

Supplementary material for “High antiferromagnetic transition temperature of new layered honeycomb oxide SrRu_2O_6 ”

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NEUTRON DIFFRACTION MEASUREMENTS

Neutron diffraction experiments were carried out on a ~ 0.4 g powder sample using the HB1, HB-1A, and HB-3A neutron spectrometers located at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory.

The HB-1A spectrometer operates with a fixed incident energy of $E_i = 14.64$ meV using a double pyrolytic graphite (PG) monochromator system. Two PG filters were placed before and after the second monochromator to reduce higher order contamination in the incident beam ($I\lambda/2 \approx 10^{-4} I\lambda$). The powder sample (in a 6 mm diameter aluminum sample can) was loaded in a top-loading closed-cycle refrigerator (CCR) and diffraction patterns over a 2θ angle range of $10^\circ < 2\theta < 90^\circ$ were collected at 40 K, 300 K and 590 K. The order parameter measurements were performed by scanning the peaks of interest at selected temperatures between 40 K and 600 K.

The same powder sample was also measured at HB-3A four-circle diffractometer in a wider Q -range for more magnetic reflections. A 2-dimensional Anger camera type detector was used for the powder measurement. A neutron wavelength of $1.542 \text{ \AA}$ was used from a bent perfect Si-220 monochromator.

Polarized neutron measurements were performed on the same sample using HB1 polarized triple-axis spectrometry, with a fixed final energy of $E_f = 13.5$ meV and a collimation of $40' \text{-} 80' \text{-} \text{sample} \text{-} 80' \text{-} 120'$. The neutron beam is polarized by using Heussler alloy monochromator and analyzer with a spin flipper in the incident beam. The sample can be maintained in a horizontal or vertical magnetic guide field such that the neutron polarization P_0 is parallel or perpendicular to the momentum transfer Q ; $P_0 \parallel Q$ when a horizontal field is applied at the sample position or $P_0 \perp Q$ when a vertical magnetic field is applied. With the spin flipper off or on, we measure the $(++)$ non-spin-flip or the $(-+)$ spin-flip scattering, re-

spectively. The HB1 experiment was performed with $P_0 \parallel Q$ at room temperature. Both $(1\ 0\ 0.5)$ magnetic peak and $(1\ 0\ 1)$ nuclear peak were measured with “Flipper Off” and “Flipper On” configurations with a horizontal magnetic guide field at the sample position.

Coherent nuclear scattering is always $(++)$ non-spin-flip scattering because it never causes a reversal or spin flip of the neutron spin direction upon scattering. On the other hand, magnetic scattering depends on the relative orientation of the neutron polarization P_0 and the scattering vector Q . Only those spin components which are perpendicular to the scattering vector are effective. Thus for a fully polarized neutron beam with the horizontal field configuration, $P_0 \parallel Q$, all magnetic scattering is $(-+)$ spin-flip scattering, and ideally no $(++)$ non-spin-flip scattering will be observed. The strong intensity shown in Fig. 2(c) was observed in the $(++)$ channel as expected for the nuclear $(1\ 0\ 1)$ peak. The strong scattering detected in the $(-+)$ spin-flip channel confirmed the magnetic origin of the $(1\ 0\ 0.5)$ peak.

REFINEMENT OF DIFFRACTION PATTERNS

The crystal and magnetic structure were analyzed by the FullProf software package. Figure 1 shows the refinement of the room temperature diffraction pattern taken at HB-3A with powder in an aluminum can. The R -factors for the nuclear and magnetic Bragg peaks are 0.037 and 0.082, respectively. The global goodness of fit is 4.17. The refinement gives the room temperature lattice parameters of $a = 5.1988(9) \text{ \AA}$, and $c = 5.2270(13) \text{ \AA}$, consistent with the previous report.

ELECTRICAL RESISTIVITY

The temperature dependence of electrical resistivity was measured in the temperature range $150 \text{ K} \leq T \leq 360 \text{ K}$

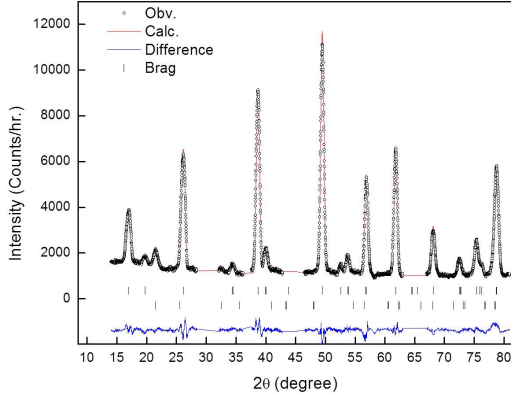


FIG. 1: (color online) Refinement of the room temperature neutron diffraction pattern taken at HB-3A. The upper and lower vertical bars show the position of nuclear and magnetic reflections, respectively. The 2θ regions with alumina can and half-lambda contaminations were excluded in the refinement.

on a cold-pressed pellet. The electrical resistivity increases upon cooling and we tried fitting to an activated model: $\rho(T) \propto \rho_0 + \exp(-\Delta/2kT)$, where ρ_0 is a constant, Δ is the band gap. The fitting gives $\Delta = 36$ meV.

