

Synthesis and defect control of Sr₂IrO₄ films by reactive sputtering

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Sr₂IrO₄ (Sr214) and related iridates have emerged as key platforms for fundamental correlated-electron physics and for potential applications such as magnonics. Here, we report the epitaxial growth of high-quality Sr214 thin films using reactive off-axis sputtering. Conventional pulsed-laser deposition of Sr214 suffers from limitations arising from the volatility and decomposition of iridates, which often result in parasitic Ruddlesden–Popper phases and iridium vacancies. By employing sputtering, we mitigate these issues and achieve stable growth conditions that allow precise mapping of the phase diagram. Systematic variation of the growth temperature reveals that Sr214 stabilizes within a window between 825 °C and 1050 °C, with optimal crystallinity and thickness obtained at 840 °C. Raman spectroscopy provides sensitive diagnostics of iridium vacancies, with defect-induced phonon modes and peak intensity ratios correlating strongly with the *c*-axis lattice parameter. Our results establish reactive sputtering as a robust route for integrating Sr214 into oxide electronic and magnonic device architectures.

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I. INTRODUCTION

Owing to the interplay between exchange interactions, crystalline electric fields, and spin-orbit coupling, iridates and other 5d-electron compounds have become important platforms for research on magnetism and correlated-electron physics [1,2]. The Mott-insulating square-lattice antiferromagnet Sr₂IrO₄ (Sr214) is a key model compound for this research field, because of its simple layered perovskite structure and electron configuration (5d⁵). The antiparallel spin (*S* = 1/2) and orbital (*L* = 1) angular momenta of the Ir⁴⁺ ions can be described in terms of a pseudospin 1/2, so that the magnetic Hamiltonian is formally analogous to that of La₂CuO₄, the progenitor of a prominent family of high-temperature superconductors. The presence of an unquenched orbital magnetization in Sr214 gives rise to subtle differences in the physical properties of both compounds [3–8] and their response to doping [9–15], which have been the subject of considerable research.

A particular consequence of the orbital component of the iridium magnetic moments is a strong and unusual interaction

between the magnetic order and excitations of the crystal lattice [16–19]. The pseudospin-lattice coupling makes the magnetic properties highly sensitive to strain, which has been demonstrated by growing epitaxial Sr214 films on substrates with varying degrees of lattice mismatch [4,20–22]. The ability to synthesize Sr214 thin films has also opened up the possibility of exploring interfacial reconstructions and device functionalities [23–31] in Sr214-based heterostructures. However, it is challenging to synthesize high-quality Sr214 films via physical vapor deposition (PVD) because of multiple chemical constraints.

First, Sr214 is an end member (*n* = 1) of the Ruddlesden–Popper phase with the chemical formula Sr_{*n*+1}Ir_{*n*}O_{3*n*+1}, which includes SrIrO₃ (Sr113) as another end member with *n* = ∞. These phases have very similar formation energies, resulting in narrow and closely spaced growth windows in parameter space [32]. Even small changes in growth parameters, such as cation ratio, temperature, or pressure, may shift the growth window, leading to the formation of parasitic phases.

Second, iridium and its oxides exhibit peculiar thermodynamic properties. Evaporation of elemental iridium requires extremely high temperatures above 2500 K. Iridium oxides, on the other hand, are volatile, yet may decompose into iridium and oxygen under vacuum [33]. This poses a challenge for widely used PVD techniques such as pulsed-laser deposition (PLD), often resulting in Sr214 films with defects such as iridium vacancies and unintended Ruddlesden–Popper phases.

The main issue in PLD is the photothermal ablation process involved in vaporizing the target. In ideal photoablation, the target is stoichiometrically ablated; however, typical PLD processes involve photothermal ablation that leads to melting

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of the target because of elevated temperatures. When iridium oxides are exposed to such high temperatures in vacuum, they decompose into iridium, leading to metallization of the target surface. This metallization continuously alters the ablation conditions, thereby seriously compromising the growth yield. This is a well-known challenge in PLD, and ruthenium oxides share the same traits with iridium oxides; therefore, both suffer from similar issues [34].

