

G-type antiferromagnetic ordering with strong anisotropic exchange interactions in $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$ Xiang Li¹, Xiaoting Li^{2,*}, Jiaqi Lin³, Zahirul Islam⁴, Mary H. Upton⁵, Vivek Thampy⁵, Yifan Jiang¹, Yuan Wan⁶, Haizhong Guo^{7,8,†} and Xuerong Liu^{1,2,‡}¹*School of Physical Science and Technology, ShanghaiTech University, Shanghai 201210, China*²*Center for Transformative Science, ShanghaiTech University, Shanghai 201210, China*³*Instrumentation and Service Center for Physical Sciences, Westlake University, Hangzhou 310024, Zhejiang Province, China*⁴*Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA*⁵*Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA*⁶*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*⁷*Key Laboratory of Materials Physics, Ministry of Education, School of Physics, Zhengzhou University, Zhengzhou 450052, China*⁸*Institute of Quantum Materials and Physics, Henan Academy of Sciences, Zhengzhou 450046, China*

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Isovalent substitution of Ir^{4+} with Sn^{4+} in perovskite SrIrO_3 induces a metal-insulator transition accompanied with strong antiferromagnetic ordering. We performed resonant x-ray scattering to investigate the magnetism in $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$. The G-type antiferromagnetic ordering with spin pointing along c axis was fully determined by azimuth-dependent elastic scattering. Dynamical magnetic excitation spectra were measured with resonant inelastic x-ray scattering. Strong magnetic exchange interactions were observed with magnon dispersing up to ~ 130 meV, and the anisotropy drives a large ~ 50 meV gap. With these results, we discuss the paramagnetic semimetallic behavior of the undoped SrIrO_3 .

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The significant spin-orbit coupling (SOC) in 5d iridium-based transition metal oxides, together with moderate effective on-site Coulomb U_{eff} and specific crystal structures, has led to many interesting observations, such as the spin nematic phase in Sr_2IrO_4 [1] and the emergence of Fermi arcs upon doping [2,3], the zigzag antiferromagnetic ordering in proximity to spin liquid phase in Na_2IrO_3 [4–6], all-in-all-out magnetic structure in pyrochlore iridates [7], etc. In these iridates, both the SOC and the crystal structure-related oxygen octahedral rotations are effective in reducing the electron bandwidth [8–11], putting the iridates in the strong electron-correlated region even with spatially quite extended 5d orbitals.

The end member of the $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ Ruddlesden-Popper series iridates, namely SrIrO_3 , also received wide attention. Earlier theoretical work suggested that SrIrO_3 is a Dirac semimetal [12,13]. Although such proposal seems to be challenged by angle-resolved photoemission spectroscopy (ARPES) measurements on films [9,14] where the linear dispersion was observed but the degeneracy was gapped, more recent transport measurements suggest that it is a semimetal

with high-mobility electrons [15]. Besides its electronic structure, SrIrO_3 is also of great interest in heterostructures to utilize its magnetic properties as thin layers [16,17]. For example, in $\text{SrRuO}_3/\text{SrIrO}_3$ [18,19] and $\text{LaMnO}_3/\text{SrIrO}_3$ [20] bilayers, the Dzyaloshinskii-Moriya (DM) interaction at the interface leads to the nanoscale skyrmions and affects the topological Hall effect.

Although the abovementioned research efforts all suggest that SrIrO_3 is at the verge of a magnetic instability and the electron correlation is a key ingredient, not much is known about its magnetic dynamics because the SrIrO_3 crystal behaves as a paramagnetic semimetal [21,22]. By thinning down SrIrO_3 into ultrathin film [23–26], or interfacing with other compound in superstructure films [27–29] or with Sn doping [30–32], the material can be driven into antiferromagnetic ordering with Néel temperature T_N around 200 K. Particularly with Sn^{4+} replacing Ir^{4+} , such isovalent substitution can drive a strong antiferromagnetic (AFM) ordering phase transition in $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$ [31]. Even $x = 0.05$ drives a Néel transition at ~ 200 K [33]. Further increasing x to 0.2 leads to a T_N saturation of ~ 280 K.

