Intrinsic nature of spontaneous magnetic fields in superconductors with time-reversal symmetry breaking

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We present a systematic investigation of muon-stopping states in superconductors that reportedly exhibit spontaneous magnetic fields below their transition temperatures due to time-reversal symmetry breaking. These materials include elemental rhenium, several intermetallic systems and Sr_2RuO_4. We demonstrate that the presence of the muon leads to only a limited and relatively localized perturbation to the local crystal structure, while any small changes to the electronic structure occur several electron volts below the Fermi energy leading to only minimal changes in the charge density on ions close to the muon. Our results imply that the muon-induced perturbation alone is unlikely to lead to the observed spontaneous fields in these materials, whose origin is more likely intrinsic to the time-reversal symmetry broken superconducting state.

A crucial issue in resolving the mechanism for unconventional superconductivity is the presence or absence of time-reversal symmetry breaking (TRSB) [1], a property that can provide a tight constraint on the symmetry of the superconducting gap. The conventional s-wave singlet BCS pairing conserves time-reversal symmetry, but triplet pairing does not. For example, a p-wave gap symmetry was ascribed [2, 3] to the superconductor Sr_2RuO_4 on the basis of its supposed triplet order parameter deduced from NMR [4] and the presence of time-reversal symmetry breaking deduced from muon-spin relaxation (μSR) [5] and polar Kerr effect [6] measurements. However, the triplet nature of Sr_2RuO_4 has recently been discounted on the basis of a new NMR investigation [7], thereby reopening the question about the nature of the gap symmetry in this compound, with several alternative singlet gap structures proposed [8–11], each of which would be consistent with TRSB. There is also a recent suggestion that the experimental signature of TRSB is not intrinsic but originates from inhomogeneous strain fields near edge dislocations [12].

The nature of TRSB superconductivity, and the need to understand how it is detected, is a question with a much wider applicability than merely the particular case of Sr_2RuO_4. This is because the appearance of spontaneous magnetic fields is found in a large collection of superconductors using μSR measurements, though importantly it is absent for most superconductors (SCs) [1]. μSR has emerged as an effective probe of superconducting properties [13], extracting the penetration depth and hence the superfluid stiffness [14], examining vortex lattice melting [15, 16] and determining the nature of the pairing [17, 18]. The superconducting vortex lattice produced by an *applied field* is, in general, incommensurate with the crystalline lattice and so the precise location of the muon site makes no difference in these studies. Wherever the muon sits inside the unit cell, it will uniformly sample the magnetic field distribution produced by the vortex lattice [19]. This is not the case for muon studies of magnetism for which the local field extracted by μSR depends sensitively on the location of the muon site (see e.g. [20]). This issue becomes extremely relevant for superconductors studied in *zero applied magnetic field* where the signature of TRSB is the appearance of a very small spontaneous local field. There is currently no accepted theory which predicts how large the spontaneous field should be, to what extent these spontaneous fields should be screened by supercurrents, whether these fields are particularly associated with defects, interfaces and domain boundaries, or indeed whether the presence of the muon itself might play the role of a defect. Nevertheless, the results of μSR experiments have been used to argue for TRSB on the basis of spontaneous fields detected in a number of unconventional superconductors, including Sr_2RuO_4[5], $LaNiC_2$ [21], SrPtAs [22], Zr_3Ir [23, 24], Re [25], and Re_6Zr [26]. In this Letter, we critically reexamine these experiments by calculating the muon site in these candidate TRSB superconductors using density functional theory (DFT), to assess the degree to which the muon perturbs its local environment.

We first consider muon stopping sites in elemental rhenium, which is the material exhibiting spontaneous magnetic fields with the simplest crystal structure. Structural relaxations of a supercell containing a muon yield two crystallographically distinct muon sites and, as an illustration of the physics relevant to the more complicated materials discussed below, we first consider the site tetrahedrally coordinated by Re atoms. For each of the muon sites found in this study, we have calculated the defect formation energy [27, 28] in order to assess how favorable it is for each of these to be realized (see Table I). We note that, while the tetrahedral site in rhenium is 0.50 eV higher in energy than the other candidate site discussed below, it is possible that it is occupied nevertheless. Re

atoms in the coordination tetrahedron of the muon can be divided into two distinct environments [Fig. 1(a)], Re1 at the apex of the tetrahedron and Re2 forming the base of the tetrahedron. Although the presence of the muon distinguishes these different environments, this tetrahedron is not regular, even in the absence of the muon. The muon is not quite at the centre of mass of the coordination tetrahedron [which has fractional coordinates ($\frac{2}{3}$, $\frac{1}{3}$, $\frac{3}{8}$)] but is instead slightly closer to Re1, which results in a larger muon-induced displacement of this Re atom than for the others in the tetrahedron [Fig. 1(b)]. As is also shown in Fig. 1(b), the displacements of the nearest-neighbour atoms due to the muon are small: 0.15 Å for Re1 and 0.09 Å for Re2.

