Competing magnetic interactions in spin-1/2 square lattice: hidden order in Sr₂VO₄

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(Dated: January 6, 2022)

With decreasing temperature Sr₂VO₄ undergoes two structural phase transitions, tetragonal-to-orthorhombic-to-tetragonal, without long-range magnetic order. Recent experiments suggest, that only at very low temperature Sr₂VO₄ might enter some, yet unknown, phase with long-range magnetic order, but without orthorhombic distortion. By combining relativistic density functional theory with an extended spin-1/2 compass-Heisenberg model we find an antiferromagnetic single-stripe ground state with highly competing exchange interactions, involving a non negligible inter-layer coupling, which places the system at the crossover between between the XY and Heisenberg picture. Most strikingly, we find a strong two-site "spin-compass" exchange anisotropy which is relieved by the orthorhombic distortion induced by the spin stripe order. Based on these results we discuss the origin of the hidden order phase and the possible formation of a spin-liquid at low temperatures.

The Heisenberg model on a square lattice is one of the most widely studied models in statistical physics. The applicability of this model in modern solid-state physics has become popular after the discovery of layered high-Tc superconductors, since the parent magnetically ordered compounds are often considered as quasi two-dimensional (q2D) systems [1], and, more recently, for the interest in the interplay between magnetism and superconductivity in Fe-based superconductors [2]. In the 2D Heisenberg model the relative strength and competition of antiferromagnetic nearest and next nearest neighbor interactions (J₁ and J₂) provides useful insights on the stability of specific types of magnetic order: J₁ favors the Néel order (e.g., cuprates [3]), J₂ favors the stripe (ST) order (e.g. few types of vanadates [4–7]), whereas a spin-liquid state is expected to emerge near the classical phase-boundary between the Néel and the ST orderings [8–10]. The inclusion of additional interactions (J₃, J₄, ... ) leads to more exotic states [11], for example the enigmatic magnetic phases recently found in Fe-based superconductors which arises from the strong competition between the ST and Néel order [12–15].

Tetragonal Sr₂VO₄, isostructural to the high-Tc parent compound La₂CuO₄ and similar to layered vanadate Li₂VOSiO₄ [4], provides an opportunity to explore the role of the various types of magnetic interaction at play in a square lattice, owing to the presumably weak interplane interaction between spin-1/2 V⁴⁺ (d⁵) layers. The complicated structural, magnetic, and electronic transitions observed in Sr₂VO₄, in fact, suggests a competition and/or coexistence of different magnetic phases [16–24]. Upon cooling, the crystal structure evolves from tetragonal to an intermediate phase (at Tc₂ ~ 140K), and again to a tetragonal phase (Tc₁ ~ 100K) with a larger c/a ratio [19]. In the intermediate regime, an anomaly in the susceptibility is observed at TM ~ 105K, which was initially thought to originate from magnetic order [16]. No signals of spin-orbital order was detected down to low temperature but, rather, a possible existence of a magnetic phase, originating from the competition between magnetic interaction and Jahn-Teller (JT) effect [23].

Several proposals have been put forwarded for the elusive low-T ground state: orbitally ordered phase due to JT distortion [16], a competition between an orbitally ordered parquet-type and a double-stripe (DS) magnetic ordering [17, 18] (DS10 in Fig. 1(a)), an octupolar order driven by spin-orbit coupling (SOC) [19], and a Néel order with muted magnetic moment, where the spin and orbital moments cancel each other [21].

The magnetic properties are highly dependent on the sample quality [20]. Recent experiments performed with high quality samples have shed some light on the physics of Sr₂VO₄. X-ray diffraction (XRD) experiments has revealed that the intermediate structure (Tc₁ < T < Tc₂) is orthorhombic phase [22, 23]. Based on a muon spin rotation and relaxation (µSR) study Sugiyama et. al found that the actual magnetic ordering temperature (TN ~ 10K) is much lower than the temperature of the susceptibility anomaly (TM ~ 105K) and claimed that the role of the SOC is marginal [24]. This is consistent with the reported persistence of inhomogeneous magnetic states down to 30 mK, where the sizeable competition of ferromagnetic and antiferromagnetic correlations prevents the system to develop a long range ordered phase [22]. The current understanding of the magnetic and structural properties of Sr₂VO₄ is thus still debated and a commonly agreed explanation is still lacking.