So far, the only microscopic information on the magnetism of $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$ is the observation of a low-temperature magnetic peak on $x = 0.2$ powder sample from neutron scattering [31], whose temperature dependence follows the magnetization curve. Based on its wave vector value, $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$ was postulated as a G-type AFM ordered insulator. In this Letter, we report our resonant x-ray scattering results on thick $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$ single-crystal film grown on SrTiO_3 substrate. With resonant elastic x-ray magnetic scattering (REMS), the AFM magnetic ordering is identified as G-type and the

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ordered spin is determined to be along the c direction. With the AFM ordering structure fully determined, resonant inelastic x-ray scattering (RIXS) was used to map the magnetic excitation dispersion along high-symmetry directions (see the Supplemental Material [34]). The magnetic excitation disperses up to ~ 130 meV at the zone boundary, with an excitation gap of ~ 50 meV at the zone center.

Since pristine $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$ is only available in powder form, high-quality perovskite $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$ single-crystal films epitaxially grown on SrTiO_3 (001) substrates with the pulsed laser deposition method were used. From a survey of multiple structural Bragg peaks on our samples [34], an octahedral rotation pattern of $a^-b^-c^+$ in Glazer notation [35] was verified, confirming a c -orientation growth on the SrTiO_3 substrate. In this work, we adopt the cubic unit cell notation for simplicity.

REMS is sensitive to the spin orderings [36,37] and has been proven to be powerful in determining the magnetic structures [4,38,39]. The measurements were carried out at beamline 6ID-B of the Advanced Photon Source (APS), Argonne National Laboratory (ANL). The incident x-ray was σ polarized, which is perpendicular to the scattering plane. A graphite (0 0 8) crystal analyzer was used to separate the scattered intensity into σ' (nonmagnetic) and π' (magnetic) channels. The data were taken on ~ 200 -nm-thick $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$ mostly at 5 K.

A well-defined structural Bragg peak was observed at $\mathbf{Q} = (-0.5 \ 0.5 \ 2.5)$ in the σ - σ' channel, associated with the oxygen octahedral rotations [40]. At the same wave vector, a peak was also observed in the σ - π' channel. The peaks in both channels show similar shape [Figs. 1(a)–1(c)], but have distinct incident x-ray energy dependence. Shown in Fig. 1(d) is the intensity evolution of several peaks across the Ir L_3 resonant edge. The $(-0.5 \ 0.5 \ 2.5)$ peak in the σ - π' channel shows a drastic enhancement at the resonant edge, which is the characteristic of resonant magnetic scattering. However, for its counterpart in the σ - σ' channel, the intensity shows a dip at the Ir L_3 edge. This suggests that the structural peak is due to oxygen or strontium displacement, while Ir coordinate is largely irrelevant. The dip is due to enhanced Ir absorption. As a comparison, another structural peak at (0 0 3), which has partial contribution from Ir, is also shown. A typical dip-hump energy dependence from anomalous scattering is observed.

To further confirm the magnetic origin of $(-0.5 \ 0.5 \ 2.5)$ peak in the σ - π' channel, the temperature dependence of the peak intensity in both channels was measured, and the results are shown in Fig. 2(a). The charge scattering in the σ - σ' channel shows an almost linear increase as the temperature reduces, while the intensity in the σ - π' channel shows typical second-order phase transition behavior, which starts around 180 K and gradually saturates around 80 K. We emphasize that the nonzero intensity in the σ - π' channel at temperature higher than ~ 200 K is the leakage from charge scattering because the polarization discrimination of the graphite analyzer is not perfect. Besides $(-0.5 \ 0.5 \ 2.5)$, other peaks were also surveyed in a large reciprocal space region. Similar resonant enhancement behavior was observed at $\mathbf{Q}$ points, where H , K , and L are half-integers, including $(-0.5 \ -0.5 \ 3.5)$ and $(-1.5 \ -0.5 \ 3.5)$ [34]. Based on these observations, we conclude

