Tuning Magnetic Coupling in Sr$_2$IrO$_4$ Thin Films with Epitaxial Strain

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We report x-ray resonant magnetic scattering (XRMS) and resonant inelastic x-ray scattering (RIXS) studies of epitaxially-strained Sr$_2$IrO$_4$ thin films. The films were grown on SrTiO$_3$ and (LaAlO$_3$)$_{0.3}$(Sr$_2$AlTaO$_6$)$_{0.7}$ substrates, under slight tensile and compressive strains, respectively. Although the films develop a magnetic structure reminiscent of bulk Sr$_2$IrO$_4$, the magnetic correlations are extremely anisotropic, with in-plane correlation lengths significantly longer than the out-of-plane correlation lengths. In addition, the compressive (tensile) strain serves to suppress (enhance) the magnetic ordering temperature $T_N$, while raising (lowering) the energy of the zone-boundary magnon. Quantum chemical calculations show that the tuning of magnetic energy scales can be understood in terms of strain-induced changes in bond lengths.

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The physics of strong spin orbit coupling (SOC) in condensed matter systems has been drawing increased interest in recent years. In particular, iridates have emerged as interesting model systems in which novel magnetism arises due to entangled spin and orbital degrees of freedom [1,2]. A prototypical example is the layered Sr$_2$IrO$_4$ (SIO), in which Ir$^{4+}$ (5d$^9$) ions form a square lattice. Due to the strong SOC, the Ir orbital moment is not quenched in this compound, and the local magnetic moment is described by a spin-orbit coupled $j_{sd} = 1/2$ instead of the usual spin only value commonly observed in lighter transition metals [1,3–6]. A main reason for the strong interest in Sr$_2$IrO$_4$ is that its magnetic properties are strikingly similar to those of the parent compounds of cuprate superconductors [2,7], raising the possibility that unconventional superconductivity could be realized in this system by doping [8,9]. Although superconductivity has not been realized so far, doping studies have shown that structural details, such as the Ir-O-Ir angle and the Ir-Ir distance, are important factors to consider when studying the magnetic properties of Sr$_2$IrO$_4$ [10–14]. However, since doping may also affect the charge concentration in addition to the crystal structure, an alternative means to tune the structure is necessary to elucidate the structure-property relation in Sr$_2$IrO$_4$.

One of the most promising approaches to the structural tuning of oxide materials is strain engineering, accomplished by growing thin films on substrates with varying degrees of lattice mismatch. This method has been successfully used to study 3d and 4d transition metal oxides. For example, it was found that strained thin film cuprates show an increase in superconducting $T_c$ [15]. Strain can also be used to tune the properties of ferroelectrics such as SrTiO$_3$ [16] and BaTiO$_3$ [17]. The study of iridate thin films is still in its early stages, and most studies to date have focused on the structural and electronic properties of thin film Sr$_2$IrO$_4$ [20–23]. Rayan Serrao et al. studied Sr$_2$IrO$_4$ films with various thicknesses grown on SrTiO$_3$ substrates (slight tensile strain), and reported that thinner samples exhibit smaller c/a ratio [21]. They also suggested that electronic anisotropy is reduced in thinner samples based on their structural and x-ray spectroscopic data. Nichols et al. grew Sr$_2$IrO$_4$ films on substrates with varying degrees of strains, ranging from highly compressive to highly tensile [22]. They found that the optical absorption peak shifts to higher energies under tensile strain. Until recently, the magnetic properties of these thin film samples have been largely unexplored [24].

In this Letter, we report complementary x-ray resonant magnetic scattering (XRMS) and resonant inelastic x-ray scattering (RIXS) studies on Sr$_2$IrO$_4$ thin film samples epitaxially grown on SrTiO$_3$ (STO) and (LaAlO$_3$)$_{0.3}$(Sr$_2$AlTaO$_6$)$_{0.7}$ (LSAT) substrates, which have been chosen to provide tensile and compressive strain, respectively. The most surprising result is that the magnetic ordering temperature $T_N$ is found to be suppressed (enhanced) in samples with compressive (tensile) strain. This observation is somewhat counter-intuitive, since both RIXS experiments and quantum chemical calculations predict magnetic interaction to strengthen when the in-plane lattice constant shrinks. The magnetic coupling energy scale determined from the zone-boundary magnon energy increases (decreases) under compressive (tensile) strain, which is also well reproduced.
in calculated values for magnetic exchange constants. We argue that the observed behaviour of \( T_N \) could be accounted for by the subtle change in inter-layer magnetic coupling due to in-plane strain. Our findings illustrate that the magnetic properties of \( \text{Sr}_2\text{IrO}_4 \) are highly sensitive to the effects of epitaxial strain.