In this Letter we investigate the peculiar magnetic behavior of Sr₂VO₄ by first-principles calculations including relativistic effects. We show that Sr₂VO₄ can be considered as a frustrated spin-1/2 Heisenberg system with highly competing exchange interactions. Our data indicate that the onset of the magnetic order is controlled by two factors with similar order of magnitude: the relativis-
To study the magnetic interactions in Sr$_2$VO$_4$ we have performed first principles calculations within the density functional theory (DFT) plus an on-site Hubbard $U$ using the Vienna Ab Initio Simulation Package [25, 26]. The experimental high-$T$ tetragonal structure [27] was modeled with a supercell containing 16 unit cells, with which we have simulated different types of spin orderings: single stripe (ST), double stripe along [100] and [110] (DS10 and DS11, respectively), ferromagnetic (FM), staggered dimer (SD), Néel, and parquet order [see Fig. 1(a)]. For the calculation of the exchange parameters we adopt the value $U=5$ eV, which is consistent with the value obtained fully ab initio within the constrained random phase approximation (cRPA). Further methodological details are available in the Supplemental Materials [28].

Fig 1(b) shows the DFT+$U$ total energies for the considered spin orderings as a function of $U$. The data for $U < 2$ eV are omitted because for these values of $U$ DFT finds a metallic ground state, in disagreement with experiment [29, 30]. We find that the ground state is the ST configuration irrespective of the value of $U$. This is surprising because in earlier literature the ST phase has never been considered as a possible ground state magnetic structure, even though other layered oxides such as Li$_2$VOSiO$_4$ exhibit the ST order [4]. Our data shows that the relative energies do not depend strongly on $U$, and with increasing $U$ the relative-energy window becomes narrower indicating that the strength of the magnetic interactions are progressively weakened. This can be expected since the $J$s are inversely proportional to $U$, $J_{ij} \sim t_{ij}^2/U$, where $t_{ij}$ is the hopping between two sites, $i$ and $j$. The shrinking of the energy window clearly implies a strong competition among the various exchange interactions in action between neighboring spins (see inset in Fig. 1).

Independently on the magnetic state and on the $U$-value DFT deliver a spin moment on V of about 1 $\mu_B$, which suggests a spin-1/2 local moment state, in agreement with the experimental Curie-Weiss behavior of the high-$T$ magnetic susceptibility which provides an effective moment close to 1.36 $\mu_B$ [24]. However, Sr$_2$VO$_4$ does not show any magnetic order down to very low-$T$ [20].

The driving mechanism that stabilizes a magnetic order at finite temperatures is one of the most important and subtle question related to 2D magnetic compounds, as stated in the famous Mermin-Wagner theorem [31]. To analyze the physical reasons for the absence of magnetic order in Sr$_2$VO$_4$ we start by mapping the different spin-ordered DFT total energies onto the following Heisenberg-like Hamiltonian,

$$
H_H = J_1 \sum_{\substack{\alpha,\langle ij \rangle}} S_{i\alpha} \cdot S_{j\alpha} + J_2 \sum_{\substack{\alpha,\langle\langle ij \rangle\rangle}} S_{i\alpha} \cdot S_{j\alpha} + J_3 \sum_{\substack{\alpha,\langle\langle\langle ij \rangle\rangle\rangle}} S_{i\alpha} \cdot S_{j\alpha} + J_4 \sum_{\substack{\alpha,\langle\langle\langle\langle ij \rangle\rangle\rangle\rangle}} S_{i\alpha} \cdot S_{j\alpha}
$$

$$
+ J_\perp \sum_{\substack{\alpha,\langle ij \rangle}} S_{i\alpha} \cdot S_{j\alpha+1} + R \sum_{\alpha, \text{plaqette}} \left[ (S_{i\alpha} \cdot S_{j\alpha})(S_{k\alpha} \cdot S_{l\alpha}) + (S_{i\alpha} \cdot S_{l\alpha})(S_{k\alpha} \cdot S_{j\alpha}) + (S_{i\alpha} \cdot S_{k\alpha})(S_{j\alpha} \cdot S_{l\alpha}) \right],
$$

where $\langle \rangle$, $\langle\langle \rangle\rangle$, $\langle\langle\langle \rangle\rangle\rangle$, and $\langle\langle\langle\langle \rangle\rangle\rangle\rangle$ represent the sum over first, second, third and fourth nearest neighbors (NNs),

FIG. 1. (a) Sketch of the different magnetic orderings used to fit the Heisenberg spin Hamiltonian. Single stripe (ST), double stripe along [100] and [110] (DS10 and DS11), ferromagnetic (FM), staggered dimer (SD), Néel, and parquet order. (b) Total energy as a function of $U$ within DFT+$U$ calculations. Inset: schematic description of the exchange interactions.
part of the paper we will consider the possibility that the stabilization of a magnetic order at finite temperature could originate from anisotropic effects. Specifically, we will analyze the anisotropy of the exchange interactions due to SOC.

