Capacitance and compressibility of heterostructures with strong electronic correlations

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Strong electronic correlations related to a repulsive local interaction suppress the electronic compressibility in a single-band model, and the capacitance of a corresponding metallic film is directly related to its electronic compressibility. Both statements may be altered significantly when two extensions to the system are implemented which we investigate here: (i) we introduce an attractive nearest-neighbor interaction as antagonist to the repulsive on-site repulsion, and (ii) we consider nano-structured multilayers (heterostructures) assembled from two-dimensional layers of these systems. We determine the respective total compressibility κ and capacitance C of the heterostructures within a strong coupling evaluation, which builds on a Kotliar-Ruckenstein slave-boson technique. Whereas the capacitance C(n) for electronic densities n close to half-filling is suppressed—illustrated by a correlation induced dip in C(n)—it may be appreciably enhanced close to a van Hove singularity. We demonstrate that C and κ can differ substantially, as κ is very sensitive to internal electrostatic energies which in turn depend on the specific set-up of the heterostructure. In particular, we show that a capacitor with a polar dielectric has a smaller electronic compressibility and is more stable against phase separation than a standard non-polar capacitor with the same capacitance.

I. INTRODUCTION

The experimental and theoretical investigation of oxide heterostructures has advanced significantly in recent years, not least with the discovery of a metallic state at the interface of two bulk band insulators, LaAlO3 and SrTiO3 [1]. It is the electronic reconstruction in the vicinity of the interface, which generates the metallic state [2, 3] and allows for the coexistence of superconductivity and magnetism [4, 5]. Furthermore, it is suggested to cause the formation of phase-separated states [6–9]; even a state with negative electronic compressibility at the interface has been identified [10, 11].

Electronic devices are generically heterostructures where functionalities are determined by interfaces and surfaces. The conventional semiconductor physics was fascinatingly successful in this respect. Heterostructures with transition metal oxides may come to rival the silicon based systems provided that electronic mobilities can be sustained sufficiently high [12]. One of their advantages is the possibility to generate strongly correlated electronic states [13]. High-temperature superconductivity, the transition to a Mott insulating state as well as exotic magnetism are particular manifestations of strong electronic correlations.

What is the impact of strong correlations on the electronic reconstruction in heterostructures? In this paper we focus on the electronic compressibility controlled by the electronic density at the respective interfaces and surfaces. The density at interfaces can be tuned by gate biases, in fact by at least an order of magnitude in the case of LaAlO3/SrTiO3. Moreover, as the electronic compressibility is intimately related to the capacitance, the question arises how the capacitance is controlled by strong electronic interactions—in dependence on the electronic density.

Long-range Coulomb interactions generate a negative compressibility in a dilute homogeneous electron system. Corresponding analytical calculations in perturbation theory are supported by the results of quantum Monte Carlo evaluations [14–16]. It is mostly the exchange interaction which is responsible for this behavior. Electronic exchange produces a negative compressibility on account of the formation of exchange holes, and capacitances with electrodes comprising such a dilute electron systems may be enhanced well beyond their geometrical capacitance value [10, 17–19]. For the case of very dilute electron systems, Wigner crystallization is suggested to explain observed capacitance enhancements [20].

In this work we want to investigate systems for intermediate filling, that is, we do not consider the dilute homogeneous electron system where exchange becomes so pronounced. We focus on a single-band lattice system and consider only on-site and nearest-neighbor interaction, and evaluate compressibility and capacitance of the electronic system. The capacitance of interacting electrons in a ring geometry pierced by a magnetic flux was introduced by Büttiker [21], and the flux-dependent capacitance was evaluated for a disordered electronic system with electron interactions represented by an extended Hubbard model [22]. Here we will not consider inhomogeneous systems; this is beyond the scope of this work. Inhomogeneities are introduced exclusively through the multilayer setup of the planar heterostructure.

An investigation of the capacitance of multilayers with strongly correlated materials has been conducted recently by Hale and Freericks [23]. They considered a het-
erostructure with non-interacting leads however with a barrier represented by a Falicov-Kimball model, and they identify an enhancement in the capacitance with increasing thickness of the barrier. We evaluate a very different, rather complementary set-up: we build the capacitance from a pair of two-dimensional (2D) metallic electrodes which represent correlated electron systems and the barrier in between is a polar or non-polar dielectric material.

In these systems with sizable band filling, the long-range Coulomb interaction is screened within the metallic plates and the exchange does not play a dominant role in the compressibility. Instead, short-range interactions control the compressibility—apart from the kinetic term. Also further one-particle contributions such as spin-orbit coupling, or transverse potential profiles can have an impact on the compressibility. We will briefly address systems with Rashba spin-orbit coupling in the last part of Sec. IV.

The repulsive on-site Coulomb interaction is screened by the polarization of neighboring atoms, for example in the cuprates by the polarization of oxygen ions. The screening depends on the excitations of the local cluster of atoms. It can be quite different for the nearest-neighbor interaction term and produce an attractive nearest-neighbor interaction, as in certain iron-pnictides. Polaronic effects from strong electron-lattice coupling may enhance an attractive nearest-neighbor interaction. For LaAlO$_3$/SrTiO$_3$ polaronic effects have been observed and investigated within a first-principles-based scheme.

A repulsive on-site interaction $U$ will be effective close to half-filling and it will generically lead to a filling-dependent suppression of the compressibility. As antagonist to the repulsive on-site interaction we introduce an attractive nearest-neighbor interaction $V < 0$. The nearest-neighbor interaction $V$ can generate a charge instability, even for large $U$ if the electronic density is not too close to half-filling. In this paper we investigate how such a scenario applies to a heterostructure and how electronic correlations modify the capacitance of the heterostructure.

Whereas the (inverse) electronic compressibility is related to the second derivative of the free energy wrt to the total number of mobile electrons, a negative electronic compressibility would signify that an increase in the electronic charge entails a decrease in the chemical potential. The electronic compressibility may attain a negative value even in an equilibrium state as long as this is compensated by a sufficiently large positive value from the ionic system. Moreover, it is to be emphasized that electronic subsystems may well display negative compressibility but the total electronic system still has a positive compressibility on account of a large electrostatic contribution, even for nanosized lateral extension of a capacitive electronic system. Here, we will discuss that in systems with polar dielectric materials this latter situation is often realized due to the electrostatic energy of the polar layers. Therefore, a scenario where a planar structure of a heterostructure has negative electronic compressibility does not necessarily imply that the system is stable and develops a phase separation—the negative compressibility rather supports a capacitance enhancement. An instructive example is the heterostructure with Rashba spin-orbit coupling in a metallic interface plane.

Eventually, an important issue arises in this respect: when is the electronic compressibility equivalent to the capacitance of a heterostructure and when does it differ appreciably? We will deal with this question by “deforming” heterostructures continuously and thereby approach different geometric limits such as a standard two-plate capacitance or a set-up known from a gated LaAlO$_3$/SrTiO$_3$ multilayer structure. Thereby we will identify a charge-transfer function which represents the increase in charge of a subsystem with the increase of the total electronic charge. This charge-transfer function parameterizes the discrepancy between differential capacitance and compressibility and allows to extract information about the stability of the electronic system. We will find that heterostructures with polar dielectrics acquire the highest electronic stability, in comparison to different non-polar heterostructures with identical capacitances.

