Lattice relaxation and ferromagnetic character of \((\text{LaVO}_3)_m/\text{SrVO}_3\) superlattices

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**Abstract** – The experimental observation that vanadate superlattices \((\text{LaVO}_3)_m/\text{SrVO}_3\) show ferromagnetism up to room temperature (Lüders U. et al., Phys. Rev. B, 80 (2009) 241102(R)) is investigated by means of density functional theory, and the band structure for \(m=5\) and \(6\) is calculated. A buckling of the interface VO\(_2\) layers is found in both cases, but subtle differences in bond length lead to very different properties for even and odd values of \(m\): in the even case, the two interface VO\(_2\) layers effectively decouple from the adjacent LaO layers due to a strong bond length enhancement. This results into a local inversion of the orbital occupancy and to the confinement of the charge carriers. In the odd case, the amplitude of the bond length variation is smaller, so that the charge carriers spill into the deeper-lying VO\(_2\) layers, and spin-polarised interfaces are obtained.

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**Introduction.** – The combination of low dimensionality and strong correlations proved fruitful in the quest for new materials with functional-oriented properties. It led to the high-\(T_c\) superconductivity [1,2], transparent conducting oxides [3], high-capacitance heterostructures [4], and large thermopower [5], to quote a few. Recently, the creation of high-mobility electron gases with reduced dimensionality has also been pursued in artificially layered oxide systems. They include i) the interface between the two insulators SrTiO\(_3\) and LaAlO\(_3\) [6-8], ii) systems with ultra-thin doping layers as Nb : SrTiO\(_3\) in SrTiO\(_3\) [9], and RO layers with R = La, Pr, Nd in SrTiO\(_3\) [10], and iii) the surface of SrTiO\(_3\) [11]. In some of these systems functional properties were found, such as superconductivity [12] and ferromagnetism [13,14].

Charge carriers with reduced dimensionality can be introduced in other complex oxides, too. For instance, ultra-thin SrVO\(_3\) layers can be intercalated in LaVO\(_3\) [15], resulting in ferromagnetism up to room temperature [16] although none of the two oxides is magnetic at this temperature. This points out the relevance of the layered geometry as the solid solution La\(_{1-x}\)Sr\(_x\)VO\(_3\) shows the typical phase diagram of a doped Mott insulator with a transition from an antiferromagnetic insulator to a non-magnetic metal at about \(x = 0.2\) [17]. On the contrary, \((\text{LaVO}_3)_m/\text{SrVO}_3\) superlattices show ferromagnetic behaviour above room temperature for \(m\) ranging from 3 to 6, with a higher magnetisation for even \(m\) values than for odd ones. Thus, the geometrically confined doping in the superlattices gives rise to a magnetic phase which is not observed in the randomly doped solid solution.

From the theoretical point of view, the reduced dimensionality of electronic states confined in a potential created by the interface of two insulators, or by geometrically confined doping, leads to a new perspective in two-dimensional systems. Studies of the three-band Hubbard model predict a ferromagnetic ground state in case of 2/3 and 3/4 band filling for a reduced bandwidth due to the enhanced correlations of the electrons [18]. Ferromagnetism was also theoretically predicted for a model specific to two \(t_{2g}\) orbitals [19], at SrTiO\(_3\)/LaVO\(_3\) interfaces [20] and in SrTiO\(_3\)/SrVO\(_3\) superlattices [21], indicating a strong coupling of the electronic, magnetic and orbital degrees of freedom to the geometry of the superlattice including the number of SrVO\(_3\) layers. Again, in both cases the
magnetic phase was largely explained by the confinement and a change in the orbital occupancies.

However, in the case of superlattices or heterostructures, the reduced bandwidth and therefore enhanced correlations are just one part of the story. As in those systems two different materials are epitaxially grown on top of each other, also the midst of the lattice parameters of both materials plays a role for the properties of the system mediated by the distortion of the structure due to strain. Especially in oxides the distortion can have a strong influence on the properties since, as mentioned above, the electronic, spin and lattice degrees of freedom are strongly coupled. This effect was shown theoretically in distorted LaVO₃ where, depending on the distortion of the lattice, different antiferromagnetic structures become energetically favourable [22].

