Suppressed fluctuation as the origin of the static order in strained \( \text{Sr}_2\text{RuO}_4 \)

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Combining first principle density functional calculations and Moriya’s self-consistent renormalization theory, we explain the recently reported counterintuitive appearance of an ordered magnetic state in uniaxially strained \( \text{Sr}_2\text{RuO}_4 \) beyond the Lifshits transition. We show that strain weakens the quantum spin fluctuations, which destroy the static order, more strongly than the tendency to magnetism. A different rate of decrease of the spin fluctuations versus magnetic stabilization energy promotes the onset of a static magnetic order beyond a critical strain.

Introduction. After many years of pursuing triplet superconductivity in \( \text{Sr}_2\text{RuO}_4 \), recent studies provide evidence in favor of a singlet state \(^1\)\(^2\). However, while most theorists believe that spin fluctuations are important for superconductivity, there is no consensus on the detailed mechanism or the pairing symmetry. Some of the proposals include \( d_{x^2-y^2} \) \(^3\), \( d_{xy} \) \(^4\), \( d_{xz} + id_{yz} \) \(^5\), or \( g_{xy}(x^2-y^2) \) \(^6\). Experiments under uniaxial strain have been instrumental in the quest for elucidating the pairing symmetry \(^7\)\(^8\). One of the most impressive pieces of evidence was the observation of a maximum in \( T_c \) at the critical strain corresponding to the Lifshitz transition \(^8\)\(^9\). The latest addition in this direction is establishing an ordered static magnetic state under strain beyond the Lifshitz transition \(^10\).

This result is counterintuitive because the magnetism is nearly universally suppressed by pressure as the atomic distances become smaller, and enhanced itinerancy eventually wins over the spin exchange splitting. Hence, understanding this paradox may bring up new and novel progress in understanding the physics of this compound, which has a nontrivial magnetic energy landscape \(^11\). The emergence of magnetism under pressure, as stated, is very uncommon and usually associated with materials close to the itinerant magnetism, where the long-range order is suppressed by the fluctuations (cf. Fe-based superconductors, Ref. \(^12\)).

Such anomalous pressure effect on magnetism can be understood within Moriya’s self-consistent renormalization (SCR) theory \(^13\). It stipulates that the magnetization in itinerant magnets is soft and fluctuates in amplitude. Assuming Gaussian fluctuations with the mean square amplitude, \( \xi \), it was shown that if the total energy is expanded with the magnetic order parameter \( M \) as

\[
E = a + bM^2 + cM^4 + dM^6 + ..., \quad (1)
\]

then, according to the fluctuation-renormalized expansion, the corresponding coefficients change as

\[
\hat{b} = b + \frac{5}{3}d\xi^2 + \frac{35}{9}d\xi^4 + ... \\
\hat{c} = c + \frac{14}{3}d\xi^2 + ... \quad (2)
\]

and so on. Obviously, this increases the value of the inverse spin susceptibility, \( 2\hbar \), and its sign changes from negative to positive for \( \xi^2 > \frac{3b}{5c} \). Hence, the spin fluctuation in an itinerant magnet can lead to \( (i) \) the reduction of spin susceptibility in paramagnetic materials, \( (ii) \) the decrease of the average magnetic moment, or \( (iii) \) the suppression of the long-range magnetic order in a system where mean-field theories, such as density functional theory (DFT), predicts to be an ordered magnet.

SCR theory has been successfully applied to various (near-)ferromagnetic materials, such as \( \text{Pd} \) \(^15\), \( \text{Ni}_3\text{Ga} \) and \( \text{Ni}_3\text{Al} \) \(^16\), \( \text{ZrZn}_2 \) \(^17\), and notable other systems where the SCR theory is somewhat more straightforward, but it was also called up in connection with Fe-based superconductors \(^14\)\(^18\). Moriya, in his book \(^13\), emphasizes that, while the entire frequency and momentum dependence of spin susceptibility, via the fluctuation-dissipation theorem, determines the value of \( \xi \), a significant role is played by the phase space, \( i.e., \) the fraction of the Brillouin zone (BZ) that is close to a magnetic instability.