II. TECHNIQUE TO INVESTIGATE CORRELATED ELECTRON SYSTEMS

In our analysis of the compressibility of a heterostructure, which is composed of two 2D metallic subsystems and further insulating layers, we resort to a technique that we already tested on a 2D extended Hubbard model in a previous work, the Kotliar and Ruckenstein slave boson technique.

A. Kotliar-Ruckenstein slave bosons

Extending Barnes’ pioneering work, slave boson representations to the most prominent correlated electron models have been introduced and studied (for a review see Ref. 37). Most relevant to the Hubbard model are
the Kotliar and Ruckenstein (KR) representation 38, as well as its rotationally invariant generalizations 39, 40. All of them possess an internal gauge symmetry group. It may be made use of to simplify the problem and to gauge away the phases of some bosons by promoting all constraint parameters to fields 39. Thereby radial slave boson fields remain 41, that may not Bose condense. In fact, their exact expectation values are generically non-vanishing 42 (see Ref. 43 in the case of Barnes’ representation to the single impurity Anderson Model), and may be approximately obtained through the saddle-point approximation that we are going to use below. The latter has been shown to compare favorably with numerical simulations in a number of cases. For instance, the comparison of the ground state energy to Quantum Monte Carlo (QMC) data on the square lattice for $U = 4t$ yields an agreement in the less than 3% range 44. On the honeycomb lattice, the location of the metal-to-insulator transition was found to be in excellent agreement with QMC data 45 (for more examples see Ref. 46).

Regarding fluctuations at one loop order, RPA forms have been obtained in the low frequency-long wavelength limit, that even agree quantitatively with perturbation theory to lowest order in $U$, in both spin and charge channels 47. In the complementary equal-time limit, quantitative agreement to QMC charge structure factors has been demonstrated for intermediate coupling 48.

It has recently been shown that the spin rotation invariant representation can be extended to incorporate longer range interactions 46. While it allowed to reveal several instabilities, both on the cubic 46 and square 34 lattices, it has not been put to such numerical tests. In fact, it is only established to be variationally controlled in the large dimensionality limit 46. Below, we show that, in the weak coupling regime, it reproduces the Hartree approximation. However it is not limited to the small $U$ regime, and we make use of its versatility to perform calculations for arbitrary parameter values in the thermodynamical limit. All necessary details of the technique and numerical results can be found in the previous studies on the homogeneous 3D and 2D systems (specifically, for the evaluation of the compressibility and Landau parameters, in Ref. 35 and 46).

B. Hartree-Fock evaluation

We will compare results of the filling dependent compressibility for strong correlations in the electronic subsystems to those in the weak coupling case. For this comparison to be reasonable, the weak coupling regime must be realized within the same approach. While non-local interactions reduce to their Hartree approximation in the large D limit, it is not obvious in the 2D case that the slave-boson technique allows to gain the weak coupling results, in particular, when a non-local (attractive) interaction is included. For this purpose we evaluate the free energy of the 2D electronic system within standard Hartree-Fock perturbation theory and compare the results to those of the slave-boson evaluation with weak interaction. The extended 2D Hubbard model, which is expected to present the physics of the metallic electronic systems, is:

$$H = \sum_{\langle i,j \rangle, \sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_{i} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + V \sum_{\langle i,j \rangle} \hat{n}_{i} \hat{n}_{j},$$

(1)

In our evaluations below, we restrict the matrix elements $t_{ij}$ to $t$ for $\langle i,j \rangle$ a pair of nearest-neighbour sites and to $t'$ for next-nearest neighbour pairs on a square lattice; each pair is counted once. We compare the filling-dependent free energies in Hartree-Fock perturbation theory and in KR slave-boson evaluation 35.

We take the Fourier transform of Hamiltonian (1) and make use of the commutation relations for fermionic operators:

$$H = H_t + H_U + H_V$$

(2)

with

$$H_t = - \sum_{\sigma, k} \left\{ 2t [\cos (ak_x) + \cos (ak_y)] \right\}$$

$$+ 4t' \cos (ak_x) \cos (ak_y) \right\} c_{k\sigma}^\dagger c_{-k\sigma}$$

(3)

$$H_U = \frac{U}{L^2} \sum_{k, p, q} c_{k+q\uparrow}^\dagger c_{q\uparrow}^\dagger c_{p\downarrow} c_{p-q\downarrow} c_{-k\uparrow} c_{-k\uparrow}$$

(4)

$$H_V = \frac{V}{L^2} \sum_{\sigma, \sigma'} \sum_{k, p} \left\{ - [\cos a(p_x - k_x) + \cos a(p_y - k_y)] \times \right\}$$

$$\times c_{\sigma p}^\dagger c_{\sigma' p}^\dagger c_{\sigma k}^\dagger c_{\sigma p}^\dagger + 2 c_{\sigma k}^\dagger c_{\sigma k} c_{\sigma p}^\dagger c_{\sigma p}^\dagger \right\}.$$  

(5)
Now the mean field approximation is performed

\[
H_{\text{MFA}}^{U} = U \sum_{\mathbf{k}, \sigma} n_{-\sigma} c_{\mathbf{k} \sigma}^\dagger c_{\mathbf{k} \sigma} - UL^2 n_{\uparrow} n_{\uparrow} \tag{6}
\]

\[
H_{\text{MFA}}^{V} = V \sum_{\mathbf{k}, \sigma} c_{\mathbf{k} \sigma}^\dagger c_{\mathbf{k} \sigma} \left\{ 4(n_{\uparrow} + n_{\downarrow}) - \frac{2}{L^2} \sum_{\mathbf{k}\sigma} \left[ \cos a(p_x - k_x) + \cos a(p_y - k_y) \right] n_{p,\sigma} \right\} + V \frac{1}{L^2} \sum_{\mathbf{k}, \sigma} \sum_{\mathbf{k}' \neq \mathbf{p}} [\cos a(p_x - k_x) + \cos a(p_y - k_y)] \times n_{\mathbf{p}, \sigma} n_{\mathbf{k}, \sigma} - 2VL^2(n_{\uparrow} + n_{\downarrow})^2. \tag{7}
\]

Here \(L\) is the linear extension of the system, \(a\) is the lattice spacing and \(n_{\sigma} = 1/L^2 \sum_{\mathbf{k}} n_{\mathbf{k}, \sigma} = 1/L^2 \sum_{\mathbf{k}} (c_{\mathbf{k} \sigma}^\dagger c_{\mathbf{k} \sigma}) \) is the electron density with spin \(\sigma\). The first and the last term of Eq. (7) constitute the Hartree term while the remaining two terms constitute the Fock term. The sign of the contribution of the Fock term to the energy is opposite to the sign of \(V\). We take the parameter set \(U = t\), \(V = -0.14 t\) or \(-0.45 t\), and \(t' = -0.45 t\).