To investigate the possible effects of the muon on the electronic structure of the system, we computed the density of states (DOS) for Re1 and Re2 for the pristine structure and for the structure with a muon at the tetrahedral site, and we show these in Figs. 1(c) and 1(d), respectively. The projected density of states (PDOS) of Re1 and Re2 show some small differences due to the symmetry-breaking effect of the implanted muon. However, the changes in the PDOS compared to the pristine system are very minor, particularly in the vicinity of the Fermi energy, where the DOS would have a significant impact on the electronic properties of the system. The PDOS of the muon has the form of a localized state lying around 10 eV below the Fermi energy and this defect state is therefore unlikely to affect the electronic properties of the system.

In the lowest-energy site in Re, shown in Fig. S1 in the Supplemental Material (SM) [29], the muon is octahedrally coordinated by Re atoms, with Re–μ^+ distances of

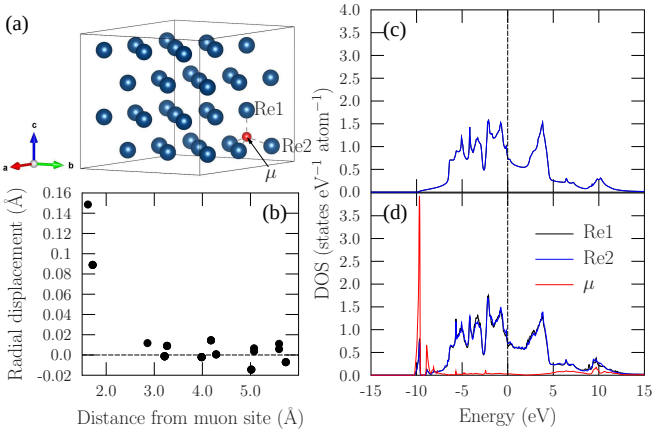

FIG. 1. Tetrahedral muon site in rhenium. (a) The two distinct nearest-neighbour Re environments, Re1 and Re2, in the coordination tetrahedron of the muon. (b) Radial displacements of the Re atoms as a function of their distances from the muon. PDOS for the nearest-neighbor Re atoms (c) without a muon and (d) with a muon. Energies are given with respect to the Fermi energy.

Material	H_f (eV)	Fractional coordinates	σ_{VV} (MHz)	σ_n (MHz)
Re	−2.16	0,0,0	0.372	0.314 [25]
	−1.67	$\frac{2}{3}$, $\frac{1}{3}$, 0.388	0.420	
Sr_2RuO_4	−2.01	0.225, 0.0, 0.184	0.048	0.02, 0.06 [5]
$LaNiC_2$	−2.59	0.004, 0.498, 0.112	0.120	0.08 [21]
SrPtAs	−2.21	0.333, 0.662, 0.034	0.073	0.12 [22]
	−1.92	0.009, 0.207, 0.245	0.086	
Zr_3Ir	−3.06	0.001, 0.000, 0.500	0.039	0.15 [24]
	−2.72	0.000, 0.000, 0.998	0.036	
	−2.44	0.285, 0.106, 0.223	0.033	
	−2.23	0.573, 0.007, 0.173	0.034	
Re_6Zr	−4.98	0.122, 0.120, 0.004	0.336	0.256 [26]
	−4.92	0.449, 0.001, 0.006	0.338	
	−4.61	0.504, 0.253, 0.001	0.379	

TABLE I. Crystallographically distinct muon stopping sites obtained from structural relaxations and their defect formation energies H_f. Fractional coordinates are given for the conventional cell. We also show the Van Vleck second moments σ_{VV} computed for each of the sites (calculated in the limit of strong quadrupolar splitting) and compare these with the measured relaxation rates due to nuclear moments σ_n.