In order to analyse the ferromagnetic state observed in (LaVO₃)ₘ/SrVO₃ superlattices, electronic band structure calculations are helpful to confirm the confinement of the electronic states and to elucidate the origin of the ferromagnetism. For this purpose, we will use density functional theory and address the distortion of the lattice. Furthermore, the superlattices show an asymmetry of the magnetic properties between odd and even numbers of LaVO₃ unit cells. Thus, one aim of the calculations is to provide evidences for a possible difference in confinement between the two geometries.

**Bulk LaVO₃.** — There are many systems for which there is a consensus about the appropriate density functional. This is not the case for the vanadates and we therefore start our considerations addressing this point. The rare-earth vanadates with perovskite structure, including LaVO₃, have the V ion in a 3+ valence state, i.e., the two electrons in the V 3d shell occupy two of the t₂g orbitals with S = 1. The crystal field splitting of the 3d orbitals in threefold degenerate t₂g and twofold degenerate e₉ orbitals is a consequence of the octahedral coordination of the V ions by O atoms. The rare-earth vanadates show simultaneous antiferro-type spin and orbital ordering [23]. The spin ordering of LaVO₃ is C-type, i.e., the alignment is antiferromagnetic within the xy-plane and ferromagnetic along the z-direction [24]. The accompanying orbital order is G-type, i.e., it alternates in all directions. Other vanadates show G-type spin ordering together with C-type orbital ordering. For an overview of the phase diagram see ref. [23]. It has been shown in ref. [22] that strain due to the coherent growth on a substrate can strongly affect the orbital ordering. For an increasing c/a ratio the G-type order is destabilised.

It is essential that the approximation to the density functional treats the lattice, magnetic, and electronic properties on an equal level. While the local density approximation (LDA) typically underestimates the bond lengths and magnetic couplings, the generalised gradient approximation (GGA) overestimates both. For bulk LaVO₃, the GGA correctly predicts the C-type spin ordering, while the LDA tends to delocalise the electronic states, which is critical in the case of narrow 3d bands. To obtain an insulating state, corrections beyond the LDA/GGA have to be considered, such as the LDA/GGA+U approach in which a local interaction is added for the 3d orbitals [25]. This enhances the separation of the 3d energy levels and opens a band gap. The magnetic ordering may be changed by this treatment [26].

A further step towards a rigorous treatment can be achieved by hybrid functionals which incorporate a non-local Fock exchange. Hybrid functionals such as PBE0 and HSE yield an electronic structure comparable to self-consistent GW calculations for much lower computational cost. Even the local exact exchange for correlated electrons approach has shown clear advantages over the LDA/GGA + U in describing the electronic structure in nickelate stripe phases [27]. However, it is known that the Fock term (without correction of the correlation term) results in a substantial overestimation of the magnetic coupling and, therefore, in an unreliable determination of the magnetic ordering, including phase transitions. A GW treatment leads to accurate electronic structures and magnetic phase transitions but is computationally too costly for superlattice systems. The best compromise therefore is the LDA/GGA+U approach, which we apply in the following. Specifically, we employ the WIEN2k augmented plane wave plus local orbitals code [28]. We find that the energies and electronic states obtained for the fully localised and around mean-field double-counting corrections show the same behaviour.

It turns out that the interaction parameter $U_{\text{eff}}$ entering the self-interaction correction (SIC) scheme [29] should be adjusted as follows: for $U_{\text{eff}} > 5$ eV the C-type spin ordering is energetically favoured over the G-type ordering.

