Establishing the physical mechanism governing exchange interactions is fundamental for exploring exotic phases such as the quantum spin liquids (QSLs) in real materials. In this work, we address exchange interactions in Sr$_2$CuTe$_{6-x}$O$_6$, a series of double perovskites that realize the spin-1/2 square lattice and were suggested to harbor a QSL ground state arising from random distribution of non-magnetic ions. Our \textit{ab initio} multi-reference configuration interaction calculations show that replacing Te atoms with W atoms changes the dominant couplings from nearest to next-nearest neighbor explained by the crucial role of unoccupied states of non-magnetic ions in the super-superexchange mechanism. Combined with spin-wave theory simulations, our calculated exchange couplings provide an excellent description of the inelastic neutron scattering spectra of the end compounds, as well as explain the magnetic excitations in Sr$_2$CuTeO$_6$ and Sr$_2$CuWO$_6$ as emerging from the bond-disordered exchange couplings. Our results provide crucial understanding of the role of non-magnetic cations in exchange interactions paving the way to further exploration of QSL phases in bond-disordered materials.

In transition metal (TM) oxides, especially composed of 3\textit{d} elements, the on-site Coulomb repulsive interactions between the electrons are strong enough to confine them to the TM sites, leading to the formation of localized spin or spin-orbital moments [1]. The manner in which these moments couple to each other is primarily governed by the underlying exchange interactions, which may be direct and/or mediated by the intermediate anions or ligands (L), the latter is also referred to as the superexchange. There are many possible ways these interactions can manifest, resulting in a plethora of magnetically ordered states such as ferromagnetic and different types of antiferromagnetic (AFM) order, magnetic spirals or more exotic topologically protected magnetic textures such as Skyrmions [1–4].

Even more fascinating ground states that stem from exchange interactions are those which do not undergo any magnetic ordering even at absolute zero temperature, e.g. spin-liquid states in low-dimensional magnetic systems [5]. Broken-symmetry valence-bond solids and QSLs where symmetry is conserved are some of the examples of such phases [5–7]. In these quantum paramagnetic phases, the long-range magnetic order is typically destroyed by frustrated exchange interactions and quantum fluctuations [8]. In the simplistic and prototypical two-dimensional spin-1/2 Heisenberg square lattice (HSL) model, the ratio of nearest-neighbor (NN) $J_1$ and AF next-nearest neighbor (NNN) $J_2$ exchange interactions of $\sim 0.5$ results in magnetic frustration and a QSL ground state [7].

The exchange mechanisms in TM compounds, principally the superexchange, are reasonably well understood in the form of the Goodenough-Kanamori-Anderson (GKA) rules [1]. The highly successful GKA rules correctly predict the sign of magnetic coupling for the 180° and 90° TM-L-TM bond angles. In double perovskite magnetic compounds like Sr$_2$CuTeO$_6$ and Sr$_2$CuWO$_6$ the magnetic ions are separated by additional non-magnetic cations Te$^{6+}$ and W$^{6+}$, respectively, and the magnetic coupling is a result of super-superexchange (SSE) mechanism. As shown in Fig. 1, there are multiple SSE paths – the NN exchange is via two identical Cu-O-Te/W-O-Cu paths involving four bridging ligands and two non-magnetic cations, with 90° Cu-Te/W-Cu angle. Alternatively, the second or NNN coupling arises from only one Cu-O-Te/W-O-Cu exchange path (180° Cu-Te/W-Cu angle) involving two ligands and a non-magnetic cation. The interesting aspect in these compounds is that the non-magnetic cation may or may not participate in the exchange.

In this paper, we primarily address the question – “Do the exchange mechanisms in double perovskite compounds depend on the non-magnetic cations and if so, how?” We compute the exchange couplings in double perovskite Sr$_2$CuTe$_{6-x}$O$_6$ with $x = \{0.0, 0.5, 1.0\}$ compounds using \textit{ab initio} many-body quantum chemistry (QC) calculations. We analyze their microscopic provenance by examining the different SSE paths in-
volved and show that the bridging non-magnetic cation plays a pivotal role in the exchange mechanism depending on whether it has empty or completely filled d manifold [9, 10]. Further, we decipher the possible physical origin of the features observed in the inelastic neutron scattering (INS) spectrum using spin-wave theory (SWT) while simulating the site disorder phenomena prevailing in Sr$_2$CuTe$_6$W$_{1-x}$O$_6$. Our study exposes double perovskites with non-magnetic cations as the ideal playground to explore bond-disordered couplings and associated QSL phenomena.