From Fig. 1, it is obvious that the Hartree approximation is not only consistent with the slave-boson result but is nearly identical in the weak coupling regime. This is a remarkable result as one expects that slave-boson theory is not appropriate for weak coupling and small band filling. Yet, the KR scheme provides suitable projection factors for the kinetic energy and the nearest-neighbor interaction so that its weak coupling limit is not only exact in the zero interaction case \((U = 0)\) but also in first order of \(U\).

The exchange (Fock) correction for the non-local interaction \(V\) is not included in the slave-boson evaluation. The free energies \(F(n_0)\), as functions of the electron density \(n_0 = \sum_\sigma n_\sigma\), differ pronouncedly close to their minima for sizable \(V\)—these minima are shifted from \(n_0 = 1\) on account of the particle-hole symmetry breaking through a finite \(t'\). However, for the compressibility we rather have to compare the second derivative of the free energies with respect to \(n_0\): the largest corrections from the Fock term arise for small densities (below the filling which displays the van Hove singularity related cusp). For long range Coulomb interaction, it is well known that the Fock term dominates the direct Coulomb interaction only in the low density limit. This statement is also confirmed in our evaluation for a nearest-neighbor attractive interaction (see insets of Fig. 1). Consequently, we argue that the weak coupling regime is attained in the slave boson approach even for finite but small interaction parameters—provided that the electronic density is not too low. We test this assertion again in Appendix B where we compare the results for the capacitance of a heterostructure in Hartree-Fock and slave-boson evaluation.

### III. Compressibility and Capacitance of a Polar Heterostructure

We first consider a setup with two metallic 2D electron systems separated by a polar dielectric material. Such a configuration is realized, for instance, in gated LaAlO$_3$/SrTiO$_3$ heterostructures where a polar LaAlO$_3$ film is placed on a SrTiO$_3$ substrate. With a top gate on the LaAlO$_3$ film and with electronic reconstruction at the interface between LaAlO$_3$ and SrTiO$_3$ into a (laterally) confined mobile electronic system, the heterostructure may be presented approximately as a multilayer system of the kind shown in Fig. 2 (left panel, the topmost plane is the gate and the lowest plane represents the interface). The charging of such a capacitance and the electronic density at the interface is tuned through the gate. The total mobile electronic density is \(n_{\text{tot}}\) and \(n_1\) denotes the electron density which is transferred from the top plane to the interface. We investigated this type of heterostructure in a recent publication with focus on Rashba spin-orbit coupling at the interface [20]. Here, we analyze the compressibility and capacitance of such a structure in dependence on electronic interaction parameters. We calculate the differential capacitance, which is the differentiation of the charge of a conducting plate with respect to the voltage, that is, the difference of the electrochemical potential with respect to the reference plate. Generally, the capacitances of the conducting subsystems define a capacitance matrix but for the simple system comprised of two conducting 2D plates the matrix elements merge into a single capacitance parameter \(C\) [49].

The electronic compressibility is to be evaluated for the entire system—assuming that no subsystem decouples. Then the compressibility signals if the electronic system is prone to develop an instability towards a charge separated state. Such a phase separated state might be formed on a nanoscale with distinct properties, such as localization. However, we will not investigate the various realizations of the symmetry broken state. We allow, right from the onset, a charge redistribution between mobile electrons on the surface electrode and the interface which is the electronic reconstruction driven by the polar dielectric in between the two metallic systems.

In our model the total mobile electron charge density in the system, \(-e n_{\text{tot}}\), is compensated by an equal amount of immobile positive background charge. The two electrodes (for example, surface plane and interface) are connected by an external voltage source, so that the difference in electrochemical potentials between the plates is given by \(V_{\text{ext}}\). A possible offset in chemical potentials between the two layers is included in this quantity. The electron density on the interface, \(n_1\), and on the surface, \(n_2 = n_{\text{tot}} - n_1\), is tuned by applying the voltage \(V_{\text{ext}}\). The free energy of the total system,

\[
F(n_{\text{tot}}, n_1) = F_1(n_1) + F_2(n_{\text{tot}} - n_1) + F_{\text{ext}}(n_{\text{tot}}, n_1)
- eV_{\text{ext}}n_1 A, \tag{8}
\]
is the sum of the free energy on the interface \(F_1(n_1)\) and surface \(F_2(n_{\text{tot}} - n_1)\), an electrostatic contribution \(F_{\text{es}}(n_{\text{tot}}, n_1)\), and the term due to the difference in electrochemical potentials between the electrode plates. Here \(A\) is the area of one plate and \(e\) the elementary charge.

The free energy of the correlated electron system consists of the electrostatic energy \(\frac{e^2 A}{\epsilon} \frac{d}{\varepsilon} (n_{\text{tot}} - n_1)^2\) and the second derivative of the free energy with respect to \(n_{\text{tot}}\) and \(n_1\), which is fixed by the values of \(F_1\) and \(F_2\). Here, we assume the configuration to be symmetric, i.e., \(d_a/d_b = d_b/d_a = d/(2\epsilon)\). In LAO/STO a conducting interface develops for a distance \(d\) of four unit cells, while the dielectric constant \(\varepsilon\) is approximately twenty. So we assumed that the effective distance between the plates is \(d/\varepsilon = 4a_B\).

The total density \(n_{\text{tot}}\) is a thermodynamic variable, and the second derivative of the free energy with respect to \(n_{\text{tot}}\) determines the stability of the system. In contrast, the density on the interface, \(n_1\), is an internal variable that is fixed by the values of \(n_{\text{tot}}\) and \(V_{\text{ext}}\) and has to minimize the free energy of Eq. (8):

\[
\frac{\partial F}{\partial n_1} = \frac{\partial F_1}{\partial n_1} + \frac{\partial F_2}{\partial n_1} + \frac{\partial F_{\text{es}}}{\partial n_1} - eV_{\text{ext}} A = 0
\]

The differential capacitance is the differential change of the charge on one plate with applied voltage:

\[
C_{\text{diff}}^{-1} = \frac{\partial V_{\text{ext}}}{eA \partial n_1} - \frac{1}{e^2 A} \left( \frac{\partial^2 F_1}{\partial n_1^2} + \frac{\partial^2 F_2}{\partial n_1^2} + \frac{\partial^2 F_{\text{es}}}{\partial n_1^2} \right)
\]

(11)

which implies that

\[
A/C_{\text{diff}} = \frac{1}{e^2 A} \frac{\partial^2 F}{\partial n_1^2}
\]

(12)

The last term in Eq. (13) constitutes the classical geometric capacitance. If the sum of the partial derivatives of \(F_1\) and \(F_2\) is negative, the differential capacitance is larger than the geometric capacitance. Note that for an isolated system, \(\partial^2 F_1/\partial n_1\) corresponds to an inverse compressibility, and a negative value would signify instability. But here the interface and surface are coupled by the electric field and voltage source, so that the stability of the system is determined by the total derivative of the total free energy with respect to \(n_{\text{tot}}\):

\[
\kappa^{-1} = n_{\text{tot}}^2 \frac{d^2(F/A)}{dn_{\text{tot}}^2}
\]