![Fig. 1: (Colour on-line) Supercell for the (LaVO₃)ₘ/SrVO₃ heterostructure for m = 5 (left) and m = 6 (right). La (Sr) atoms are represented by blue (green) spheres and V⁴⁺ (V⁵⁺) ions are represented by orange (pink) spheres. The grey squares indicate the oxygen octahedra. The oxygen atoms themselves are omitted, except for the bottom interface layer for m = 5 where black spheres are shown to highlight the buckling of the VO₂ layer. In the bottom of the m = 6 structure the (101) plane of the tetragonal (orthorhombic) cell is indicated in black.](image-url)
A band gap of 1.5 eV is obtained for $U_{\text{eff}} = 6.12$ eV in the case of C-type spin ordering, while the same gap is found in the case of G-type spin ordering for $U_{\text{eff}} = 4.8$ eV. In addition, the C-type orbital ordering under G-type spin ordering depends only weakly on $U_{\text{eff}}$ and on the lattice strain. On the other hand, the G-type orbital ordering under C-type spin ordering is very sensitive to strain. Disorder or ferro-type orbital ordering is found for $c/a > 1$ (which is fulfilled in our superlattice system). Ferromagnetism and A-type antiferromagnetism result in higher energies than the C- and G-type spin orderings for all approximations under study. While the G-type density of states (DOS) indicates very narrow bands, the bands are much broader for C-type spin ordering despite the higher U value. For C-type spin ordering, the $d_{xy}$ orbital, forming a double peak, is occupied, while the $d_{yz}$ orbitals are partially occupied and therefore subject to orbital ordering, in agreement with ref. [22]. In contrast, for G-type spin ordering, the $d_{xy}$ orbital is occupied, with a much less pronounced DOS structure, and the orbital ordering affects the $d_{x^2-y^2}$ orbitals.

Within GGA the ground state of bulk SrVO$_3$ is antiferromagnetic with $\mu_{\text{V}} = 0.2 \mu_B$, but the energy difference to the non-magnetic solution is small, amounting to 0.2 meV per V atom [30]. For the GGA+$U$ method, the C-type antiferromagnet is favoured by 30 meV per V atom over the A-type antiferromagnet and by 140 meV per V atom over the ferromagnet. A local hybrid approach with 25% Fock mixing would favour G-type spin ordering, since a C-type starting configuration changes to G-type during the self-consistency calculation. The on-site interaction does not open a band gap in bulk SrVO$_3$ but the compound stays metallic. Around the Fermi energy, we find the $t_{2g}$ orbitals equally partially occupied, while the $e_g$ orbitals are empty. Even though the spin-orbit coupling has little impact on the ground state of LaVO$_3$ it must be included in the case of SrVO$_3$ [21]. Indeed it splits the $t_{2g}$ states which in the end leads to a band gap of 0.61 eV. In this case C-type spin ordering (magnetisation in the z-direction) comes along with filled $d_{xy}$ and half-filled $d_{x^2-y^2}$ orbitals, yielding ferro-type orbital ordering. Accordingly, we include the spin-orbit coupling in our calculations. They are performed using the SIC scheme with $U_{\text{eff}} = 6.12$ eV. For the wave function expansion inside the atomic spheres we set $\ell_{\text{max}} = 10$ and define the plane-wave cutoff as $R_{\text{mt}}K_{\text{max}} = 7$. We use fine $10 \times 10 \times 1$ and $11 \times 11 \times 1$ k-meshes for the $m = 5$ and $m = 6$ supercells, respectively.

$m = 6$ superlattice. – The structure setup for the calculations of the superlattice starts from the tetragonal phase of LaVO$_3$, neglecting an orthorhombic lowering of the symmetry due to rotations of the oxygen octahedra. In addition, ideal atomically sharp interfaces are studied. We put one SrVO$_3$ layer between $m = 5$ or $m = 6$ LaVO$_3$ layers, representative of the odd and even superlattices, respectively. The lattice constants are taken from ref. [16] to be $a = 3.88 \text{Å}$ and $c = (m + 1) \cdot 3.95 \text{Å}$, incorporating the influence of the substrate.

The experimental $c/a$ ratio in the superlattice deviates from the bulk ratio of LaVO$_3$ [15]. In bulk LaVO$_3$ it is smaller than 1, $c/a = 0.98$ [31], but the strain exerted by the SrVO$_3$ layers distorts the LaVO$_3$ sub-layers in the superlattice. In case of the odd superlattice, six VO$_2$ layers lie between the SrO layers and the LaO forms the inversion layer, while in the even superlattice with its seven VO$_2$ layers, the central VO$_2$ layer is the inversion layer (see fig. 1). We neglect the antiferromagnetic order in the $ab$-plane and restrict ourselves to a $1 \times 1$ super-cell in the $ab$-plane. In the $c$-direction, ferromagnetic alignment is proposed, as expected for C-type spin order.