The \( \text{Sr}_2\text{IrO}_4 \) thin films, 20 unit-cells thick (\( \approx 50 \) nm), were grown using pulsed laser deposition, as described in Ref. [22]. The films were deposited on two different substrates: STO (100) (SIO-STO), with a 0.45 % nominal tensile strain, and LSAT (100) (SIO-LSAT), with a 0.45 % nominal compressive strain. The XRMS measurements were conducted at the Advanced Photon Source (APS) using beamline 6-ID-B. The data were collected at the Ir \( L_3 \) (11.215 keV) absorption edge. A graphite (008) polarization analyzer was used to select outgoing photon polarization for XRMS. Ir \( L_3 \)-edge RIXS measurements were carried out using the MERIX spectrometer on beamline 30-ID-B at the APS. Measurements were performed using a spherical (2 m radius) diced Si (844) analyzer and a channel-cut Si (844) secondary monochromator to give an energy resolution (FWHM) of 45 meV. The RIXS data was collected in horizontal scattering geometry, with a scattering angle close to \( 2\theta = 90^\circ \), to minimize the background contribution from elastic scattering. To maximize the signal, measurements were performed near glancing incidence, with an angle of incidence \( \alpha < 1^\circ \), for both the thin films and bulk sample.

The tetragonal structure of bulk \( \text{Sr}_2\text{IrO}_4 \) gives rise to two distinct magnetic domains. The first domain is characterized by magnetic reflections observed at \((1, 0, 4n + 2)\) and \((0, 1, 4n)\), with integer \( n \). The second domain gives rise to peaks at \((0, 1, 4n + 2)\) and \((1, 0, 4n)\) [25]. With an x-ray beam spot much larger than the size of the magnetic domains, we expect to observe both types of domains and thus find peaks at all the even \( L \) positions of \((1, 0, L)\) and \((0, 1, L)\). Magnetic peaks have also been observed for odd \( L \) values when small magnetic field is applied or when doping occurs on the Ir sites [1, 26, 27].
Fig. 2 (c)-(d) for SIO-STO. Both in-plane scans (H and K scans) are quite similar and only one of these are shown. Unlike the SIO-LSAT film, which shows monotonous temperature dependence, the SIO-STO shows anomalous temperature dependence in the 30 – 100 K range. These anomalies are presumably due to a variation in strain with temperature caused by structural transitions in the STO substrate. STO adopts several distinct structural phases at low temperatures: with a cubic to tetragonal transition at (T = 110 K) and several other transitions at lower temperatures [28]. The temperature dependence illustrates the close relation between strain and magnetism in this material. However, a further quantitative characterization of the SIO-STO film structure at low temperatures is beyond the scope of this paper, and we will only focus on T > 110 K data here.

The peak profiles were fitted with a n-th power Lorentzian function: \( I(q) = I_{\text{max}}[(q - q_0)^2/(\zeta_n^2 + 1)]^{-n} \), where \( \zeta_n = \sqrt{2^{1/n} - 1} \) is a constant set to keep \( \kappa \) the half-width at half maximum (HWHM). The parameter \( q \) is either H, K, or L, and \( q_0 \) is the respective peak position. The fitting procedure is detailed in the Supplemental Material [29]. The fitting results are presented in Fig. 3(a).

