Nitride Multilayers as a Platform for Parallel Two-Dimensional Electron-Hole Gases: MgO/ScN(111)

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At interfaces between insulating oxides $LaAlO_3$ and $SrTiO_3$, a two dimensional electron gas (2DEG) has been observed and well studied, while the predicted hole gas (2DHG) has not been realized due to the strong tendency of holes in oxygen $2p$ orbitals to localize. Here we propose, via ab initio calculations, an unexplored class of materials for the realization of parallel two dimensional (2D), two carrier (electron+hole) gases: nitride-oxide heterostructures, with (111)-oriented ScN and MgO as the specific example. Beyond a critical thickness of five ScN layers, this interface hosts spatially separated conducting Sc-$3d$ electrons and N-$2p$ holes, each confined to ~two atomic layers – the transition metal nitride provides both gases. A guiding concept is that the N^{3-} anion should promote robust two carrier 2D hole conduction compared to that of O^{2-}; metal mononitrides are mostly metallic and even superconducting while most metal monoxides are insulating. Our results, including calculation of Hall coefficient and Seebeck coefficient for each conducting layer separately, provide guidance for new exploration, both experimental and theoretical, on nitride-based conducting gases that should promote study of long sought exotic states viz. new excitonic phases and distinct, nanoscale parallel superconducting nanolayers.

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The unexpected magnetic and electronic phases appearing at the interface between two perovskite oxides have been studied extensively during the last decade [1–5].The discovery of a two dimensional electron gas (2DEG) at the interface between $LaAlO_3$/$SrTiO_3$(LAO/STO) by Ohtomo and Hwang[1] stimulated excitement for the design of similar heterostructures and their possible use in electronic devices [6, 7]. The further realization that electronic reconstruction could provide a parallel, nanoscale 2DEG and 2DHG (2D hole gas) provided a more exotic vista [8, 9], including two band, two carrier magnetic or superconducting systems, as well as long sought excitonic condensates [10, 11]. Substantial theorizing has been waiting favorable experimental platforms for progress. Unfortunately, at oxide interfaces the hole gas has never materialized. Due to the proclivity of holes to occupy O $2p$ orbitals, it is always observed to be non-conducting, eliminating the many possibilities provided by two, or many, 2DEGs and 2DHGs separated by only 2-3 nm [12, 13].

The difficulty in obtaining conducting p-type interfaces in oxides suggests using related but less electronegative ions, bringing to mind nitrides. Whereas $3d$ transition metal monoxides are mostly strongly correlated (Mott) insulators [14], the existing mononitrides are usually conducting and even superconducting [15, 16]. This tendency toward conductivity while forming similar or even identical structures brings a new dimension to the design of heterostructures of insulators having active interfaces.

(001) layers of transition metal mononitrides (or monoxides) with rocksalt structure are charge neutral, thus being ineffective in providing the 2D gases of interest here. This layer neutrality, and the challenges provided by localized O $2p$ states, invites innovative material design, especially because layer-by-layer growth provides the experimental capability of concerted theory plus experiment design of novel materials with tailored properties at the nanoscale.

The simplest polar structure in mononitrides is provided by (111) orientation, where oppositely charged metal and anion-atom layers alternately stack along the [111] direction (see Fig. 1). In spite of the widely discussed instability of ideal polar surfaces in a semi-infinite geometry, MgO films with atomically flat (111) surface regions have been reported using layer-by-layer growth on a variety of substrates [17–25]. These atomically flat surfaces are promising to explore the possibility of polar interface engineering using simple metal monoxides and mononitrides with rocksalt structure.