Let us start our investigations with the $m = 6$ superlattice and first focus on the orbital occupancy reconstruction at the interface. Deep in the LaVO$_3$ region a V$^{3+}$ valency is realised, with the $3d_{xz}$ and $3d_{yz}$ orbitals occupied and the V $3d_{xy}$ orbitals empty, as revealed by fig. 2. On the contrary, for the interface layers (the two VO$_2$ layers adjacent to the SrO layer) the V $3d_{xy}$ orbital is occupied and the V $3d_{x^2-y^2}$ orbitals are partially occupied, see fig. 2, together with a small admixture of the $e_g$ orbitals. This results in a smaller valency of the V ions, which provides us with a first indication that the doping of LaVO$_3$ is geometrically confined. This statement is further confirmed.
Table 1: Displacements (in Å) off SrO layers of the V and O atoms in the interface and middle/sub-interface layers for the heterostructures with m = 5 and m = 6. In the first LaO layers for m = 5 (6), the motion towards the SrO layer of the La ions amounts to 0.118 Å (0.093 Å), and the one of the O ions amounts to 0.012 Å (0.010 Å).

|                  | m = 5 | m = 6 |
|------------------|-------|-------|
| Interface layer  | −0.022| 0.096 |
| Middle/sub-interface | −0.005| 0.003 |

Fig. 3: Left: band structure of the m = 6 superlattice with non-magnetic interfaces. Right: band structure of the m = 5 superlattice with ferromagnetically polarised interfaces. The interface V 3dz² + 3dyz orbitals are highlighted.

Fig. 4: (Colour on-line) V-O and O-V bond lengths along the z-direction for m = 5 (top) and m = 6 (bottom). The vertical dashed (dotted) line indicates a/2 (c/2).

by the band structure shown in fig. 3. It exhibits four interface-based bands crossing the Fermi energy, two of them giving rise to fairly symmetrical Fermi surfaces (in the kzk_y-plane) centred around the Γ-point. These bands are characterised by rather large Fermi velocities. In addition, small hole pockets centred around the M-point are found. They are characterised by clearly smaller Fermi velocities and are due to bulk states. Therefore, the fast charge carriers are mostly confined to the interface layers, though in states also containing a small admixture from the sub-interface layer. Additional features of the band structure point to an interesting two-dimensional character of the bands crossing the Fermi energy. Principally constituted by d_{xz} (d_{yz}) orbitals which disperse along the Γ-X (Γ-Y) direction, they unexpectedly show considerable dispersion along X-M (Y-M), pointing to an admixture of other orbitals to the d_{xz} (d_{yz}) orbitals. Also, the dispersion around the Γ-point is rather flat and not cosine-like as would be expected for pure d_{xz}/d_{yz} bands.

A partial occupancy of the d_{xz} and d_{yz} orbitals establishes the prerequisite of an orbitally ordered state. However, this is neither observed in our calculations nor in the experiments, where these systems resemble rather good metals at low temperature [15]. An explanation for this behaviour may be found in the observed structure relaxations, where the most important ones are summarised in table 1. In the interface layers the V (O) ions move towards (off) the SrO layers, resulting in a sizeable buckling of the interface layers with an amplitude close to 0.11 Å. The first LaO layers move quite homogeneously towards the SrO layer, with a buckling amplitude smaller than 0.01 Å, while the buckling amplitude of the sub-interface VO₂ layer is close to 0.04 Å.

