Electronic superlattice revealed by resonant scattering from random impurities in Sr$_3$Ru$_2$O$_7$

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Resonant elastic x-ray scattering (REXS) is an exquisite element-sensitive tool for the study of subtle charge, orbital, and spin superlattice orders driven by the valence electrons, which therefore escape detection in conventional x-ray diffraction (XRD). Although the power of REXS has been demonstrated by numerous studies of complex oxides performed in the soft x-ray regime, the cross section and photon wavelength of the material-specific elemental absorption edges ultimately set the limit to the smallest superlattice amplitude and periodicity one can probe. Here we show – with simulations and REXS on Mn-substituted Sr$_3$Ru$_2$O$_7$ – that these limitations can be overcome by performing resonant scattering experiments at the absorption edge of a suitably-chosen, dilute impurity. This establishes that – in analogy with impurity-based methods used in electron-spin-resonance, nuclear-magnetic resonance, and Mössbauer spectroscopy – randomly distributed impurities can serve as a non-invasive, but now momentum-dependent probe, greatly extending the applicability of resonant x-ray scattering techniques.

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RuO₂ planes of this system. The problem of measuring REXS at the underlying instabilities of the Ru 4f electrons is known as a metal on the verge of ferromagnetism, due to the presence of strong ferromagnetic fluctuations. More recently, magnetic field tuned quantum criticality and electronic nematic fluid behavior have been proposed for this compound and associated with a metamagnetic transition. In Sr₃(Ru₁₋ₓMnx)₂O₇, it has been shown that Mn impurities display an unusual crystal-field level inversion, due to the interplay between localized Mn 3d and delocalized Ru 4d–O 2p valence states. Upon lowering the temperature, a Mott-type metal-insulator (MIT) phase transition driven by electronic correlations has been observed for 5% Mn substitution at T_{MIT} ≈ 50 K, and as progressively higher T_{MIT} upon increasing the Mn concentration. Following the onset of local antiferromagnetic (AF) correlations at T_{MIT}, for Mn concentrations x = 0.025–0.2 an unusual long-range AF superstructure emerges at T_{order} < T_{MIT}^{16–18}, with magnetic wave vector Q = (1/4, 1/4, 0) as revealed by electron neutron scattering experiments. Since pure Sr₃Ru₂O₇ does not show any long-range magnetic order, it is clear that the latter is induced by the S = 2, 3d-Mn³⁺ impurities; we emphasize however that the ordering – and particularly the Q = (1/4, 1/4, 0) magnetic wave vector – are independent of the precise 5–10% Mn concentration, indicating that the role of Mn is merely that of triggering and/or stabilizing a tendency already incipient in the parent compound Sr₃Ru₂O₇.

Results

Choice of material system. As a study case for the electronic scattering from random impurities, let us introduce Sr₃(Ru₁₋ₓMnx)₂O₇, in which Mn is being randomly substituted on the Ru site within the RuO₂ planes of the parent compound (see Fig. 1). Pure Sr₃Ru₂O₇ is known as a metal on the verge of ferromagnetism, due to the presence of strong ferromagnetic fluctuations. More recently, magnetic field tuned quantum criticality and electronic nematic fluid behavior have been proposed for this compound and associated with a metamagnetic transition. In Sr₃(Ru₁₋ₓMnx)₂O₇, it has been shown that Mn impurities display an unusual crystal-field level inversion, due to the interplay between localized Mn 3d and delocalized Ru 4d–O 2p valence states. Upon lowering the temperature, a Mott-type metal-insulator (MIT) phase transition driven by electronic correlations has been observed for 5% Mn substitution at T_{MIT} ≈ 50 K, and as progressively higher T_{MIT} upon increasing the Mn concentration. Following the onset of local antiferromagnetic (AF) correlations at T_{MIT}, for Mn concentrations x = 0.025–0.2 an unusual long-range AF superstructure emerges at T_{order} < T_{MIT}^{16–18}, with magnetic wave vector Q = (1/4, 1/4, 0) as revealed by electron neutron scattering experiments. Since pure Sr₃Ru₂O₇ does not show any long-range magnetic order, it is clear that the latter is induced by the S = 2, 3d-Mn³⁺ impurities; we emphasize however that the ordering – and particularly the Q = (1/4, 1/4, 0) magnetic wave vector – are independent of the precise 5–10% Mn concentration, indicating that the role of Mn is merely that of triggering and/or stabilizing a tendency already incipient in the parent compound Sr₃Ru₂O₇.