Here, we explore the use of the narrow gap semiconductor ScN,[26–28] sandwiched between highly insulating MgO [29, 30], as a material to support bilayer electron+hole conducting gases and
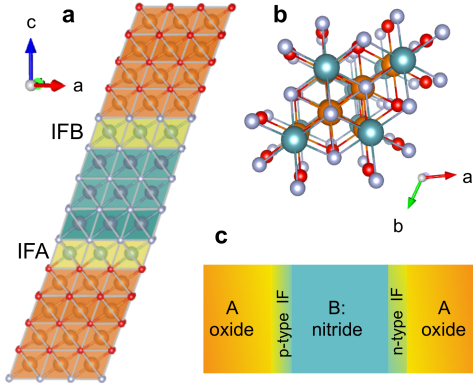
FIG. 1: Structure of ScN/MgO (111) multilayers. (**a**) Side view of the unit cell of ScN/MgO(111) multilayers five ScN layers thick with Sc atoms in blue, N atoms in gray, Mg atoms in orange, and O atoms in red. Yellow indicates the IF region: IFA, n-type (O-Sc) and IFB, p-type (N-Mg) (**b**) Honeycomb-like structure of the multilayers in the ab plane. (**c**) Sketch of the heterostructure configuration consisting on a thin slab of a transition metal nitride (B, ScN) sandwiched between an oxide (A, MgO)
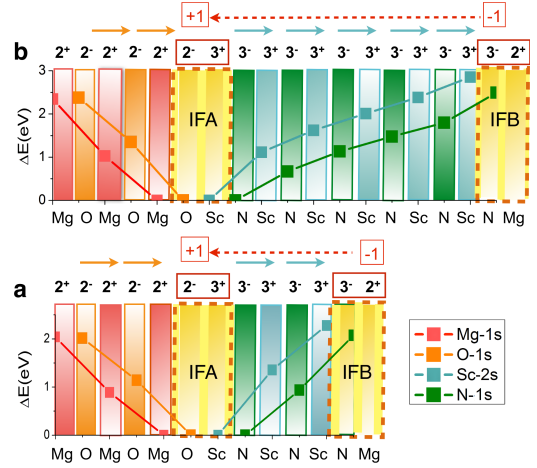

FIG. 2: Potential gradient in the multilayers. Layer-by-layer shifts for Mg-, N-, and O-1s (red, green and orange squares, respectively) and for Sc-2s (blue squares) core state energies for multilayers three (**a**) and six (**b**) ScN layers thick (ΔE represents the value of the shift in eV). Yellow indicates the interface O-Sc (IFA) and N-Mg (IFB) regions. The zero is set at IFA. An electric field is formed from IFB to IFA producing a potential build up across the multilayer of 2-3 eV. In the ScN side of the multilayer, there are N (green) and Sc (blue) planes with formal charges 3- and 3+. In the MgO side, Mg (red) and O (orange) layers have formal charges 2+ and 2-.

all the unusual phases that have been anticipated in such heterostructures. By means of first principles calculations, we have designed and explored MgO/ScN(111) superlattices with varying ScN thickness containing two charge imbalanced interfaces: one n-type and one p-type. Beyond a ScN thickness threshold of five layers these interfaces host conducting electron+hole gases, both gases lying within but on opposite edges of the ScN layer. Use of the N anion should promote robust two carrier 2D conduction compared to oxides, where holes are prone to localization.

Potential gradient. The DFT calculations were performed using the full-potential linearized augmented plane-wave code WIEN2K,[31] with the Wu-Cohen[32] and modified Becke-Johnson[33] exchange-correlation potential. See Supplemental Material for technical details. The basic structure of the multilayers is shown in Fig. 1. When growing periodically arranged superlattices two polar interfaces arise (see Fig. 2): the so-called interface A (IFA, n-type) $(2^-/3^+)$ and interface B (IFB, p-type) $(3^-/2^+)$. The polar nature of these multilayered systems will give rise to an intrinsic electric field formed as indicated by the arrows in Fig. 2. Induced by the polar discontinuities, one expects a large electrostatic potential offset between the two interfaces causing a sharp potential gradient. This effect can be seen by tracking the energy of the layer-by-layer core levels shown in Fig. 2 for multilayers

with different ScN thickness. A potential difference of 2 to 3 eV on each block of the multilayer develops. This gradient corresponds to a local electric field of 1.1×10^7 V/cm acting across the interface region, on the order of that obtained in LAO/STO heterostructures [8, 34]. The layer-by-layer shifts in the core levels are about 1 eV in the thinner multilayer and are reduced when the interfaces metalize. The superlattice periodicity forces the potential to return to zero after it has ramped up the ScN slab.