2.0 Å. The displacements due to this site are small, with Re atoms in the coordination octahedron each being displaced by around 0.04 Å away from the muon. These displacements are significantly smaller than those associated with the tetrahedral site. This is likely due to the smaller space available for the muon in a tetrahedral vacancy as compared to an octahedral case, which might also explain the higher total energy of this site. We also do not observe any significant changes to the electronic structure due to the implanted muon in this case. To compare the computed sites to the measured spectra [25] we computed the expected relaxation rate for each distinct muon stopping site using the Van Vleck second moment in the limit of strong quadrupolar splitting [30]. The calculated relaxation rates are reported in the final column of Table I and take into account the repulsion of the nearby Re atoms by the muon. We see that both sites give rise to a relaxation rate that is slightly higher than, though broadly consistent with, the value $\sigma = 0.314$ MHz observed experimentally.

We now turn to the the layered perovskite superconductor Sr_2RuO_4. In the lowest energy muon site, the muon is bonded to an oxygen (O2) with bond distance 0.973 Å, as shown in Fig. 2(a). This is consistent with muon sites in other oxides including high-temperature superconducting cuprates [31, 32] and pyrochlores [33], where the muon stops ≈1 Å from an O anion. The radial displacements of the ions due to the implanted muon are shown in Fig. 2(b) as a function of their distances from the muon site. The most significant displacements are experienced by the Sr and O1 atoms nearest the muon [labelled in Fig. 2(a)], with magnitudes in the range of 0.16 to 0.21 Å. The O2 that forms a bond with the

muon is repelled by 0.06 Å away from the muon, while the nearest Ru atom [Ru–μ distance of 2.54 Å, also indicated in Fig. 2(a)] experiences a radial displacement of 0.04 Å. For all species, the muon-induced displacement vanishes rapidly as a function of distance from the muon site, such that significant distortions are observed only for atoms within 6 Å of the muon site.

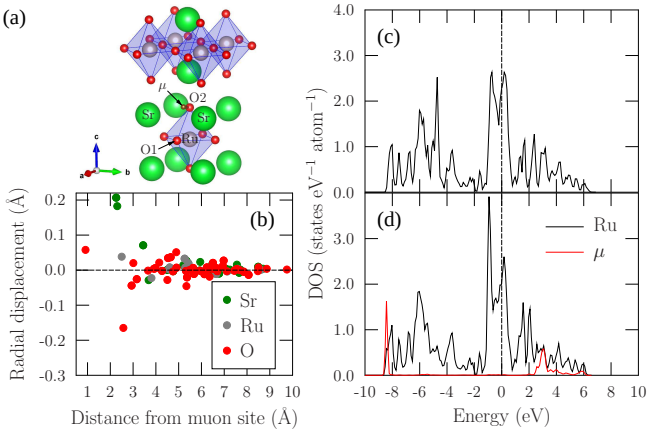

FIG. 2. The lowest energy muon site in Sr_2RuO_4. (a) The local geometry of the muon site. (b) Radial displacements of atoms as a function of their distances from the muon site. PDOS for the Ru atom closest to the muon site for the structures (c) without a muon and (d) with a muon. Energies are given with respect to the Fermi energy.

The dominant contribution to the DOS close to the Fermi energy is that from the Ru atoms [29]. The effect of muon implantation on the PDOS of Ru atom closest to the muon site is shown in Figs. 2(c-d). There is a significant increase in the DOS at around 1 eV below the Fermi energy caused by small changes in the splitting of the Ru $4d_{zy}$ and $4d_{zx}$ states at the Fermi level, which are not observed for Ru atoms further away from muon. After summing the d-state contributions from all of the Ru ions in the supercell, the small state splitting is no longer resolvable. Similar to Re, the PDOS corresponding to the muon lies well below the Fermi energy (around 8 eV below in this case). These results suggest that the implanted muon does not have a significant effect on the local electronic structure of Sr_2RuO_4.

Muon sites obtained for other materials in this study are summarized in Table I and we show the radial displacements of atoms as a function of their distances from the muon for the lowest-energy site in Fig. 3. We summarize the important features of the muon sites in each of these systems below. (See the Supplemental Material [29] for further details.)