It is worth mentioning that the electrical resistivity was measured on a cold-pressed pellet without high temperature treatment. Measuring the electrical resistivity is rather challenging because (1) the SrRu_2O_6 powder synthesized by hydrothermal technique contains small crystallites with the largest dimension around $50\mu\text{m}$. With the regular die/press, we cannot press the fine crystallites into a dense pellet even after grinding for an extended period. With the help of a pair of WC anvils, we have now managed to make a dense pellet that has allowed us to perform resistivity measurements. (2) SrRu_2O_6 decomposes around 800 K. Thus, no high temperature heat treatment of the cold-pressed pellet can be performed to relieve the residual stress. As a result, we expect grain boundaries and residual stress would prevent an accurate measurement of the intrinsic resistivity of SrRu_2O_6 . Future work on single crystals is desired.

DFT CALCULATIONS

The muffin-tin radii r for Sr, Ru and O were set to 2.36, 1.93, and 1.72 a.u., respectively. We used the maximum modulus for the reciprocal lattice vectors K_{max} was chosen so that $r_{\text{O}} \times K_{\text{max}} = 9.0$. In the calculation of the partially screened Coulomb interaction, we took 120 unoccupied bands and used a $4 \times 4 \times 4$ grid. The double Fourier transform of the constrained susceptibility was done with the cutoff of 5 (1/a.u.).

Fig. 3 presents the band dispersions for the NM and

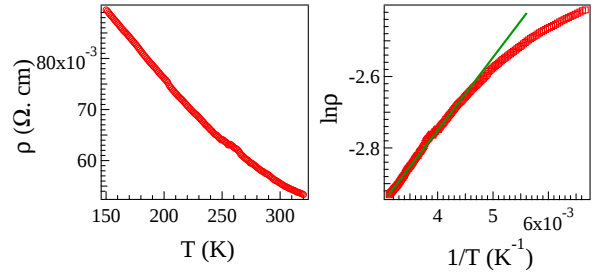


FIG. 2: Temperature dependence of electrical resistivity measured on a cold-pressed SrRu_2O_6 pellet. The right panel shows the data plotted as $\ln\rho$ vs $1/T$.

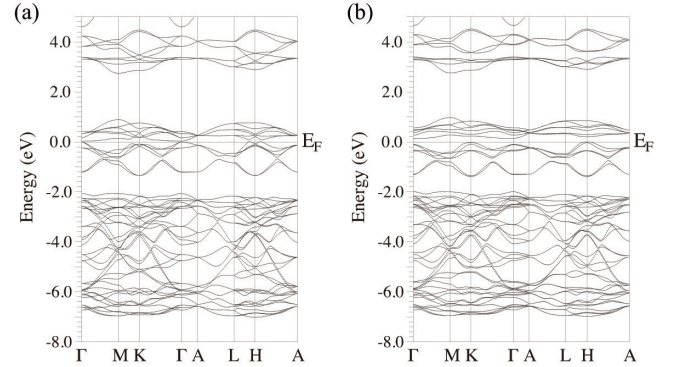


FIG. 3: Band dispersion for the (a) NM and (b) G-type AF phases using a doubled unit cell.

G-type AF phases using a doubled unit cell. Please note that the number of bands is doubled from that for the NM band dispersion using a primitive cell presented in the main text. The band dispersions of the NM and AF phases are very similar, except that the AF order enlarges the band gap from 0.05 eV to 0.14 eV. Density of states (DOS) projected on Ru sites polarized with up spin is shown in Fig. 4. Oxygen bands below -2 eV show almost no spin polarization, which validates our strategy to concentrate on the t_{2g} bands lying around the Fermi level. DOS projected on O sites is not shown here because spin polarization for up and down spins are exactly canceled in the AF phase.

EFFECTIVE PARAMETERS AFTER DOWNFOLDING

Starting with the DFT calculation for the NM state, we first construct the Wannier functions and calculate the transfer integrals between them. Next, we evaluate the interaction parameters, the Coulomb repulsion $U(\mathbf{r})$ and the exchange coupling $J(\mathbf{r})$ by the constrained RPA. For simplicity, we neglect the spin-orbit interaction since it is irrelevant in the following analysis. The Coulomb parameters from constrained RPA and transfer integrals

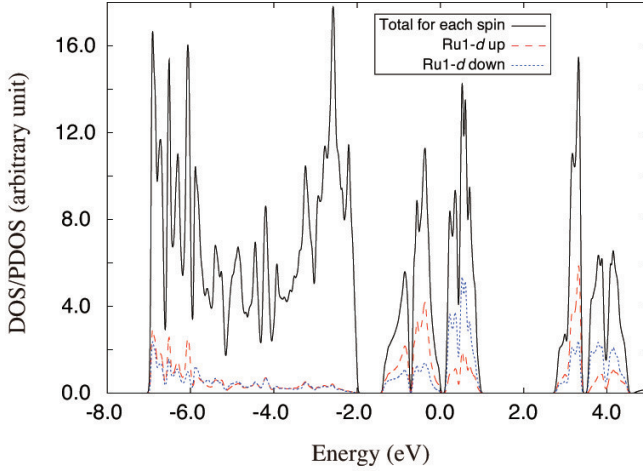


FIG. 4: Density of states (DOS) for the G-type AF phase. Total DOS for each spin and projected DOS (PDOS) for d states on Ru sites polarized with up spin (denoted Ru1 sites here) are shown.

after applying the downfolding procedure are given below. The first three basis states correspond to effective orbitals centered on the first Ru site in the unit cell, and the second three correspond to those centered on the second site. All values are reported in eV.