As a consequence of this potential gradient (that scales with thickness) there is an insulator-to-metal transition as the number of ScN layers is increased. For multilayers 3 ScN layers thick the gap (1.5 eV) is widened by confinement with respect to the value in the bulk (see the bulk band structure in the Supplementary Fig. 1), for 4 ScN layers the gap is reduced to 0.34 eV, and from a critical thickness of 5 ScN layers the band structures are metallic. Insulator-to-metal transitions with thickness have been observed in a number of oxide-based heterostructures in the literature, both polar and non-polar, including open-shell and closed-shell materials, such as multilayers of VO_2/TiO_2 [35], $SrVO_3/SrTiO_3$ [36–38], $LaVO_3/SrTiO_3$ [39], and the above mentioned $LaAlO_3/SrTiO_3$ [2, 34].
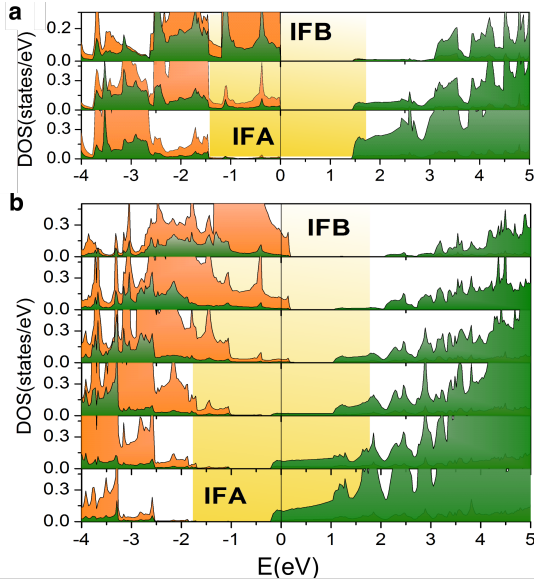
FIG. 3: Layer resolved density of states for Sc/N atoms across the ScN slab from IFB to IFA for ScN/MgO(111) multilayers three **(a)** and six **(b)** ScN-layers-thick. N-p states in orange, Sc-d states in green. The insulator-to-metal transition with thickness and the shift in the N-p and Sc-d states (of almost 1 eV/layer) as moving from IFA to IFB can be observed.

Electronic structure. The Sc and N projected density of states (DOS) for three and six ScN layers are shown in Fig. 3 where the the insulator-to-metal transition with thickness is evident. The nearly rigid upward shift consistent with the above described potential gradient can be seen by focusing on the N-$2p$ bands (orange) that move towards the Fermi level from IFA to IFB. The insulating state obtained for thinner ScN layers is shown in Fig. 3(a). The gap is formed between occupied N-p states and unoccupied Sc-d states, widened with respect to that in the bulk, an effect attributed to perpendicular confinement.

For multilayers with a thicker ScN block both interfaces host metallic states as shown in Fig. 3(b). N-p and Sc-d bands overlap in energy at IFB and IFA, respectively, leading to spatially separated hole and electron conducting states. The band structure plots and Fermi surfaces are shown in Supplementary Figs. 2 and 3, respectively. This band overlap provides a multilayer realization of the polar discontinuity scenario of electronic reconstruction, originally found for LAO overlayers on an STO substrate. Upon metallization electrons are transferred from one interface of the unit cell to the other, thus reducing the internal field although a substantial gradient persists (of the order of 10^7 V/cm). The result is 2D bilayer conductors each confined to two atomic layers (IF and IF-1), constituting a periodic array of alternating electron and hole 2DGs. The N-derived 2DHG should promote robust two carrier 2D conduction instead of insulating p-type interfaces.