Resonant scattering at the Ru L-edge. We have performed REXS experiments on Sr₃(Ru₁₋ₓMnx)₂O₇ on both Ru and Mn L₂,₃ edges, for a range of Mn concentrations. On a 10% Mn-substituted sample at 20 K, REXS at the Ru L-edge detects a forbidden superlattice diffraction peak appearing at Q = (1/4, 1/4, 0). The Ru L-edge measurements provide a critical information: while the Q = (1/4, 1/4, 0) modulation observed by neutron scattering points toward an electronic modulation or order in the system^{15–18}, one needs to exclude that this is not merely associated with a Mn clustered phase or a structural ordering of the Mn impurities, in which case the host Ru valence electrons would not participate. As revealed by the photon-energy dependence of the (1/4, 1/4, 0) peak in Fig. 2(a), Mn than Ru absorption edge. This paves the way to the use of dilute, substitutional impurities as a ‘marker’ of electronic ordering phenomena in other oxides and complex systems.
the intensity enhancement observed at both \(L_{2,3}\) Ru absorption edges – with onset at \(T_{\text{order}}\) of the AF phase as shown in Fig. 2(b) – demonstrates that the new order is electronic and not chemical/structural, and pertains to the system as a whole. In addition, the magnetic character of the order is confirmed by the azimuthal dependence of the order parameter in REXS experiments. This sets the stage to investigate the possibility of studying the magnetic superstructure by REXS measurements performed at the Mn-impurity L edge.

**Resonant scattering at the Mn L-edge.** At the Mn \(L_{2,3}\)-edge, REXS measurements were performed on 5 and 10% Mn-substituted \(\text{Sr}_3\text{Ru}_2\text{O}_7\) and indeed exhibit superlattice diffraction peak. For clarity, only the data from 10% Mn substitution are shown in Fig. 3(a), which presents the energy dependence of the Q = (1/4, 1/4, 0) superlattice peak intensity at the Mn \(L_{2,3}\) resonance profile at \(T = 20\) K (its full 2-dimensional reciprocal space map is shown in the inset). Also in the Mn case the forbidden superstructure reflections are detected only below \(T_{\text{order}}\); and as for the Ru L-edge measurements, the magnetic character of the Mn L-edge superlattice peak has again been confirmed by the azimuthal dependence of the order parameter. The magnetic order progressively gain strength upon reducing temperature and increasing Mn concentration. This is shown as a function of temperature in Fig. 3(b) based on the raw data for the (1/4 + \(\Delta q\), 1/4 + \(\Delta q\), 0) momentum scans measured on the 10% Mn sample at 641 eV (\(L_{2}\)-edge). A summary for both 5 and 10% Mn substituted samples is presented in Fig. 3(c), where the integrated peak intensities of the longitudinal momentum scans (1/4 + \(\Delta q\), 1/4 + \(\Delta q\), 0) are plotted versus temperature. Following the experimental evidence given above, in the remainder of the paper we will demonstrate – both analytically and computationally – how resonant scattering at the impurity edge can be observed, and what deeper insights might this provide.

**Scattering from random impurities.** To understand how it is possible to observe superlattice diffraction signal from random impurities, we can start from the idealized case of 100% Mn substitution. Fig. 4 presents such \(\text{MnO}_2\) square lattice with an up-up-down-down zig-zag spin order (red/up, blue/down). This magnetic pattern satisfies the symmetries obtained in the azimuthal dependence measured by REXS in Ref. 17. For the 100% magnetic pattern satisfies the symmetries obtained in the up-up-down-down zig-zag spin order (red/up, blue/down). This demonstrates that the new order is electronic and not chemical/structural, and pertains to the system as a whole. In addition, the magnetic character of the order is confirmed by the azimuthal dependence of the order parameter. This sets the stage to investigate the possibility of studying the magnetic superstructure by REXS measurements performed at the Mn-impurity L edge.

\[
\begin{align*}
\Delta q (x10^{-3}) & \quad -8 & \quad -6 & \quad -4 & \quad -2 & \quad 0 & \quad 2 & \quad 4 & \quad 6 & \quad 8 \\
\text{Temperature (K)} & \quad 20 & \quad 40 & \quad 60 & \quad 80 \\
\text{Integrated Intensity} & \quad 0 & \quad 0.1 & \quad 0.2 & \quad 0.3 & \quad 0.4 & \quad 0.5 & \quad 0.6 & \quad 0.7 & \quad 0.8 & \quad 0.9 & \quad 1
\end{align*}
\]

**Figure 3** (a) Mn resonance profile for the (1/4, 1/4, 0) superlattice diffraction peak measured at 20 K on \(\text{Sr}_3(\text{Ru}_{1-x}\text{Mn}_x)_2\text{O}_7\) with \(x = 0.1\) (the arrow at 641 eV indicates the energy used in the Mn-edge REXS experiments). Inset: full reciprocal space map of the superlattice peak. (b) Temperature dependence of the Mn \(L_2\) edge (1/4 + \(\Delta q\), 1/4 + \(\Delta q\), 0) momentum scans for \(x = 0.1\). (c) Temperature dependence of the integrated intensity of the (1/4, 1/4, 0) peak for 5% and 10% Mn-substitution.

