\xi_{i\sigma}(z)^2$:

$$\left.\frac{\delta\mu}{\delta V_{s\sigma}(z)}\right|_{\eta} = \frac{1}{N_s}\sum_{i}\theta_{i\sigma}\xi_{i\sigma}(z)^2. \tag{32}$$

Inserting Eq. (32) in (31) we have

$$\left.\frac{\delta n_{i\sigma'}}{\delta V_{s\sigma}(z)}\right|_{\eta} = -\frac{1}{2\pi}\sum_{j}\left[\delta_{\sigma\sigma'}\delta_{ij} - \frac{\theta_{j\sigma}}{N_s}\right]\xi_{j\sigma}(z)^2. \tag{33}$$

Finally, substituting in Eq. (30)

$$\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\eta} = \sum_{i}\int dz'\frac{\delta\mathcal{F}}{\delta\xi_{i\sigma}(z')}\xi_{i\sigma}(z)G_{i\sigma}(z',z) - \frac{1}{2\pi}\sum_{ij\sigma'}\left[\delta_{\sigma\sigma'}\delta_{ij} - \frac{\theta_{j\sigma}}{N_s}\right]\frac{\partial\mathcal{F}}{\partial n_{i\sigma'}}\xi_{j\sigma}(z)^2. \tag{34}$$

By comparing Eqs. (34) and (28), we obtain

$$\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\eta} = \left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\mu} + \frac{1}{2\pi N_s}\sum_{i\sigma'}\frac{\partial\mathcal{F}}{\partial n_{i\sigma'}}\sum_{j}\theta_{j\sigma}\xi_{j\sigma}(z)^2, \tag{35}$$

in agreement with Eq. (A3), as can be verified taking into account additionally Eqs. (29) and (32).

### 4. *Calculation of $\partial\mathcal{F}/\partial\eta|_{V_{s\updownarrow}}$*

With the same considerations previous to Eq. (29) and applying the chain rule, we have

$$\left.\frac{\partial\mathcal{F}}{\partial\eta}\right|_{V_{s\updownarrow}} = \sum_{i\sigma}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}\left.\frac{\partial n_{i\sigma}}{\partial\eta}\right|_{V_{s\updownarrow}}.$$

The partial derivatives of the occupation factors $n_{i\sigma}$ can be readily obtained from their definition in Eq. (4) and the result

$$\left.\frac{\partial\mu}{\partial\eta}\right|_{V_{s\updownarrow}} = \frac{2\pi}{N_s},$$

that can be obtained from Eq. (5) and the fact that $\partial\varepsilon_{i\sigma}/\partial\eta|_{V_{s\updownarrow}} = 0$. Combining everything we get

$$\left.\frac{\partial\mathcal{F}}{\partial\eta}\right|_{V_{s\updownarrow}} = \frac{1}{N_s}\sum_{i\sigma}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}. \tag{36}$$

Comparing Eq. (36) with (29) we obtain, additionally,

$$\left.\frac{\partial\mathcal{F}}{\partial\eta}\right|_{V_{s\updownarrow}} = \frac{2\pi}{N_s}\left.\frac{\partial\mathcal{F}}{\partial\mu}\right|_{V_{s\updownarrow}}.$$

This result shows that Eqs. (16) and (22) are equivalent.

## B. Density- and spin-density response functions

### 1. *Density-density response function*

As an application of the results obtained in Sec. IV A, we derive here explicit orbital expressions for the density response function $\chi^{r}_{\sigma\sigma'}(z,z')$ as defined in Eq. (9), for $r = \mu,\eta$. Setting $\mathcal{F} = n_{\sigma}(z)$ in Eq. (28), we obtain

$$\chi^{\mu}_{\sigma\sigma'}(z,z') = \sum_{i}\int dz_1\frac{\delta n_{\sigma}(z)}{\delta\xi_{i\sigma'}(z_1)}\xi_{i\sigma'}(z')G_{i\sigma'}(z_1,z') - \frac{1}{2\pi}\sum_{i}\frac{\partial n_{\sigma}(z)}{\partial n_{i\sigma'}}\xi_{i\sigma'}(z')^2. \tag{37}$$

From Eq. (3), we obtain

$$\frac{\delta n_{\sigma}(z)}{\delta\xi_{i\sigma'}(z_1)} = 2\,\delta_{\sigma\sigma'}\delta(z-z_1)\theta_{i\sigma}n_{i\sigma}\xi_{i\sigma}(z),$$

$$\frac{\partial n_{\sigma}(z)}{\partial n_{i\sigma'}} = \delta_{\sigma\sigma'}\theta_{i\sigma}\xi_{i\sigma}(z)^2$$

to find, after corresponding replacements in Eq. (37),

$$\chi^{\mu}_{\sigma\sigma'}(z,z') = 2\,\delta_{\sigma\sigma'}\sum_{i}\theta_{i\sigma}n_{i\sigma}\xi_{i\sigma}(z)G_{i\sigma}(z,z')\xi_{i\sigma}(z') - \frac{\delta_{\sigma\sigma'}}{2\pi}\sum_{i}\theta_{i\sigma}\xi_{i\sigma}(z)^2\xi_{i\sigma}(z')^2. \tag{38}$$

For the fixed $\eta$ case, we make analogous replacements to Eq. (34) with the result

$$\chi^{\eta}_{\sigma\sigma'}(z,z') = 2\delta_{\sigma\sigma'}\sum_{i}\theta_{i\sigma}n_{i\sigma}\xi_{i\sigma}(z)G_{i\sigma}(z,z')\xi_{i\sigma}(z') - \frac{1}{2\pi}\sum_{i,j}\left[\delta_{\sigma\sigma'}\delta_{ij} - \frac{\theta_{j\sigma'}}{N_s}\right]\theta_{i\sigma}\xi_{i\sigma}(z)^2\xi_{j\sigma'}(z')^2 \tag{39}$$

or, in a more compact form,

$$\chi^{\eta}_{\sigma\sigma'}(z,z') = \chi^{\mu}_{\sigma\sigma'}(z,z') + \frac{1}{2\pi N_s}\sum_{ij}\theta_{i\sigma}\theta_{j\sigma'}\xi_{i\sigma}(z)^2\xi_{j\sigma'}(z')^2. \tag{40}$$

For an arbitrary perturbation of the KS potential $\delta V_{s\sigma}(z)$, there will be a charge transfer between the quantum well and the particle reservoir at fixed $\mu$ in the open system. In contrast, in the closed system such a perturbation will cause a charge transfer between subbands. From Eq. (40), we see that $\chi^{\eta}_{\sigma\sigma'}(z,z')$ is nondiagonal in spin, therefore, the perturbation will cause in general a net charge transfer between subbands with different spin component. In terms of the density response function, the change on the total charge for the spin projection $\sigma$ can be written as

$$\delta^r\eta_\sigma = \sum_{\sigma'}\int dz\,dz'\,\chi^{r}_{\sigma\sigma'}(z,z')\delta V_{s\sigma'}(z').$$

For the open case $r=\mu$, using Eq. (38) we get

$$\delta^\mu\eta_\sigma = -\frac{1}{2\pi}\sum_i \theta_{i\sigma}\overline{\delta V}_{si\sigma}, \tag{41}$$

where we have used that $\int dz\,\xi_{i\sigma}(z)G_{i\sigma}(z,z') = 0$ and $\overline{\delta V}_{si\sigma}$ is defined by

$$\overline{\delta V}_{si\sigma} = \int dz\,\xi_{i\sigma}(z)^2\delta V_{s\sigma}(z). \tag{42}$$

Consistently with Eq. (38), that is diagonal in spin, we find from Eq. (41) that a change in the KS potential for spin component $\sigma$ does not affect the occupation of the opposite spin component. For the closed case $r=\eta$ we find, resorting now to Eq. (39),

$$\delta^\eta\eta_\sigma = \frac{N_{s\sigma}}{2\pi N_s}\sum_i\theta_{i\underline{\sigma}}\overline{\delta V}_{si\underline{\sigma}} - \frac{N_{s\underline{\sigma}}}{2\pi N_s}\sum_i\theta_{i\sigma}\overline{\delta V}_{si\sigma},$$

which implies

$$\delta^\eta\eta_\sigma = -\,\delta^\eta\eta_{\underline{\sigma}},$$

i.e., a flow of charge $\delta^\eta\eta_\sigma$ from one spin component to the other, as expected for a closed system. In the equations above, $N_{s\sigma}=\sum_i\theta_{i\sigma}$ is the number of occupied subbands with spin component $\sigma$.