(13)

For isolated (sub-)systems the quantities compressibility and capacitance are (up to a constant factor) the same. However, for the considered coupled system they can differ quite substantially (see Appendix C):

\[
\frac{1}{n_{\text{tot}}^2 \kappa^{-1}} = \frac{e^2 A}{C_{\text{diff}}} \left( 1 - \frac{\partial n_1}{\partial n_{\text{tot}}} \right) \frac{\partial n_1}{\partial n_{\text{tot}}} + \partial F_{\text{es}}
\]

(14)
where the differential operator \( D \) is defined through \( D \equiv \left( \frac{\partial^2}{\partial n_1 \partial n_{\text{tot}}} \right) \). In the case of the considered polar layout (see Fig. 2(a) and (b)), \( D F_{\text{es}} = 0 \), and the compressibility is negative if the “charge-transfer function” \( \partial n_1 / \partial n_{\text{tot}} \) is smaller than 0 or larger than 1, assuming a positive differential capacitance. Eq. (14) represents a generalization of a relation with \( D F_{\text{es}} = 0 \) that was derived in Ref. [26]. There, the polar layout was considered exclusively; here we will investigate more general layouts in Sec. [IV].

The weak coupling results for the compressibility and capacitance are discussed in Appendix [A]. Here, we focus on the strong coupling analysis (Fig. 3) for the heterostructure of Fig. 2. For comparison, we provide the inverse compressibility of an isolated 2D planar electron system (see Ref. [35]) in the topmost line of panels. In this section we assume that the electron systems and their respective parameters are the same for both plates.

First, for \( t' = 0 \), the density dependence of the compressibility of the heterostructure (panel (d)) is similar to that of the single 2D plane (panel (a)): strong correlations \( (U = 9t) \) produce a significant decrease in compressibility close to half-filling. We characterize the peak structure in the inverse compressibility as “correlation peak”. Here we took \( n_{\text{tot}} = 2.0 \) for the heterostructure. Consequently, \( n_1 = 1.0 \) signifies that the single bands of the two plates (interface and surface) are each half-filled, and the correlation peak has its maximum at this value of \( n_1 \). Other values of \( n_1 \) are obtained by applying a corresponding voltage. Whereas the compressibility of the isolated 2D system becomes negative away from half-filling for sufficiently attractive nearest neighbor interaction, this is not the case for the heterostructure. The polar dielectric material in between the plates provides...
an additional electrostatic energy and thereby stabilizes the system. The density dependence of the capacitance of the heterostructure (panel (g)) reflects the density dependence of the compressibility. The slight decrease of the capacitance for small and large \( n_1 \) (either most of the charge at the interface or at the surface) is caused by the fact that the attractive interaction is less effective at the band edges.

A finite value of \( t' \) moves the van Hove singularity (vHs) from the center of the band. We take a filling of \( n_{\text{tot}} = 1.75 \) for \( t' = -0.15t \) so that for equal filling in the two plates, the corresponding Fermi energies are placed at the vHs. The proximity to the vHs for both plates for \( n_1 \approx n_{\text{tot}}/2 \) makes the structure in the compressibility more pronounced (panel (e)). At \( n_{\text{tot}}/2 \) the inverse compressibility \( \kappa^{-1} \) displays a cusp on account of the vHs. For \( n_1 \) larger than \( n_{\text{tot}}/2 \), the interface approaches half-filling and the strong correlations lead to an increase in \( \kappa^{-1} \), producing the correlation peak. Similarly, for \( n_1 \) smaller than \( n_{\text{tot}}/2 \), the surface electron system approaches half-filling and \( \kappa^{-1} \) increases. The capacitance has (correlation induced) minima at fillings where either plate is in an electronic state close to half-filling (panel (h)). In the center we find a peak in the capacitance that originates from the attractive interaction \( V \) augmented by the vHs which, for this filling, is positioned at the Fermi energies of both plates. This leaves a broad filling range where the capacitance is considerably enhanced with respect to its \( U = 0 = V \) value. This happens when the filling in both plates is not close to half-filling.

Eventually, for \( t' = -0.45t \) and \( n_{\text{tot}} = 0.93 \) the correlation effects from the on-site repulsion \( U \) are of minor importance because neither plate is close to half-filling. The nearest neighbor interaction \( V \) dominates the behavior of the compressibility and capacitance (see panels (f) and (i)). A strongly attractive \( V \) increases the compressibility and capacitance. The strongest enhancement of the capacitance is at the van Hove filling (both plates have fillings so that the Fermi energies are placed at the vHs) at \( n_1 = n_{\text{tot}}/2 \). At the largest and smallest value of \( n_1 \) one of the plates carries an electron system with filling close to \( n_{\text{tot}} = 0.93 \), and the on-site repulsion reduces the capacitance enhancement.

For weak coupling—we present the results for \( U = t \) in Appendix A—the correlation peak in the inverse compressibility at half-filling is absent. Instead one observes a dip in the inverse compressibility at the position of the van Hove singularity.

IV. COMPRESSIBILITY AND STABILITY IN DISTINCT LAYOUTS

Even though the 2D electronic system may display a negative compressibility and consequently a tendency towards phase separation (see, e.g., Fig. 3(a)–(e)), the entire heterostructure, which is composed of one or two of these 2D layers, is not necessarily instable towards charge separation (see, e.g., Fig. 3(d)–(f)). It is the electrostatics that stabilizes the homogeneous state for the considered setup of Fig. 2. The question arises if other layouts—with the same mobile electronic systems, \( F_1 \) and \( F_2 \), and the same distance \( d \) between the electrodes—are more prone to phase separation. It is evident that a different distribution of the positive background charge alters the electrostatic terms and thereby the total compressibility.

In order to resolve this issue we introduce a generalized effective layout of positive background charge, characterized by the set of three parameters \( \{ \alpha, \beta, m_1 \} \) (c.f. Fig. 4). The distance \( d \) between the two electrodes, which comprise the electron systems, is kept fixed. Two 2D layers with positive charge densities \( +m_1n_{\text{tot}}e \) and \( +(1 - m_1)n_{\text{tot}}e \) (where \( 0 \leq m_1 \leq 1 \)) are located at a distance \( ad \) and \( \beta d \) (where \( 0 \leq \alpha \leq \beta \leq 1 \)) from the bottom electrode. This effective model covers a variety of physical configurations, inter alia:

The specific choice \( \alpha = 0 \) and \( \beta = 1 \) refers to the standard two-plate capacitor, where the positive charge populates the same layers as the electrons. The parameter \( m_1 \) allows for tuning the allocation of the positive charge between the plates.