\[
A(Q) = \sum_{R_n} f_{R_n} e^{iQ \cdot R_n} \delta_{R_n,R_{\text{Mn}}},
\]

where \(R_n\) denotes the atomic positions, \(f_{R_n}\) is the atomic form factor, and \(\delta_{R_n,R_{\text{Mn}}}=1\) at the Mn impurity sites and zero elsewhere. It is clear from the above expression that while \(\delta_{R_n,R_{\text{Mn}}}\) removes all the non-Mn sites, the phase relation between the Mn sites – encapsulated in the exponential term – remains the same; hence, the signal from these sites interferes constructively for the same \(Q\) superlattice vector. It follows that the scattering intensity from random impurities can be nonzero and is proportional to the square of the number \(N_{\text{Mn}}\) of Mn atoms, \(I(Q) = |A(Q)|^2 \cdot N_{\text{Mn}}^2\). This demonstrates that by reducing the number of Mn impurities in \(\text{Sr}_3(\text{Ru}_{1-x}\text{Mn}_x)_2\text{O}_7\) one only loses signal proportionally to the reduced occupation of the Mn sublattice, and the rest of the Mn atoms still contribute to the diffraction superstructure as long as they have the same \(Q = (1/4, 1/4, 0)\) and \((-1/4, -1/4, 0)\) phase relationship. Since the structure factor is the Fourier transform of the two-point correlation function, removing Mn atoms also does not change the correlation length.

**Comparison between Ru and Mn edge.** Further insights can be obtained from the direct comparison of the temperature
dependence of Ru- and Mn-edge magnetic peak intensity, and corresponding correlation length (defined as $2\pi$/FWHM, where FWHM is the full-width half-maximum of the momentum scans). This is presented in Fig. 5(a,b) for the 10% Mn-substituted sample. Fig. 5(a) shows an intensity onset $\sim 5$ K higher on Ru than Mn (i.e., 65 versus 60 K), as well as a different shape of the temperature dependence (with a steeper increase at the Ru edge). This is accompanied by the smooth increase of Ru- and Mn-edge correlation lengths upon reducing the temperature [Fig. 5(b)]. Overall, the Mn order parameter is lagging behind the Ru one. The further decrease of the correlation length below 30 K has also been seen in reentrant spin glasses, due to the interplay of competing interactions and disorder. It is important to note that the order is never truly long-range at any temperature, with the highest value for the intrinsic correlation length being only 160 nm, as determined at both Ru and Mn L edges; in addition, as shown in Ref. 17, the correlation length is even shorter for lower doping levels.

Overall, these data indicate that the antiferromagnetic correlations first appear in the RuO$_2$ islands surrounding the Mn impurities; however, since initially these islands do not overlap, the Mn impurity spins are not correlated, and a somewhat incoherent magnetic signal is observed only at the Ru edge. When these islands begin to overlap, the RuO-mediated exchange interaction can energetically favor a coherent spin arrangement between the islands; as a result, the Mn impurities will interfere constructively and long-range order is observed at both edges. This behavior cannot be explained based on the assumption of a 100% spin correlation between Mn impurities as in Fig. 4, and should instead be discussed in the context of a percolative phase transition, with an effective percolation threshold achieved at $\lesssim 5\%$ Mn concentration.

This case is presented in Fig. 6(a), which shows a 64 $\times$ 64 RuO$_2$ lattice in real space, with 5% Mn sites each inducing a 4 $\times$ 4 unit of the magnetic zigzag stripe pattern. As in Fig. 4, blue and red squares in Fig. 6(a) represent up and down spins along the c-axis, while the white patches are regions of the RuO$_2$ plane where no magnetism has been induced. Here, however, no correlation between the Mn spins has been imposed ($T_{MTT} > T > T_{oder}$); the position of the Mn impurities and their corresponding spins are shown in Fig. 6(b). Even though there is no spin correlation between the 4 $\times$ 4 magnetic islands, the reciprocal space map of the scattering intensity from the Ru sites in Fig. 6(e) shows extremely broad and weak scattering peaks stemming from the onset of short-range order below $T_{MTT}$. Due to the lack of correlation between the Mn spins, no Mn-edge peaks are visible in Fig. 6(f). The situation is very different for correlated Mn spins ($T < T_{oder}$); this is shown in Fig. 6(c,d), which presents the same sequence of Fig. 6(a,b) but now for an average ‘magnetically correlated’ domain of 68 $\times$ 68 Å$^2$ ($\sim 16 \times 16$ lattice sites), as observed experimentally once long-range order is established below $T_{oder}$. As shown in Fig. 6(g,h), the superlattice ($\pm 1/4, \pm 1/4, 0$) peaks are clearly discernible above the noise – with a well defined correlation length – at both Ru and Mn edges.

