Editors' Suggestion

Fragility of the magnetic order in the prototypical altermagnet RuO_2

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(Received 11 October 2023; revised 29 February 2024; accepted 20 March 2024; published 16 April 2024)

Altermagnetism is a topic that has lately been gaining attention and the RuO_2 compound is among one of the most studied altermagnetic candidates. However, the survey of available literature on RuO_2 properties suggests that there is no consensus about the magnetism of this material. By performing density functional theory (DFT) calculations, we show that the electronic properties of stoichiometric RuO_2 are described in terms of a Hubbard U, within DFT + U, smaller than the value required to have magnetism. We further argue that Ru vacancies can actually aid the formation of a magnetic state in RuO_2. This in turn suggests that a characterization of the amount of Ru vacancies in experimental samples might help the resolution of the controversy between the different experimental results.

DOI: 10.1103/PhysRevB.109.134424

I. INTRODUCTION

In recent years the topic of altermagnetism has been gaining attention, with significant efforts directed towards finding new altermagnetic materials [1,2]. Altermagnetism is defined as a magnetic phase with symmetry-driven compensated net magnetization, where the symmetry operation responsible for this magnetic phase is neither inversion nor translation. A material exhibiting these properties combines characteristics of both ferromagnetism and antiferromagnetism. Furthermore, in regards to the electronic band structure, the bands in this phase are non-spin-degenerate, leading to intriguing applications.

For a metallic system, altermagnetism would imply a possibility of generating spin-polarized currents, as in ferromagnets. Moreover, generating spin-transfer torque and observing giant/tunneling magnetoresistance effects in such a system should be possible. These effects are utilized to construct magnetic memory devices and the benefits that altermagnets bring in comparison to ferromagnets are the vanishing stray magnetic fields and the THz range of switching [1].

Among the various proposed materials as altermagnetic candidates, RuO_2 is attracting much attention. However, the magnetism in this system is still in itself a controversial topic. On the one hand, the absence of a discernible phase transition in the heat capacity [3,4], the magnetic susceptibility [5–7], and the resistivity data [8,9] suggests that RuO_2 is a Pauli paramagnet. On the other hand, the existence of an antiferromagnetic configuration has been reported by resonant x-ray scattering [10] and neutron diffraction [11]. However, the latter measurements have shown a rather small local magnetization value (0.05 μ_B). At the same time, no magnetic hyperfine field was detected on Ru in an NMR experiment [12], usually a very sensitive probe. Additionally, there have been observations of a sizable anomalous Hall effect, consistent with a considerably larger magnetization [13,14].

The magnetic configuration suggested in Zhu *et al.* [10] is where the magnetic axis is along the c axis (see Fig. 1). However, they report a better fit to the measured scattered intensity of x rays when the magnetic moments are slightly canted. Zhu *et al.*'s [10] findings and the conclusion about the long-range magnetic order are questioned by Lovesey *et al.* [15]. In turn, they proposed three different motifs (including quadrupoles into the consideration) and attempted to fit the abovementioned experimental data but have been unable to achieve a high-quality fit to any of those models. For one of the motifs, Ref. [16] further provides an extended set of calculated diffraction patterns that could be used to test the presence of assumed magnetic order. Thus, to draw an unambiguous conclusion, one would require more experimental data.

Unfortunately, the available neutron diffraction data on RuO_2 [11] are not sufficient to confidently resolve the controversy on the magnetization, for the reasons described below. The main issue is that the quality of the magnetic component of the fit in these experiments depends on the quality of the structural refinement. In Ref. [11], the authors mention the