For \( \alpha = \beta \), all positive background charge of the effective model is placed in one plane and the setup is that of the polar heterostructures (c.f. Fig. 2(a) and (b)). The distance between the positively charged (effective) plane and the top electrode can be tuned either by using polar materials with different distances between the atomic layers \( d_{\text{a}} \) and \( d_{\text{t}} \), or by adding an insulating film and a terminating electrode on top of the polar heterostructure. Note that \( \alpha = \beta = 1 \) (or \( \alpha = \beta = 0 \)) corresponds to the case where electrons from an electrode with neutralizing positive background ions are transferred into empty bands of a charge neutral electrode (by minimizing the free energy in the presence of a bias).
The electrostatic energy of this generalized effective layout is

$$F_{es}^\alpha = 2D\left[\alpha n_1^2 + (\beta - \alpha) (m_1 n_{tot} - n_1)^2\right] + (1 - \beta)(n_{tot} - n_1)^2,$$

where

$$D = \pi e^2 A d/\varepsilon = \frac{1}{4} e^2 A/C_{geo}$$

is closely related to the inverse geometric capacitance $C_{geo}$. With the free energy of Eq. (8), we rewrite the second derivative with respect to the total electron density (see Appendix C):

$$\frac{d^2 F}{d n_{tot}^2} = \frac{1}{F_{1} + F_{2} + 4D} \left[ (F_{1}'' + F_{2}'' + 4D) \left( F_{2}'' + \frac{\partial^2 F_{es}^\alpha}{\partial n_{tot}^2}\right) \right] - \left( \frac{\partial F_{es}^\alpha}{\partial n_{tot}} - F_{2}''\right)^2$$

where $F_{1,2}'' = \partial^2 F_{1,2}/\partial n_1^2$. The inverse compressibility can be easily extracted from this quantity via Eq. (13). We emphasize that the capacitance is not affected by the distribution of the positive charge and is given (up to a factor of $e^2 A^2$) by the fraction in front of the brackets.

We now analyze three special configurations of the positive background charge:

- the symmetric polar heterostructure (SPH) with $\alpha = \beta = 1/2$, as shown in Fig. 5(a1),
- the symmetric standard capacitor (SSC) with $\alpha = 0$, $\beta = 1$, $m_1 = 1/2$, displayed in Fig. 5(a2),
- the asymmetric standard capacitor (ASC) with $\alpha = \beta = 1$, see Fig. 5(a3)); the positive charge is on one electrode.

Corresponding setups of capacitors are displayed in Fig. 5, jointly with the respective results for the inverse compressibility and the charge transfer function $\partial n_1/\partial n_{tot}$.

We can rewrite Eq. (17),

$$A_{i}^{-1} = n_{tot}^2 F_{1''} F_{2''} + DA_i$$

with $i = \{\text{SPH, SSC, ASC}\}$ and

$$A_{\text{SPH}} = 2 (F_{1''} + F_{2''} + 2D)$$
$$A_{\text{SSC}} = F_{1''} + F_{2''}$$
$$A_{\text{ASC}} = 4 F_{2''}.$$
For nanoelectronics applications an enhancement of the capacitance beyond the geometrical value is often desirable (see the discussion in Refs. [19] and [13]). A stable solution to $C > C_{\text{geo}}$ requires that $F''_1 + F''_2 < 0$, with a positive compressibility of the total system. However, the SSC system becomes unstable for $F''_1 + F''_2 < 0$ (see Appendix C, Eq. (C13)). Consequently, an enhancement of the capacitance above the geometrical value is not possible for the SSC layout (provided the electronic compressibility $\kappa_{\text{SSC}}$ stays positive). For $F''_1 + F''_2 > 0$ the compressibility $\kappa_{\text{SSC}}$ is always larger than the compressibility in the SPH system. This implies that the SPH is better suited to realize an enhanced capacitance in a thermodynamically stable state. In fact, the electrostatic energy in the SPH stabilizes the thermodynamic state.

The stability of the ASC layout depends on the sign of $F''_2$, i.e., it is best for a capacitance enhancement to pair the positive background layer with the electronic system, that is represented by a free energy with a positive second derivative. The electronic system of the second electrode should have a free energy with a negative second derivative in order to enhance the capacitance.

The strikingly different compressibilities of the three layouts with the same capacitance are shown in Fig. 5(a1)–(a3). For this plot we took the same parameters for the two electron systems which were introduced in Sec. II A. The total density is adjusted in such a way, that both electron systems on the two electrodes are at the van Hove singularity for $n_1 = n_2$. The density $n_1$ on the lower electrode is adjusted by an external voltage between the plates. In the thermodynamically unstable regime with negative compressibility, $\kappa^{-1}$ is represented by dashed lines. The SSW result is shifted to lower values of $\kappa^{-1}$ with respect to the SPH result; this observation is explained below.

The relation between compressibility and capacitance is controlled by the charge-transfer function $\partial n_1/\partial n_{\text{tot}}$ and by $DF_{\text{es}}$, as introduced in Eq. (14). In Appendix C we show that $\partial n_1/\partial n_{\text{tot}}$ is in fact equal for the SPH and the SSW layout, so that the compressibilities of these two systems differ only by $DF_{\text{es}}$ (see Eq. (14)). For the three model capacitors the $DF_{\text{es}}$ values are:

$$DF_{\text{es}}^{\text{SPH}} = 0, \quad DF_{\text{es}}^{\text{SSC}} = -D, \quad DF_{\text{es}}^{\text{ASC}} = 0 \quad (20)$$

This shift by $D$ of the inverse compressibility in the SSW (with respect to $\kappa^{-1}$ in the SPH) is visible in Fig. 5(c)–compare Fig. 5(a1) and (a2). The ASC layout displays a negative inverse compressibility in the $V = -0.9t$ case for densities larger than $n_{\text{tot}}/2$ (see Fig. 5(a3)). This results from $F''_2$ being negative for this filling (c.f. Fig. 3(c) and Eq. (18), (19)).

In systems with $DF_{\text{es}} = 0$, the compressibility is positive for $0 < \partial n_1/\partial n_{\text{tot}} < 1$, as is apparent from the comparison of Fig. 5(b1) and Fig. 5(b3) with the corresponding upper panels (a1) and (a3), respectively. For the SSC $DF_{\text{es}} = -\pi e^2 A \varepsilon/\varepsilon$ and one verifies for $n_1 = \frac{1}{2} n_{\text{tot}} = n_2$ that $\partial n_1/\partial n_{\text{tot}} = \frac{1}{2}$ (compare Fig. 5(b2)) and that, with Eq. (14), the well-known relation $\kappa^{-1}/n_1^2 + 4\pi e^2 d/\varepsilon = e^2 A/C_{\text{diff}}$ holds (see, for example, Ref. 19).

In experimental setups the top electrode is often a metallic electrode for which correlation effects can be neglected. Layouts with unequal electrodes are investigated in Appendix B. There the top electrode is chosen to be an uncorrelated 2D metallic system and the bottom electrode a correlated 2D metal.