From Eqs. (38) and (39), the following symmetry properties for the density response functions are verified:

$$\chi^{\mu}_{\sigma\sigma'}(z,z') = \chi^{\mu}_{\sigma\sigma'}(z',z) = \chi^{\mu}_{\sigma'\sigma}(z,z'),$$
$$\chi^{\eta}_{\sigma\sigma'}(z,z') = \chi^{\eta}_{\sigma'\sigma}(z',z).$$

We also derive

$$\sum_{\sigma'}\int dz'\chi^{\mu}_{\sigma\sigma'}(z,z') = -\sum_i\frac{\theta_{i\sigma}}{2\pi}\xi_{i\sigma}(z)^2,$$
$$\sum_{\sigma'}\int dz'\chi^{\eta}_{\sigma\sigma'}(z,z') = 0,$$

that is, the density response function for the Q2DEG closed system is singular, as in the case of solids [18,19]. In contrast, for the open Q2DEG system, it is nonsingular and can be inverted [22,36]. The issue of the invertibility of the density response function is discussed further in the following. The fact that the density response function for finite systems in closed and open environments is different has been already discussed in Ref. [37].

### 2. *Spin-density and macroscopic magnetic susceptibility*

Up to now, we have used only spin-resolved variables in the representations of the Q2DEG. Additional convenient variables for spin-polarized systems are the total density $n(z)$ and the magnetization density $m(z)$, which relate to the spin-resolved density through

$$n(z) = \sum_\sigma n_\sigma(z), \tag{43}$$

$$m(z) = -\mu_B\sum_\sigma \sigma\, n_\sigma(z), \tag{44}$$

with $\mu_B$ being the Bohr magneton. In the KS system, $n(z)$ and $m(z)$ couple, respectively, to the scalar potential $V_s(z)$ and the magnetic field $B_s(z)$:

$$V_s(z) = \sum_\sigma V_{s\sigma}(z)/2, \tag{45}$$

$$B_s(z) = -\sum_\sigma \sigma\, V_{s\sigma}(z)/(2\mu_B), \tag{46}$$

which can be equally used, together with the areal density $\eta$ (chemical potential $\mu$), to build alternative representations of the Q2DEG for closed (open) systems. In analogy to Eq. (9), we can define the spin-density response function

$$\chi^{r}_{mm}(z,z') = \left.\frac{\delta m(z)}{\delta B_s(z')}\right|_{V_s,r} \tag{47}$$

with $r=\mu,\ \eta$. The response function in the equation above relates to their spin-density counterparts in Eq. (9) by

$$\chi^{r}_{mm}(z,z') = \mu_B^2\sum_{\sigma\sigma'}\sigma\sigma'\chi^{r}_{\sigma\sigma'}(z,z'). \tag{48}$$

The magnetic susceptibility $\chi^r_{mm}$ is defined by Eq. (47). In terms of it, the macroscopic magnetic susceptibility, representing the total magnetic polarization $m$ induced by a homogeneous (i.e., independent of $z$ and $\boldsymbol{\rho}$) magnetic field $B_s$, is given by

$$\left.\frac{\partial m}{\partial B_s}\right|_{V_s,r} = \int dz\,dz'\,\chi^{r}_{mm}(z,z'). \tag{49}$$

In order to obtain the macroscopic magnetic susceptibility, we begin by replacing Eq. (48) in (49) and then replace $\chi^{r}_{\sigma\sigma'}(z,z')$ in the resulting expression by Eqs. (38) and (39) for open and closed systems, respectively. The calculation is straightforward, with the result

$$\left.\frac{\partial m}{\partial B_s}\right|_{V_s,\mu} = \frac{N_s\mu_B^2}{2\pi}, \tag{50}$$

$$\left.\frac{\partial m}{\partial B_s}\right|_{V_s,\eta} = \frac{N_s\mu_B^2}{2\pi}\left[1-\left(\frac{N_{s\uparrow}-N_{s\downarrow}}{N_s}\right)^2\right] \tag{51}$$

$$= \frac{2\mu_B^2}{\pi}\left(\sum_\sigma\frac{1}{N_{s\sigma}}\right)^{-1}. \tag{52}$$

From Eq. (50), we see that the KS magnetic susceptibility for an open system is independent of the distribution of electrons between the up and down subbands. This contrasts with the closed-system case: From Eq. (51) we see that the result depends on how electrons distribute in the different spin components. When $N_{s\uparrow} = N_{s\downarrow}$ (e.g., the spin-unpolarized configuration) the susceptibilities for open and closed Q2DEGs coincide. However, in the ferromagnetic case ($N_{s\uparrow} = N_s$, $N_{s\downarrow} = 0$), while the susceptibility for the open system remains finite, it becomes zero for the closed system. This is the expected result because a closed system with a ferromagnetic configuration is saturated: incrementing $B$ will not change the total magnetization because the number of electrons can not increase in the majority spin component. In contrast, in the open case, the reservoir can always provide electrons to increase the induced magnetization. This notable difference between the static noninteracting susceptibilities for open and closed systems is a consequence of the respective constraints and points to the importance of accurately accounting for them into the theory. It also suggests analogous differences for the interacting susceptibilities. For $N_s = 2$, Eq. (50) reproduces the Pauli paramagnetic susceptibility of a strict 2D noninteracting electron gas [38]. In the limit $N_s \gg 1$, one recovers the expression for the 3D homogeneous noninteracting electron gas [39].

### C. OEP equations in terms of the orbital shifts

The OEP equations can be conveniently formulated in terms of the orbital shifts $\psi^{r}_{\mathcal{F}i\sigma}(z)$. The orbital shift for subband $i\sigma$ represents the first-order variation of the KS orbital $\xi_{i\sigma}(z)$ due to a perturbation $\Delta V^{r}_{\mathcal{F}i\sigma}(z) = \theta_{i\sigma} V^{r}_{\mathcal{F}\sigma}(z) - u_{\mathcal{F}i\sigma}(z)$ where $u_{\mathcal{F}i\sigma}(z)$ is the orbital potential given by [40]

$$u_{\mathcal{F}i\sigma}(z) = \frac{1}{2An_{i\sigma}}\frac{1}{\xi_{i\sigma}(z)}\frac{\delta\mathcal{F}}{\delta\xi_{i\sigma}(z)}. \tag{53}$$

Accordingly, from Eq. (25), in terms of the Green's function $G_{i\sigma}$ the orbital shifts are given by

$$\psi^{r}_{\mathcal{F}i\sigma}(z) = \int dz'\,\Delta V^{r}_{\mathcal{F}i\sigma}(z')G_{i\sigma}(z',z)\xi_{i\sigma}(z'), \tag{54}$$

from which the orthogonality property $\int dz\,\psi^{r}_{\mathcal{F}i\sigma}(z)\xi_{i\sigma}(z) = 0$ is derived.

Replacing Eqs. (28) and (38) on the left and right sides of Eq. (13), respectively, and rearranging terms, we obtain the OEP equation for the open system in terms of the shifts:

$$\sum_i n_{i\sigma}\xi_{i\sigma}(z)\psi^{\mu}_{\mathcal{F}i\sigma}(z) = \frac{1}{4\pi}\sum_i C^{\mu}_{\mathcal{F}i\sigma}\xi_{i\sigma}(z)^2, \tag{55}$$

where the scalars $C^{r}_{\mathcal{F}i\sigma}$ are defined by

$$C^{r}_{\mathcal{F}i\sigma} = \theta_{i\sigma}\overline{V}^{r}_{\mathcal{F}i\sigma} - \frac{1}{A}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}, \tag{56}$$

$$\overline{V}^{r}_{\mathcal{F}i\sigma} = \int dz\,\xi_{i\sigma}(z)^2 V^{r}_{\mathcal{F}\sigma}(z). \tag{57}$$

Similarly, if we replace Eqs. (34) and (39) on the left and right of Eq. (17), respectively, the OEP equation for the closed system is obtained:

$$\sum_i n_{i\sigma}\xi_{i\sigma}(z)\psi^{\eta}_{\mathcal{F}i\sigma}(z) = \frac{1}{4\pi}\sum_i\left[C^{\eta}_{\mathcal{F}i\sigma} - \theta_{i\sigma}\frac{N_{s\sigma}\overline{C}^{\eta}_{\mathcal{F}\sigma} + N_{s\underline{\sigma}}\overline{C}^{\eta}_{\mathcal{F}\underline{\sigma}}}{N_s}\right]\xi_{i\sigma}(z)^2. \tag{58}$$