To date, PLD has been the most widely used method to make single-phase Sr214 epitaxial films [32,35–38]. However, iridium deficiency in Sr214 thin films has been rarely discussed in the literature in contrast to the closely related metallic Sr–Ru–O compounds [39–42], where ruthenium vacancies act as scattering centers and degrade physical properties such as ferromagnetism and superconductivity. The volatility of IrO₃ has been mostly discussed in the context of pyrochlore rare-earth iridates RE₂Ir₂O₇ [43,44], because parasitic phases, RE₃IrO₇ and rare-earth oxides, form in the iridium-poor regime. Since Sr214 is an end member of the Ruddlesden–Popper series, the center of attention was mainly on preventing iridium-rich phases [32,37]. Nevertheless, it is reasonable to assume that iridium deficiency may exist in Sr214 thin films, given that its chemical properties are analogous to those of ruthenates.

Another major PVD method for growing complex oxide thin films is molecular-beam epitaxy (MBE). The gentle evaporation process and the ability to control stoichiometry during synthesis have facilitated the growth of high-quality thin films [45]. MBE has been used to produce stoichiometric Sr214 thin films, albeit the number of reports is limited [46–48]. This can be attributed to the fact that the number of oxide MBE systems is far smaller than that of PLD systems, and that evaporating elemental iridium is challenging because of its extremely low vapor pressure even at 2000 °C, which is a typical maximum temperature achievable with a thermal evaporation cell.

Sputtering relies on physical collisions of process gas ions with the target surface, rather than thermal processes. Atoms and molecules are knocked out of the target by momentum transfer during sputtering. Thus, metals that cannot be thermally evaporated because of extremely high evaporation temperatures can still be deposited via sputtering. Nevertheless, sputtering has traditionally been used mainly for growing thin films of metals and alloys. In recent years, an increasing number of complex oxides have been synthesized using sputtering [49–52]. Especially, various ruthenium oxides have been successfully synthesized using reactive sputtering, overcoming their chemical complications [39,42,53]. Nevertheless, there have been no reports demonstrating the successful growth of Sr214 films.

In this work, we report the epitaxial growth of high-quality Sr214 thin films in an argon–oxygen atmosphere using a reactive off-axis sputtering technique. Here, the low energy of sputtering involved in the vaporization process mitigates the problems caused by the instability of iridium oxides. Owing to the stability of the sputtering process, we were able to accurately determine the boundaries of the phase diagram and identify optimized growth parameters. Within the growth window of Sr214, we observed slight changes in the *c*-axis lattice parameter, which are related to iridium vacancies. By analyzing these films, we further discovered that Raman

spectroscopy provides clear signatures of iridium vacancies that commonly elude simple diagnostics.

II. EXPERIMENTAL DETAILS

A series of strontium iridate thin films was grown using a home-built sputtering system equipped with a stoichiometric Sr214 pressed-powder target supplied by Toshima Manufacturing Co. The target, with a diameter of 33 mm, was positioned off-axis relative to the substrate, separated by a distance of 12 cm. The radio frequency power supplied to the target was 25 W. All films were deposited on (LaAlO₃)_{0.3}-(SrAl_{0.5}Ta_{0.5}O₃)_{0.7} (LSAT) (001) single crystalline substrates (CrysTec GmbH), which impose a compressive strain of approximately −0.4%. Prior to deposition, each substrate was mounted on a pure Ni block with carbon paste and heated with an infrared laser. The growth was performed over a broad substrate temperature ranging from 650 °C to 1050 °C in an Ar/O₂ atmosphere with a 1:1 ratio and a total pressure of 0.1 mbar. These values of oxygen content and total pressure were found to be optimal for Sr214 film growth. X-ray diffraction (XRD) with a Cu K- α source ($\lambda \approx 1.5406$ Å) was used to identify the crystalline phases and evaluate the structural quality of the films. The film thicknesses were determined using X-ray reflectivity (XRR).