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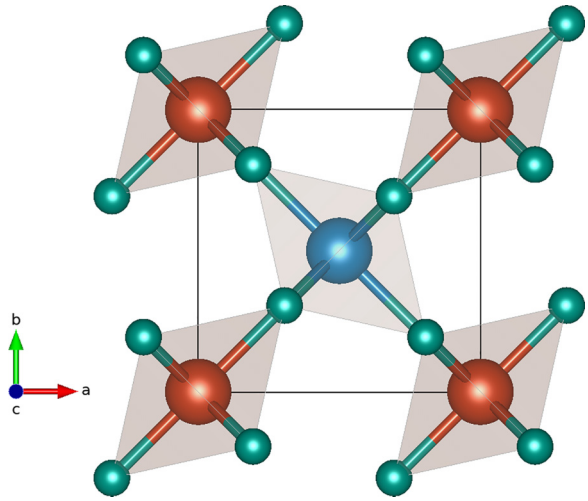

FIG. 1. Crystal structure of RuO_2: Ru atoms are shown in red and blue (different colors denote different spin orientations), and O atoms are shown in teal.

possibility of a structural distortion in the rutile phase accompanied by antiferromagnetic order. However, Ref. [11] was unable to find a distorted structure that would fit both unpolarized and polarized neutron diffraction data, while the powder x-ray diffraction patterns are consistent with the undistorted rutile structure (see the crystal structure depicted in Fig. 1). To address this problem, Ref. [11] employed density functional theory (DFT) calculations in an attempt to find such a structure. A distorted $2 \times 2 \times 2$ rutile supercell was optimized in both the nonmagnetic and antiferromagnetic states, and the rutile structure was obtained as the ground state. The available computational data on the lattice dynamics in RuO_2 [17,18] confirm that the rutile structure is dynamically stable.

The absence of a structural phase transition is indirectly confirmed by other measurements. Two independent electron transport measurements, one conducted up to 300 K [8] and one up to 1000 K [9], show no changes in resistivity that could be caused by a structural phase transition. Both data sets are well described by a model that has three contributions to the resistivity: the electron-phonon interaction with acoustic (Bloch-Grünesien) and optical modes, along with a term arising from electron-electron scattering. Moreover, no indications of a structural phase transition were found in calorimetry experiments, where the heat capacity was measured up to ∼340 K [3] and ∼1050 K [4]. This same conclusion is supported by the available measurements of thermal expansion [19].

Based on the refinement using the rutile structure, the extracted magnetic moment per Ru atom is 0.23 μ_B for unpolarized neutron diffraction and 0.05 μ_B for polarized neutron diffraction measurements [11]. Furthermore, there is no evidence of a phase transition to an antiferromagnetic phase, neither in the susceptibility data of Ref. [11] nor in earlier measurements [5–7]. Additionally, nuclear magnetic resonance measurements strongly suggest the absence of long-range magnetic order. This conclusion is supported by the absence of any contribution from Ru d electrons in both the Knight shift and the relaxation rate, as well as the absence of any hyperfine splitting [12]. The authors of this paper point out that, overall, the resonant magnetic properties closely resemble those of nonmagnetic Ru metal.

The controversy among the different experiments suggests that the existence of antiferromagnetic (and hence altermagnetic) order in RuO_2 is rather fragile, likely sample dependent, and possibly present in only a fraction of the sample volume. In order to gain a better microscopic understanding of the magnetism (or lack thereof) in this material, we have systematically investigated the magnetic states of RuO_2 employing DFT, both with and without a Hubbard U correction applied to the Ru d orbitals. Our tentative conclusion is that the perfectly ordered, stoichiometric RuO_2 is likely nonmagnetic, consistent with numerous experiments above. On top of that, a modest hole doping, for instance, by creating Ru vacancies (a common defect in this class of materials [20], cf. Ref. [21] that found 5% of vacancies in their RuO_2 samples, Ref. [22] reporting 5.3% of vacancies, and Ref. [23] reporting 1% of vacancies) promotes the RuO_2 to a magnetic state of exactly the same symmetry as suggested in Ref. [11] and utilized in Refs. [24,25].