Integrating over $z$ both sides of Eqs. (55) and (58) and using the orthogonality between the shifts and the KS orbitals, we obtain the following conditions on the scalars $C^{r}_{\mathcal{F}}$:

$$\overline{C}^{\mu}_{\mathcal{F}\sigma} = 0, \tag{59}$$

$$\overline{C}^{\eta}_{\mathcal{F}\sigma} = \overline{C}^{\eta}_{\mathcal{F}\underline{\sigma}} \equiv \overline{C}^{\eta}_{\mathcal{F}}, \tag{60}$$

where the overline indicates $\overline{C}^{r}_{\mathcal{F}\sigma} = \sum_i C^{r}_{\mathcal{F}i\sigma}/N_{s\sigma}$, which in the case of occupied-orbital functionals (i.e., functionals that depend exclusively on occupied KS orbitals and occupations) is simply an average of the $C^{r}_{\mathcal{F}i\sigma}$'s over occupied subbands. In the strict ferromagnetic case $N_{s\sigma} \neq 0$ and $N_{s\underline{\sigma}} = 0$, the optimization of Eq. (12) is performed only for the relevant spin component $\sigma$, while the component $\underline{\sigma}$ is ignored. This leads, for the closed case, to an OEP equation identical to Eq. (58) but without the term proportional to $\overline{C}^{\eta}_{\mathcal{F}\underline{\sigma}}$. Therefore, in the strict ferromagnetic case the condition of Eq. (60) plays no role. Returning to the general case $N_{s\sigma}, N_{s\underline{\sigma}} \neq 0$, the spin independence of $\overline{C}^{\eta}_{\mathcal{F}\sigma}$ given by Eq. (60) allows to define a spin-independent scalar $\overline{C}^{\eta}_{\mathcal{F}}$. With this a more compact form of the OEP equation for closed systems [Eq. (58)] can be written:

$$\sum_i n_{i\sigma}\xi_{i\sigma}(z)\psi^{\eta}_{\mathcal{F}i\sigma}(z) = \frac{1}{4\pi}\sum_i\left[C^{\eta}_{\mathcal{F}i\sigma} - \theta_{i\sigma}\overline{C}^{\eta}_{\mathcal{F}}\right]\xi_{i\sigma}(z)^2. \tag{61}$$

Upon addition of a spin-independent constant $K$ to $V^{r}_{\mathcal{F}\sigma}(z)$, the shifts remain unchanged, while the scalars transform according to $\overline{C}^{r}_{\mathcal{F}\sigma} \rightarrow \overline{C}^{r}_{\mathcal{F}\sigma} + K$ and $C^{r}_{\mathcal{F}i\sigma} \rightarrow C^{r}_{\mathcal{F}i\sigma} + \theta_{i\sigma}K$. Then, if $V^{\eta}_{\mathcal{F}\sigma}(z)$ is a solution of Eq. (61), $V^{\eta}_{\mathcal{F}\sigma}(z) + K$ is a solution too, in agreement with the conclusions of Sec. III B for the closed-system potential. On the other side, if a spin-dependent constant $K_\sigma$ is added to the solution $V^{\eta}_{\mathcal{F}\sigma}(z)$, the scalars transform according to $\overline{C}^{\eta}_{\mathcal{F}\sigma} \rightarrow \overline{C}^{\eta}_{\mathcal{F}\sigma} + K_\sigma$. Therefore, by virtue of Eq. (60), the shifted potential $V^{\eta}_{\mathcal{F}\sigma}(z) + K_\sigma$ will not be a solution unless $K_\sigma = K_{\underline{\sigma}} = K$. In other words, the condition of Eq. (60) fixes the *relative* position of the potentials $V^{\eta}_{\mathcal{F}\sigma}(z)$ and $V^{\eta}_{\mathcal{F}\underline{\sigma}}(z)$. For the open system, in contrast, if $V^{\mu}_{\mathcal{F}\sigma}(z)$ is a solution of Eq. (55), $V^{\mu}_{\mathcal{F}\sigma}(z) + K$ will not be a solution of it because a term proportional to $K$ appears on the right-hand side. Thus, $V^{\mu}_{\mathcal{F}\sigma}(z)$ is completely determined by Eq. (55). Finally, let us define $\widetilde{V}_\sigma(z) \equiv V^{\eta}_{\mathcal{F}\sigma}(z) - \overline{C}^{\eta}_{\mathcal{F}}$. By following similar arguments as before, it can be readily verified that $\psi^{\eta}_{\mathcal{F}i\sigma}(z) = \widetilde{\psi}_{i\sigma}(z)$ and $C^{\eta}_{\mathcal{F}i\sigma} = \widetilde{C}_{i\sigma} + \theta_{i\sigma}\overline{C}^{\eta}_{\mathcal{F}}$, where $\widetilde{\psi}_{i\sigma}(z)$ and $\widetilde{C}_{i\sigma}$ are obtained, respectively, from Eqs. (54) and (56) with $V^{r}_{\mathcal{F}\sigma} = \widetilde{V}_\sigma(z)$. Replacing these expressions for $\psi^{\eta}_{\mathcal{F}i\sigma}(z)$ and $C^{\eta}_{\mathcal{F}i\sigma}$ in Eq. (61) we see that $\widetilde{V}_\sigma(z)$ satisfies an equation that is formally identical to Eq. (55). This means that $V^{\mu}_{\mathcal{F}\sigma}(z) = \widetilde{V}_\sigma(z)$, i.e., the two self-consistent solutions for closed and open systems are intrinsically linked by the relation

$$V^{\mu}_{\mathcal{F}\sigma}(z) = V^{\eta}_{\mathcal{F}\sigma}(z) - \overline{C}^{\eta}_{\mathcal{F}}, \tag{62}$$

in agreement with Eq. (21). Since $V^{\eta}_{\mathcal{F}\sigma}(z)$ is determined up to a floating constant, choosing the condition $\overline{C}^{\eta}_{\mathcal{F}} = 0$ to fix this constant leads to an exact matching of both OEP potentials. This condition is equivalent to the condition of Eq. (22) which fixes the floating constant for the open-system potential when calculated in the closed-system representation. The fact that $V^{\mu}_{\mathcal{F}\sigma}(z)$ and $V^{\eta}_{\mathcal{F}\sigma}(z)$ are so simply related as above is one of the main findings of this work since it opens the way to perform calculations for a given Q2DEG closed system using OEP equations for a Q2DEG open system, and vice versa, depending just on convenience. For instance, the noninvertibility of the density response function for closed Q2DEG systems is an obstacle for the numerical evaluation of $V^{\eta}_{\mathcal{F}\sigma}(z)$, starting from Eq. (17). The same problem arises in solid-state applications of the OEP method, where it is solved, for example, by expressing the OEP equations in a restricted reciprocal space $\mathbf{G}$ excluding the $\mathbf{G} = 0$ component [18,19]. We offer here an alternative solution to this problem: Solve the OEP equations for the system of interest in an open environment, for which the density-response function is invertible [36], and then return to the physical closed situation using Eq. (62).

Equation (62) tells us that for given values of $\eta$ and $\mu$, as determined from Eq. (5), the corresponding OEP potentials differ by a constant. This constant is arbitrary because $V^{\eta}_{\mathcal{F}\sigma}(z)$, which enters in the definition of the constant in Eq. (56), is defined up to an arbitrary constant. Nevertheless, as the right-hand side of Eq. (62) is invariant upon addition of a spin-independent constant to $V^{\eta}_{\mathcal{F}\sigma}(z)$, this arbitrariness of $\overline{C}^{\eta}_{\mathcal{F}}$ does not affect the important result that the open-system potential is completely determined. A few remarks regarding its relation to finite systems and solids are in order here: As explained above, setting $\overline{C}^{\eta}_{\mathcal{F}} = 0$ makes the two potentials identical, therefore the *choice* $\overline{C}^{\eta}_{\mathcal{F}} = 0$ is equivalent to the *exact condition* $\overline{C}^{\mu}_{\mathcal{F}} = 0$ of Eq. (59) which fixes the open-system potential. This exact condition is in full analogy to the Eq. (13) of Ref. [35] for finite systems, as discussed above in relation to Eq. (22). Also, the exact condition $\overline{C}^{\mu}_{\mathcal{F}} = 0$ leads to a discontinuity in the open-system exchange (exchange correlation) OEP potential every time a new subband begins to be occupied, as shown in Ref. [40] (Ref. [22]). For closed Q2DEGs, as those studied in Refs. [23–25], the same types of discontinuities appear. This is in analogy to the discontinuities in finite systems [31] and solids [32] when the total number of electrons $N$ passes through integer values. The issue of the discontinuities in finite systems of the derivative of the exchange correlation energy with respect to the particle number, and of the exchange correlation potential, and of the static and dynamic exchange correlation kernel when passing through integer values of $N$ has been exhaustively discussed recently in Refs. [35,41,42].