The widths of the film magnetic peaks are approximately 2-3 times broader than the (1,0,18) bulk magnetic peak along the H and K-direction, and an order of magnitude broader along the L-direction, as shown in Fig. 2. The magnetic correlation length, \( \xi \), can be estimated by inverting \( \kappa: \xi = \kappa^{-1} \). Both the tensile and the compressive strain films show similar magnetic correlation lengths, with a considerable anisotropy characterized by very small correlation lengths along the c-axis, \( \xi \approx 10 – 20 \) Å (approximately one unit-cell) and much larger correlation lengths in the ab-plane, \( \xi \approx 300 – 400 \) Å. Such a short-range magnetic correlation along L is in stark contrast to the magnetic ordering in bulk \( \text{Sr}_2\text{IrO}_4 \), which develops into a full 3D long-range order [7].

In Fig. 3(b) we present the temperature dependence of the integrated intensity for SIO-LSAT (red triangles) and SIO-STO (blue squares). The magnetic transition temperatures in the films are very different from that of bulk (\( T_N = 240 \) K) [1][30]. For the compressive strain (LSAT), the transition temperature is lower than the bulk, with a \( T_N \) of only 210 K. The tensile strain (STO) has an increased \( T_N \) of 270 K. This trend is illustrated in the inset of Fig. 3(b), which shows the transition temperature as a function of strain.

Representative RIXS measurements performed on bulk SIO, SIO-STO, and SIO-LSAT are presented in Fig. 4. They highlight the strain effect on the magnon mode in \( \text{Sr}_2\text{IrO}_4 \), which is observed at energy transfers of \( \sim 170 – 200 \) meV [2]. The spectra presented were collected at the \((\pi, 0)\) zone boundary position, where the magnon energy is at its maximum. The RIXS excitation spectra for the SIO, SIO-STO and SIO-LSAT samples, including the spin-orbit exciton mode at \((\pi, 0)\) and \((\pi/2, \pi/2)\) zone boundary wave-vectors, can be found in Fig. 2 of the Supplemental Material [29]. The strain dependence of the zone boundary magnon energy, \( E_{\pi,0} \), is very different from that of \( T_N \), with compressive strain (SIO-LSAT) driving the magnon to higher energies, and tensile strain (SIO-STO) driving it lower. Quantitative values for \( E_{\pi,0} \), extracted from multi-Gaussian data fits, are provided in Table I.

On a qualitative level, the strain-induced tuning of magnetic energy scales in SIO can be understood as follows. The application of compressive epitaxial strain results in a reduction of the in-plane lattice parameters \((a \text{ and } b)\) and an enlargement of the out-of-plane lattice parameter \((c)\). The magnetic exchange interactions between neighbouring Ir ions are very sensitive to bond geometry, so a decrease of Ir-O/Ir-Ir bond lengths will enhance the interaction strength and vice-versa. Hence, we expect compressive strain to strengthen the in-plane
FIG. 4: Epitaxial strain effect on the low-lying magnetic excitations of Sr$_2$IrO$_4$. A comparison of Ir L$_3$-edge RIXS spectra collected at room temperature for bulk SIO, SIO-LSAT, and SIO-STO demonstrates that compressive (tensile) strain significantly raises (lowers) the energy of the ($\pi$,0) zone boundary magnon. The solid lines represent Gaussian fits to the data (described in the Supplemental Material [29]). The dashed vertical lines represent the fitted values for the magnon energies.

This argument is supported by \textit{ab initio} multireference configuration-interaction (MRCI) calculations on embedded clusters of two nearest-neighbour IrO$_6$ octahedra. The \textit{ab initio} wave function approach has been shown to yield results in good agreement with experiments measuring the magnetic interactions in 3$d$ [44, 45] and 5$d$ [49] oxides, as well as determining the dependence of the effective coupling constants on strain in cuprates [40] and on additional distortions in a few other 5$d$-metal compounds [49, 51, 52]. Singlet-triplet splittings for cluster models of bulk SIO and strained films of SIO are listed in Table I. The results were obtained by MRCI calculations including SOC’s (MRCI+SOC) [53]. Two different structural models were employed for the SIO films. Since the precise structural details are experimentally difficult to access in the films, we assumed in a first set of calculations that the Ir-O-Ir bond angles are the same as in bulk [3] and only the interatomic distances change with strain (Model I). At the other extreme, we considered a structural model for which the in-plane Ir-O bond lengths are fixed to the values measured in bulk [3] and for reproducing the strain induced variation of the lattice parameters [22] we modified the Ir-O-Ir angles (Model II). For Model I, the variations of the average energy of the triplet terms with respect to the singlet state, denoted in Table I as $\Delta E_{ST}$, are large, 3.5 to 7 meV. Since the structure of the triplet components is always the same, with two of them nearly degenerate and the splitting between the lowest and highest triplet terms taking values in a narrow interval between 0.9 to 1.2 meV (see the Supplemental Material [29]), we can safely conclude that for Model I the most important changes with strain concern the variation of the isotropic Heisenberg exchange $J$. The overall trend observed in the RIXS spectra for $J$ is in this case nicely reproduced. In contrast, for Model II, the variations of $\Delta E_{ST}$ are much smaller and do not follow the trend observed for $J$ by RIXS. This suggests that the most significant structural change that occurs in the epitaxial thin films is the tuning of the Ir-Ir and Ir-O bond lengths. Further detailed investigations of the local structure by local probes such as EXAFS would be extremely useful.