The amount of Ru vacancies is liable to vary from sample to sample, and even from one batch to another, depending on the growth procedure, and may even be nonuniform over a sample. This could explain the discrepancy between different experiments and leads us to conclude that a characterization of the Ru vacancies in the samples may be key to knowing about the magnetic character of RuO_2.

II. RESULTS

A. Stoichiometric RuO_2

The first concern to cover is whether stoichiometric RuO_2 is magnetic or not. To account for the possible effects of electronic correlations in this system, we performed DFT + U computations. In Fig. 2, we plot the dependency of the local magnetic moment at the Ru site for an antiparallel spin orientation (a parallel orientation, as well as various magnetic arrangements with $q \neq 0$, are invariably higher in energy) over a range of U values. Two sets of calculations were performed to mitigate the problem of multiple local minima inherent to DFT + U: one starting from $U = 0$ eV and progressively increasing U in each subsequent calculation and the other with decreasing U (the former calculation is trapped in the nonmagnetic state for the displayed range of U values). As seen from the overlap of the magnetic moment for small values of U in the plot, RuO_2 is nonmagnetic up to a critical value, $U_{\text{eff}} = U - J \sim 1.06$ eV. However, in Fig. 2 at $U_{\text{eff}} \sim 1.23$ eV the altermagnetic state becomes lower in energy than the nonmagnetic state: it is metastable between 1.06 and 1.23 eV. There is a discontinuous jump (see below the explanation) of the value of the magnetic moment to ∼0.5 μ_B. This jump is an order of magnitude larger than the 0.05 μ_B obtained from polarized neutron scattering measurements [11] and more than twice larger than the 0.23 μ_B value fitted to unpolarized data (claimed to be less reliable and contaminated by unknown structural factors). The next issue is to focus on the magnetic ground state when the system is in the regime

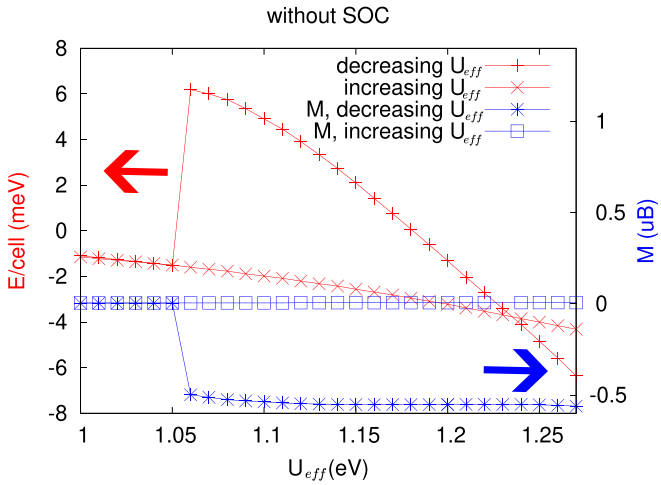

FIG. 2. Total energy (left) and local magnetization at the Ru site (right) as a function of $U_{\text{eff}} = U - J$ are explored in two sets of calculations. In one set ("increasing U_{eff}"), denoted with $\times$ (energy results) and $\square$ symbols (magnetization results) the calculations were done starting from $U = 0$ eV and progressively increasing U in each subsequent calculation. In the other set ("decreasing U_{eff}"), denoted with $+$ and $*$ symbols, the direction of the calculation was reversed. The calculations are without SOC contributions.

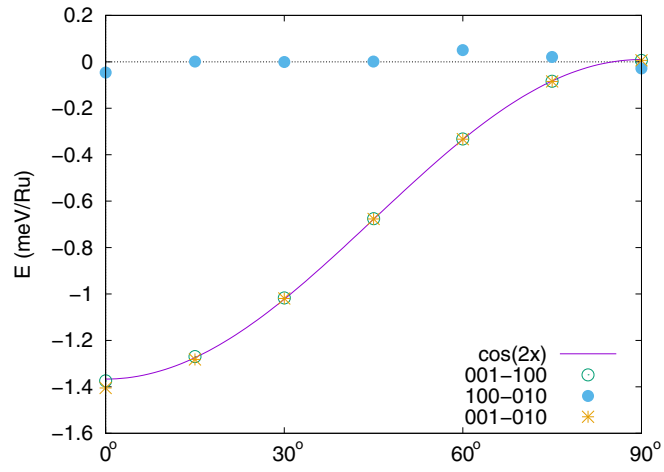

FIG. 3. Magnetic anisotropy as a function of the angle in degrees, when rotating the Neel vector in the indicated planes.