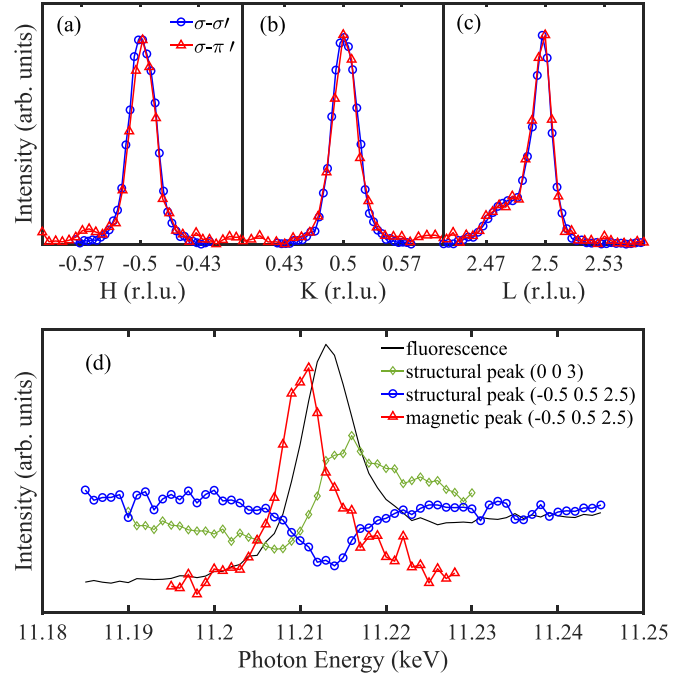


FIG. 1. (a)–(c) Reciprocal space scans of the $\mathbf{Q} = (-0.5 \ 0.5 \ 2.5)$ peak along the H , K , and L directions, respectively. (d) The energy dependence of the $(-0.5 \ 0.5 \ 2.5)$ peak in the σ - σ' and σ - π' channels across Ir L_3 edge. Solid black line shows the fluorescence signal and green diamond shows the energy dependence of the (0 0 3) structural peak for comparison.

that the AFM ordering is of G type, consistent with earlier theoretical suggestions [12].

A detailed azimuth dependence of the σ - π' component of the $(-0.5 \ 0.5 \ 2.5)$ peak was measured to identify the direction of the isospin of the AFM ordering. The dipole resonant magnetic x-ray scattering cross section varies as $|\hat{\mathbf{k}}_f \cdot \hat{\mathbf{m}}|^2 \sim \cos^2 \alpha$ [4,37], where $\hat{\mathbf{k}}_f$ is the direction of the scattered beam, $\hat{\mathbf{m}}$ is the isospin orientation, and α is the angle between these two vectors. As shown in Fig. 2(b), the azimuth-dependent intensity variation in the σ - π' channel (charge scattering leakage subtracted) follows a $\cos^2 \alpha$ curve, when the isospin direction $\hat{\mathbf{m}}$ is placed along the crystal c direction. Thus, the antiferromagnetic ordering structure in $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$ is fully solved with our REMS experiments.

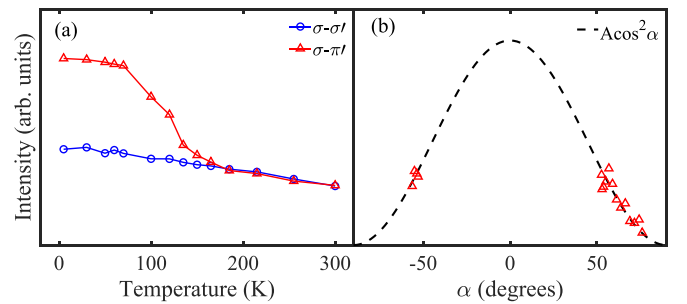


FIG. 2. (a) Temperature-dependent intensities of σ - π' and σ - σ' channels at $\mathbf{Q} = (-0.5 \ 0.5 \ 2.5)$, scaled for comparison. (b) The azimuth dependence of the $(-0.5 \ 0.5 \ 2.5)$ magnetic channel intensity.