TABLE I: Effective singlet-triplet splittings $\Delta E_{ST}$ in SIO bulk and SIO films for two adjacent Ir ions (meV). MRCI+SOC results, see text. Experimental RIXS values for the zone boundary magnon energy (proportional to $J$) are also provided. Strain-induced relative changes in the energy scales are listed in parentheses.

| Sample  | RIXS: $E_{(\pi,0)}$ | Model I: $\Delta E_{ST}$ | Model II: $\Delta E_{ST}$ |
|---------|---------------------|--------------------------|---------------------------|
| SIO-STO | 172 ± 4 (−3.4%)     | 49.8 (−12%)              | 56.4 (−0.4%)              |
| SIO-Bulk| 178 ± 4              | 56.6                     | 56.6                      |
| SIO-LSAT| 196 ± 6 (+10.0%)     | 60.1 (+5.8%)             | 55.0 (−2.8%)              |

In conclusion, we have explored the magnetic properties of Sr$_2$IrO$_4$ thin films, with tensile (STO) and compressive (LSAT) epitaxial strain, using x-ray resonant magnetic scattering and resonant inelastic x-ray scattering. The films show a quasi-two-dimensional magnetic order for both substrates, with magnetic ordering vectors reminiscent of the bulk magnetic structure. Compared to bulk Sr$_2$IrO$_4$, the film magnetic correlation lengths show a large anisotropy, with in-plane correlation lengths of 300 – 400 Å and very small (10 – 20 Å) correlation lengths along the c-axis. We have observed that the magnetic ordering temperature $T_N$ is suppressed for the compressive strain (LSAT) and enhanced for the tensile strain (STO). In contrast, the RIXS experiments show that the magnetic exchange interactions, determined from the zone-boundary magnon energy, increases (decreases) under compressive (tensile) strain. These results are supported by quantum chemistry calculations, which suggest that the most significant structural change taking place in the films is a tuning of the Ir-O bond length. Other applied perturbations in Sr$_2$IrO$_4$ seem to change the mag-
netic structure of Sr$_2$IrO$_4$ at fairly moderate levels (i.e., relatively low applied fields and dopant concentrations).

In comparison, a significant epitaxial-strain has no effect on the magnetic ordering wave-vector, while it is altering the energy scales associated with $T_N$ and $J$. This illustrates that epitaxial strain is an excellent knob for studying the magnetic properties of iridates.