The dependence of the compressibility on the electrostatic layout is also relevant for heterostructures with electronic systems other than those represented by the extended Hubbard model: which configurations allow for a negative compressibility with phase separation? In Refs. [25] and [27] an electron system at the interface of LaAlO$_3$ and SrTiO$_3$ was suggested to display electronic phase separation on account of a finite Rashba spin-orbit coupling. In Ref. [26] this system was supplemented by electronic surface states, resembling the SPH, and the possibility of a state with negative compressibility was analyzed. For values of the spin-orbit coupling consistent with experiments, the compressibility of the system was found to be positive [24].

Here we investigate two realizations of the electrostatic layout which are variants of the general scheme in Fig. 1 in the first, the total positive background charge is split in half and both charge fractions are moved from the center plane by the same distance towards the electrodes (Fig. 6(a) and (c)). The second variation keeps the background charge in plane (Fig. 6(b) and (d)). The spin-orbit coupling and all electronic system parameters are
fixed.

For the first alternative the layout of the positive background charge is symmetric. Then the charge-transfer function is identical for all values of the distance parameter $\alpha$ (see Appendix C) and the compressibilities differ only by the (density independent) electrostatic term $\Delta F_{\text{es}}(\alpha)$ (c.f. Fig. 6(a)). For $\alpha = 0$ the SSC is recovered and we find that the inverse compressibility of the total system can become negative. In the case of the layout shown in Fig. 6(d), a reduction of $\alpha$ merges the layout finally with the ASC layout (for $\alpha = 0$) which may also acquire a negative electronic compressibility. We argue that the electrostatic energy in the polar heterostructure prevents a phase separation—the SPH is represented by the purple curves with $\alpha = 0.5$ in Fig. 6. Other configurations, however, have a stronger tendency towards phase separation—such as the SSC (Fig. 6(a) and (c)) with $\alpha = 0$ or the ASC (Fig. 6(b) and (d)) with $\alpha = 0$.

V. CONCLUSIONS

The electronic compressibility characterizes the electronic state of capacitive heterostructures through its dependence on the gate bias; this voltage controls the electronic density in the system. For an isolated 2D layer, the qualitative density dependence of the compressibility can be readily understood (compare Figs. 3(a)–(c) and Figs. 10(a)–(c) for the strongly and weakly interacting 2D systems, respectively): for weak coupling the compressibility represents the density of states and one observes a pronounced dip in $\kappa_0^{-1}$ for the density where the Fermi energy is tied to the van Hove singularity. For strong coupling, electronic correlations are responsible for the peak structure in $\kappa_0^{-1}$ in a sizable filling range around $n_0 = 1$ (“correlation peak”). For a negative nearest-neighbor interaction $V$, introduced as antagonist to the repulsive on-site interaction $U$, one observes a shift of $\kappa_0^{-1}(n_0)$ towards smaller or even negative values.

These findings are to be re-examined for a heterostructure with at least two coupled metallic 2D systems (see Figs. 3(d)–(f) for the strongly interacting 2D system with a polar film between the metallic plates, as in Fig. 3). One result is of particular importance: The total electronic compressibility $\kappa$ of the heterostructure stays positive even if the compressibilities of the metallic subsystems are negative, that is the heterostructure is more stable with respect to the formation of a phase-separated electronic state. It is the interlayer electrostatic term which, apart from the intralayer electronic interaction, influences the electronic reconstruction in the heterostructure and keeps the total compressibility positive.

We investigated different layouts and identified the symmetric polar heterostructure (SPH setup in Fig. 5) as the configuration in a large class of systems (continuously characterized by three parameters) which is most robust (smallest positive $\kappa$) with respect to other configurations with the same capacitance. A system that is more susceptible to phase separation is the standard capacitor with two electrodes and no polar dielectric, provided that an electronic coupling such as an attractive nearest-neighbor interaction or a Rashba spin-orbit coupling (see Figs. 5 and 6) drives the metallic plates into a negative compressibility state. The standard capacitor always displays a negative total compressibility if the two electrodes are in a negative compressibility state, that is, it is thermodynamically unstable if no further terms (e.g. from the lattice) keep the total compressibility positive.

The capacitance is distinct from the compressibility of the heterostructure. It is the response of the charge density of a plate to a voltage difference applied to the two electrodes—whereas the compressibility is the response of the total charge density to the chemical potential of the electronic system. In our model system, the capacitance can achieve a large enhancement with respect to its geometric value. In particular, this is realized for sizable $t'$ so that the van Hove singularity, which may drive the enhancement for attractive $V$, is moved from the band center. Otherwise, the repulsive on-site interaction $U$ is very effective to suppress the capacitance when the electronic systems are close to half-filling. In order to identify heterostructures with large capacitance enhancement one should avoid electronic systems with strong on-site interaction on the electrodes. However it is also necessary to work with a sufficiently stable system—and we find that the heterostructures with polar dielectrics abide by this characterization.

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Appendix A: Weak coupling results

We analyze the same setup as described in Fig. 3 here with $U = t$ instead of $U = 9t$, that is, the electron systems are characterized by weak correlations. Since for smaller on-site repulsion it is also reasonable that the inter-site attraction is smaller, we used the same ratio of $U/V$ as in Fig. 3. As this means smaller values of $|V|$, the inverse compressibilities of the single isolated Hubbard systems shown in Fig. 4 (a)–(c) are larger than in Fig. 3. Moreover, the inverse compressibilities do not display “correlation peaks”, in marked contrast to the results for $U = 9t$.

The absence of the correlation peak is reflected in the inverse compressibilities of the total system (Fig. 3(d)–(f)), the structure of which is controlled by the single layer compressibilities. Again, the chosen densities $\rho_{\text{tot}}$
FIG. 7. (Color online) (a–c) Inverse compressibilities of an isolated 2D weakly interacting electron system with parameters $t = 0.5\, eV, U = t$ and $a = 10\, a_B$ for three different values $t'$ (panels) and five different $V$ (see color code of (f) or (i)) as function of the electron density. (d–f) Inverse total compressibilities $\kappa^{-1}$ of a system composed of two identical interacting electron systems, as presented in (a–c), at a mutual effective distance $d/\varepsilon = 4\, a_B$. The inverse compressibilities are displayed as functions of the electron density $n_1$ on the lower electrode. The electrostatic layout is illustrated in Fig. 2. The total charge $n_{\text{tot}}$ (orange) is chosen in such a way that at $n_1 = n_{\text{tot}}/2$ both systems have their Fermi energies at their respective van Hove singularities. (g–i) Differential capacitances of the above system normalized to the capacitance of two 2D electron systems with $U = 0$ and $V = 0$.

for different values of $t'$ ensure that at $n_1 = n_{\text{tot}}/2$ the Fermi energy of both systems is at the van Hove singularity. This is accomplished by $n_{\text{tot}} = 2.0$ for $t' = 0$, $n_{\text{tot}} = 1.75$ for $t' = -0.15\, t$ and $n_{\text{tot}} = 0.93$ for $t' = -0.45\, t$.