X-ray photoelectron spectroscopy (XPS) was performed with a Kratos Axis Ultra equipped with a monochromatic Al K- α x-ray source. The charge neutralizer was used to compensate for surface charging. The Ir/Sr ratio was determined from the Ir 4*f* and Sr 3*d* peak areas obtained from the survey spectra measured with a pass energy of 80 eV, after Tougaard background subtraction and normalization by the respective sensitivity factors as implemented in CasaXPS [54].

Raman spectra were acquired with a WITec alpha300-R confocal Raman microscope using a 100x super-long-working-distance objective and a WITec UHTS VIS-optimized spectrometer equipped with a 1800 grooves/mm blazed grating. The excitation laser was a diode-pumped solid-state laser with a wavelength of $\lambda = 532$ nm. Experiments were conducted at room temperature in backscattering geometry with circularly polarized light (bidirectional $\lambda/4$ waveplate) propagating along the crystallographic *c* axis and without an additional analyzer to collect the full Raman response within a single measurement. Using the Porto notation, we denote this as $z(\sigma^+U)\bar{z}$. To allow for a clearer comparison, we also measured the Raman spectrum of a bulk Sr214 single crystal grown by the flux method [55,56].

III. RESULTS AND DISCUSSION

Figure 1 shows XRD curves of samples grown at different temperatures. Here, we refer to the Ruddlesden–Popper phases of strontium iridates with simple abbreviations of the form Sr(*n* + 1)(*n*)(3*n* + 1), following the example of Sr214. As shown in Fig. 1(a), high temperatures in the range 825–1050 °C stabilize the pure Sr214 phase.

A reduction of the growth temperature below 825 °C results in the formation of the Sr327 phase [Fig. 1(b)], with a minor contribution from Sr214 marked by diamonds. The broad diffraction peaks and the absence of thickness fringes

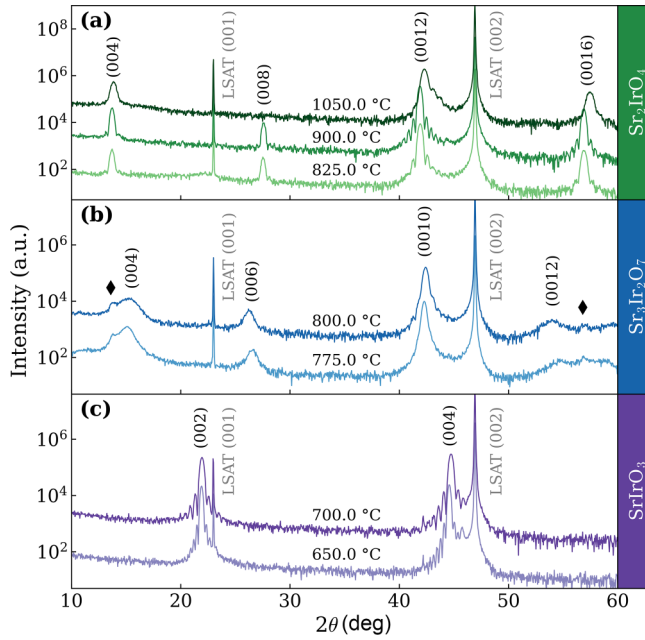


FIG. 1. XRD patterns of films grown at different temperatures on LSAT (001). (a) Temperature window for stabilization of single-phase Sr_{214} (825–1050 °C). (b) Mixed-phase growth of Sr_{327} and Sr_{214} (775–800 °C). The diamonds indicate the peak positions of Sr_{214} . (c) Formation of the single Sr_{113} phase (650–700 °C).

suggest a high density of defects, which indicates that the present condition using the Sr_{214} target is not optimal for the growth of Sr_{327} . Unlike the Sr_{214} and Sr_{113} , which are the end members of the Ruddlesden–Popper phases, there is a large number of possible phases in the vicinity of Sr_{327} . Therefore, intergrowth of multiple phases occurs under conditions, which degrades the crystal quality.