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[1] B. J. Kim, H. Ohsumi, T. Komesu, S. Sakai, T. Morita, H. Takagi, and T. Arima, Science 323, 1329 (2009).
[2] J. Kim, D. Casa, M. H. Upton, T. Gog, Y.-J. Kim, J. F. Mitchell, M. van Veenendaal, M. Daghofer, J. van den Brink, G. Khalilullin, et al., Phys. Rev. Lett. 108, 177003 (2012).
[3] M. K. Crawford, M. A. Subramanian, R. L. Harlow, J. A. Fernandez-Baca, Z. R. Wang, and D. C. Johnston, Phys. Rev. B 49, 9198 (1994).
[4] G. Cao, J. Bolivar, S. McCall, J. E. Crow, and R. P. Guertin, Phys. Rev. B 57, R11039 (1998).
[5] B. J. Kim, H. Jin, S. J. Moon, J.-Y. Kim, B.-G. Park, C. S. Leem, J. Yu, T. W. Noh, C. Kim, S.-J. Oh, et al., Phys. Rev. Lett. 101, 076402 (2008).
[6] M. Moretti Sala, S. Balseggia, D. F. McMorrow, and G. Monaco, Phys. Rev. Lett. 112, 026403 (2014).
[7] S. Fujiyama, H. Ohsumi, T. Komesu, J. Matsuno, B. J. Kim, M. Takata, T. Arima, and H. Takagi, Phys. Rev. Lett. 108, 247212 (2012).
[8] F. Wang and T. Senthil, Phys. Rev. Lett. 106, 136402 (2011).
[9] H. Watanabe, T. Shirakawa, and S. Yunoki, Phys. Rev. Lett. 110, 027002 (2013).
[10] T. F. Qi, O. B. Korneta, L. Li, K. Butrouna, V. S. Cao, X. Wan, P. Schlottmann, R. K. Kaul, and G. Cao, Phys. Rev. B 86, 125105 (2012).
[11] R. J. Cava, B. Batlogg, K. Kiyono, H. Takagi, J. J. Krajewski, W. F. Peck Jr., L. W. Rupp Jr., and C. H. Chen, Phys. Rev. B 49, 11890 (1994).
[12] M. V. Rama Rao, V. G. Sathe, D. Sornadurai, B. Panigrahi, and T. Siriphati, J. Phys. Chem. Solids 61, 189 (1990).
[13] A. J. Gatiimu, R. Berthelot, S. Muiir, A. W. Sleight, and M. A. Subramanian, J. Solid State Chem. 190, 257 (2012).
[14] T. Shimura, Y. Inaguma, T. Nakamura, M. Itoh, and Y. Morii, Phys. Rev. B 52, 9143 (1995).
[15] Y. Klein and I. Terasaki, Journal of Physics: Condensed Matter 20, 295201 (2008).
[16] C. Cosio-Castaneda, G. Tavizon, A. Baeza, P. de la Mora, and R. Escudero, Journal of Physics: Condensed Matter 19, 446210 (2007).
[17] J.-P. Locquet, J. Perret, J. Fompeyrine, E. Machler, J. W. Seo, and G. Van Tendeloo, Nature 394, 453 (1998).
[18] J. H. Haeni, P. Irvin, W. Chang, R. Uecker, P. Reiche, Y. L. Li, S. Choudhury, W. Tian, M. E. Hawley, B. Craigo, et al., Nature (London) 430, 758 (2004).
[19] K. J. Choi, M. Biegalski, Y. L. Li, A. Sharan, J. Schubert, R. Uecker, P. Reiche, Y. B. Chen, X. Q. Pan, V. Gopal, et al., Science 306, 1005 (2004).
[20] J. S. Lee, Y. Krockenberger, K. S. Takahashi, M. Kawasaki, and Y. Tokura, Phys. Rev. B 85, 035101 (2012).
[21] C. Rayan Serrao, L. Liu, J. T. Heron, G. Singh-Bhalla, A. Yadav, S. J. Suresha, R. J. Paull, D. Yi, J.-H. Chu, M. Trassin, et al., Phys. Rev. B 87, 085121 (2013).
[22] J. Nichols, J. Terzic, E. G. Bittle, O. B. Korneta, L. E. D. Long, J. W. Brill, G. Cao, and S. S. A. Seo, Appl. Phys. Lett. 102, 141908 (2013).
[23] J. Nichols, O. B. Korneta, J. Terzic, L. E. D. Long, G. Cao, J. W. Brill, and S. S. A. Seo, Appl. Phys. Lett. 103, 131910 (2013).