The relevance of Eqs. (59) and (60) should be emphasized here, as follows. (i) Both constraints are already contained in the defining OEP equations for open [Eq. (55)] and closed [Eq. (61)] systems. In other words, if the solution to any of these two equations is found, the respective constraints of Eqs. (59) or (60) are automatically satisfied. It is important to stress the fact that Eq. (59) is a consequence of the OEP equations for open systems, and should not be confused with the so-called “HOMO constraint” as applied to finite systems (see Ref. [43] and Sec. IV F). Although the constraints of Eqs. (59) or (60) are already contained in the OEP equations, knowing about their existence is extremely useful for the numerical solution of the OEP equations, either as a means to verify the accuracy of the numerical results, or by forcing their satisfaction through the self-consistent iterative numerical loop. We have found crucial this last application for achieving the self-consistent numerical results presented in the following. More discussions about these points are given in Sec. V, devoted to the numerical results. (ii) The one-subband limit (either $N_{\uparrow} = N_{\downarrow} = 1$ or $N_{\sigma} = 1, N_{\underline{\sigma}} \neq 1$) for occupied-orbital functionals of Eqs. (55) and (61) is interesting since it admits an analytical solution, and it is physically relevant [44]. In this situation, the right-hand side of both equations becomes zero, and the analytical solutions are $V^{r}_{\mathcal{F}\sigma}(z) = u_{\mathcal{F}1\sigma}(z)$, with the constraints $C^{\mu}_{\mathcal{F}1\sigma} = 0$ and $C^{\eta}_{\mathcal{F}1\uparrow} = C^{\eta}_{\mathcal{F}1\downarrow} = C^{\eta}_{\mathcal{F}1}$. As in the general case, the solution $V^{\eta}_{\mathcal{F}\sigma}(z)$ is defined up to a floating constant. For $\mathcal{F} = E_x$, the analytical expression for $u_{\mathcal{F}1\sigma}(z)$ is given in Eq. (46) of Ref. [45]. (iii) Equation (60) admits an interesting physical interpretation. As discussed in Appendix B, its satisfaction guarantees that the total energy of an interacting closed system be an extremal with respect to density magnetization variations $\delta m$.

As a final remark to this section, let us emphasize that the OEP equations for open [Eq. (55)] and closed [Eq. (61)] situations derived here are valid both for the case where the functional $\mathcal{F}$ depends on occupied orbitals alone (as is the case of the exchange functional), and also for the case where it depends on occupied and unoccupied orbitals (as is the case of the correlation energy functional derived in Refs. [22,45] for the Q2DEGs in semiconductor quantum wells).

### D. OEP equations as first-order density shifts

A known property of the OEP potential for occupied-orbital functionals is that the density is unchanged to first order upon the orbital-dependent perturbation $\Delta V_{xci\sigma}(\mathbf{r}) = V_{xc\sigma}(\mathbf{r}) - u_{xci\sigma}(\mathbf{r})$ (see, e.g., Ref. [46]). Here we show, also for occupied-orbital functionals [47], that the potentials defined by the OEP Eqs. (55) and (58) fulfill this important property. In the present case the derivation is more involved because the right-hand side in Eqs. (55) and (58) is not zero. This term, that comes from taking into account variations in the occupations, is exactly zero in the usual OEP approach in which orbital occupations are kept fixed [8,9].

Using similar arguments as those to derive Eqs. (38) and (39), it can be shown that for an orbital-dependent perturbation $\Delta V^{r}_{\mathcal{F}i\sigma}(z)$ of the KS potential, the first-order change in the density is

$$\delta^{\mu} n_{\sigma}(z) = 2\sum_{i} \theta_{i\sigma} n_{i\sigma} \xi_{i\sigma}(z) \psi^{\mu}_{i\sigma}(z) - \frac{\theta_{i\sigma}}{2\pi} \overline{\Delta V}^{\mu}_{\mathcal{F}i\sigma} \xi_{i\sigma}(z)^{2} \tag{63}$$

for an open system, and

$$\delta^{\eta} n_{\sigma}(z) = 2\sum_i \theta_{i\sigma} n_{i\sigma} \xi_{i\sigma}(z)\psi^{\eta}_{i\sigma}(z) - \frac{1}{2\pi}\sum_i \theta_{i\sigma} \times \left[\overline{\Delta V}^{\eta}_{\mathcal{F}i\sigma} - \frac{N_{s\sigma}\overline{\Delta V}^{\eta}_{\mathcal{F}i\sigma} + N_{s\underline{\sigma}}\overline{\Delta V}^{\eta}_{\mathcal{F}i\underline{\sigma}}}{N_s}\right]\xi_{i\sigma}(z)^2 \qquad (64)$$

for a closed system. The overline indicates an integral analogous to Eq. (42). For occupied-orbital functionals $\mathcal{F}$ that can be explicitly written as functionals of the set $\{k_{\mathrm{F}i\sigma}\xi_{i\sigma}(z)\}$ (with $k_{\mathrm{F}i\sigma} = \theta_{i\sigma} n^{1/2}_{i\sigma}$ the subband Fermi momentum), as is the case, for example, for all explicit density functionals as well as the exact-exchange energy functional, it can be shown (see Appendix C) that

$$\overline{u}_{\mathcal{F}i\sigma} = \frac{1}{A}\frac{\partial \mathcal{F}}{\partial n_{i\sigma}}. \qquad (65)$$

This implies that the scalars in Eq. (56) fulfill

$$C^{r}_{\mathcal{F}i\sigma} = \overline{\Delta V}^{r}_{\mathcal{F}i\sigma}. \qquad (66)$$

Replacing Eq. (66) in Eqs. (63) and (64), we see immediately that the fulfillment of the OEP equations (55) and (58) for an occupied-orbital functional $\mathcal{F}$ implies that

$$\delta^{r} n_{\sigma}(z) = 0. \qquad (67)$$

This equation admits a very appealing physical interpretation: The potential $V^{r}_{\mathcal{F}\sigma}(z)$ that optimizes the energy of the interacting Q2DEG is such that the first-order change in the density vanishes when the KS system is perturbed with the orbital-dependent perturbation $\Delta V^{r}_{\mathcal{F}i\sigma}(z)$.

If the restriction over occupied-only functionals is lifted, one arrives at

$$\frac{\delta^{r} n_{\sigma}(z)}{2} = -\sum_i^{unocc} n_{i\sigma}\xi_{i\sigma}(z)\psi^{r}_{\mathcal{F}i\sigma}(z) + \frac{1}{4\pi}\sum_i^{unocc}\left[C^{r}_{\mathcal{F}i\sigma} - \theta_{i\sigma}\overline{C}^{r}_{\mathcal{F}}\right]\xi_{i\sigma}(z)^2, \qquad (68)$$

where the summations on the right-hand side run over unoccupied orbitals. In this case, an interpretation of the OEP potential as made after Eq. (67) does not seem to be possible because there is not an obvious reason for the right-hand side of Eq. (68) to be exactly zero.

### E. Explicit expressions for $V^{r}_{\mathcal{F}\sigma}(z)$ in terms of the shifts

By virtue of the completeness of the basis formed by the single-particle KS orbitals, it can be shown using Eqs. (26) and (54) that the shifts satisfy the following differential equation:

$$[H_{s\sigma}(z) - \varepsilon_{i\sigma}]\psi^{r}_{\mathcal{F}i\sigma}(z) = -\left[\Delta V^{r}_{\mathcal{F}i\sigma}(z) - \overline{\Delta V}^{r}_{\mathcal{F}i\sigma}\right]\xi_{i\sigma}(z), \qquad (69)$$

where the KS Hamiltonian $H_{s\sigma}(z)$ is identical to the expression in square brackets on the left-hand side of Eq. (2). Using this result and Eq. (2), after some algebra it is possible to find that the potential $V^{r}_{\mathcal{F}\sigma}(z)$ can be written as the sum of two terms

$$V^{r}_{\mathcal{F}\sigma}(z) = V^{r,\mathrm{KLI}}_{\mathcal{F}\sigma}(z) + V^{r,\Delta}_{\mathcal{F}\sigma}(z), \qquad (70)$$

where the first term is to be identified with the Krieger-Li-Iafrate (KLI) approximation [48]

$$V^{r,\mathrm{KLI}}_{\mathcal{F}\sigma}(z) = \sum_i \frac{\theta_{i\sigma} n_{i\sigma}\xi_{i\sigma}(z)^2}{n_{\sigma}(z)}\left[u_{\mathcal{F}i\sigma}(z) + \overline{\Delta V}^{r}_{\mathcal{F}i\sigma}\right], \qquad (71)$$

and the second term is

$$V^{r,\Delta}_{\mathcal{F}\sigma}(z) = \sum_i \frac{\theta_{i\sigma} n_{i\sigma}}{2n_{\sigma}(z)}\left[\xi_{i\sigma}(z)\psi^{r}_{\mathcal{F}i\sigma}(z)'' - \xi_{i\sigma}(z)''\psi^{r}_{\mathcal{F}i\sigma}(z)\right], \qquad (72)$$

where the prime denotes derivation with respect to $z$. Up to now, we have not taken into account the OEP equation and therefore Eq. (70) is trivial in the sense that it is verified for arbitrary $V^{r}_{\mathcal{F}\sigma}(z)$. In order to incorporate the OEP equation, we derive twice Eqs. (55) and (61) with respect to $z$ and use the conditions on the $\overline{C}^{r}_{\mathcal{F}\sigma}$ from Eqs. (59) and (60). Restricting to the case of occupied-orbital functionals $\mathcal{F}$ one gets, after some algebra,

$$V^{r,\Delta}_{\mathcal{F}\sigma}(z) = -\sum_i \frac{\theta_{i\sigma} n_{i\sigma}}{n_{\sigma}(z)}\frac{d}{dz}\left[\psi^{r}_{\mathcal{F}i\sigma}(z)\xi_{i\sigma}(z)'\right] + \sum_i \theta_{i\sigma}\frac{C^{r}_{\mathcal{F}i\sigma} - \overline{C}^{r}_{\mathcal{F}\sigma}}{8\pi n_{\sigma}(z)}\frac{d^2\xi_{i\sigma}(z)^2}{dz^2}. \qquad (73)$$

Equations (70), (71), and (73) together with the corresponding conditions from Eq. (59) or (60) completely determine the potential $V^{r}_{\mathcal{F}\sigma}(z)$. They must be solved self-consistently. It should be clear at this point that the set of Eqs. (55) and (61) is fully equivalent to Eqs. (70) plus the constraints of Eqs. (59) and (60). The use of one set of equations or the other is just a question of convenience; in our case, we have used the form (70) for the numerical calculations, and the forms (55) or (61) for obtaining analytical results, like the asymptotic limits to be discussed following.