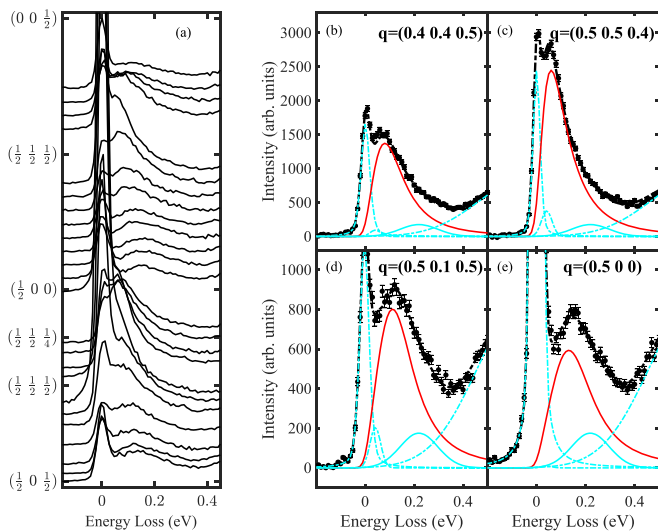


FIG. 3. (a) Stacking plot of RIXS spectra along high-symmetry paths. The total momentum transfer $\mathbf{Q}$ is given by $\mathbf{Q} = (HKL) + \mathbf{q}$, where (HKL) is either $(3\ 1\ 3)$ or $(3\ 2\ 3)$, and $\mathbf{q}$ is the momentum shown in the figure. (b)–(e) Fitting at several $\mathbf{Q}$ points. The paramagnon is represented by the red lines.

With the magnetic ordering structure of $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$ determined, we proceeded to study its dynamics by measuring the magnon dispersion with RIXS at the Ir L_3 edge. The experiments were performed at beamline 27ID-D of APS, ANL with an instrumental resolution of ~ 40 meV (FWHM). The stacking plot of the RIXS spectra measured at 20 K is shown in Fig. 3(a). The magnon excitation is clearly observed, while its line shape is quite broad, indicating strong damping from the Sn^{4+} dopants. Accordingly, we employed a damped harmonic oscillator model [41] to fit the magnon. The fitting results at a few typical $\mathbf{Q}$ points are shown in Figs. 3(b)–3(e).

Shown in Fig. 4 is the extracted magnon dispersion along high symmetry directions measured. The magnon disperses

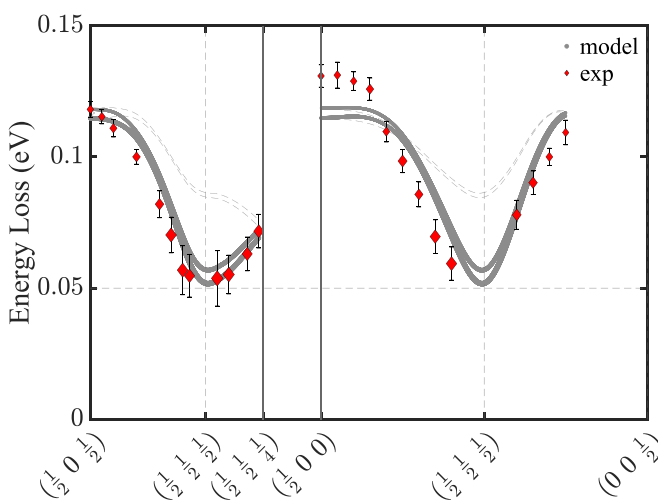


FIG. 4. Magnon dispersion. Red diamonds represent extracted excitation energy from RIXS measurements. Gray lines are the simulated magnon dispersion. The magnon intensity is represented by the dot size and linewidth. The intensity of the branches represented by dashed lines is marginal.