The isostructural double perovskite copper oxides Sr$_2$CuTeO$_6$ [11] and Sr$_2$CuWO$_6$ [12] realise a quasi-two-dimensional spin-1/2 HSL antiferromagnet, despite their three-dimensional crystal structures [13–15]. However, the magnetic order in the two systems is different. While a Néel AFM (NAF) ordering is observed in Sr$_2$CuTeO$_6$ with large AFM $J_1$ and small $J_2$ of the same sign [14, 16], a columnar AFM (CAF) order is stabilized in Sr$_2$CuWO$_6$ with small $J_1$ and large $J_2$, both AFM [15]. Interestingly, the reported $J_2/J_1$ ratio in these two compounds differs by two orders of magnitude, 0.03 and 7.92 for Sr$_2$CuTeO$_6$ and Sr$_2$CuWO$_6$ respectively.

It has been anticipated that a solid solution with equal quantities of Te and W may result in a ratio $J_2/J_1$ close to 0.5, leading to strong magnetic frustration and possibly producing a spin-liquid ground state [10, 17, 18]. Interestingly, the macroscopic magnetic features of Sr$_2$CuTe$_x$W$_{1-x}$O$_6$ for $x = 0.5$ show no signs of magnetic ordering, which may indicate its proximity to the highly frustrated $J_2/J_1 = 0.5$ region. Furthermore, the specific heat behavior at low temperatures is reminiscent of a gapless QSL state with collective excitations of entangled spins [10]. Fascinatingly, the suppression of long range magnetic order is observed in a wide region of $x \approx 0.1 - 0.6$ [19].

To begin with, Table I compares the Heisenberg exchange couplings defined in Fig. 1 for the end compounds of Sr$_2$CuTe$_x$W$_{1-x}$O$_6$ solid solution – Sr$_2$CuTeO$_6$ and Sr$_2$CuWO$_6$ – obtained from ab initio multireference dedicated configuration interaction (MR-DCCI) calculations [20, 21]. Those obtained from INS measurements are also shown in the same table. The calculations were done on three different embedded clusters for $J_1$, $J_2$ and $J_c$ (see Fig. 1), respectively. For computational details see Ref. [14] and Supplementary material [22]. In contrast to conventional density functional theory (DFT) and correlated calculations in conjunction with dynamical mean field theory (DFT + DMFT), our calculations are parameter free and accurately describe correlations within the cluster of atoms in a systematic manner. The virtual hopping processes necessary to capture the exchange interactions are well described in this approach and this makes it the only ab initio method that has sufficient predictive capability for estimating magnetic couplings [23–26].

To extract the isotropic exchange couplings, the ab initio magnetic spectrum of two unpaired electrons in two Cu$^{2+}$ ions is mapped onto that of a two-spin Heisenberg Hamiltonian $H_{ij} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$. In the QC computations, a complete active space reference wavefunction with two electrons in two Cu$^{2+}$ ground state $d_{x^2−y^2}$-type orbitals was first constructed for the singlet and triplet spin multiplicities [27], state-averaged [28]. In the MR-DCCI calculations the electrons in the doubly occupied Cu 3d orbitals and the O 2p orbitals of the bridging (Te/W)O$_6$ octahedron were correlated. To account for size-consistency errors, we adopted Davidson correction scheme [29]. All calculations were done using the MOLPRO quantum chemistry package [30].

We have shown in an earlier publication [14] that in Sr$_2$CuTeO$_6$ the dominant Heisenberg coupling is the NN $J_1$, see columns one and two in Table I. The SSE path that gives this large coupling is Cu$^{2+}$−O$^{2−}$−O$^{2−}$−Cu$^{2+}$ along the two bridging TeO$_6$ octahedra and does not include the Te$^{6+}$ ions explicitly. On the other hand, the NNN $J_2$ coupling with 180° Cu−Te−Cu angle is significantly smaller and is through the bridging Te atom – Cu$^{2+}$−O$^{2−}$−Te$^{6+}$−O$^{2−}$−Cu$^{2+}$.