Since our OEP equations and the associated constraints are different from the ones used usually in deriving the KLI approximation, some justification is needed for the label "KLI" in Eqs. (70) and (71). In the first place, the explicit expression for $V^{r,\mathrm{KLI}}_{\mathcal{F}\sigma}(z)$ in Eq. (71) is identical to the one in previous works [9]. Second, by following the steps in Ref. [8], it can be shown that

$$\int dz\, n_{\sigma}(z)\, V^{r,\Delta}_{\mathcal{F}\sigma}(z) = 0. \qquad (74)$$

In words, the meaning of this equation is that the weighted average of the terms in $V^{r}_{\mathcal{F}\sigma}$ beyond the KLI approximation is zero. This is exactly the same that has been found in previous works, and used as argument to justify both the approximation and its well-documented good performance for localized systems such as atoms and molecules [8], and also for extended systems [26,27].

### F. Asymptotic limits for $x$-only OEP

Here, we will be mainly concerned with the asymptotic limit of $V^{\eta}_{x\sigma}(z)$, that is, in the special case when $\mathcal{F} = E_x$ is the exact-exchange energy functional. Solving Eq. (62) for $V^{\eta}_{x\sigma}(z)$ and taking the limit $z \to \infty$,

$$V^{\eta}_{x\sigma}(z \to \infty) = V^{\mu}_{x\sigma}(z \to \infty) + \overline{C}^{\,\eta}_{x}. \tag{75}$$

Now, in previous works we have already obtained that [22,40]

$$V^{\mu}_{x\sigma}(z \to \infty) \to \overline{\Delta V}^{\,\mu}_{xm_\sigma\sigma} = \overline{V}^{\,\mu}_{xm_\sigma\sigma} - \overline{u}_{xm_\sigma\sigma}, \tag{76}$$

with $i = m_\sigma$ being the last occupied (the more energetic) energy level corresponding to spin $\sigma$. Using Eq. (62) once more,

$$\overline{V}^{\,\mu}_{xm_\sigma\sigma} = \overline{V}^{\,\eta}_{xm_\sigma\sigma} - \overline{C}^{\,\eta}_{x}\,, \tag{77}$$

and replacing in the previous equation,

$$V^{\mu}_{x\sigma}(z \to \infty) \to \overline{V}^{\,\eta}_{xm_\sigma\sigma} - \overline{u}_{xm_\sigma\sigma} - \overline{C}^{\,\eta}_{x}. \tag{78}$$

Returning to Eq. (75),

$$V^{\eta}_{x\sigma}(z \to \infty) \to \overline{\Delta V}^{\,\eta}_{xm_\sigma\sigma} = \overline{V}^{\,\eta}_{xm_\sigma\sigma} - \overline{u}_{xm_\sigma\sigma}. \tag{79}$$

Since in the closed system $V^{\eta}_{x\sigma}(z)$ is defined up to a floating constant, one can choose (either for $\sigma = \uparrow$ or $\sigma = \downarrow$, but not for both) that $V^{\eta}_{x\sigma}(z \to \infty) \to 0$ by imposing the condition $\overline{\Delta V}^{\,\eta}_{xm_\sigma\sigma} = 0$, that implies that $\overline{V}^{\,\eta}_{xm_\sigma\sigma} = \overline{u}_{xm_\sigma\sigma}$. On the other side, the other spin component of the exchange potential, $V^{\eta}_{x\underline{\sigma}}(z)$, is *fully* determined by the constraint in Eq. (60), and has then the asymptotic limit $V^{\eta}_{x\underline{\sigma}}(z \to \infty) \to \overline{\Delta V}^{\,\eta}_{xm_{\underline{\sigma}}\underline{\sigma}} = \overline{V}^{\,\eta}_{xm_{\underline{\sigma}}\underline{\sigma}} - \overline{u}_{xm_{\underline{\sigma}}\underline{\sigma}}$, which in general will be different from zero. Regarding the open system, its asymptotic limit is given by Eq. (76). In this case, if $n_\uparrow \neq n_\downarrow$ and both densities are different from zero, the spin-dependent OEP exchange potential tends to *two* different constant values far in the vacuum region. In the spin-unpolarized case, the asymptotic limit $V^{\eta}_{x\sigma}(z \to \infty) = V^{\eta}_{x\underline{\sigma}}(z \to \infty) := V^{\eta}_{x}(z \to \infty) \to \overline{\Delta V}^{\,\eta}_{xm_\sigma\sigma} = \overline{\Delta V}^{\,\eta}_{xm_{\underline{\sigma}}\underline{\sigma}} := \overline{\Delta V}^{\,\eta}_{xm}$. Fixing the floating constant by the standard choice $\overline{\Delta V}^{\,\eta}_{xm} = 0$, then $V^{\eta}_{x}(z \to \infty) \to 0$. The spin-unpolarized asymptotic limit in the open system has been discussed in Ref. [40]. It has been shown there that $V^{\mu}_{x\sigma}(z \to \infty) = V^{\mu}_{x\underline{\sigma}}(z \to \infty) := V^{\mu}_{x}(z \to \infty) \to \overline{\Delta V}^{\,\mu}_{xm_\sigma\sigma} = \overline{\Delta V}^{\,\mu}_{xm_{\underline{\sigma}}\underline{\sigma}} := \overline{\Delta V}^{\,\mu}_{xm}$. And since $\overline{\Delta V}^{\,\mu}_{xm} \neq 0$ in general, the OEP exchange potential of the spin-unpolarized open system tends to a well-determined single finite value in the asymptotic region.

In the context of finite systems such as atoms and molecules, the equivalent to the condition $\overline{V}^{\,\eta}_{xm_\sigma\sigma} = \overline{u}_{xm_\sigma\sigma}$ has been obtained in several works, either by appealing to fractional particle numbers [48], from scaling arguments [49], or from analyzing the asymptotic limit of the corresponding $x$-only OEP equations [8]. This constraint, denoted in this context as the "HOMO constraint," is very useful in helping for solving the intrinsic invertibility problem of the density-response function for this type of system [43]. An important difference, however, is that this "HOMO constraint" is applied in finite systems to both spin components, while in our Q2DEG case we have found that it only can be applied to one of the two spin components (closed case) or to none of them (open case), for a spin-polarized situation. As discussed above, this has the consequence that the asymptotic value of the "unconstrained" spin component of the OEP potential has in general a nonzero asymptotic value. A numerical example of this behavior is given in the next section.

## V. NUMERICAL EXAMPLE FOR A JELLIUM METALLIC SLAB

The jellium-slab model for a metallic surface represents an example of Q2DEG [50,51]. In this model, the positive ions inside the metal are replaced by a uniform 3D slab charge distribution $n_+(z)$ (the jellium):

$$n_+(z) = n_0\, \theta\left(\frac{d}{2} - |z|\right), \tag{80}$$

where $d$ is the width of the slab with edges at $z = -d/2$ and $z = d/2$ and $n_0 = \eta/d$ [52]. The attractive jellium confines the movement of electrons in the $z$ direction, while they are free to move along the translational invariant $x$-$y$ plane. Defined this way, the slab has global (but not local) neutrality. The same model has been studied in the past for the spin-unpolarized case [23–25]. We want here to give a numerical example of the utility of the spin-dependent OEP formalism for closed Q2DEGs developed in the previous sections, for a concrete spin-polarized situation.

The calculation of the $x$-only OEP ($x$-OEP) spin-dependent exchange potential for the closed system $V^{\eta}_{x\sigma}(z)$ amounts to the numerical solution of Eqs. (70), (71), and (73), imposing the fulfillment of Eq. (60). In the case of the $x$-only KLI ($x$-KLI) approximation, the last term in the right-hand side of Eq. (70) was removed. In all the results shown following, the correlation potential has been neglected.