This point was clearly demonstrated in Ref. \(^19\), where \( \xi \) was estimated from first principle calculations for two similar compounds, \( \text{Ni}_3\text{Al} \) and \( \text{Ni}_3\text{Ga} \). Within DFT, both systems are ferromagnetic, but, in reality, only the former is, while \( \text{Ni}_3\text{Ga} \) is a strongly renormalized paramagnet. Counterintuitively, the calculated magnetic moment and magnetic stabilization energy were higher in \( \text{Ni}_3\text{Ga} \). This apparent paradox was resolved in Ref. \(^14\) by the fact that the instability in \( \text{Ni}_3\text{Ga} \), while stronger, is also considerably less localized in the momentum space (the characteristic volume of the unstable part of the BZ is more than twice larger), which leads to a larger \( \xi \),...
and a stronger suppression. As a result, from the view of mean-field theory, the magnetic instability is entirely suppressed in the more ferromagnetic material, while in the less one, the magnetism survives \[16\].

In this Letter, we argue that \(\text{Sr}_2\text{RuO}_4\) represents a similar case with the spin-density wave (SDW) type instability: The tendency toward SDW-antiferromagnetism is stronger for the unstrained material, but the static order is not established due to the even more substantial spin fluctuations. We show that when uniaxial stress is applied, the fluctuations are suppressed more strongly than the tendency towards magnetism which causes the emergence of a static magnetic phase.

Methods. We employed the Vienna \textit{ab initio} simulation package (VASP) \[19, 20\] within the Generalized gradient approximation (GGA) by Perdew-Burke-Ernzerhof functional \[21\]. The energy cut for the plane waves of 600 eV was used with a Monkhorst-Pack \(k\)-mesh of \(17 \times 17 \times 10\) for the primitive unit cell. For the spin spiral calculations with various \(q\)-values, we employed the generalized Bloch theorem\[22\], allowing spiral calculations for an arbitrary wave vector without using the supercells. We have further used a very stringent convergence criteria of \(10^{-7}\) eV in most cases.

To obtain a series of strained structures, we fixed the \(a\) lattice parameter and fully optimized \(b\) and \(c\) parameters and the internal positions. Due to the well-known fact that GGA overestimates the equilibrium volume at a given pressure, the critical stress (but not the critical strain) in our calculations is likely overestimated (we get the Lifshitz transition at \(\sigma = 1.5\) GPa, see Fig. 1 about twice the value estimated in Ref. \[10\]), but the important part is that we trace the evolution of magnetic properties well past this transition.

Results. \(\text{Sr}_2\text{RuO}_4\) does not order magnetically down to low temperatures; however, it was predicted from DFT calculations\[23\] and later confirmed by the neutron diffraction that it features strong spin fluctuations with the wave vector \(q \sim (0.3,0.3,0)\) \[24, 25\] (here and below, we give the wave vectors in the orthorhombic reciprocal lattice units, \(i.e., 2\pi/a\)). This SDW feature, from the nesting of 1D \(\alpha\) and \(\beta\) Fermi surfaces, can be well-captured from the DFT approach despite the overestimating tendency toward magnetism \[27\]. In our case, we obtained \(q_{SDW} = (0.29,0.29,0)\) as the ground state for the unstrained case, which gives excellent agreement with the neutron diffraction study (from now on, we will omit \(q_0 = 0\) for brevity).

First, to insist the correlation between the leading magnetic instability and uniaxial pressure in \(\text{Sr}_2\text{RuO}_4\), we have calculated the total energy and Ru local magnetic moment for representative magnetic orderings in a wide range of stress levels and pressures. In addition to the \(q_{SDW}\) phase, we have considered the \(q_{SDW} = (1/2,1/4)\) order suggested by Roemer \textit{et al.}\[26\] and the checkerboard ordering \(q = (1/2,1/2)\), as well as \(q = (1/2,0)\) and \((0,1/2)\) configurations \[27\], based on the observation that only the first nearest neighbor exchange responds to uniaxial stress \[27\]. In Fig. 1, we display the energy difference of each configuration with the non-magnetic one \((\Delta E)\) and Ru local moment for uniaxially strained \(\text{Sr}_2\text{RuO}_4\). We find that the SDW phase is the most stable state up to 3.6 GPa. As shown in Fig. 1(c), the van Hove singularity crosses the Fermi energy at around 1.5 GPa, and the SDW phase remains the ground state well beyond that. Uniaxial strain breaks the \(C_4\) symmetry and bifurcates the first nearest neighbor exchange interactions between Ru ions \[27\]. Hence, \(q = (1/4,1/2)\) and \(q = (1/2,1/4)\) are progressively split in energy. We see that at the highly-strained limit of 5.7 GPa, \(q = (1/2,1/4)\) is lower than \(q_{SDW}\) in energy, which, however, does not indicate the \(q = (1/2,1/4)\) is the ground state, as discussed in detail later.