TABLE I. Magnetic interaction parameters (meV) extracted from magnon dispersion using determined magnetic structure and LSWT.

J	$ D $	G	J^c	$ D^c $	G^c
45	15	6	12	4	2

up to ~ 130 meV at the zone boundary, indicating that strong exchange interaction similar to other corner shared iridates of more 2D nature [42–48]. A large band gap of ~ 50 meV at zone center is also observed, suggesting the existence of strong magnetic anisotropy.

We constructed a phenomenological minimum exchange interaction model to describe the determined *c*-orientation *G*-type AFM structure and the measured magnon dispersion. Full symmetry analysis was also attempted [34]. Due to the low symmetry from strong Ir-O octahedron rotation and tilting [31,49], there are 15 independent exchange interaction parameters, which are far beyond the dispersion information we collected in the reciprocal space. Thus, our modeling is limited to be the nearest-neighbor Heisenberg term J , DM-interaction $\vec{D}$ and Ising term G [50]. As a result, the in-plane and out-of-plane exchange interaction Hamiltonian are written as

$$H_{\text{in}} = \sum_{\langle i,j \rangle} J_{ij}(\vec{S}_i \cdot \vec{S}_j) + \vec{D}_{ij}(\vec{S}_i \times \vec{S}_j) + G_{ij}S_i^z S_j^z,$$

$$H_{\text{out}} = \sum_{\langle i,j \rangle} J_{ij}^c (\vec{S}_i \cdot \vec{S}_j) + \vec{D}_{ij}^c (\vec{S}_i \times \vec{S}_j) + G_{ij}^c S_i^z S_j^z.$$

A set of exchange interaction parameters in this model is chosen under the guidance of our measured isospin structure, magnon dispersion, and its intensity variation from linear spin wave theory (LSWT) calculation. The spin is taken to be $S^z = 1/2$. The parameters listed in Table I produce a canted AFM ordering with the isospin mainly pointing toward the c direction. The orientations of DM vectors are determined by oxygen octahedral rotations [34,51].

In the RIXS process, the magnon scattering intensity is sensitive to the measurement geometry [52] as $I_n(\mathbf{Q}) \propto \sum_{\epsilon_{out}}^{\pi, \sigma} |\sum_i \epsilon_{out} \times \epsilon_{in} \cdot \mathbf{M}_{n,i}^{\mathbf{Q}}|^2$, where ϵ_{in} and ϵ_{out} are the polarizations of the incident beam and the scattered signal, and $\mathbf{M}_{n,i}^{\mathbf{Q}}$ is the eigenvector of the n th magnon mode for the i th site within one magnetic unit cell. As shown in Fig. 4, the resulted dispersion and the intensity distribution from the parameters in Table I agree well with our experimental data. Two excitation modes (dashed lines) have marginal scattering cross section in our experimental geometry. The measured RIXS signal is dominated by the lower two branches in the measured reciprocal space region.

Although our modeling is simplified, it points to a few key aspects. First, tens of meV in-plane nearest-neighbor Heisenberg exchange is needed in defining the overall magnon dispersion, emphasizing the significance of electron correlation. Second, the interplane Heisenberg term is much smaller than the in-plane Heisenberg term, emphasizing the crystal structure anisotropy from oxygen octahedral rotations in this 3D system. Third, the determined AFM c orientation is dictated by the Ising term G , while the competing DM interaction tends to twist the isospin away from the c direction.

With a few meV in strength, the Ising term G opens a large gap, and the anisotropic DM interaction actually tends to close the gap.

With its magnetic structure determined and exchange interaction strength extracted, we discuss the nature of the AFM order and the metal-insulator transition in $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$. First of all, we rule out the Fermi surface instability resulted spin density wave transition. The band structure and the corresponding Lindhard function of SrIrO_3 are calculated. The strongest response is at $(1/4, 1/4, 1/4)$, which differs from the ordering wave vector $(1/2, 1/2, 1/2)$ of G -type AFM structure [34].