A further reduction in temperature stabilizes the Sr_{113} phase, as shown in Fig. 1(c). Notably, Sr_{113} films exhibit smooth surfaces despite the off-stoichiometric deposition. There is a report that Sr_{113} thin films can be grown by PLD using a Sr_{214} target [57], and it is not uncommon for perovskites to host a large amount of B-site vacancies. Stabilization of the 113 phase at low temperatures has also been reported in other Ruddlesden–Popper phases, such as the Sr–Ru–O system. It is attributed to their high tolerance to B-site vacancies, and the formation of a flat SrO layer at the surface [58].

In order to identify the optimal growth parameters for Sr_{214} , we explored various temperatures within the growth window. Figure 2(a) presents XRD specular scans for a series of samples grown with small temperature increments between 825 °C and 1050 °C. In this temperature range, most samples show all the out-of-plane reflections. Well-defined Laue fringes around the (0 0 12) and (0 0 16) reflections indicate a smooth surface, a sharp interface, and high crystallinity.

All the samples listed were grown under identical conditions, except for the growth temperature. Nevertheless, the variation in thickness obtained from XRR measurements is large (see Table I). The thickest sample (grown at 840 °C) is nearly twice as thick as the thinnest one (grown at 1000 °C). The high growth rate suggests that 840 °C represents the optimal growth temperature for Sr_{214} . Notably, the sample grown at 840 °C also exhibits the clearest Laue fringes around the (0 0 4) reflection. Since Laue fringes are superimposed on Bragg reflections from thin films, their clear visibility indicates that the (0 0 4) reflection from this sample is significantly stronger than those of the others. As a result, most of the oscillatory features rise above the noise level of the measurement setup, further underscoring the excellent crystalline quality of this sample.