Using a transition metal nitride provides yet other opportunities. The superconductivity observed in many samples of STO/LAO may be simply the state observed in n-doped STO (confined somewhat to reflect a 2D superconducting behavior), or it might be a new phenomenon due to an interfacial pairing processes. Ti, Zr, V, and Nb mononitrides with a rocksalt structure were among the first discovered superconductors with T_c's up to 16 K [15, 16]. Also, electron-doped transition metal chloronitrides MNCl (M = Hf, Zr, Ti) are superconducting in the 15-26 K range [40], putting them in the top six classes of high temperature superconductors. The observation of superconductivity at a transition metal nitride interface (as in this ScN/MgO system) is an exciting possibility, and if discovered would support that superconductivity observed at STO/LAO interfaces is derivative of bulk STO superconductivity.

Excitons (electron-hole bound states) are a topic of great importance in condensed matter physics and play a central role in solar energy conversion [10, 11, 39, 41]. Bristowe *et al.*[42] noted the possibility of exciton formation in the context of $LaAlO_3$/$SrTiO_3$ multilayers with alternating p and n interfaces, stymied because hole conduction is very difficult to achieve. The route proposed here for the formation of proximal electron and hole gases should provide the desired platform.

Bilayer transport. Parallel electron-hole bilayers were pioneered in GaAs/AlGaAs heterostructures [43]. In LAO/STO (001) overlayers a parallel electron-hole conduction has been reported only when an STO capping layer is added to protect the uppermost 2DEG[8, 9] – the 2DHG relies on a surface state rather than an interfacial layer, making it repeatable in multilayer form. It also provides a different regime of conducting bilayers, with thicknesses and separations down to one nm and the possibility of moving through various (correlated) phases with doping or gating.

To allow for additional experimental tests of the origin and nature of the conduction mechanism in these multilayers, we have calculated the temperature dependence of the thermopower $S(T)$. The expression for $S(T)$ within Bloch-Boltzmann transport theory, the Mott formula, is

$$S(T) = \frac{\pi^2 k_B^2 T}{3e} \frac{dlog\sigma(\varepsilon)}{d\varepsilon}\bigg|_{\varepsilon=\mu(T)}, \qquad (1)$$
$$\sigma(\varepsilon) \propto N(\varepsilon)v^2(\varepsilon)\tau(\varepsilon),$$

in terms of the energy-dependent conductivity $\sigma(\varepsilon)$, where k_B is the Boltzmann constant, e is the fundamental charge, N the density of states, v the group velocity, and τ the scattering time. The energy dependence of τ depends on the scattering mechanism and is usually treated as unimportant on a fine energy scale, as we do here.

For parabolic bands in 2D, $N(\varepsilon)$ is constant and the only energy dependence is from $v^2(\varepsilon) \propto \varepsilon$, hence $S(T) \propto \frac{\pi^2 k_B^2 T}{3e\mu(T)}$ where $\mu(T)$ is the chemical potential that is usually weakly T-dependent. When a two-carrier system develops, hole and electron contributions always add in the conductivity, but S(T) is determined by the sign of $d\sigma/d\varepsilon$, providing opportunities for engineering behavior. One outcome is that electron-hole systems may display a small and almost temperature independent thermopower. The Bloch-Boltzmann expressions [44],which include thermal excitation and involve only near Fermi surface quantities versus those from throughout the occupied bands (such as the carrier density), give the results shown in Fig. 4.

Values of the thermoelectric power for electrons and holes are linear with T and virtually identical in size ($|dS/dT| = 0.11\mu$V/K^2). These functions had to be calculated separately from the electron and hole bands. The net thermopower including electron- and hole-like Fermi surfaces is weighted by the respective conductivities ($\sigma_{xx,i}$) in the following way: $S_{xx} = \sum_i \sigma_{xx,i} S_{xx,i} / \sum_i \sigma_{xx,i}$ with i representing the band index. The two contributions compensate almost perfectly even though there is higher conductivity from the electron-like (Sc d) Fermi surface versus the hole-like one (N p), because $d\sigma/d\varepsilon$ also differs. Such clean compensation is desirable in application where decoupling between temperature gradients and potential differences is important. Fig. 4(b) shows the density of states (DOS) of electron and hole bands separately, indicating a nearly flat, 2D-like electron DOS over a range of several $k_B T$ (even at 400 K), while the hole DOS has a negative derivative.