Interestingly, the size of the Ru moment strongly varies for different \(q\) values. The \(q_{SDW}\) moment is the largest for all studied ranges, while the one for \(q = (1/2,0)\) is negligibly small. The sizes of the moment for \(q_0 = (1/4,1/2)\) and \(q = (1/2,1/4)\) also bifurcate, and, at 3.6 GPa, the moment suddenly vanishes for \(q = (1/4,1/2)\).

For the unstrained case, while strong \(q_{SDW}\) tendency exists, the long-range magnetic order does not set in due to the strong spin-fluctuations. As the system undergoes uniaxial strain, \(\mu\text{SR}\) experiments have found the stabilization of a magnetic order beyond the Lifshitz transition point \[19\]. However, the actual magnetic pat-
tions. For materials close to ferromagnetism, first consider the case beyond the Lifshitz transition, and 5.7 GPa to the highly strained limit. Here, the grid calculation is performed with varying $q_x$ and $q_y$ values with the interval of $\Delta q = 0.02 \pi / a$ at $q_z = 0$. In (b), the nesting vector positions are marked. (c) The corresponding Fermi surface plots. The diagonal dashed arrows indicate the nesting vectors for unstrained and 5.7 GPa cases.

Our calculations, consistently with $\mu$SR, show that the DFT (that is to say, mean field) ground state is always an incommensurate SDW, even though its wave vector slowly shifts from $(0.29, 0.29)$ toward $q = (1/2, 0, 0)$ [Fig. 2(b)]. As expected, while SDW is always stable, at the mean field level, the stabilization energy [Fig. 2(b)] and the equilibrium magnetic moment [Fig. 2(a)] are considerably reduced by the strain.

The next step is to estimate the effect of the fluctuations. For materials close to ferromagnetism, first principle calculations of the $\xi$ parameter (Eq. [2]) are tedious but feasible. For those close to antiferromagnetism, we do not have a similar recipe. Therefore, we have adapted “a poor man’s” approach, estimating $\xi^2$ as $\langle M^2(q) \rangle$, where the averaging is over the entire Brillouin zone. While this estimate may be quantitatively inexact, it gives the correct trends upon increasing the strain.

To this end, we have calculated, for three stresses of 0.0 GPa, 2.3 GPa, and 5.7 GPa, the moments and magnetic stabilization energies on a full 2D-grid in $q_x$ and $q_y$, while keeping $q_z = 0$. The results are shown in Fig. 2 (a) and (b). For the unstrained case, a large moment ($> 0.8 \mu_B$) can be found in a broad region centered at $q_{SDW} = (0.29, 0.29)$. The size of the moment strongly depends on $q$, and almost vanishes close to $q = (1/2, 0)$ and $q = (0, 1/2)$. The large $q$ range where a strong instability occurs demonstrates the highly fluctuating nature of the spin moment in Sr$_2$RuO$_4$. As the uniaxial stress is imposed, the lowest energy position is gradually moved asymmetrically to $q = (0.32, 0.24)$ for 2.3 GPa, and $q = (0.34, 0.20)$ for 5.7 GPa (See Fig. 2(b)). According to our calculation, the magnetic instability is of SDW-type, not the commensurate one suggested in Ref. [20] and not checkerboard $q = (1/2, 1/2)$. While there is an apparent tendency of the ground-state $q$ to shift from the original SDW one, $q_{SDW} = (0.29, 0.29)$, toward $q = (1/2, 1/4)$, the magnetism of the system remains incommensurate. Our calculations show that, while proximity to an SDW instability in unstrained Sr$_2$RuO$_4$ is definitely related to nesting [17], the exact position is not given by the nesting vector (which is related to the imaginary, not the real part of susceptibility [28]), and in fact the evolution of the SDW vector with strain cannot be attributed to the evolution of nesting (Fig. 2).

Estimating $\xi^2 \approx \langle M^2(q) \rangle$, we obtain for 0.0 GPa, 2.3 GPa and 5.7 GPa stress, respectively, 0.43, 0.36, and 0.22 $\mu_B^2$, demonstrating the progressive suppression of the spin fluctuations. The apparent shrinking of the area where the moment survives (Fig. 2(a)) demonstrates the dramatic reduction of the phase space available for fluctuations under strain.