In the present work, we performed QC calculations for

![FIG. 1. (a) The crystallographic unit cell of Sr$_2$Cu(Te/W)O$_6$. (b) The ab-plane view of Sr$_2$CuWO$_6$ along with the in-plane Cu 3$d_{x^2−y^2}$ (red and blue) and W 5$d_{x^2−y^2}$ (yellow and orange) orbitals. The different exchange couplings are shown by arrows connecting two Cu$^{2+}$ ions.]({

| Table I. A comparison of the Heisenberg exchange couplings obtained from ab initio MR-DCCI calculations (QC) and experimentally using inelastic neutron scattering (INS) for Sr$_2$CuTeO$_6$ [14] and Sr$_2$CuWO$_6$[15]. The couplings obtained from INS are from fits to SWT with a renormalization factor $Z_c = 1.18$ as the first-order correction [31] to calculated magnetic dispersion. All values are given in meV. |
| --- |
| Sr$_2$CuTeO$_6$ | Sr$_2$CuWO$_6$ |
| QC | INS [14] | QC | INS [15] |
| $J_1$ | 7.38 | 0.68 | 1.02 |
| $J_2$ | 0.05 | 0.60(3) | 8.33 | 8.50 |
| $J_c$ | 0.003 | 0.04 | 0.005 | - |


Sr$_2$CuWO$_6$. We find a strong NNN $J_2$ and a small NN $J_1$ resulting in the ratio $J_2/J_1 \sim 12$. The coupling along the $c$-axis is estimated to be two orders of magnitude smaller, but nevertheless larger than in Sr$_2$CuTeO$_6$. These results are consistent with the couplings extracted from the INS data [15]. We note that the spin-wave renormalization, not included in the linear SWT employed in Ref. [15], could have the same effect as a small NN exchange coupling. Therefore, further corrections to $J_1$ and $J_2$ values extracted from the INS might be necessary to account for this. However, we expect this to be small, e.g. in Sr$_2$CuTeO$_6$, this corresponds to $(J_1, J_2)$ values being renormalized from $(7.60,0.60)$ to $(7.18,0.21)$ [14].

Given the qualitative similarity of the crystal structures [11, 12] as well as the electronic states near the Fermi level [32], it seems surprising to find the dominant $ab$-plane exchange couplings opposite in the two compounds – $J_1$ in Sr$_2$CuTeO$_6$ and $J_2$ in Sr$_2$CuWO$_6$. It is important to note that the states above (unoccupied) and below (doubly occupied) the Fermi level play an active role in the superexchange process, particularly, if these states belong to the ions bridging the two magnetic sites. In this respect, there is a considerable difference in the unoccupied manifold near the Fermi level in the two compounds. While there is a large density of W 5$d$ unoccupied states in Sr$_2$CuWO$_6$ at 4 eV above the Fermi level [32], in Sr$_2$CuTeO$_6$ the relatively small density of unoccupied states near the Fermi level consists of Te 5$p$ character [32]. Further, owing to the delocalized nature of W 5$d$ orbitals, there is a considerable $dp$-hybridization with the bridging O 2$p$ orbitals leading to appreciable hopping matrix element across the W$^{6+}$ ions. In contrast, the Te 5$p$ orbitals are compact and little or zero $pp$-hybridization is expected with O 2$p$ orbitals. Thus, in Sr$_2$CuWO$_6$ the W$^{6+}$ ions actively participate in the superexchange mechanism.

One might ask “why $J_1$ is small in Sr$_2$CuWO$_6$ ?”, given the arguments brought forward in the previous paragraph. To gain more insight into the SSE paths involved in NN and NNN couplings, we have computed $J_1$ and $J_2$ by restraining the virtual hopping processes involving the W$^{6+}$ unoccupied orbitals. This can be achieved in QC calculations by setting the coefficients of these orbitals to zero. Although this is unphysical, it gives direct information about the role of W virtual orbitals in the exchange mechanisms.

The NN $J_1$ coupling along two 90° Cu-W-Cu paths, see Fig. 1(b), decreases to 0.49 meV (∼25% reduction) when the unoccupied orbital coefficients of the two W$^{6+}$ ions are eliminated. This implies that the contribution to $J_1$ from the configurations involving W virtual orbitals is not the leading one. It turns out that the other exchange paths, particularly the Cu-O-O-Cu path, has larger contribution to $J_1$ just as in the case of Sr$_2$CuTeO$_6$ [14]. It should be noted that there are three different W 5$d$ orbitals that participate in the SSE mechanism. While the in-plane 5$d_{x^2-y^2}$ orbitals, see Fig. 1(b), have $\sigma$-overlap with the bridging oxygen 2$p$ orbitals and result in an AF coupling, the out-of-plane degenerate 5$d_{xz}$ and 5$d_{yz}$ orbitals with $\pi$-type overlap contribute to ferromagnetic exchange that is governed by the Hund’s rule coupling of the W 5$d$ orbitals. A competition of these two mechanisms result in an overall small AF exchange.