With no correlation peak in the inverse compressibility, the differential capacitance (Fig. 3(g)–(i)) does not display a dip at $n_1 = n_{\text{tot}}$. We normalized the capacitance to that of a $U = 0$ and $V = 0$ system. It is evident that for weak inter-site attraction (black and red curves) the finite on-site repulsion $U = t$ induces a differential capacitance smaller than $C_{U=0, V=0}$. Since $|V|$ is taken to be smaller, the isolated single layer inverse compressibilities Fig. 7(a)–(c) are larger (and mostly positive) and, hence, the achieved increase of the capacitance is smaller than in Fig. 3(g)–(i).

Appendix B: Heterostructure with unequal electrodes

In experimental setups the surface electrode is typically distinct and comprises a weakly correlated electron system. Here, we consider a heterostructure with an uncorrelated electron system at the surface electrode (labeled “2”) whereas the interface electrode (labeled “1”) comprises a strongly correlated electron system.

For plate 2 (surface), a two-dimensional metal with free electrons of effective mass ratio $m_2/m_e$ yields

$$F_2(n_{\text{tot}} - n_1) = \frac{\pi a_B e^2}{m_2/m_e} (n_{\text{tot}} - n_1)^2$$

for the free energy, where $a_B$ is the bare Bohr radius. We take $m_2/m_e$ to be 5 throughout this section. A (more realistic) effective Bohr radius can be integrated into a
modified effective mass $m_2$. For plate 1 (interface) we take strongly correlated electrons with $U = 9t$ and $t' = -0.15t$.

The second derivative with respect to $n_1$ does not depend on the density,

$$F'_2 = \frac{2\pi a_B e^2}{m_2/m_e},$$

so that the only density-dependent quantity in the compressibility and capacitance enters via $F''_2$ (c.f. Eq. (17) and Eq. (12)). This is reflected in Figs. 8 and 9 where we analyzed the system described in Fig. 3 and replaced the surface by a two-dimensional metal with free electrons. The total electron density is fixed to $n_{\text{tot}} = 2.0$.

The more complex structure of a double peak and double dip around $n_1 = n_{\text{tot}}/2$ present in Fig. 3(e) and (h), respectively, is not observed for the asymmetric system of this section; the structure of the inverse compressibility in Fig. 8 is more reminiscent to that of Fig. 3(b).

The capacitance in Fig. 9 is normalized to $C_{U=0,V=0}$ and shows clearly the “correlation dip” when the density of the interface electrons is close to half filling.

The capacitance for an extended Hubbard system with $U = t$ and $t' = -0.45t$ coupled to a two-dimensional free electron gas is shown in Fig. 10. As before, since the contribution of $F''_2$ to the capacitance is constant, the density dependence is dominated by $F''_1$ (c.f. Fig. 7(c)). The normalization to the geometric capacitance reveals
a slight increase of $C$ for $V = -0.4 t$ above $C_{\text{geo}} = 4\pi d / \varepsilon$ when the system is at one of the van Hove singularities for fillings $n_1 = 0$ or $n_1 \approx 0.47$. Elsewhere the compressibility of the single layer $\kappa_0$ is positive and hence the capacitance is reduced below the geometric value. The compressibility of the total system (Fig. 11) is approximately that of Fig. 7 (c), up to a constant.

Fig. 10 and Fig. 11 include also a comparison between the slave-boson, Hartree and Hartree-Fock approximations. The slave-boson technique agrees excellently with the Hartree calculations. The difference between Hartree and Hartree-Fock is due to the second derivative of the Fock term. This contribution is most notable for empty and full bands, and at the vHs at intermediate filling.

**Appendix C: Compressibility and Capacitance**

For the derivation of a general relation between total compressibility and differential capacitance we assume two conditions to be fulfilled: Firstly, the free energy of the total system can be written in the form of Eq. (8):

$$F(n_1, n_2) = F_1(n_1) + F_2(n_1 - n_2) + F_{\text{es}}(n_1, n_2)$$

where we introduced the abbreviation $n_1 \equiv n_{\text{tot}}$ for this section. Secondly, the internal variable $n_1$ minimizes the free energy and is not a boundary value, i.e., $n_1 \neq 0$ and $n_1 \neq n_2$. This stays valid for a differential change of the total charge $n_1$:

$$\frac{d}{dn_1} \frac{\partial F}{\partial n_1} = 0$$

$$\iff \frac{\partial^2 F}{\partial n_1 \partial n_1} = -\frac{\partial^2 F}{\partial n_1^2} \frac{\partial n_1}{\partial n_1}$$

The inverse compressibility of the system is proportional to the second derivative with respect to the total charge density:

$$\kappa^{-1} A / n_1^2 = \frac{\partial^2 F}{\partial n_1^2}$$

$$= -\frac{\partial^2 F}{\partial n_1^2} + 2 \frac{\partial^2 F}{\partial n_1 \partial n_1} \frac{\partial n_1}{\partial n_1} + \frac{\partial^2 F}{\partial n_1^2} \left( \frac{\partial n_1}{\partial n_1} \right)^2$$

$$= \frac{\partial^2 F}{\partial n_1^2} - \frac{\partial^2 F}{\partial n_1^2} \left( \frac{\partial n_1}{\partial n_1} \right)^2$$

We make use of the form of the free energy,

$$\frac{\partial^2 F}{\partial n_1 \partial n_1} = -F''_1 + \frac{\partial^2 F_{\text{es}}}{\partial n_1 \partial n_1} - \frac{\partial^2 F}{\partial n_1^2} \frac{\partial n_1}{\partial n_1}$$

$$\frac{\partial^2 F}{\partial n_1^2} = F''_1 + \frac{\partial^2 F_{\text{es}}}{\partial n_1^2}$$

Last line is inserted in the relation for the compressibility (C5):

$$A \frac{\kappa^{-1}}{n_1^2} = \frac{\partial^2 F}{\partial n_1^2} \frac{\partial n_1}{\partial n_1} \left( 1 - \frac{\partial n_1}{\partial n_1} \right) + \left( \frac{\partial^2 F}{\partial n_1 \partial n_1} + \frac{\partial^2 F}{\partial n_1^2} \right) F_{\text{es}}$$

$$= e^2 A^2 \frac{\partial n_1}{\partial n_1} \left( 1 - \frac{\partial n_1}{\partial n_1} \right) + D F_{\text{es}},$$

where we introduced the differential operator $D = (\partial^2_{n_1} + \partial^2_{n_1} \partial n_1)$. This relation corresponds to Eq. (14) in Sec. III.