While the leading instability has, of course, its origin in the momentum space, it is instructive to look at it also from the real space point of view. As discussed in our previous paper [27], the Fermi-surface driven instability, when mapped onto the nearest-neighbor Heisenberg Hamiltonian, results in three sizeable exchange parameters, corresponding to the ferromagnetic coupling along the 110 bond, $J_{110}$ and two antiferromagnetic couplings,
along the 100 and 200 bonds, so that $|J_{200}| > |J_{110}| > |J_{100}|$. Note that single $J_{100}$ generate a $q = (1/2, 0, 0)$ order, and $J_{200}$ a $q = (1/4, 0, 0)$ order, etc. In Ref. [27] we estimated, using the real-space Disordered Local Moments method, the effect of uniaxial strain (without reoptimizing the atomic position) on the parameters $J$s, and found that only $J_{100}$ is affected, by splitting into different $J_{100}$ and $J_{010}$. For sufficiently large strain, $J_{100}$ dictates the antiferromagnetic order along $x$, that is, $q_x = 1/2$, while (still the largest) $J_{200}$ forces $q_y$ to get close to 1/4, thus promoting the $(1/2, 1/4)$ order. However, as discussed above, direct calculations show that this limit is never achieved in the considered stress range, albeit the leading instability shifts from $(1/3, 1/3, 0)$ to this general direction. Thus, the evolution of the instability $q$ vector can be roughly described as the competition between the Fermi surface driven instability and the nearest neighbor superexchange.

We can illustrate the physics of the recovery of magnetism under uniaxial strain on a simple toy model (Fig. 3). Let us assume that the energy of the SDW in the unstrained state in the mean field theory (MFT) is a simple quadratic polynomial of its amplitude: $E(m) = -m^2 + 2m^4$. The MFT ground state is then $m = 1/2$. If we take Gaussian spin fluctuations of $ξ = 0.6$, then, per Eq. [2] these will destabilize the static order and stabilize a nonmagnetic ground state. Let us now assume that under strain, the quadratic coefficient got reduced by a factor of two (a reduced tendency towards magnetism), $E_{\text{strain}}(m) = m^2/2 + 2m^4$. This would shift the MFT solution to $m = 1/2\sqrt{2}$, and the magnetic stabilization energy will be reduced from $1/8$ ($E(m = 1/2)$) to $1/32$ ($E_{\text{strain}}(m = 1/2\sqrt{2})$). If we now assume that $ξ$ has also been reduced by a factor of two, from 0.6 to 0.3, the SCR solution will still be magnetic, $m ≈ 0.42$, $E_{\text{strain}} ≈ −0.06$.

**Summary and discussions.** Our DFT calculations show that, as expected, the stress generally weakens the tendency toward magnetic order in Sr$_2$RuO$_4$. That is to say, the magnetic ground state is less stable in the uniaxially strained case than in the unstrained one on the mean-field level. However, at the same time, the propensity of magnetic order to be destroyed by quantum spin fluctuations becomes weaker and allows the establishment of long-range order in the strained system. The competition between the two effects can be understood as follows: The mean square amplitude of spin fluctuations $⟨m^4⟩$ decreases much faster with applied strain than static magnetic moment formation energy. In the parlance of Moriya’s SCR theory, the coefficient $b$ in Eq. [1] is negative, indicating the magnetic tendency, and becomes less negative with increasing the uniaxial strain. However, the strength of the spin fluctuation quantified by the parameter $ξ$, also decreases with strain and, apparently, varies faster than $b$. As a result, up to some critical stress, determined experimentally as $~0.8$ GPa, $b = b + 5/3ξ^2$ remains positive and becomes negative afterward.

A corollary of this picture is that under further straining, the ordered magnetism will be suppressed again due to the further reduction of the magnetic tendency.

It is also of note that the position of the MFT instability (or, equivalently, of the maximum in spin susceptibility) shifts with strain from its unstrained position of $q = (0.29, 0.29)$ toward $q = (1/2, 1/4)$, but remains strongly incommensurate, $q ≈ (0.34, 0.20)$, even at the strain of several GPa, and this evolution does not reflect the changes in nesting.

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