where magnetization is permitted. To address this question, we initially calculated, using VASP [26–29], the energy difference between the ferromagnetic (FM) and altermagnetic (AM) stoichiometric RuO_2 configurations. The calculations were done without considering spin-orbit-coupling (SOC) effects while setting the value of U_{eff} to 1.3 and 1.4 eV and we analyzed the magnetization of both configurations. Interestingly, the AM configuration converged to $M_{\text{Ru}} = 0.66$ and 0.78 μ_B for each Ru atom in the cell, respectively. In contrast, the FM ones essentially collapsed, yielding a total magnetization of $M_{\text{tot}} = 0.015$ (0.038) μ_B per Ru atom. Correspondingly, the AM energy was lower than the FM one by 3.3 (9.3) meV/Ru. Altogether, these results indicate that the altermagnetic configuration has the lowest energy. Besides that, the spin spirals with $\vec{q} = (0, 0, q)$ and $\vec{q} = (q, 0, 0)$ were checked, leading to $q = 0$ as the lowest-energy state in both cases.

We also checked the calculated magnetic anisotropy and compared it with the experiment. Including spin-orbit coupling, we found that the c axis is the easy axis along the direction, in agreement with experiment [11], as seen in Fig. 3.

Thus, the magnetic ground state is characterized by an antiparallel alignment along the c axis of the magnetic moments of two Ru atoms. This state can be described by the magnetic space group $P4'_2/mnm'$ (BNS 136.499).

However, $U_{\text{eff}} > 1$ eV is rather large for this good metallic, strongly hybridized, 4d system. For comparison, first-principles calculations of U_{eff} for the ruthenium-based spin-orbit Mott insulators α-$RuCl_3$, $RuBr_3$, and RuI_3 gave estimates of 2 to 1 eV [30]. Considering the metallic screening occurring in RuO_2, it is expected that its U_{eff} will be noticeably smaller than the values given above. This leads us to conclude that for stoichiometric RuO_2 a smaller U_{eff} is likely more realistic to describe its properties than the required one to have magnetism, and therefore, stoichiometric RuO_2 is most probably nonmagnetic.

B. Density of states

Moving on to analyzing the projected density of states (DOS) (see Fig. 4), we observe that the main contribution to the DOS around the Fermi level comes from the xz and yz Ru d orbitals. The value of the DOS at the Fermi level is relatively low and flat in its vicinity. This causes the Stoner criterion for ferromagnetism to be very hard to fulfill.

Zone-center antiferromagnetism, as in RuO_2, obeys a modified Stoner criterion, where, instead of the uniform susceptibility, $\chi(\mathbf{q} = 0) = N(0)$ at some finite reciprocal lattice vectors appears, $\chi(\mathbf{G} \neq 0)$, but it is quite obvious that highly dispersive bands at the Fermi level and low DOS are rather unfavorable in this case as well.