A generalized distribution of the positive background charge is depicted in Fig. 4. We assume that the distance between the electrodes is given by $d$. The fraction $m_1$ of the total positive charge resides at distance $ad$ from the lower electrode, while the rest of the positive charge is at a distance $\beta d$ from the lower electrode. For $\alpha = \beta = 0.5$ we recover the polar heterostructure of Fig. 2 (with $d^+ = d_1^+$ and $\varepsilon_a = \varepsilon_a$), and for $\alpha = 0$, $\beta = 1$ the standard configuration where the layers of positive charge are identical with the layers of negative charge. The electrostatic energy of the general layout is

$$F_{\text{es}} = 2D [m_1 (n_1^2 + (\beta - \alpha) (n_1 n_2 - n_2)^2 + (1 - \beta) (n_1 - n_2)^2]$$

with $D = \pi e^2 A d / \varepsilon$. This generalized model allows to analyze the effect of a variety of electrostatic configurations on the compressibility in one framework. The respective contribution to the differential capacitance,

$$\frac{\partial^2 F_{\text{es}}}{\partial n_1^2} = e^2 A \frac{C_{\text{geo}}}{C_{\text{diff}}} = 4D,$$

is independent of the layout of the positive charge and determined by the distance between the electrodes. The differential operator,

$$DF_{\text{es}} = 4D (\beta - \alpha) m_1 (m_1 - 1),$$

is zero if all positive charge resides in one plane ($\alpha = \beta$) and otherwise negative. These two results can be combined with Eq. (C8),

$$\kappa^{-1} A \frac{m_1}{n_1^2} = \frac{e^2 A^2 \partial n_1}{C_{\text{diff}} \partial n_1} \left( 1 - \frac{\partial n_1}{\partial n_1} \right)$$

$$- \frac{e^2 A^2}{C_{\text{geo}}} (\beta - \alpha) m_1 (1 - m_1).$$

Eq. (C12) yields the condition for a system to be stable ($\kappa > 0$):

$$\frac{C_{\text{diff}}}{C_{\text{geo}}} (\beta - \alpha) m_1 (1 - m_1) < \frac{\partial n_1}{\partial n_1} \left( 1 - \frac{\partial n_1}{\partial n_1} \right).$$

This relation limits the enhancement of the differential capacitance over its geometric value, since the right hand side of the last inequality is $\leq 1/4$.

We now select three special cases for the distribution of positive background charge:
• \( \beta^\alpha = \alpha^\alpha \)  
  The asymmetric case \( \alpha \), where the whole positive charge is concentrated in one layer,

• \( \beta^\alpha = 1 - \alpha^\alpha, \ m_1^\alpha = 1/2 \)  
  the symmetric distribution \( s \) of the positive charge and

• \( \alpha^t = 0, \ \beta^t = 1, \ 0 \leq m_1^t \leq 1 \)  
  the standard configuration \( t \) with the planes of positive charge coinciding with the electrodes.

For \( \alpha^a = \alpha^s = 1/2 \) the first two cases are equal and recover the symmetric polar heterostructure (SPH) introduced in section IV. The symmetric standard capacitor (SSC) is obtained for \( \alpha^s = 0 \) and \( m_1^s = 1/2 \) for the last two cases and the asymmetric standard capacitor (ASC) for \( \alpha^a = 1 \) and \( m_1^a = 0 \).

For the asymmetric layout \( \alpha \) the differential operator \( [C11] \) vanishes so that there is no limitation to the capacitance enhancement. In the standard configuration, on the other hand, a necessary condition for its stability is given by

\[
\frac{C_{\text{diff}}}{C_{\text{geo}}} \leq \frac{1}{4m_1^t (1 - m_1^t)}. \quad (C14)
\]

This implies that for \( m_1^t = 1/2 \), which is the symmetrical standard configuration, no enhancement above the geometrical capacitance is possible. The more asymmetric the positive charge is distributed, the larger the allowed capacitance enhancement is.

The differential capacitance of the symmetric configuration is limited by

\[
\frac{C_{\text{diff}}^s}{C_{\text{geo}}} \leq \frac{1}{1 - 2\alpha^s}. \quad (C15)
\]

In this layout the condition used to determine \( n_1 \),

\[
\partial_{n_1} F_s^a = \partial_{n_1} F_1 + \partial_{n_1} F_2 + 4D \left( n_1 - \frac{n_t}{2} \right) - eV_{\text{ext}}A \, \frac{d}{2} = 0,
\]

is independent of \( \alpha^s \). Hence the solution \( n_1^t \) and the charge transfer function \( \partial_{n_1} n_1^t \) are equal for all \( \alpha^s \). Note that, due to

\[
D F_{\text{es}}^a = -D(1 - 2\alpha^s) \quad (C16)
\]

and Eq. (C8), the inverse compressibilities for different symmetric layouts differ by a constant proportional to \( 1 - 2\alpha^s \). The nearer the positive charge is to the electrodes, the more compressible the total system becomes.

Finally, we derive Eq. (18) which specifies the compressibility for the SPH, SSC and ASC layouts. Firstly, we can replace \( \partial_{n_1}/\partial n_1 \) in Eq. (C5) by Eq. (C3),

\[
\frac{d^2 F}{dn_1^2} = \frac{\partial^2 F}{\partial n_1^2} - \left( \frac{\partial^2 F}{\partial n_1^2} \right)^2 \frac{\partial^2 F}{\partial n_1^2}
\]

\[
= \left( \frac{\partial^2 F}{\partial n_1^2} \right)^{-1} \frac{\partial^2 F}{\partial n_1^2} \left[ \frac{\partial^2 F}{\partial n_1^2} - \frac{\partial^2 F}{\partial n_1^2} \right] \quad (C17)
\]

and then make use of the special form of the free energy, Eq. (8):

\[
\frac{d^2 F}{dn_1^2} = \frac{1}{F_1'' + F_2'' + 4D} \left[ (F_1'' + F_2'' + 4D) \left( F_2'' + \frac{\partial^2 F_{\text{es}}}{\partial n_1^2} \right) \right]
\]

\[
- \left[ -F_2'' + \frac{\partial^2 F_{\text{es}}}{\partial n_1^2} \right]^2, \quad (C18)
\]

where \( F_1''_t = \frac{\partial^2 F_{\text{es}}}{\partial n_1^2} \). The electrostatic energies for the different layouts are

\[
F_{\text{sp}}^{\text{SPH}} = D[n_1^2 + (n_t - n_1)^2]
\]

\[
F_{\text{sp}}^{\text{SSC}} = 2D \left( \frac{n_t}{2} - n_1 \right)^2 \quad (C19)
\]

\[
F_{\text{sp}}^{\text{ASC}} = 2Dn_1^2
\]

and the corresponding derivatives yield

\[
\frac{\partial^2 F_{\text{es}}^{\text{SPH}}}{\partial n_1^2} = 2D, \quad \frac{\partial^2 F_{\text{es}}^{\text{SPH}}}{\partial n_1^2} = -2D
\]

\[
\frac{\partial^2 F_{\text{es}}^{\text{SSC}}}{\partial n_1^2} = D, \quad \frac{\partial^2 F_{\text{es}}^{\text{SSC}}}{\partial n_1^2} = -2D \quad (C20)
\]

\[
\frac{\partial^2 F_{\text{es}}^{\text{ASC}}}{\partial n_1^2} = 0, \quad \frac{\partial^2 F_{\text{es}}^{\text{ASC}}}{\partial n_1^2} = 0.
\]

Inserting these expressions into Eq. (C18) yields Eq. (18).

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