This set of equations has to be solved in a self-consistent procedure. The standard way, which we have applied, consists of the following steps: (i) Solve the Kohn-Sham equations [Eq. (2)] for each spin projection with an initial approximated KS potential. We have used as initial potential $V^{\eta,\mathrm{KLI}}_{s\sigma}(z) = V_e(z) + V_{\mathrm{H}}(z) + V^{\eta,\mathrm{KLI}}_{x\sigma}(z)$ in the case of $x$-OEP. (ii) Use the resulting orbitals $\xi_{i\sigma}(z)$ and energies $\varepsilon_{i\sigma}$ to obtain the orbital potentials $u_{xi\sigma}(z)$ and then solve the differential equation Eq. (69) for the orbital shifts $\psi^{\eta}_{xi\sigma}(z)$. (iii) Insert the orbitals, subband occupations, orbital potentials, and orbital shifts in Eqs. (70), (71), and (73) to obtain an improved $V^{\eta}_{x\sigma}(z)$. (iv) Update the KS potential using a mixture of the initial KS potential and the new one. (v) Self-consistently solve the Kohn-Sham equations with the improved approximation. (vi) Repeat these steps until convergence is achieved. Step (ii), where the orbital potentials and orbital shifts are numerically determined, is by far the more demanding step of the $x$-OEP self-consistent loop regarding the computational time. This step is the main difference with the self-consistent $x$-KLI loop, where the calculation starts from an $x$-LDA seed for the initial KS potential and there is no need of solving the differential equation for the shifts [53]. This, together with the fact that the mixing factor used in step (iv) may be greater in the self-consistent $x$-KLI loop, combines in yielding great savings of computational cost, and as we will show, with only a small loss of accuracy as compared to the $x$-OEP result. More details on the numerical technique for solving the Q2DEG's OEP equations are given in Refs. [23–25].

Inside the iterative self-consistent loop we have used the invariance upon the addition of a spin-independent constant to

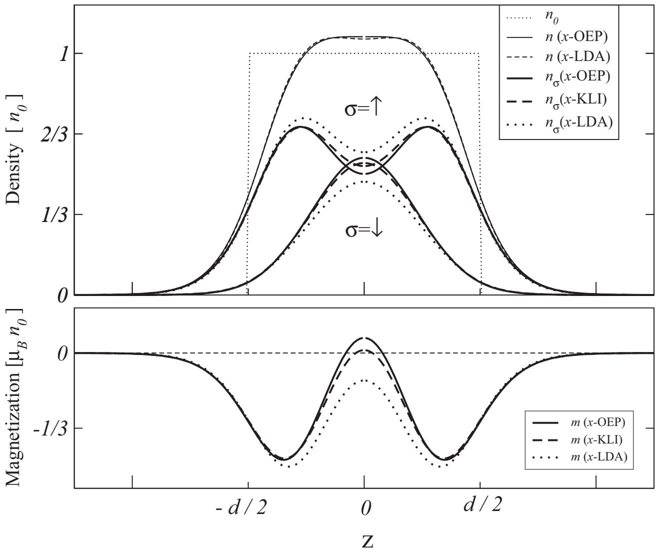

FIG. 1. Upper panel: total $[n(z)]$ and spin-dependent densities $[n_\sigma(z)]$. Full lines are for the $x$-OEP case, dashed and dotted lines correspond to the $x$-KLI and $x$-LDA approximations, respectively. Lower panel: magnetizations $m(z)=\mu_B[n_\downarrow(z)-n_\uparrow(z)]$. The slab parameters are $r_s=5.0$ and $d=0.8\,\lambda_F$.

$V^{\eta}_{x\sigma}(z)$ in order to enforce the limit $V^{\eta}_{s\uparrow}(z\to\infty)\to 0$ for the most populated spin component, chosen here (arbitrarily) as the up-spin component; this is equivalent to the assumption that $V^{\eta}_{x\uparrow}(z\to\infty)\to 0$. As discussed above, if $i=m_\uparrow$ is the last occupied slab discrete level corresponding to the most populated spin component, this assumption is enforced by the constraint $\Delta\overline{V}^{\eta}_{xm_\uparrow\uparrow}=\overline{V}^{\eta}_{xm_\uparrow\uparrow}-\overline{u}_{xm_\uparrow\uparrow}=0$. With this choice and the condition given by Eq. (60), the less populated spin component ($\sigma=\downarrow$) of the $x$-OEP and $x$-KLI exchange potentials is *fully* determined.

Densities and magnetizations for a narrow slab with $r_s=5$ and $d=0.8\,\lambda_F$ (with $\lambda_F$ as defined in Ref. [30]) are shown in Fig. 1, for three different approximations: $x$-LDA, $x$-KLI, and $x$-OEP. While the total densities are essentially the same in the three approximations, spin-discriminated densities are much more sensitive to which approximation is used, leading to the different magnetizations shown in the lower panel. It is interesting to compare $m(z)$ for the $x$-LDA and the $x$-OEP calculations. In the $x$-LDA case (dotted line), $m(z)<0$ in the full $z$ range, corresponding to a ferromagnetic ground state. Conversely, the $x$-OEP gives a slightly antiferromagnetic configuration with $m(z)<0$ on the sides of the slab and $m(z)>0$ close to the center. This result is counterintuitive: As compared to $x$-OEP, the $x$-LDA potential underestimates the exchange interaction, therefore, it is natural to expect that $x$-OEP would favor a global ferromagnetic configuration more strongly than LDA.

Nevertheless, this result can be understood on the basis that, separately for each spin component, the $x$-OEP potential favors spatial charge accumulation more effectively than $x$-LDA, in order to maximize the negative exchange interaction energy. Under this basic fact, we can analyze the different potentials and densities for each spin component.

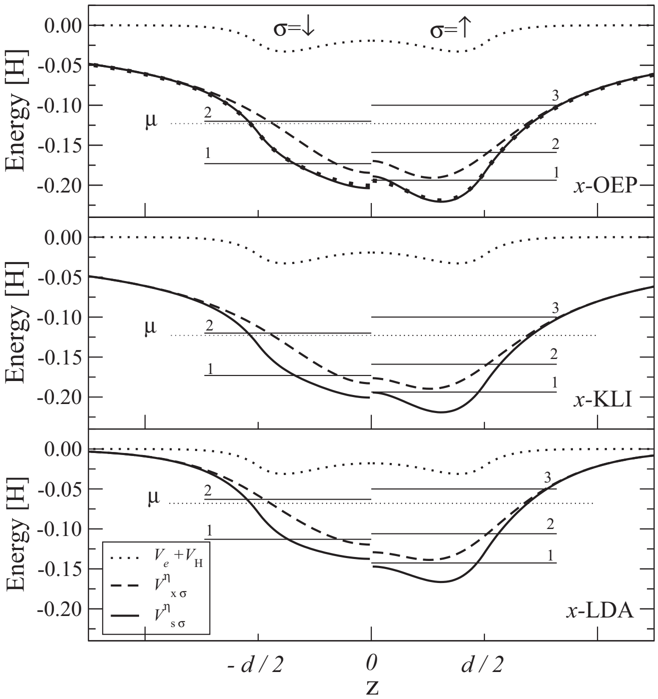

FIG. 2. External plus Hartree (dotted lines), exchange (dashed lines), and Kohn-Sham (full lines) self-consistent potentials for the $x$-OEP, $x$-KLI, and $x$-LDA approximations (upper, middle, and lower panels, respectively), for the same slab of Fig. 1. For $z<0$, the $\sigma=\downarrow$ (minority-spin component) electronic structure is shown; for $z>0$, the $\sigma=\uparrow$ (majority-spin component) electronic structure is shown. $\mu$ is the chemical potential, and the horizontal straight lines represent the spin-dependent KS eigenvalues $\varepsilon_{i\sigma}$. In the upper panel, the $x$-KLI KS potential has been included, for comparison.

As displayed in Fig. 2, for $\sigma=\downarrow$ (minority-spin component), the $x$-OEP potential is deeper than the $x$-LDA one, as can be seen from the difference $\Delta=V_{x\downarrow}(-d/2)-V_{x\downarrow}(0)$: $\Delta(\mathrm{OEP})\simeq 0.0716$ H $>\Delta(\mathrm{LDA})\simeq 0.0602$ H. Therefore, the spin-down $x$-OEP potential favors charge accumulation in the center more effectively than $x$-LDA. The case for $\sigma=\uparrow$ is more involved because there are two occupied subbands. Due to symmetry considerations, the subband of lowest energy $\xi_{1\uparrow}(z)$ is symmetrical around the center of the slab, while $\xi_{2\uparrow}(z)$ is antisymmetrical and therefore exactly zero in the slab center. As a consequence, it turns out to be more favorable to accumulate charge on the sides of the slab through the building of a barrier in the middle. The height of this barrier is roughly given by $\Delta_b=V_{x\uparrow}(0)-V_{x\uparrow}(\mathrm{min})$, where $V_{x\uparrow}(\mathrm{min})$ is the potential's minimum. We have $\Delta_b(\mathrm{OEP})\simeq 0.0211$ H $>$ $\Delta_{\mathrm{b}}(\mathrm{LDA})\simeq 0.0095$ H. In sum, the combined effect of a deeper potential for $\sigma=\downarrow$ and a higher middle barrier for $\sigma=\uparrow$ leads to a slight antiferromagnetic configuration for $x$-OEP, not observed in the $x$-LDA result. The $x$-KLI magnetization lies in-between the $x$-LDA and $x$-OEP.