For the lowest energy muon site in $LaNiC_2$, the muon is triangularly-coordinated by three La atoms in the bc plane and sits between two Ni atoms along the a axis, with two approximately equal Ni–μ^+ distances of $\approx$ 1.86 Å. The Ni atoms are displaced by around 0.11 Å

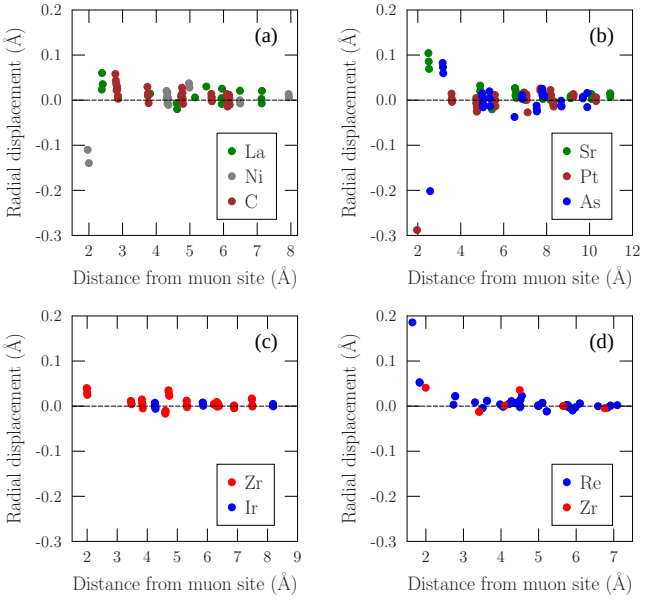

FIG. 3. Radial displacements of atoms as a function of their distances from the muon site for the lowest energy muon sites in (a) $LaNiC_2$, (b) SrPtAs, (c) Zr_3Ir, and (d) Re_6Zr.

and 0.14 Å towards the muon, as shown in Fig. 3(a). The La atoms are displaced radially outwards, though by a smaller distances (between 0.02 Å and 0.06 Å). The nuclear relaxation rate computed for the site is very similar to the value $\sigma = 0.08$ MHz obtained experimentally [21].

For SrPtAs, structural relaxations result in three distinct symmetry-inequivalent muon stopping sites. The energy difference of 0.29 eV between the two lowest energy sites (shown in Table I) is not large enough to rule out all but the lowest energy site and we therefore consider both of these sites as candidate muon sites, and refer to these as site 1 and site 2. Both sites make shorter bond distances with Pt than with Sr and As atoms. As seen in Fig. 3(b), for site 1, the Pt atom nearest the muon experiences the largest displacement, with magnitude 0.3 Å. For site 2, the displacement of the nearest Pt atom is much smaller ($\approx$ 0.12 Å), but significant displacements persist to larger distances away from the muon site [29].

For Zr_3Ir, structural relaxations result in 10 distinct muon sites, which we cluster into four groups by considering the proximity of their positions within the unit cell, and we include a representative member of each group in Table I. In the lowest energy site, the muon is tetrahedrally coordinated by Zr atom, with Zr–μ bond lengths of 2.03 Å. For this site, the muon does not introduce any significant distortions to its host, with all displacements being 0.04 Å or smaller, as seen in Fig. 3(c). In fact, for all sites in Zr_3Ir, the maximum displacement remains below 0.1 Å.

For the Re-Zr alloy Re_6Zr, obtaining the precise Re:Zr composition would require a prohibitively large supercell, so we instead considered compositions $Re_{49}Zr_9$ and

$Re_{50}Zr_8$ as close approximations. The distinct sites for $Re_{50}Zr_8$ are reported in Table I. For the lowest energy site, the muon is tetrahedrally coordinated by 3 Re atoms and 1 Zr atom, with Re–μ distances of 1.83 Å, 1.88 Å and 1.88 Å and a Zr–μ distance of 2.04 Å. The coordination tetrahedron of the muon in this site (and for other tetrahedral sites in this system) is therefore highly irregular, with this being the case even before the addition of a muon. As seen in Fig. 3(d), the displacement of each of the ions in the coordination tetrahedron reflects their proximity to the muon site; atoms that are closer to the muon are displaced by a greater amount, with the displacement of the Re atom closest to the muon being significantly larger than for any of the other atoms in the system.