As a result, the bond lengths are strongly reduced between the V ions in the interface VO₂ layers and the O ions in the SrO layer, becoming smaller than a/2, i.e., the distance of the V ion to the O ions in the x-y-plane of the sample positioned at a/2 is larger than that to the O ion in the z-direction. Consequently, the non-degenerate V energy level ε_{xy} lies below the ε_{xz,yz} levels in the interface layer, leading to an enhancement of the dispersion in those layers. On the other hand, the enlarged bond length between the interface layer and the adjacent LaO layer suppresses the orbital overlap in the z-direction and leads to an effective electronic decoupling of the interface VO₂ layer with respect to the other VO₂ layers, as shown in fig. 4. The same tendency of the buckling can still be seen in the sub-interface layer, but with a much smaller amplitude. The O-V bond length in this case is near to c/2 as for the other VO₂ layers. Thus, the larger distance between the V ions and the surrounding O atoms is to be found along the z-direction, inducing the bulk orbital occupancy and, hence, an orbitally ordered insulating state like in the bulk.

Thus, the calculated band structure accounts for a two-dimensional metallic state, but it cannot account for the experimentally observed ferromagnetism. In a Stoner picture this would be understood as the result of a rather small DOS at the Fermi energy. Therefore, should this system be ferromagnetic then ferromagnetism would likely be caused by further correlation effects, as taken into
account in calculations of multi-orbital Hubbard models [18,19]. Moreover, we have verified that the above electronic structure is stable against the employed $U_{\text{eff}}$ value and lattice relaxation, as is the establishment of an ultra-thin two-dimensional electron gas.

$m = 5$ superlattice. — The $m = 5$ superlattice hosts different physics. Again, lattice relaxation plays an important role. The obtained buckling of the interface layers is summarised in table 1. The interface VO$_2$ layer shows a buckling with the same displacement directions as for $m = 6$ (the V ions move towards the SrO layer and the O ions towards the LaO layer) but the amplitude is smaller. Notably, as shown in fig. 4, while for the $m = 6$ case the distance between the V ion in the interface layer and the O ion in the SrO layer becomes smaller than $a/2$, inducing the inversion of the orbital occupancy, in the $m = 5$ case the bond lengths stay larger than $a/2$ in all the VO$_2$ layers. The smaller amplitude of the buckling therefore does not induce an inversion of the orbital occupancy and the confinement of the charge carriers is clearly less effective than in the even case. Furthermore, the reduction of the bond lengths reaches further into the LaVO$_3$ layer, where the bond length of the V ion with the second LaO layer counted from the SrO layer is still strongly reduced. In comparison, in the $m = 6$ case the same bond length is larger than $c/2$.

The change of the structural relaxation is visible in the resulting layer-projected densities of states in fig. 2. They consist of the spin-majority density of states, as no sizeable contribution from the spin-minority bands is obtained. All layers are clearly off stoichiometry and all V ions are in a mixed valency state, yet closer to the 3+ valency in the bulk layers than in the interface layers. Moreover, all V 3$d$ electrons occupy the 3$d_{xz}$ and 3$d_{yz}$ orbitals, while all 3$d_{xy}$ orbitals are empty, in contrast to the $m = 6$ superlattice.

The band structure of the $m = 5$ superlattice is shown in fig. 3. As there are no spin-minority bands close to the Fermi level, only spin-majority bands are shown. There are two narrow bands formed by the interface V 3$d_{xz,yz}$ orbitals around the Fermi level. The four next lower-lying bands are due to the middle and bulk layers with a larger bandwidth. The bands at the Fermi level are separated by a band gap of about 1 eV (measured at the Γ-point) from the parabolic bands of the bulk states. In addition, the bands with the largest Fermi velocities mostly trace back to the middle layer V 3$d_{xz,yz}$ orbitals, with some admixtures of the bulk and interface layers. Therefore, transport is no longer confined to the interface layers but there is a distinct modulation associated to it. There are fast charge carriers mostly confined to the next-to-interface layer, with a small admixture of their two neighbours, and slow charge carriers mostly confined to the interface layer. Narrow $d$ bands with high DOS at the Fermi level often explain ferromagnetism in the 3$d$ transition metal oxides. Here the high DOS follows from the slow charge carriers that are confined to the interface layers.

| Interface layer | Middle/sub-interface layer | Bulk layer/middle layer | −/bulk layer |
|-----------------|---------------------------|------------------------|--------------|
| 1.6             | 1.6                       | 1.4                    | 1.6          |