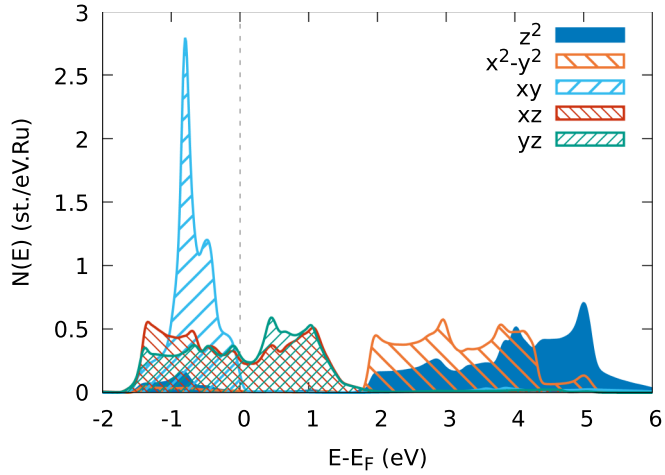

FIG. 4. Projected nonmagnetic density of states onto Ru d orbitals: blue, orange, cyan, red, and teal are used to depict z^2, x^2-y^2, xy, xz, and yz orbitals, respectively. The coordinate system is aligned with Ru-O bonds.

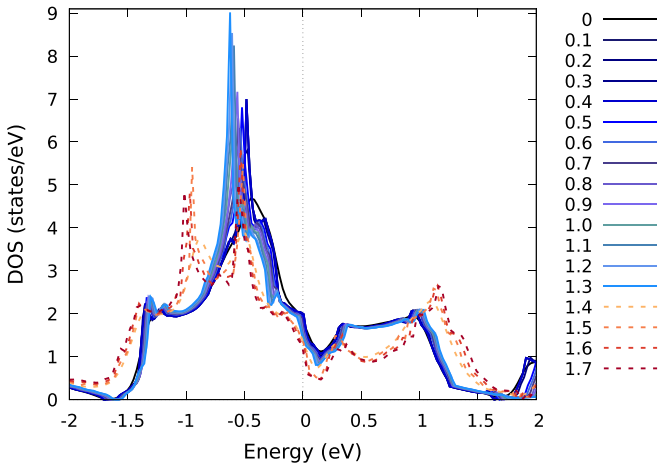

FIG. 5. Spin-polarized total density of states for majority spin as a function of U_{eff} (in eV). The color corresponds to various values of U_{eff}, where solid lines in blue shades correspond to the nonmagnetic ground state and dashed lines in red shades correspond to the AM order. Note that here the calculations were done using WIEN2K; thus, the value of U_{eff} is not directly comparable with other calculations where VASP was employed due to the differences in implementations.

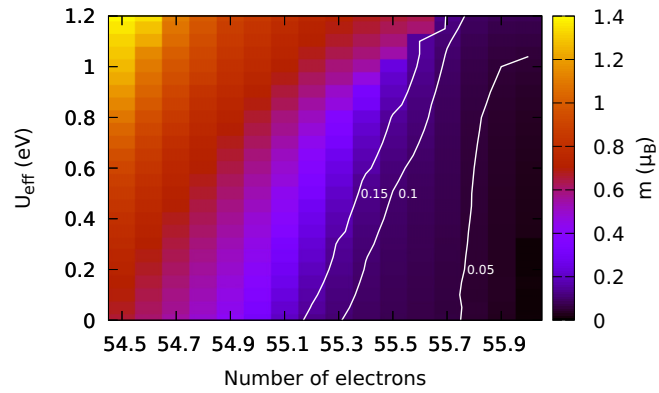

FIG. 6. Dependence of the local atomic magnetization m on hole doping (an undoped case corresponds to 56 electrons per cell, i.e., per two formula units) and the effective Hubbard parameter $U_{\text{eff}} = U - J$. Isolines for $m = 0.05, 0.10$, and $0.15\ \mu_B$ are depicted by white lines (with no attempts to smooth the plotted lines).

Another interesting aspect in the DOS is the narrow peak below the Fermi level coming from the xy Ru orbitals. If the Fermi level were shifted closer to this peak, it could potentially trigger a magnetic transition from the current nonmagnetic state to a state with a significant magnetic moment.

Overall, the electronic structure presented here is consistent with previously reported studies [11,31].