We emphasize that in QC calculations all the virtual orbitals of the W$^{6+}$ ions participate in the SSE process and estimating the contributions from a particular virtual orbital is impractical. However, one can understand the SSE from a simplified Hubbard model (SSE-H) that contains two oxygen $p$ orbitals and an additional single W 5$d_{x^2-y^2}$ virtual orbital $(d-p-d-p-d)$ compared to a conventional $d-p-d$ model applied for charge-transfer insulators [1]. In Fig. 2, the SSE processes in the $J_2$ coupling within the SSE-H model are shown. There are three different possible virtual hopings, represented schematically in Fig. 2, that lift the spin degeneracy and hence contribute to the AFM exchange coupling. In scheme I, Fig. 2(a), the electron from one Cu$^{2+}$ ion can hop to the other and back through the intermediate configurations with a single hole (electron) in O 2$p$ (W 5$d$) orbitals at a particular instance. This scheme has dominant

![FIG. 2. Super-supercoupling mechanisms involved in NNN $J_2$ coupling in a four band Hubbard model. The three possible intermediate states (see text) that contribute to exchange interaction are shown as three schemes. The Cu 3$d_{x^2-y^2}$, O 2$p$ and W 5$d_{x^2-y^2}$ levels are represented in blue, green and orange, respectively. The sequence of virtual electron hoppings (arrows) are marked by numbers 1 to 8.](image-url)
contribution to the $J_2$ coupling. Two other possibilities that are also viable are shown in schemes II and III in Fig. 2(b) and 2(c). Here, configurations where both Cu $d_{x^2-y^2}$ orbitals are doubly occupied are active. While in scheme II both oxygen atoms can contain two holes and the W $5d_{x^2-y^2}$ orbital holds two electrons, in scheme III only one of the oxygen atoms contains two holes. The last scheme contributes twice as either of the two oxygen atoms can accommodate two holes.

The coupling arising from scheme I can be written as

$$J_2^1 = 2 \frac{t_{dd}^2}{U_{Cu}^{dx}} , \text{ with } t_{dd} = \frac{t_{pdCu}^2}{\Delta_{pdCu}^2} ,$$

where $t_{pdCu}$ and $t_{pdW}$ are the hopping matrix elements from O 2p to Cu 3$d_{x^2-y^2}$ and from O 2p to W 5$d_{x^2-y^2}$, respectively, $U_{dd}^{Cu}$ is the on-site Coulomb interaction on the Cu sites, and $\Delta_{pdCu}$ and $\Delta_{pdW}$ are the charge-transfer energies from O 2p to Cu 3d and W 5d orbitals, respectively. Schemes II and III would involve $U_{pp}^{O}$ ($U_{dd}^{W}$), the Coulomb interactions when two holes (electrons) are accommodated in O 2p (W 5d) orbitals, and yield minor contributions.

Next, we discuss the effect of Te/W atom disorder on the exchange coupling constants. Let us consider Sr$_2$CuTe$_2$W$_{1-x}$O$_6$ and assume that Te and W atoms are perfectly ordered such that every Cu$^{2+}$ ion is surrounded by two Te$^{6+}$ and two W$^{6+}$ ions. In such a scenario, there are three NN ($J_a$, $J_b$, and $J_c$) and two NNN ($J_d$ and $J_e$) exchange couplings as shown in Fig. 3. Four of the five couplings, $J_b$, $J_c$, $J_d$, and $J_e$, remain the same as in the end compounds Sr$_2$CuWO$_6$ and Sr$_2$CuTeO$_6$ as the exchange paths are the same. On the other hand, the exchange channels corresponding to $J_a$ are different compared to the end compounds of the solid solution. We estimated the coupling $J_a$ from our ab initio singlet-triplet energy separation for two Cu$^{2+}$ ions with the neighboring environment as shown in Fig. 3. We find this coupling AMF with a magnitude, 0.3 meV, much smaller than the dominant coupling in the end compounds. To summarize, our calculations show that the average $J_1$ and $J_2$ can be tuned from effectively 0 to 8 meV through substitution of Te for W, opening up an interesting arena to explore bond disorder of a spin-1/2 square lattice antiferromagnet.

Now we turn to the INS experiments and SWT calculations. Measurements on Sr$_2$CuTeO$_6$ were performed using the IN4 spectrometer at the ILL utilizing an incident neutron energy of $E_i = 25.2$ meV [33]. The Sr$_2$CuTe$_2$W$_{1-x}$O$_6$ for $x = 0.5$ and 1.0 samples were studied using MERLIN at ISIS with $E_i = 45$ meV [34]. Further details on Sr$_2$CuTeO$_6$ and Sr$_2$CuWO$_6$ INS measurements are reported elsewhere [14, 15].