Hubbard interaction

$$\begin{pmatrix} 2.732 & 2.186 & 2.167 & 1.133 & 1.150 & 1.186 \\ & 2.680 & 2.185 & 1.149 & 1.156 & 1.163 \\ & & 2.731 & 1.185 & 1.163 & 1.162 \\ & & & 2.731 & 2.185 & 2.167 \\ & & & & 2.680 & 2.186 \\ & & & & & 2.732 \end{pmatrix}$$

Direct exchange interaction

$$\begin{pmatrix} 0 & 0.280 & 0.282 & 0.010 & 0.016 & 0.029 \\ & 0 & 0.280 & 0.016 & 0.020 & 0.016 \\ & & 0 & 0.029 & 0.016 & 0.013 \\ & & & 0 & 0.280 & 0.282 \\ & & & & 0 & 0.280 \\ & & & & & 0 \end{pmatrix}$$

Transfer integrals

$$\begin{pmatrix} 5.552 & 0 & 0 & 0.130 & 0.049 & 0.279 \\ & 5.386 & 0 & 0.049 & 0.222 & 0.028 \\ & & 5.552 & 0.279 & 0.028 & -0.193 \\ & & & 5.552 & 0 & 0 \\ & & & & 5.386 & 0 \\ & & & & & 5.552 \end{pmatrix}$$

FINITE SIZE SCALING

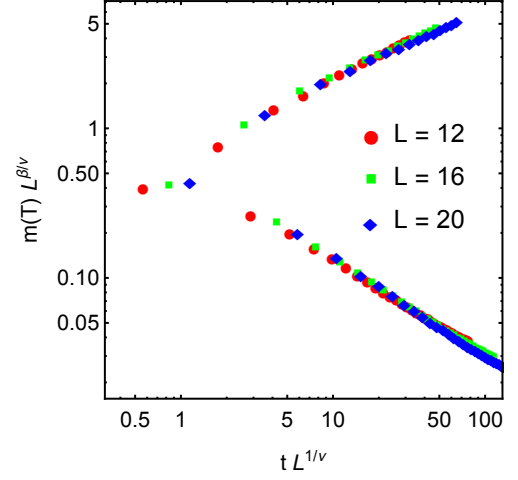


FIG. 5: (color online) Finite size scaling has been performed for $J_{\perp}/J_{\parallel} = 0.03$. The reduced temperature is denoted $t = (T - T_N)/T_N$.

The transition temperature in a system of infinite size is signaled by a divergence of the correlation length ξ , a divergence of the susceptibility χ , and a sharp change in the behavior of the staggered magnetization M . These sharp transitions are rounded off in finite systems, and the transition temperature must therefore be determined using finite size scaling methods. Near the transition, the divergence of the correlation length is cut off by the finite size of the system, L , hence the staggered magnetization M , for example, now scales with L according to $M(t, L) \sim t^{\beta} \mathcal{F}(L/\xi)$, where $\mathcal{F}(x)$ is a universal scaling function, β is a critical exponent and $t = (T - T_N)/T_N$ is the reduced temperature measured relative to the infinite system transition temperature T_N . Using the definition $\xi \sim t^{-\nu}$, where ν is a critical exponent, the dependence of staggered magnetization can be written as $M(t, L) \sim L^{-\beta/\nu} \tilde{\mathcal{F}}(L^{1/\nu} t)$ in terms of a new scaling function $\tilde{\mathcal{F}}(x)$. From this scaling relation, it is evident that the staggered magnetization must be independent of the system size at $t = 0$. Fig. 4(a) in the main text shows the extraction of the transition temperature obtained from the intersection of the magnetization curves at different system sizes. Having determined T_N , the scaling relation further suggests that if we plot the scaled staggered magnetization $L^{\beta/\nu} M(t, L)$ against a scaled temperature variable $L^{1/\nu} t$, we can determine the exponents. In this case, the critical exponents belong to the Heisenberg universality class with $\nu = 0.70 \pm 0.01$ and $\beta = 0.69 \pm 0.01$ that are well known. Our data for the scaled staggered magnetization curves lie on a universal curve as shown in Fig. 5 of this supplement; we obtain the best fit for

$\nu \sim 0.7$ and $\beta \sim 0.5$, which is somewhat lower than expected because of crossovers in the intermediate coupling regime.