To this aim, we have computed the total energy of the ST phase with the moments oriented along the high-symmetry directions [001], [010] and [001] by means of magnetically constrained DFT+U+SOC calculations [39] (see Fig.2. We found that $E_\perp$ is 2.75 meV and 2.63 meV (per V ion) lower than $E_\parallel$ and $E_z$, respectively, implying strong anisotropic effects. As these differences cannot be explained by single-site anisotropy (since we have a good spin-1/2 system) they must be attributed to symmetric anisotropy exchange, which can be taken into account by including additional terms in the spin Hamiltonian (Eq. 1). Within a first NN approximation the resulting spin Hamiltonian reads:

$$H_{TH} = H_H + \sum_{<nn>} J^{zz} S_i^z S_j^z \pm \sum_{<nnxy>} J^{xy} (S_i^x S_j^y - S_i^y S_j^x),$$

where $J^{zz}$ is the two-site Ising anisotropy (Kitaev term), and the "spin-compass" exchange $J^{xy}$. By mapping the total energies onto the above generalized spin Hamiltonian, we obtain $J^{xy} = -0.71$ meV and $J^{zz} = -1.21$ meV, of the order of 10 K, similar to the case of 4d Sr$_2$RuO$_4$ [32]. The emergence of anisotropic terms demonstrates the considerable role played by SOC even in 3d oxides. However, we find a negligible orbital moment ($<0.05 \mu_B$), which shows that SOC contributes to the anisotropy of the system but does not form a spin-orbit entangled phase [19, 21].

The very strong Ising term $J^{zz}$ favors easy-plane anisotropy, putting the system close to the Heisenberg/XY-model threshold since it does not lift the frustration of the ST order. Thus Sr$_2$VO$_4$ is quite different from Sr$_2$TeO$_4$ [38] and other similar q2D systems, where the magnetic order appears to be stabilized by small negative $J^{zz}$ and a weak dipole-dipole interaction [38]. What is surprising and peculiar in Sr$_2$VO$_4$ is the existence of a strong spin-compass anisotropy $J^{xy}$

| TABLE I. Total energies of the considered spin-ordered configurations and magnetic interactions of the ST ground state evaluated from the Heisenberg Hamiltonian (Eq. 1). Data given in meV for $U=5$ eV. |
|---|---|---|---|---|---|---|
| ST | DS10 | Néel | Parquet | FM | SD | DS11 |
| -8.39 | -5.56 | -1.35 | -0.28 | 0 | 1.21 | 6.26 |

| $J_1$ | $J_2$ | $J_3$ | $J_4$ | $J_{\perp}$ | $R$ | $J^{xy}$ | $J^{zz}$ |
|---|---|---|---|---|---|---|---|
| -0.65 | 1.02 | -0.54 | 0.46 | -0.12 | -0.28 | -0.71 | 1.21 |
which lifts the frustration of the ST phase and favors the onset of magnetic order at finite temperatures.

From the calculated values of the dominant interactions we can now make some estimates and provide a comparison with experiment. At high temperatures the system exhibits a Curie-Weiss (CW) behavior of the susceptibility with slightly different CW temperatures $\theta_{CW}$ reported in literature: $-24$ K [24], $+24$ K [40], and $+47$ K [41]. The mean-field value of $\theta_{CW}$ derived from our data (Tab.1) is $+10$ K falling exactly in the middle of the experimental scattered data range. If we estimate a mean-field ordering temperature of the ST phase we obtain $+74$ K, which is somewhat below, but still close to the narrow temperature window, 100-140 K, covering the two structural ($T_{c2}$ and $T_{c1}$) transitions and the magnetic susceptibility anomaly at ($T_M = 105$ K) [23]. Thus, the ST short-range order and critical fluctuations are expected to develop exactly in the temperature interval where the orthorhombic distortion exists. But, there is a reason for $\text{Sr}_2\text{VO}_4$ to avoid a transition to ST order.