Discussion and conclusions. — Our calculations show that the doping resulting from the charge carriers donated by the SrO layers in (LaVO$_3$)$_m$/SrVO$_3$ superlattices leads to rich physics built on fast and slow charge carriers together with lattice relaxation. For both types of superlattices the lattice relaxation plays an important role, demonstrated by the obtained buckling of the VO$_2$ interface layers and alternation of short and long V-O bond lengths along the c-axis. Although the buckling of the interface VO$_2$ layers is qualitatively the same in the two studied cases, the critical difference in the amplitude leads to two different band structures. In the odd $m = 5$ case, slow charge carriers are mostly confined to the interface layers in which they order ferromagnetically. The confinement of the fast charge carriers is less effective and their distribution peaks in the middle layers. For even superlattices the relaxation confines the fast charge carriers to largely decoupled interface layers in which they form an ultra-thin two-dimensional electron gas. No magnetic order is found and the distribution of slow charge carriers peaks in the sub-interface layers. Therefore, coming back to our original question, it seems unlikely that the geometric confinement of the charge carriers alone is the origin of the experimentally observed ferromagnetic state. Instead, the fact that the even and odd superlattices show a different behaviour points out the relevance of the number of unit cells of the LaVO$_3$ layers. This also holds true for the distribution of the magnetic moments carried by the V ions. Indeed, table 2 shows that they are quite homogeneously distributed over all layers, with the notable exception of the interface layer, for $m = 6$. Their distribution is quite homogenous for $m = 5$ as well, though with a suppressed magnetic moment in the bulk layers.

In our calculations we did not take into account the orthorhombic structure of LaVO$_3$ [31], generated by the octahedral tilts which were shown to be present in pure LaVO$_3$ thin films as well [32]. The general features of bulk LVO, as the magnetic and the orbital ordering, can be theoretically reproduced based on a cubic description of the lattice [24], and the orthorhombic distortions enhance further the stability of these features [22] without being critical. However, the observed relaxation of the structure of the superlattices may be a reminiscence of these energetically favourable orthorhombic distortions of the bulk. The related unit cell is larger than the tetragonal cell used in the calculations, in particular along the tetragonal c-axis, where the out-of-phase rotation of the
octahedra leads to a doubling of the unit cell, see fig. 1. Therefore, in an even superlattice it is possible to establish the bulk structure including the octahedral rotations in the LaVO$_3$ layer. On the other hand, in the odd superlattice the thickness of the LaVO$_3$ layer cannot be established. The compensation by the structure of the lack of the energetically favourable out-of-phase rotation may lead to the small but critical differences of the lattice relaxations and therefore the orbital occupancies between the even and odd case. The combination of the individual reactions of the structural, orbital, and electronic degrees of freedom leads in the end to a ferromagnetic ground state in the odd superlattices.

Our calculations confirm therefore the experimentally observed magnetisation oscillations [16], although in the experiment the even superlattices were found to show a higher magnetisation than the odd ones. This difference can be explained by the fact that the thickness of the LaVO$_3$ in the experimental study is a nominal one, where the announced thickness does not take into account any intermixing of La and Sr during the growth. It was shown that the interface between the LaVO$_3$ and SrVO$_3$ layers is abrupt but exhibits steps of typically one perovskite unit cell [33]. The steps lead to the presence of Sr atoms in the first LaO layer next to the SrO layer. Taking into account the growth direction, it can be argued that the thickness of the pure LaVO$_3$ layer will thus be one unit cell less than the nominal one, resulting in an inversion of the nominally odd and nominally even superlattices. Based on the band structure calculations, we also expect to see a difference in the resistive properties of the superlattices, especially concerning the mobility of the charge carriers.

In conclusion, two scenarios for the even and odd superlattice have been found. In both cases, the $d_{xz}$ and $d_{yz}$ orbitals form bands crossing the Fermi level. In the even superlattice, the orbital occupancy changes from the interface to the bulk layers, while we find a homogeneous orbital occupancy in the odd superlattice. The room temperature magnetism is related to the incomplete formation of the orthorhombic unit cell of LaVO$_3$ in odd superlattices. A next step could be a comparison of theoretical and experimental macroscopic transport properties of the confined fast charge carriers.

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