In Fig. 5 we present the dependency of the total density of states for spin-polarized calculation on the value of U_{eff}. For high values of U_{eff}, the xy band is split but not polarized. That is to say, magnetic instability here is achieved not due to increased DOS, as in the below-discussed case of hole doping, but due to enhancement of the effective Stoner factor. Indeed, as discussed in Ref. [32], in the first approximation DFT + U (in its Fully Localized Limit (FLL) flavor, used in this and other papers) is equivalent to renormalizing the DFT Stoner parameter as $I_{\text{eff}} = I_0 + \frac{U-J}{n}$, where I_0 is the DFT Stoner parameter and n is the number of orbitals at the Fermi level [32]. For the $4d$ metals, typically, $I_0 \sim 0.5$ eV. The density of states for RuO_2 at the Fermi level is $D_\uparrow(E_f) = 0.7$ (st/eV spin), thus $D_\uparrow(Ef) \cdot I_0 = 0.35 < 1$, so the material is stable against the formation of a ferromagnetic state. As a rough estimate of the effect of U, we take $n = 1$, and for $U_{\text{eff}} = 1.4$ eV we get $I_{\text{eff}} \approx 1.9$ eV, thus fulfilling the Stoner criterion; for $n = 2$ it is just a bit below instability. Of course, these are just order-of-magnitude estimates, since in reality we consider instability against antiferromagnetism, not ferromagnetism, and should instead of $D(E_F)$ use the (unknown) unrenormalized spin susceptibility at the corresponding wave vector, but it gives us the general feeling of how DFT + U generates magnetism without using the xy orbital.

C. Nonstoichiometric RuO_2

The shift of the Fermi level can be accomplished through hole doping, changing the electron occupation. The only other alternative for generating a magnetic order *without doping* is to increase U, and thus the effective Stoner parameter $I_{\text{eff}} = I + (U - J)/5$ (see Ref. [32]) until the Stoner criterion ($I_{\text{eff}} > J$) is satisfied. To check this hypothesis we did a series of DFT + U calculations varying both the value of U_{eff} and the number of electrons. Figure 6 shows the value of the local magnetic moment at the Ru site as a function of U_{eff} and the number of electrons per unit cell (two formula units). The isoline with $m = 0.05\ \mu_B$, corresponding to the measured value from Ref. [11], is highlighted. One can see that there is a rather stable ground state for ~0.1 hole/Ru doping, within a reasonable range of $U_{\text{eff}} \lesssim 1$ eV. For the same U_{eff}, a larger doping of 0.4 hole/Ru, corresponding to 10% of Ru vacancies, generated a local magnetic moment of $m = 0.2\ \mu_B$. It is worth pointing out that the discontinuous jump in the calculated magnetic moment for the undoped compound as a function of U_{eff} is immediately understood from the DOS in Fig. 4. That is because, in order to get a stable magnetic solution, the exchange splitting (proportional to U_{eff}) must reach the threshold (around 1 eV, from Fig. 4) corresponding to the separation between the xy band and the Fermi level.

Note that the data in Fig. 6 merely show a trend of the system to attain a magnetic moment with hole doping, but the preferred magnetic orientation may depend on the doping as well. In particular, when SOC is accounted for, our DFT + U calculations with $U_{\text{eff}} = 1.4$ eV show that the magnetic anisotropy energy $E_{100} - E_{001}$ changes linearly with doping and there is a transition from the easy axis along the c direction towards an easy plane at around 0.2 hole/Ru. However, the full sampling of magnetic ground states as a function of U_{eff} and hole doping is out of the scope of this work.