An interesting feature regarding the results displayed in Fig. 2 is the much slower decay of the $x$-OEP and $x$-KLI exchange potentials, as compared with the $x$-LDA. This has been already discussed for the spin-unpolarized situation, were it has been found that $V^{\eta}_{x\uparrow}(z\to\infty)=V^{\eta}_{x\downarrow}(z\to\infty)\to$

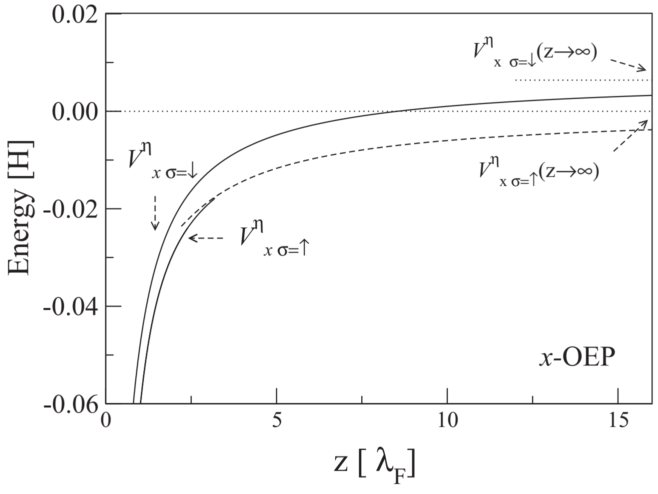

FIG. 3. Asymptotic behavior of the spin-dependent exchange potential in the $x$-OEP approximation, for the same slab of Fig. 1; the slab center is at $z = 0$. Full curves represent $V^{\eta}_{x\sigma}(z)$, with $\sigma = \uparrow$ ($\sigma = \downarrow$) for the majority- (minority-) spin component. The dashed curve is an approximate asymptotic limit for the many-subband $x$-OEP exchange potential (see text for more details), and the two arrows on the right signal the two *different* asymptotic limits for the $\uparrow$ and $\downarrow$ exchange potentials.

$-e^2/z$, both for the $x$-OEP and $x$-KLI approximations [23–27]. The $x$-LDA exchange potential, on the other side, decays much more rapidly, following the exponential decay of the density displayed in Fig. 1.

We have included in the top panel of Fig. 2 the KLI results for the spin-dependent KS potentials of the middle panel; clearly the KLI potentials are hardly distinguishable from the full OEP exchange potentials, suggesting once more the accuracy of the KLI approximation as applied to Q2DEGs [40].

We display in Fig. 3 an enhanced view of the upper panel of Fig. 2, for small enough energies, and for values of $z$ deep in the vacuum region ($z \gg d$). The asymptotic limit of the minority-spin component (with just one occupied subband) is given by $V^{\eta}_{x\downarrow}(z \to \infty) \to \overline{V}^{\eta}_{x1\downarrow} - \bar{u}_{x1\downarrow} - e^2/z \simeq 0.0065\,\mathrm{H} - e^2/z$. On the other side, the asymptotic limit for the majority-spin component (with two occupied subbands) is given by $V^{\eta}_{x\uparrow}(z \to \infty) \to \overline{V}^{\eta}_{x2\uparrow} - \bar{u}_{x2\uparrow} - e^2/z = -e^2/z$, according to our choice of the floating constant for $V^{\eta}_{x\sigma}(z)$ discussed above ($\overline{V}^{\eta}_{x2\uparrow} - \bar{u}_{x2\uparrow} = 0$). For $z = 15\,\lambda_F$, we obtain that $V^{\eta}_{x\downarrow}(z = 15\,\lambda_F) \simeq 0.0024\,\mathrm{H}$, while $V^{\eta}_{x\uparrow}(z = 15\,\lambda_F) \simeq -0.0041\,\mathrm{H}$, in good agreement with the results displayed in Fig. 3. The important point here is that $V^{\eta}_{x\uparrow}(z \to \infty) \neq V^{\eta}_{x\downarrow}(z \to \infty)$, and there is no possible choice of the unique floating constant that allows the matching of the two asymptotic limits. This is a consequence of the crucial constraint imposed by Eq. (60). This is true, of course, if $n_\uparrow \neq n_\downarrow$. If $n_\uparrow = n_\downarrow$, the spin-unpolarized case, $V^{\eta}_{x\uparrow}(z) = V^{\eta}_{x\downarrow}(z)$, so both potentials approach the same asymptotic limit automatically, and the choice for the floating constant defines the (arbitrary) zero of energy or vacuum level. It should be noted that the dashed curve in Fig. 3 corresponds to an accurate analytical approximation for $V^{\eta}_{x\uparrow}(z)$, valid in the asymptotic regime; the explicit expression for it is given by Eq. (35) in Ref. [24].

## VI. CONCLUSIONS

In summary, we have generalized the optimized effective potential implementation of ground-state DFT, as applied to quasi-two-dimensional electron gases in two important directions: (a) open versus closed systems, and (b) spin-unpolarized versus spin-polarized configurations. Regarding the first item, we have found that the OEP potentials for the open and closed possible representations of a given physical system are just related by a simple spin-independent constant: $V^{\eta}_{\mathcal{F},\sigma}(z) = V^{\mu}_{\mathcal{F},\sigma}(z) + \overline{C}^{\eta}_{\mathcal{F}}$. This essentially means that both descriptions lead to the same results for the density, as for any other ground-state physical magnitude, and that the choice of which representation to use is mainly a question of convenience. For instance, quasi-two-dimensional electron gases at the interface between two different semiconductors are usually represented as open systems, while metallic jellium slabs are usually represented as closed systems. This work suggests that these choices are not unique, and that both types of systems may be represented equivalently as open or closed. As a consequence, a calculation scheme is advanced to avoid the noninvertibility problem of the density-response function for closed Q2DEG systems.

Regarding the second issue, we have found the fundamental conditions of Eqs. (59) and (60). In the case of spin-polarized systems, their satisfaction implies that both spin-up and -down OEP potentials are different for *all* values of the position coordinate, even far in the vacuum region for the jellium slab model of a metallic surface. This results from the fact that for a spin-polarized system we have *two* different asymptotic limits for the spin-up and -down OEP potentials, but just a *single* spin-independent floating constant for the closed system, and *no floating constant at all* for the open case. This crucial point has been illustrated through the numerical solution of a spontaneously spin-polarized narrow metallic slab, for which the $x$-OEP exchange potential has been obtained exactly. The $x$-KLI approximation for the same system has been also tested, and found to be quite accurate, similarly to its well-documented accuracy found in non-condensed-matter systems, such as atoms and molecules.

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## APPENDIX A: RELATION BETWEEN OPEN- AND CLOSED-SYSTEM POTENTIAL FUNCTIONAL DERIVATIVES

Consider a variation $\delta V_{s\sigma'}(z') = \epsilon\delta_{\sigma\sigma'}\delta(z - z')$, $\delta\eta = 0$ in the $\{V_{s\sigma}, \eta\}$ representation, with $\epsilon$ a small real number. For this variation, the change in a density functional $\mathcal{F}$ will be

$$\delta\mathcal{F} = \epsilon \left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\eta}. \tag{A1}$$

Now, consider an equivalent variation in the $\{V_{s\sigma},\mu\}$ representation, that is, one with the same $\delta V_{s\sigma'}(z')$ as above and a variation $\delta\mu$ that keeps the areal density unchanged. Such $\delta\mu$ is easily found by applying Eq. (A1) to $\mathcal{F}=\mu-V_{s\sigma'}(z')$: $\delta\mu=\epsilon\delta\mu/\delta V_{s\sigma}(z)|_{\eta}$. With this result, we can express the variation $\delta\mathcal{F}$ in Eq. (A1) in the representation $\{V_{s\sigma},\mu\}$ as

$$\delta\mathcal{F}=\epsilon\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\mu}+\epsilon\left.\frac{\partial\mathcal{F}}{\partial\mu}\right|_{V_{s\updownarrow}}\left.\frac{\delta\mu}{\delta V_{s\sigma}(z)}\right|_{\eta}. \tag{A2}$$

We can now equate Eqs. (A1) and (A2) to find the following relation between fixed-$\eta$ and fixed-$\mu$ functional derivatives:

$$\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\mu}=\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\eta}-\left.\frac{\partial\mathcal{F}}{\partial\mu}\right|_{V_{s\updownarrow}}\left.\frac{\delta\mu}{\delta V_{s\sigma}(z)}\right|_{\eta}. \tag{A3}$$