In addition to demonstrating that impurity-edge superlattice peaks can be measured even in case of a complex percolative phase transition, the results in Fig. 6 reveal the origin of the difference in temperature evolution of Ru- and Mn-edge intensity in Fig. 5(a).
can be summarized as follows: the Ru-edge intensity has an earlier onset and also faster rise than that of Mn. Our modelling shows that in the crossover from $T_{\text{MIT}}$ to $T_{\text{order}}$, Mn impurities begin to get correlated, which defines the onset of the Mn-edge intensity; however, the number of magnetically correlated sites increases much faster for Ru than Mn, resulting in a much faster REXS intensity rise. Eventually, in the saturation regime, the correlation length is the same at both edges since this is defined by size of the macroscopically magnetically-ordered domains, which is dependent on the Mn-concentration.

**Discussion**

This study proves that REXS signal from dilute and randomly distributed impurities can give rise to superlattice diffraction peaks, which can be used to probe the electronic order in an element sensitive manner. Interestingly, the underlying spin order in $\text{Sr}_3(\text{Ru}_{1-x}\text{Mn}_x)_2\text{O}_7$ is more difficult to detect at the Ru than at the Mn $L$-edge, where a much larger resonance is observed. In the case of 10% Mn substitution, as evidenced by the signal-to-noise ratio and corresponding error bars in Fig. 3(c), the scattering signal from Ru is already much weaker than that of Mn, although Ru occupies 90% of the lattice sites. Once Mn substitution is reduced to 5%, the Ru signal is swamped in the background while REXS at the Mn $L$-edge still is too small to be detectable at the strong L$_{2,3}$ resonances, because of the too long photon wavelength at those energies. Substitution with an element of the same valence and similar size – but with a much higher absorption-edge resonance energy, corresponding to much shorter wavelength photons – could be used to gain access to the electronic ordering, enabling the study of the associated phases and phase transitions. If in addition one can scatter from both impurity and host atoms selectively, this approach would become extremely powerful in the study of spatially inhomogeneous orders in bulk, surfaces, interfaces, and heterostructures. At an interface or heterostructure, new electronic states can occur due to interaction between interfacial atoms; the role of electronic order and interaction across the interface might be clarified by element selective and/or impurity-edge REXS performed right at the interfacial region.

Finally, these ideas and approaches can also be generalized to other techniques, such as e.g. resonant inelastic x-ray scattering (RIXS) and time-resolved resonant x-ray scattering. With the development of brighter x-ray sources and higher resolution momentum-resolved capabilities, one can take advantage of impurity edge RIXS experiments to study momentum-resolved elementary excitations (photons, magnons, orbitons), involving host and impurity degrees of freedom and also theoretical predictions on subtle issues like whether noninteracting atoms contribute coherently to the inelastic scattering cross section$^{22}$. In addition, with the availability of coherent two-color time-resolved probes at the next generation free-electron laser sources, such as LCLS, one can envision experiments performed simultaneously at the impurity and host edges, to disentangle the causal relationships between lattice, charge, spin, and orbital orders in a variety of complex materials.

**Methods**

**Light scattering experiments.** REXS measurements were performed at beamlines 8.0.1 at ALS in Berkeley (Mn $L$-edges) and KMC-1 at BESSY in Berlin (Ru $L$-edges). In both cases we used a two-circle ultra-high-vacuum diffractometer in horizontal scattering geometry, with the incident photon beam polarized parallel to the...
diffraction plane (\(s\)). The scattered signal contained polarization components both parallel (\(\parallel\)) and perpendicular (\(\perp\)) to the diffraction plane. The correlation lengths in Fig. 5 are defined as 2FWHM, where FWHM is the full-width half-maximum of the Lorentzian fit to the momentum scans (with \(x\)-axis converted to \(\AA^{-1}\) units). At the Mn and Ru \(L\) edges, our angular resolutions are \(\leq 1^\circ\) and 0.4°, respectively.

**Sample preparation.** \(\text{Sr}_3(\text{Ru}_{1-x}\text{Mn}_x)\text{O}_7\) single crystals grown by the floating zone technique\(^{67}\) were cut and polished along the (110) direction. The samples were mounted on cryogenic manipulators, which allow a polar (\(\theta\)) and azimuthal (\(\phi\)) angle rotation of the sample about the scattering vector, in the temperature range 20–300 K. Note that the diffraction peaks are indexed with respect to the undistorted tetragonal \(I\bar{4}m\bar{m}\) unit cell with axes along the RuO bond directions (\(a_0 = b_0 \geq 3.9a\)).

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