III. DISCUSSION

As presented in the previous section, pristine DFT calculation characterizes stoichiometric RuO_2 as nonmagnetic. Accounting for correlations with DFT + U one can stabilize a nonmagnetic or magnetic state depending on the value of U_{eff}, with a magnetic state stabilized for $U_{\text{eff}} > 1.06$ eV. However, the stabilized magnetic state for high-U values is not without

an issue: the value of the local magnetic moment is way higher (~0.5 μ_B vs 0.05 μ_B) if compared with the only available value from neutron scattering measurements [11]. The report on DFT + DMFT study of this compound predicts larger magnetic moments than DFT + U calculations [31]. Besides that, we argue that the reasonable range of U_{eff} values is when $U_{\text{eff}} < 1$ eV, leading us to conclude that stoichiometric RuO_2 is nonmagnetic.

Unfortunately, the available pool of experimental data does not provide a unanimous answer about RuO_2 being magnetic or not. Clearly, more measurements are needed. For example, multiple experiments claim that RuO_2 is antiferromagnetic [10,11], but the transition temperature has never been detected. The temperature dependence of the local magnetic moment, if any, is unknown as well. Furthermore, some experiments detect small but finite magnetic moments, some detect no long-range magnetic order, and some indirectly imply a large magnetic moment of the order of 0.6–1 μ_B.

In this article, we want to bring the attention of the scientific community to another missing piece of the puzzle: a characterization of RuO_2 stoichiometry. Our DFT calculations show that hole-doped RuO_2 can be magnetic. This finding, if confirmed experimentally, may be used to reconcile contradicting measurements: they are simply measuring different objects.

IV. CONCLUSIONS

Our DFT calculations show that, for a realistic value of U_{eff} (using Ru-based insulators as reference), the stoichiometric RuO_2 compound is nonmagnetic. However, hole doping due to Ru vacancies can induce a phase transition to the antiferromagnetic phase even for small values of U_{eff}. This observation may be a key to reconciling different, strongly mutually contradicting experiments. If our conjecture is correct, every experimental work on RuO_2 must begin with a careful characterization of the O and Ru content. Moreover, a systematic experimental investigation of magnetic properties as a function of the O and Ru content is absolutely necessary. One verifiable corollary is that, with controllable Ru vacancies, one should be able to observe the antiferromagnetic transition in thermodynamics, transport, and magnetometry.

V. COMPUTATIONAL DETAILS

Computations were done using density functional theory in the generalized gradient approximation with the Perdew-Burke-Ernzerhof [33,34] functional as implemented in the VASP [26–29] package employing the projector augmented wave method (PAW) [35,36]; Ru_sv and O (or O_h, for structural optimization) pseudopotentials were used. The energy cutoff was set to 400 eV (for the test purpose, selected calculations were performed with a 900-eV cutoff) and an $8 \times 8 \times 12$ ($12 \times 12 \times 18$ for testing) k-point Monkhorst-Pack grid [37,38] was used. The hole doping was achieved by varying the number of electrons (NELECT flag in VASP) and the calculations were done using a pristine rutile structure with $a = 4.480$ Å, $c = 3.105$ Å, Ru at the $2a$ Wyckoff position, and O at the $4f$ position with $x = 0.304\,79$ [39]. For selected calculations, a cross-check using WIEN2K was used. WIEN2K was also used to compute the total and projected density of states.

Note added. Recently, we became aware of an experimental work investigating the muon spin relaxation in bulk RuO_2, where it was reported that no static magnetic order had been observed in the temperature range 5–400 K [40].

The data needed to reproduce and verify the results presented in this article are publicly available on the TU Wien Research Data Repository [41].

ACKNOWLEDGMENTS

We thank Libor Šmejkal, Huibo Cao, Dmitry Khalyavin, Jan Kuneš, and Robert Svagera for the discussions. A.S. was supported by the Austrian Science Fund (FWF) through Project No. P33571 "BandITT." L.G-G. and R.V. were supported by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) for funding through TRR 288-422213477 (Project No. B05). I.M. was supported by the Army Research Office under Cooperative Agreement No. W911NF- 22-2-0173. He also acknowledges the Heraeus Foundation for supporting his visits to University of Frankfurt. Some of the images in the paper were created using VESTA software [42].

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