In an analogous fashion, considering first a variation with $\delta\mu=0$ and the equivalent variation $\delta\eta$ that leaves $\mu$ unchanged in the $\{V_{s\sigma},\eta\}$ representation, the related equation is obtained:

$$\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\eta}=\left.\frac{\delta\mathcal{F}}{\delta V_{s\sigma}(z)}\right|_{\mu}-\left.\frac{\partial\mathcal{F}}{\partial\eta}\right|_{V_{s\updownarrow}}\left.\frac{\delta\eta}{\delta V_{s\sigma}(z)}\right|_{\mu}. \tag{A4}$$

## APPENDIX B: PHYSICAL INTERPRETATION OF EQ. (60)

We begin by writing the total energy of the interacting system as

$$E=T_s+A\int dz\,V_e(z)n(z)+E_{\mathrm{H}}+\mathcal{F} \tag{B1}$$

$$\begin{aligned}&=E_s+E_{\mathrm{H}}-A\int dz\,V_{\mathrm{H}}(z)n(z)\\&\quad+\mathcal{F}-A\sum_{\sigma}\int dz\,V_{\mathcal{F}\sigma}(z)n_{\sigma}(z).\end{aligned} \tag{B2}$$

The ground-state density is obtained from optimizing Eq. (B1) with respect to the density. To perform an equivalent optimization using Eq. (B2) it is essential to realize that the KS potential $V_{s\sigma}$ in $E_s=T_s+A\sum_{\sigma}\int dz\,V_{s\sigma}(z)n_{\sigma}(z)$, as well as its components $V_{\mathrm{H}}$ and $V_{\mathcal{F}\sigma}$, must be kept fixed when varying the density (or equivalent representing variables). Otherwise, the optimized functional is not the same as the fundamental one of Eq. (B1).

Taking as independent variables the set $\{V_s,B_s,\eta\}$, the optimization of Eq. (B2) with respect to $B_s$ is written

$$\begin{aligned}\left.\frac{\partial E}{\partial B_s}\right|_{V_s\eta}&=\left.\frac{\partial E_s}{\partial B_s}\right|_{V_s\eta}+\Bigg[\left.\frac{\partial\mathcal{F}}{\partial B_s}\right|_{V_s\eta}\\&\quad-A\sum_{\sigma}\int dz\,V_{\mathcal{F}\sigma}(z)\left.\frac{\partial n_{\sigma}(z)}{\partial B_s}\right|_{V_s\eta}\Bigg]=0.\end{aligned} \tag{B3}$$

The analogous term to the one in square brackets for the Hartree component is zero, therefore, it is not written in the above equation. Only the macroscopic component has been taken in writing Eq. (B3). By definition, both $E$ and $E_s$ are optimized at the same density, therefore, the term in square brackets in Eq. (B3) must be zero:

$$\left.\frac{\partial\mathcal{F}}{\partial B_s}\right|_{V_s,\eta}-A\sum_{\sigma}\int dz\,V_{\mathcal{F}\sigma}(z)\left.\frac{\partial n_{\sigma}(z)}{\partial B_s}\right|_{V_s,\eta}=0. \tag{B4}$$

This is just a form of the OEP equation: The optimized potential $V_{\mathcal{F}\sigma}(z)$ must satisfy Eq. (B4) in order to have a common density (equivalently a common set $\{V_s,B_s,\eta\}$) optimizing both $E$ and $E_s$.

For an orbital-based functional $\mathcal{F}$, we can apply now the chain rule to the magnetic field functional derivative $\partial\mathcal{F}/\partial B_s|_{V_s,\eta}$. We do not include derivatives with respect to orbital shapes, as they remain fixed for a uniform (i.e., $z$ and $\rho$ independent) change of $B_s$:

$$\left.\frac{\partial\mathcal{F}}{\partial B_s}\right|_{V_s,\eta}=\sum_{i\sigma}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}\left.\frac{\partial n_{i\sigma}}{\partial B_s}\right|_{V_s,\eta}, \tag{B5}$$

subsequent application of the chain rule with respect to $V_{s\sigma}$ gives, for the magnetic field derivative on the right-hand side,

$$\left.\frac{\partial n_{i\sigma}}{\partial B_s}\right|_{V_s,\eta}=\sum_{\sigma'}\int dz\,dz'\left.\frac{\delta n_{i\sigma}}{\delta V_{s\sigma'}(z')}\right|_{\eta}\left.\frac{\delta V_{s\sigma'}(z')}{\delta B_s(z)}\right|_{V_s,\eta}. \tag{B6}$$

The first derivative on the right-hand side is given by Eq. (33). The second is easily obtained from $V_{s\sigma}(z)=V_s(z)-\mu_B\sigma B_s(z)$, with the result $\delta V_{s\sigma'}(z')/\delta B_s(z)=-\mu_B\sigma'\delta(z-z')$. In sum, after integrating the resulting expression and taking into account the value of the magnetic susceptibility for the closed system [Eq. (51)], we obtain

$$\left.\frac{\partial\mathcal{F}}{\partial B_s}\right|_{V_s,\eta}=\frac{\mu_B}{2}\left.\frac{\partial m}{\partial B_s}\right|_{V_s,\eta}\sum_{i\sigma}\frac{\sigma}{N_{s\sigma}}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}. \tag{B7}$$

Applying Eq. (B7) to the first term of Eq. (B4) and to the derivative of $n_{\sigma}(z)$ in the second term, we obtain, after using (38) and integrating on $z$,

$$\begin{aligned}&\left.\frac{\partial\mathcal{F}}{\partial B_s}\right|_{V_s,\eta}-A\sum_{\sigma}\int dz\,V_{\mathcal{F}\sigma}(z)\left.\frac{\partial n_{\sigma}(z)}{\partial B_s}\right|_{V_s,\eta}\\&\quad=\frac{\mu_B}{2}\left.\frac{\partial m}{\partial B_s}\right|_{V_s,\eta}\sum_{\sigma}\sigma\overline{C}^{\eta}_{\mathcal{F}\sigma}=0\end{aligned} \tag{B8}$$

with $\overline{C}^{\eta}_{\mathcal{F}\sigma}$ as defined in Sec. IV C. The second equality in Eq. (B8) is the same as the condition (60) for closed systems. Taking into account the first equality in Eq. (B8), together with Eq. (B3) and the definition $\delta\mathcal{G}|_{V_s\eta}\equiv\partial\mathcal{G}/\partial B_s|_{V_s\eta}\delta B_s$, for $\mathcal{G}$ an arbitrary functional of $\{V_s,B_s,\eta\}$, we can write

$$\delta E|_{V_s,\eta}=\delta E_s|_{V_s,\eta}+\frac{\mu_B}{2}\left(\sum_{\sigma}\sigma\overline{C}^{\eta}_{\mathcal{F}\sigma}\right)\delta m|_{V_s,\eta}. \tag{B9}$$

Thus, the second equality of Eq. (B8) (or equivalently [60]) expresses the stability of the closed *interacting* system with respect to the total magnetization $m$.

### APPENDIX C: RELATION BETWEEN $\bar{u}_{\mathcal{F},i\sigma}$ AND $\partial\mathcal{F}/\partial n_{i\sigma}$ FOR ORBITAL FUNCTIONALS $\mathcal{F}$

For functionals that depend explicitly on the set $\{p_{i\sigma}(z)=k_{\mathrm{F}i\sigma}\xi_{i\sigma}(z)\}$, we can apply the chain rule for functional derivatives to obtain

$$\begin{aligned}\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}&=\sum_{j\sigma'}\int dz\,\frac{\delta\mathcal{F}}{\delta p_{j\sigma'}(z)}\frac{\partial p_{j\sigma'}(z)}{\partial n_{i\sigma}}\\&=\frac{1}{2n_{i\sigma}^{1/2}}\int dz\,\xi_{i\sigma}(z)\frac{\delta\mathcal{F}}{\delta p_{i\sigma}(z)}\end{aligned}\qquad\text{(C1)}$$

and

$$\frac{\delta\mathcal{F}}{\delta\xi_{i\sigma}(z)}=\sum_{j\sigma'}\int dz'\,\frac{\delta\mathcal{F}}{\delta p_{j\sigma'}(z)}\frac{\delta p_{j\sigma'}(z)}{\delta\xi_{i\sigma}(z)}=n_{i\sigma}^{1/2}\frac{\delta\mathcal{F}}{\delta p_{i\sigma}(z)}.\qquad\text{(C2)}$$

From Eqs. (C1) and (C2) we conclude that

$$\frac{\partial\mathcal{F}}{\partial n_{i\sigma}}=\frac{1}{2n_{i\sigma}}\int dz\,\xi_{i\sigma}(z)\frac{\delta\mathcal{F}}{\delta\xi_{i\sigma}(z)},\qquad\text{(C3)}$$

which, taking into account Eq. (53), implies Eq. (65). A relation analogous to Eq. (C3) was obtained for the case of the exact-exchange energy functional in Ref. [48]. Here, we see that this property is generally valid for all functionals that admit an explicit representation in terms of $\{p_{i\sigma}(z)\}$.

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