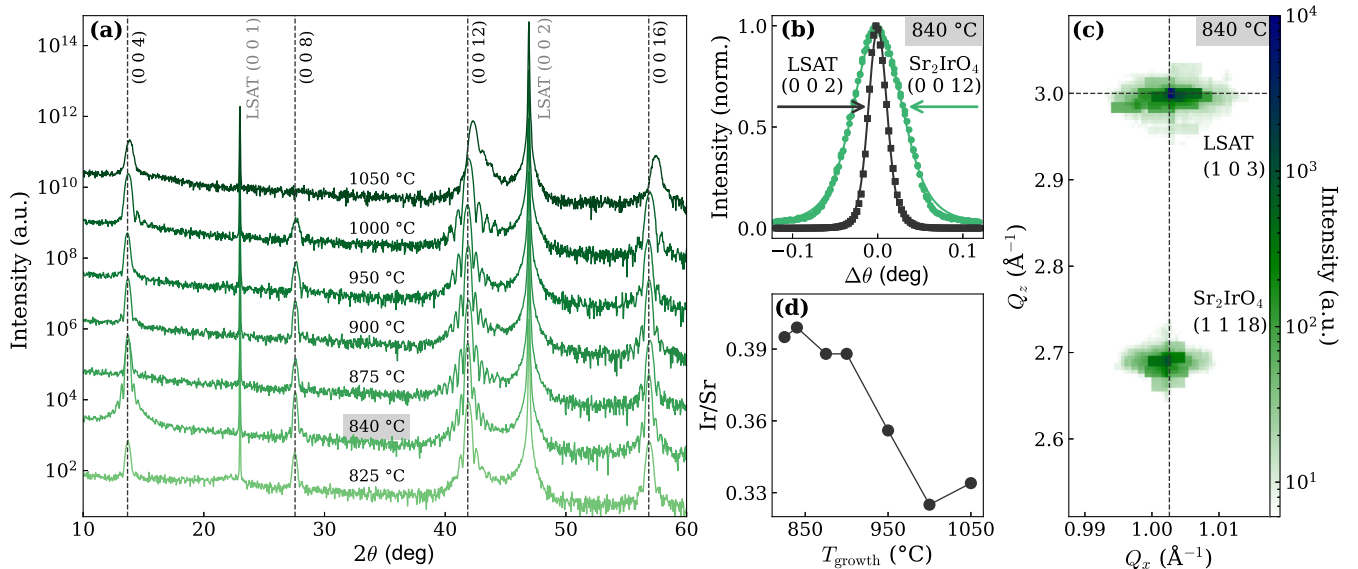


FIG. 2. (a) XRD patterns of Sr_{214} films grown on LSAT (001) at different temperatures. The dashed lines indicate the (0 0 *l*) reflection positions of the highest quality film in the series (840 °C). (b) Rocking curves of the Sr_{214} (0 0 12) and LSAT (0 0 2) reflections for the film grown at 840 °C. (c) Reciprocal-space map around the Sr_{214} (1 1 18) and LSAT (1 0 3) reflections for the same film as in (b). (d) Ratio of Ir and Sr atomic concentrations extracted from XPS measurements as a function of growth temperature.

TABLE I. List of thin film parameters, arranged in order of increasing film growth temperature.

T_{growth} (°C)	Thickness (nm)	FWHM _{film} (deg)
825	18.7	0.063
840	24.3	0.069
875	20.3	0.069
900	20.95	0.068
950	18.4	0.070
1000	13.8	0.069
1050	14	0.071

The high quality of the film grown at 840 °C is further confirmed by the rocking curve and the reciprocal-space map, shown in Figs. 2(b) and 2(c), respectively. The narrow peak from the rocking curve around the (0 0 12) reflection [Fig. 2(b)] exhibits a full width at half maximum (FWHM) of approximately 0.069°, indicating a well-aligned out-of-plane film orientation. FWHM values of all rocking curves are summarized in Table I. A reciprocal space map [Fig. 2(c)] was measured around the in-plane peaks of Sr214 (1 1 18) and LSAT (1 0 3). The two reflections share the same Q_x , indicating that the film, grown coherently on the substrate, remains fully strained. Despite the high growth temperature, which can sometimes lead to rapid strain relaxation, the film stays fully strained up to a thickness of 24.3 nm.

Further analysis of the XRD patterns [Fig. 2(a)] reveals a shift of some peaks toward higher angles, which becomes more pronounced in thin films grown at higher temperatures. The shift indicates a shrinkage of the c -axis lattice constant and is most likely caused by iridium vacancies, as iridium oxides become volatile at the growth temperatures used here.

To track the trend in the Ir/Sr ratio, we carried out XPS. It should be noted that the peak shapes in XPS spectra from epitaxial transition-metal-oxide films can deviate from the standard XPS line shapes, which may introduce inaccuracies in quantitative analysis [59]. To minimize these effects, the Ir/Sr ratio was used as an indicator rather than an exact value for stoichiometry, as shown in Fig. 2(d). The analysis reveals that the film grown under optimal deposition conditions exhibits the highest iridium concentration, whereas the Ir/Sr ratio shows a slight decrease up to 900 °C and then rapidly drops upon further increase of the growth temperature. A similar phenomenon was observed in Sr₂RuO₄ thin films, where the c -axis lattice constant shrinks upon growth at higher temperatures [39]. The authors attributed the origin of the shrinkage of the c -axis lattice constant to the formation of ruthenium vacancies as well.