The thermopower obtained in this ScN/MgO(111) multilayer is different than some examples discussed elsewhere. 2DEGs have been obtained in related interfaces but involving only one type of carrier [45].Quantum confinement, which narrows (some) bands, may produce a thermopower enhancement [46, 47]. Interestingly, ScN itself has been found to yield an anomalously large thermopower albeit at high doping level [48–50], suggested to result from localized impurity states close to the Fermi level [51].

The calculated Hall coefficient for electron and hole-like bands gives $R_h^H = 0.11 \times 10^{-7}$ m^3/C, R_e^H
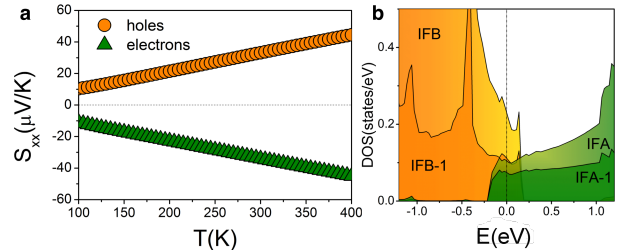

FIG. 4: *Bilayer conduction.* (a) Temperature dependence of the in-plane thermopower for MgO/ScN multilayers six ScN layers thick for electrons and holes. The contributions from electron and hole-like bands to $S_{xx}(T)$ compensate. The calculations were done by summing over only the valence Fermi surfaces for the hole properties (orange), only over the conduction Fermi surfaces for the electron properties (green). (b) Density of states near the Fermi level: hole-like N-p states at IFB and IFB-1 (orange) and electron-like Sc-d at IFA and IFA-1 (green).

= -0.16 $\times 10^{-7}$ m^3/C, both T-independent above 200 K, corresponding in a standard but simplistic interpretation to an effective carrier density (Hall number) n_e of 3×10^{13}carriers/cm^2 and $n_h = 4\times10^{13}$carriers/cm^2, in the same range as in oxide-only interfaces but without implying additional electrons that may reside in localized states [3, 8]. These densities are not equal because the bands are not parabolic as assumed in the interpreted from use of the Hall number. The classic textbook expression for R^H for a multi-carrier/multiband system demonstrates that electron and hole contributions are however not simply additive. The Bloch-Boltzmann semiclassical generalization demonstrates that for non-elliptical energy surfaces (as are two of the Fermi surfaces in this superlattice), the electron and hole contributions are entwined.

However, parallel superlattice 2DGs of the type being discussed here constitute a new type of alternating metal-insulator multilayer: the electron and hole 2DGs are distinct 2D systems, but intimately so. As such, in evaluating $S(T)$ and R^H above, the expressions from Bloch-Boltzmann theory (and the BoltzTraP code) have to be reconsidered. If in the definition $R_{xyz}^H = E_y/j_x B_z$ the current j_x from *both* electron and hole 2DGs is used, the result incorrectly incorporates quantities from two separate subsystems. These superlattices correspond to alternately layered n- and p-type 2DEGs, each with its own Hall voltage given by its own current density, *i.e.* velocities and DOSs, and the common electric field. The contributions must be calculated separately, as we have done.

We have proposed a novel approach to achieve 2D,

two carrier bilayer conducting systems by moving away from oxygen holes for hole conduction. Designing by choice of nitride and orientation of the interface, and studying with first principles calculations, hybrid oxide-nitride multilayers are proposed as a platform for the realization of parallel two dimensional, two carrier (electron+hole) gases. The basic concept builds on a slab of a narrow gap transition metal nitride (B) sandwiched between a wide gap insulating oxide (A). The polar discontinuity at the A and B interfaces provides an internal electric field that raises the N-p valence band maximum above the Sc-d conduction band minimum, with the charge transfer metalizing both interfaces. The use of the narrow gap nitride ScN means that both signs of carriers live in ScN. Electrically MgO is inert, shifting the active component from an oxide to a nitride.

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