In q2D compounds with ST order, like Fe-pnictides and layered $\text{Li}_2\text{VOSiO}_4$ [4], the transition to the ST phase is associated with an orthorhombic distortion, which is believed to break the frustration intrinsic to the ST phase [42]. The orthorhombic distortion in the Fe-pnictides [43] appears at a temperature considerably higher than the magnetic phase transition implying that the short-range ST order is responsible for the orthorhombic distortion even though long-range magnetic order is absent. In the following we show that the orthorhombic distortion in $\text{Sr}_2\text{VO}_4$ is a consequence of the magnetostriction effects in the ST phase.

To inspect the relation between magnetic and structural order we have fully relaxed the high-$T$ tetragonal phase within DFT+U by imposing the ST order. As a result, we obtained a bifurcation of the lattice parameters ($a=3.971$ Å, $b=3.960$ Å) and the formation of an orthorhombic phase, in agreement with XRD measurements [23]. This transition is connected to the magnetostriction effect, namely the coupling between non-relativistic isotropic exchange and crystal structure. However, the inclusion of anisotropic effects via SOC leads to a very surprising result: in the fully relaxed orthorhombic phase the anisotropic terms are considerably reduced, $J^{xy} = 0.00$ meV and $J^{zz} = 0.38$ meV, meaning that the two-site compass term almost vanishes ($E_x = E_y$). The orthorhombic distortion caused by magnetostriction in the ST phase quenches the spin-compass anisotropy, which is the leading mechanism for the stabilization of ST phase (lifting of the frustration) at finite temperature.

Based on our analysis we can now interpret the complicated transitions in $\text{Sr}_2\text{VO}_4$: (i) Starting well above the magnetic ordering temperature and decreasing the temperature a strong short-range order, characteristic of a ST phase, develops within a tetragonal symmetry.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig2}
\caption{(a) Schematic diagram showing the transition temperatures and structures. The system has a tetragonal structure with active anisotropic exchange interactions for $T > T_{c2}$, while for the orthorhombic structure for $T_{c2} > T > T_{c1}$ the anisotropy vanishes. Tetragonal symmetry is recovered below $T_{c1}$. Stripe order is shown with moments along (b) (100) and (c) (010), respectively. The squares denotes unit cell used.}
\end{figure}

(ii) This short-range ST order causes a tetragonal to orthorhombic structural transition and simultaneously kills the spin-compass anisotropy; the system avoids a magnetic transition to the ordered ST phase since the latter remains fully frustrated. (iii) When the temperature is lowered further, the system re-enters the tetragonal phase, since other phases, like DS10, supported also by a non-frustrating inter-plane FM coupling, become energetically equally favorable and partially destroy the short-range ST order. (iv) At this point the system enters an intermediate temperature range with different energetically competing magnetic orders, in line with the complex behavior of the measured magnetic susceptibility; (v) Finally, at low temperature the system falls into some spin-liquid or spin-glass states, where the FM phase becomes quasi-stable due to strong short-range FM interactions in the 1NN shells both in-plane and inter-plane, in agreement with recent $\mu+\Sigma$SR experiment [22].

In summary, we have shown that $\text{Sr}_2\text{VO}_4$ exhibits a peculiar magnetism, distinctively different from other layered oxides. It displays a frustrated magnetic ground state with significant inter-plane interaction (placing it in the 2D/3D crossover regime), and strong in-plane anisotropy (Heisenberg/XY model crossover), in particular the "spin-compass" exchange term, which lifts the frustration of the ST ground state. Anisotropic magnetic interactions, like the one observed in $\text{Sr}_2\text{VO}_4$, are expected be a pervasive phenomenon not only in 4d and 5d, but also in 3d compounds, in particular transition metal oxides, and could play a key role in the onset of hidden magnetic orderings. Our proposed scenario is compatible with experimental data and call for a full verification by future experiments. For instance, we expect that nuclear magnetic resonance or inelastic neutron scattering experiments on $\text{Sr}_2\text{VO}_4$ single crystals could allow for a conclusive identification of the origin of the low-$T$ phases [23] and should reveal the character of the magnetic short-range order effects in the three distinctive temperature intervals.
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ACKNOWLEDGEMENTS

We thank J. Sugiyama for fruitful discussions. This work was supported by the Austrian Science Fund (FWF) projects ViCom (F4109-N28) and INDOX (11490-N19). The computational results presented have been achieved using the Vienna Scientific Cluster (VSC).