From the perspective of defect formation, the most disordered sample is the one grown at 1050 °C, the boundary of our growth window for Sr214. In this sample, the out-of-plane peaks are broader and somewhat asymmetric [Fig. 2(a)]. In addition, the significant peak shifts and the absence of the (0 0 8) reflection clearly indicate that the structure of the sample is degraded by a substantial concentration of iridium vacancies, whose formation is thermodynamically preferred at elevated temperatures.

TABLE II. Mode assignment of the observed peaks in the Raman spectra in accordance with literature [62,63]. The values were obtained from the fit of the 840 °C spectrum.

Label	Symmetry	$\omega_{i/j}$ (cm ⁻¹)	Mode description
M1	A _{1g}	181	Sr stretching
M2	A _{1g}	265	Ir–O–Ir bending
M3	B _{2g}	393	O displacement in IrO ₆ -plane
M4	A _{1g}	552	O stretching
M5	A _{1g}	703	O displacement in IrO ₆ -plane

We carried out Raman spectroscopy to further investigate the Sr214 thin films. Raman spectroscopy in a confocal geometry is a powerful technique for probing the vibrational and electronic features of thin films [60]. The use of a high-magnification lens in the confocal setup effectively suppresses the substrate signal, enabling the acquisition of Raman spectra primarily from the thin film. Previous studies have identified Raman spectroscopy as an effective method for probing point defects, such as oxygen and cation vacancies, in thin films [39,61].

Figure 3(a) presents dark-subtracted Raman spectra of the films exhibiting the pure Sr214 phase, as confirmed by XRD analysis. All spectra show contributions from both Sr214 and the substrate. Peaks corresponding to the phonon modes of Sr214 are marked by red triangles. The three sharp peaks at 151, 470, and 879 cm⁻¹, along with a broad hump at 603 cm⁻¹, correspond to the phonon modes of the LSAT substrate.

The spectrum from the sample grown at the optimized temperature closely resembles that of a high-quality single crystal [Fig. 3(a)] and is consistent with previously reported spectra of bulk Sr214 in the literature [18], confirming the optimal growth temperature. As the growth temperature deviates from the optimum, the intensity of the Sr214 phonon peaks decreases. Notably, the intensities of the peaks at 190 and 240 cm⁻¹ diminish more rapidly than those of the others, suggesting that they are highly sensitive to defects. However, extra care must be taken when interpreting confocal Raman spectra of thin films, as the optical penetration depth exceeds the film thickness. Consequently, the peak intensities are proportional to the film thickness, making it essential to compare peak ratios rather than absolute intensities among different samples.

To extract the integrated peak intensities quantitatively, the spectra were fitted to a series of Fano profiles on top of a broad background. Figure 3(b) shows the results of this fit for a specific film. Individual components are highlighted, and the assignment of peaks M1–M5 to various Sr214 phonon modes is listed in Table II.

Before quantitative analysis of the Raman spectra, one apparent change is noticeable in the sample with the highest defect concentration. The samples grown at 1000 °C and 1050 °C exhibit a depletion of the M2, M3, and M5 peaks, along with the emergence of new phonon modes near 320 cm⁻¹ and 750 cm⁻¹ [see Figs. 3(c) and 3(d)]. When the local symmetry is lowered by defects, IR-active phonons or backscattering-forbidden modes can appear in the Raman