There are no significant changes to the DOS in the vicinity of the Fermi energy as a result of implanting a muon in any of the materials studied. The significant contributions from muon PDOS typically lie between 6 and 10 eV below the Fermi energy, and the PDOS corresponding to the other species remains basically unchanged near the Fermi energy. While we often find evidence for some hybridization between the muon states and the states of ions in the system, the fact that these muon states lie so far below the Fermi energy means that this is unlikely to affect the electronic properties of the system, even locally. We have found that the electronic behavior of the muon in these systems is very similar to that in the conventional superconductor Nb [29], with no notable differences that could be responsible for muon-induced internal fields.

Another possible way in which the muon could perturb its host is by altering the charge states of nearby atoms. For Zr_3Ir, it was found that, in the vicinity of the muon site, the occupation of the d states and charges of the neighbouring Zr atoms change. This was particularly true for the lowest energy site, which has four nearest-neighbour Zr atoms. This is due to the redistribution of the charge on the atoms bonding with the muon, since the charge on the muon remains the same for calculations with muonium as it does for μ^+. In fact, the muon exists in a charge state resembling muonium in all of materials in this study, which is consistent with the fact that the muon PDOS lies well below the Fermi energy and hence these states would be expected to be occupied. In general, the overall (+1) charge of the unit cell is maintained by the having the charges on all of the other atoms in the host system become very slightly more positive, rather than through significant changes in the charge state of particular atoms.

We also carried out a series of spin-polarized calculations to investigate the possibility of muon-induced spin density which could, in principle, act as a source of a non-zero local magnetic field. For $LaNiC_2$, we found no appreciable spin density, both for the pristine structure and for the structure including an implanted muon ($< 0.01\hbar/2$ per atom according to a Mulliken analysis). Similar spin-polarized calculations on Zr_3Ir do not show the presence of spin density at the muon site. After muon implantation, the the small (0.0122 a.u.) spin density on each of the Zr atoms is altered (by 0.004 a.u.) and vanishes for the nearest-neighbor Zr atoms, confirming that the muon does not induce magnetization. It is sometimes possible to form local moments on impurities in metals through their resonant interaction with conduction electrons [34, 35]. However, the facts that the muon is a light impurity and that the muon PDOS lies so far below the Fermi energy makes moment formation on the muon extremely unlikely here (a criterion for this is provided in the SM [29]).

In summary, we have carried out a systematic investigation of the muon sites in a variety of superconducting compounds which purportedly exhibit TRSB in order to assess the extent of any muon-induced effect that could, in itself, give rise to the observation of a spontaneous field. Because the muon acts like a charged impurity, the most significant effects it could have are (i) on the local structural arrangement of atoms or ions close to the muon and (ii) on the local electronic structure. For point (i), our results show that in all cases studied the structural distortion involves only a modest alteration in the positions of nuclei that is rapidly suppressed with distance. Point (ii) is potentially more important since many superconductors that are candidates for TRSB have several bands crossing the Fermi energy, leading to multiple Fermi-surface sheets; if the muon were to appreciably alter the electronic structure near E_F this might conceivably provide a mechanism for the muon to couple to some muon-induced spin density (although one would then need an explanation for why this effect tracks the order parameter in the superconducting state). However, our results show that in these materials the changes to the local electronic structure resulting from muon implantation occur several eV below E_F, well away from the superconducting gap (which is a few meV around E_F), precluding any direct effect of the muon on the local superconducting state. This contrasts with results on hydrogenic impurity states in several semiconductors (e.g. ZnO [36, 37] and HfO [38]) in which the muon level is found within the gap and close to E_F [39]. Moreover, since the muon level is deep below E_F in all the compounds considered in this paper, it acts as a neutral defect and so the only perturbation of the local charge density is caused by the (very small) movement of the nearby ions that drag their charge density with them. In these systems, we find that the calculated change in the electronic charge on nearby ions is typically $< 0.4\%$. This puts a tight constraint on any models which attempt to explain the spontaneous fields as being due to the suppression of the superconducting order parameter by an imagined screening cloud of charge density around the muon [40, 41]. The calculations show that in these systems the muon is instead a rather benign defect that produces minimal effect on the

local charge density. We therefore conclude that the observation of spontaneous local fields in superconductors exhibiting TRSB is an effect which is intrinsic to these compounds and not a result of a muon-induced effect.

Finally, we note that the techniques demonstrated here are applicable well beyond the question of muons in superconductors exhibiting TRSB. These results suggest such that systematic calculations of muon sites in materials is a promising, and necessary, means to assess the influence of the stopped muon on any exotic physics for which it is being used to act as an experimental probe.

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