[24] L. Miao, H. Xu, and Z. Q. Mao, Phys. Rev. B 89, 035109 (2014).
[25] C. Dhillon, T. Hogan, Z. Yamani, C. de la Cruz, X. Chen, S. Khadka, Z. Ren, and S. D. Wilson, Phys. Rev. B 87, 144405 (2013).
[26] S. Calder, G.-X. Cao, M. D. Lumsden, J. W. Kim, Z. Gai, B. C. Sales, D. Mandrus, and A. D. Christianson, Phys. Rev. B 86, 220402 (2012).
[27] J. F. Clancy, A. Lupascu, H. Grettarsson, Z. Islam, Y. F. Hu, D. Casa, C. S. Nelson, S. C. LaMarra, G. Cao, and Y.-J. Kim, Phys. Rev. B 89, 054406 (2014).
[28] F. W. Lytle, J. Appl. Phys. 35, 2212 (1964).
[29] See Supplemental Material [url], which includes 31–41.
[30] G. Jackeli and G. Khalilullin, Phys. Rev. Lett. 102, 017205 (2009).
[31] Q. Huang, J. Soubeyroux, O. Chmaissem, I. Sora, A. Santoro, R. Cava, J. Krajewski, and W. P. Jr., J. Solid State Chem. 112, 355 (1994).
[32] C. de Graaf, C. Sousa, and R. Broer, J. Mol. Struct. (Theochem) 458, 53 (1998).
[33] L. Hozoi, L. Siurakshina, P. Fulde, and J. van den Brink, Sci. Rep. 1, 65 (2011).
[34] H.-J. Werner, P. J. Knowles, G. Knizia, F. R. Manby, and M. Schütz, MOLPRO 2012, see http://www.molpro.net.
[35] D. Figgen, K. A. Peterson, M. Dolg, and H. Stoll, J. Chem. Phys. 130, 164108 (2009).
[36] T. H. Dunning, J. Chem. Phys. 90, 1007 (1989).
[37] N. A. Bogdanov, V. M. Katukuri, H. Stoll, J. van den Brink, and L. Hozoi, Phys. Rev. B 85, 235147 (2012).
[38] N. A. Bogdanov, R. Maurice, I. Rousochatzakis, J. van den Brink, and L. Hozoi, Phys. Rev. Lett. 110, 127206 (2013).
[39] H. Stoll (2013), (unpublished).
[40] P. Fuentealba, L. von Szentpaly, H. Preuss, and H. Stoll, J. Phys. B 18, 1287 (1985).
[41] K. Fink, R. Fink, and V. Stammmler, Inorg. Chem. 33, 6219 (1994).
[42] A. B. van Oosten, R. Broer, and W. C. Nieuwpooort, Chem. Phys. Lett. 257, 207 (1996).
[43] C. J. Calzado, S. Evangelisti, and D. Maynau, J. Phys. Chem. A 107, 7581 (2003).
[44] B. H. Kim, G. Khalilullin, and B. I. Min, Phys. Rev. Lett. 109, 167205 (2012).
[45] N. B. Perkins, Y. Sizyuk, and P. Wölfle, Phys. Rev. B 89, 035143 (2014).
[46] T. Helgaker, P. Jørgensen, and J. Olsen, Molecular Electronic-Structure Theory (Wiley, Chichester, 2000).
[47] D. Muñoz, F. Illas, and I. de P. R. Moreira, Phys. Rev. Lett. 84, 1579 (2000).
[48] J. P. Malrieu, R. Caballol, C. J. Calzado, C. de Graaf, and N. Guihry, Chemical Reviews 114, 429 (2014).
[49] V. M. Katukuri, H. Stoll, J. van den Brink, and L. Hozoi, Phys. Rev. B 85, 220402 (2012).
[50] M. Minola, L. Hozoi, D. DiCastro, R. Felici, M. Moretti Sala, A. Tebano, G. Balestrino, G. Ghiringhelli, J. van den Brink, and L. Braicovich, Phys. Rev. B 87, 085124 (2013).
[51] L. Hozoi, C. Presura, C. de Graaf, and R. Broer, Phys. Rev. B 67, 035117 (2003).
[52] E. Bordas, C. de Graaf, R. Caballol, and C. J. Calzado, Phys. Rev. B 71, 045108 (2005).
[53] A. Berning, M. Schweizer, H.-J. Werner, P. J. Knowles, and P. Palmieri, Mol. Phys. 98, 1823 (2000).