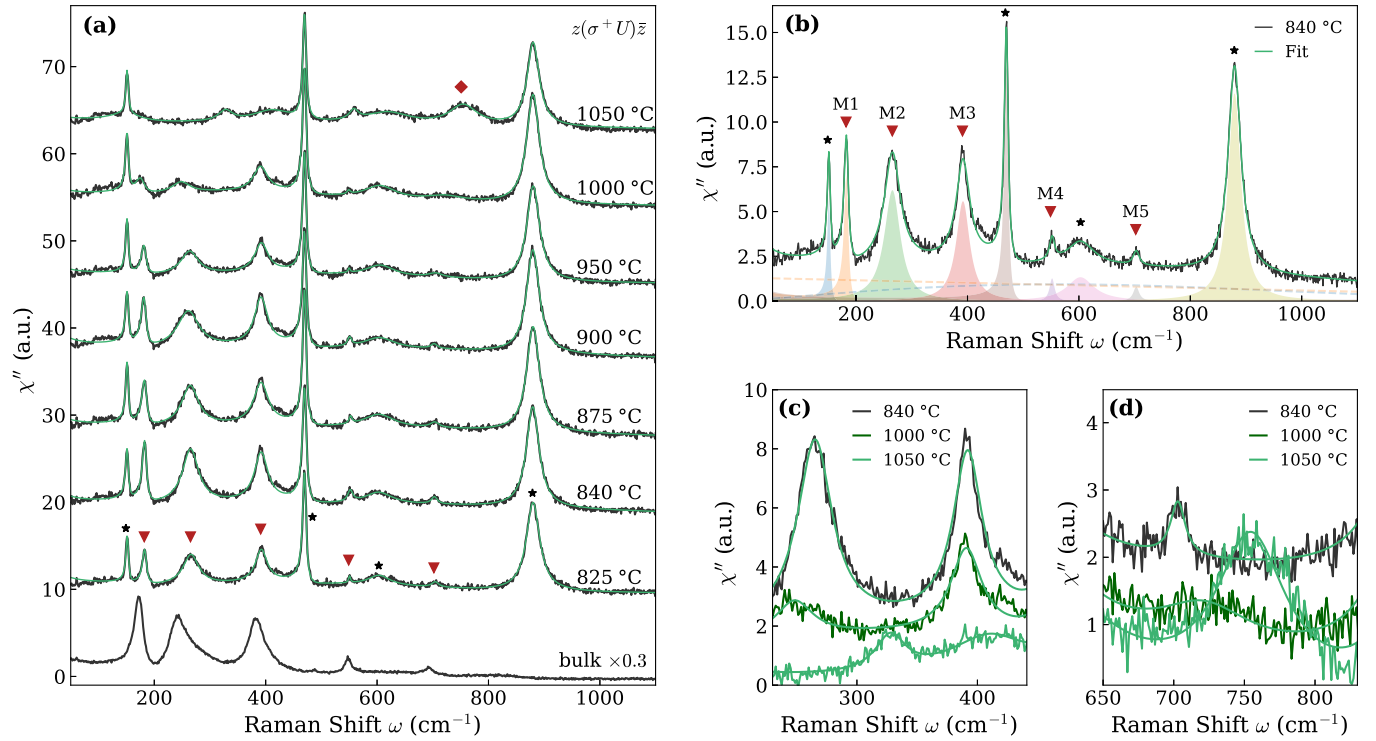


FIG. 3. (a) Room-temperature Raman spectra for Sr214 thin films on LSAT substrates for different growth temperatures and a bulk single crystal of Sr214 taken in backscattering geometry and using circularly polarized light without an additional analyzer. Green curves show fits overlaid on the gray raw data. Spectra are offset for clarity. Red triangles indicate thin film related modes, and black stars indicate substrate related modes. A red diamond indicates a new mode appearing associated with strong disorder in the system. (b) Raw spectrum of a Sr214 film grown at 840 °C and the corresponding fit to the data. Individual Lorentzian functions are shown in opaque colors below the spectrum. Dashed lines indicate the background. Red triangles labeled M1-M5 indicate the mode peaks related to Sr214. Black stars mark the phonon modes of the LSAT substrate. (c)(d) Detailed views of the Raman features associated with Ir vacancies in films grown at 1000 °C and 1050 °C, shown in comparison with the spectrum of the highest quality film in this series. Fits are overlaid as a guide.

spectra via the Fröhlich interaction [64]. IR spectroscopy and computational chemistry studies show that there are IR-active modes matching the energies of the new defect-induced phonon modes [63,65]. These phonon peaks in the Raman spectra can be used as a clear diagnostic feature of defective samples.