Figure 4 shows the inelastic neutron scattering spectra of powder Sr$_2$CuTe$_2$W$_{1-x}$O$_6$ that have been collected on $x = \{0, 0.5, 1.0\}$ compounds. A background, adjusted for the Bose thermal population factor, recorded at >100 K has been subtracted from the spectra to remove the phonon contribution at larger $|Q|$. The end-compounds of Sr$_2$CuWO$_6$ and Sr$_2$CuTeO$_6$ show spin waves dispersing from the CAF and NAF zone centres, respectively. A strong band of scattering around 15-17 meV is found in both compounds. This corresponds to a van Hove singularity from the top of the spin-wave dispersion. The INS spectrum of the intermediate Sr$_2$CuTe$_{0.5}$W$_{0.5}$O$_6$ compounds is dramatically different. There appears to be a significant smearing of the spectrum in momentum and energy transfer. The band of scattering corresponding to the van Hove singularity is absent. Weak excitations are observed up to around 20 meV. This scattering decreases with increasing $|Q|$, as would be expected for magnetic
scattering. Magnetic modes emerge from $|Q| = 0.65$ and $1.4 \text{ Å}^{-1}$, much like in $\text{Sr}_2\text{CuWO}_6$, which would suggest the dominant interactions in $\text{Sr}_2\text{CuWO}_6$ persist in the $x = 0.5$ compound.

To simulate the INS spectra, we need to construct an appropriate magnetic ground state from which magnetic fluctuations can be calculated. We define a $10 \times 10$ square lattice with randomly populated W and Te atoms. The strengths of the $J_1 - J_2$ exchange parameters are as given in Table I and the different possible exchange pathways in the mixed $x = 0.5$ compound are according to Fig. 3. Therefore, $J_1$ can take values of $7.60 \text{ meV}$ or $1.02 \text{ meV}$ depending on whether two Te or W atoms are involved in the exchange process with similar arguments applying to $J_2$. In the case of one W and one Te atom, we take $J_1 = 0$. From this construction, we find the classical spin configuration which minimises the total energy and calculate the spin-wave dispersion. To account for truncation of the spin Hamiltonian at the quadratic terms when calculating the one-magnon energy we rescale the magnon energy by a constant factor of $Z_c = 1.18$ [31].

The calculation is repeated with different distributions of Te and W and the resulting spin-wave pattern is averaged. Figures 4(d)-4(f) show the calculated powder averaged spectra for each composition. Comparing the calculated spectra for $x = 0.5$ to the end compounds, we observe that the simulation predicts a rather broad spectrum. The intense and sharp scattering at the top of the bandwidth in $\text{Sr}_2\text{CuWO}_6$ and $\text{Sr}_2\text{CuTeO}_6$ is no longer present for the intermediate compound. Apart from excitations emerging from $|Q| \approx 0.7$ and $1.4 \text{ Å}^{-1}$, the spectrum is featureless. This appears to be in good agreement with the measured powder spectrum, indicating that the bond-disordered exchange couplings reproduce the INS spectrum of $\text{Sr}_2\text{CuTeO}_{0.5}\text{W}_{0.5}\text{O}_6$.

To summarize, we have computed the NN and NNN Heisenberg exchange couplings in $\text{Sr}_2\text{CuWO}_6$ and $\text{Sr}_2\text{CuTeO}_{0.5}\text{W}_{0.5}\text{O}_6$ finding excellent agreement with available experimental observations. We established that the non-magnetic cation bridging the magnetic sites play a significant role in the SSE process. In the case of completely filled $d$-manifold ($\text{Te}^{6+}$) cation, the exchange path does not include any of its orbitals, but for the $d^0$ ($\text{W}^{6+}$) bridging cation, the SSE process via these empty orbitals is pivotal. We further provided the rationale for the observed exchange interactions, justifying with numerical evidence. Our simulated INS spectra for $\text{Sr}_2\text{CuTeO}_6$ and $\text{Sr}_2\text{CuWO}_6$ compare very well with experimental data, and they give a good understanding of the measured powder spectrum for $\text{Sr}_2\text{CuTeO}_{0.5}\text{W}_{0.5}\text{O}_6$. Although further neutron scattering studies are necessary to examine the latter compound, our calculations provide a deep insight into the nature of the interactions within the complex ground state of this system. Our work thus establishes the theoretical background for describing bond-disorder exchange couplings highlighting site-disordered materials as a new playground for exploring QSL states.

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