The Slater versus Mott transition and also Slater-Mott crossover are much discussed recently [53,54], especially for iridates [55–57]. The proposed topological band structure from earlier theoretical work [13] and the large exchange interaction revealed in our work make $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$ a special case to consider. For $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$, it is obvious that the on-site Coulomb repulsion U alone is insufficient to open a clear and strong gap, evidenced by the paramagnetic semimetal behavior in SrIrO_3 . On the other hand, this does not necessarily put $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$ in a weakly correlated region. Our extracted large J value evidences significant electron correlation in this system.

As pointed out by earlier theoretical work [9,12] and also our calculations [34], the Ir-O octahedral rotation with the presence of strong SOC is very efficient in reducing the Ir $5d$ bandwidths. As a result, the bands near the Fermi level is merely ~ 300 meV in width in most of the reciprocal region in this three-dimensional system, and the Fermi energy is pinned near the nodal line [13]. Zeb and Kee [12] proposed that the pinned small Fermi pockets in return help to stabilize the nodes against magnetic fluctuations, unless the exchange interaction is strong to surpass a critical value. Our observed magnetic dispersion band top of ~ 130 meV and the extracted J of 45 meV suggest that this should be the case. As our magnetic dispersion is measured in a spin-diluted system with significant Sn^{4+} substitution, it is reasonable to speculate that the exchange interaction in the parent compound SrIrO_3 is even stronger [58]. Once the AFM ordering is established, it renormalizes the band structure [57], leading to a Slater metal-insulator transition in a general sense. In the case of $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$, such a renormalization is expected to be specially efficient since the AFM ordering breaks the time reversal symmetry and strongly modifies the dispersion slope near the nodal line region, where the Fermi surface is pinned. This conjecture explains the sensitivity of SrIrO_3 to the fine tunings mentioned in the introduction and the associated high T_N in the 130–280 K range. Given the above reasonable arguments, an important question remains: Why the pristine SrIrO_3 is a paramagnetic semimetal?

We speculate two possible scenarios. First, starting from a local picture and assuming that the nearest-neighbor

exchanges in pristine SrIrO_3 are highly frustrated by further neighbor magnetic interactions, the Sn^{4+} substitution disrupts the magnetic exchange pathways [59], with longer-range exchange interactions being more strongly suppressed than nearest-neighbor coupling [60]. Thus, a few percent Sn^{4+} tips the frustration balance and drives the system into long-range ordered AFM state, i.e., order by disorder.

Another more appealing explanation emphasizes the itinerancy of this non-Fermi-liquid system hosting the pinned Dirac node and electron correlation [61]. She and Bishop suggest that stronger electron correlation leads to longer-range Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which gives rise to enhanced frustration when the spins form a lattice [62]. The Sn^{4+} substitution breaks the mirror symmetry and releases the Fermi surface pinning, thereby turning off the frustration from the RKKY interaction in $\text{SrIr}_{1-x}\text{Sn}_x\text{O}_3$. Experimentally, strong itinerant electron scattering from localized spins was indeed observed in SrIrO_3 films on different substrates, appearing as resistivity curvature turnover around ~ 200 K [63]. Such consideration suggests SrIrO_3 to be a special case where both topological band structure and strong electron correlation are at play.

In summary, the magnetism in $\text{SrIr}_{0.8}\text{Sn}_{0.2}\text{O}_3$, an isovalent doped system from paramagnetic topological semimetal SrIrO_3 , is studied with resonant elastic magnetic x-ray scattering and resonant inelastic x-ray scattering. Its magnetic ordering is determined to be of G -type with the isospin pointing along the c direction. Together with the strong SOC, the observed strong magnetic dispersion up to ~ 130 meV with a large gap of 50 meV explains its uniqueness and robustness for spintronics applications in various forms. From the physics perspective, one might expect that such strong exchange interaction shall also drive an AFM ordering in the parent compound. In this regard, the persistence of the topological band structure, paramagnetism, and semimetallicity in pristine SrIrO_3 remains to be explored.

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Data availability. The data are available from the authors upon reasonable request.

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