However, it is notable that the samples from the intermediate temperature range do not exhibit additional phonon modes. Instead, the ratio between the peaks seems to vary as the growth temperature deviates from the optimal value. Especially, I_{M2}/I_{M3} decreases upon growth at higher temperatures, which can be attributed to the fact that a strong Ir–O–Ir bending mode (M2) response requires the presence of Ir over an extended range since it is related to the connection of IrO₆ octahedra. The O displacement modes (M3) on the other hand, are not directly dependent on linked IrO₆ octahedra and are thus more robust against Ir vacancies. Figure 4 displays the integrated intensity ratio I_{M2}/I_{M3} along with the c -axis lattice constant, as the c -axis lattice parameter should be directly related to lattice defects. As a reference point, the I_{M2}/I_{M3} value from the high-quality bulk crystal was measured to be 1 ± 0.05 , which is very close to the value we obtained from our best thin film. The plot clearly demonstrates the strong correlation between I_{M2}/I_{M3} and the c -axis lattice parameter, indicating that this value from Raman spectroscopy can be a sensitive indicator of the iridium vacancies.

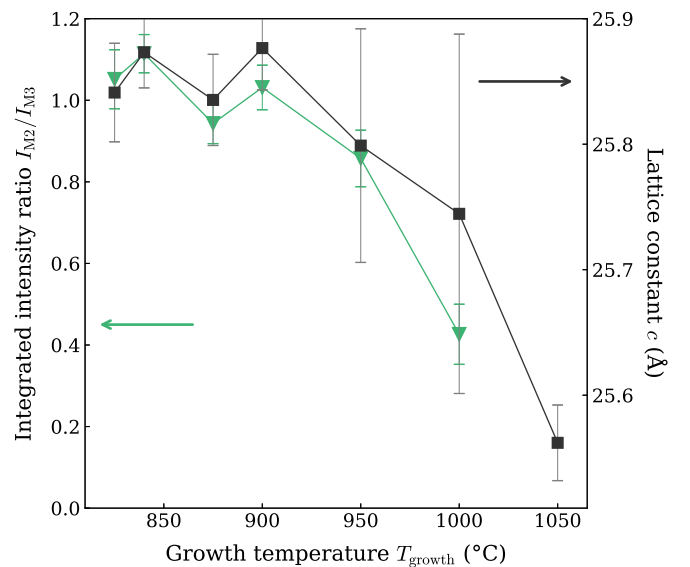


FIG. 4. Growth temperature-dependent quantities extracted from the XRD and Raman characterization measurements. Specifically the integrated intensity ratio obtained from the Raman fit of the M2 mode associated with Ir–O–Ir bending and the M3 mode associated with O displacement within the IrO₆ octahedral plane, and the c -axis lattice constant calculated from Fig. 2(a). Error bars indicate the standard deviation.

IV. CONCLUSIONS

In this work, we demonstrated a reliable method for the epitaxial growth of Sr214 thin films by reactive off-axis sputtering that circumvents the instability of iridium oxides. Through systematic temperature-dependent studies, we identified 840 °C as the optimal growth condition with minimal Ir vacancies. In addition, we demonstrate the sensitivity of Raman spectroscopy to Ir-vacancy-related disorder, as detected by XRD and XPS. This opens interesting perspectives for operando monitoring of electrochemical devices based on Sr214 [66,67] and other iridates [68,69].

Although sputtering is extensively used in the thin-film industry, which requires large-area growth, it has not been widely adopted for complex oxides. As research on complex oxides has matured, exploring their synthesis via sputtering has become increasingly important. Thus, our findings open a

path toward the integration of Sr214 into future devices, such as magnonic devices, that exploit the large strain sensitivity of THz magnons in Sr214 [25] via coupling with piezoelectric oxides.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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