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14. ABSTRACT
In this report we will provide a technical description of three femtosecond laser-based optical spectroscopic techniques that we have constructed with DURIP support for probing novel electronic properties of 5d transition metal oxide compounds. 1) We demonstrate a new method to perform nonlinear optical rotational anisotropy experiments on small single crystals at low temperatures, which enables the crystallographic and magnetic symmetries of many correlated electron phases to be resolved on micron length scales for the first time. We also discuss a scheme to increase the data collection frequency of these measurements by over three orders of magnitude

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Second harmonic generation, nonlinear optical microscopy, time-resolved reflectivity, iridates

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Report Title

Final Report: Instruments for Optical Spectroscopy and Imaging of Correlated Spin-Orbit Phases

ABSTRACT

In this report we will provide a technical description of three femtosecond laser-based optical spectroscopic techniques that we have constructed with DURIP support for probing novel electronic properties of 5d transition metal oxide compounds. 1) We demonstrate a new method to perform nonlinear optical rotational anisotropy experiments on small single crystals at low temperatures, which enables the crystallographic and magnetic symmetries of many correlated electron phases to be resolved on micron length scales for the first time. We also discuss a scheme to increase the data collection frequency of these measurements by over three orders of magnitude to greatly improve the sensitivity to small changes in symmetry. 2) We describe the construction of a nonlinear optical and Kerr microscope to image the spatial distribution of both crystallographic and time-reversal symmetry broken domains with micron scale resolution. We show that magnetic ordering in a 5d transition metal oxide compound generates a large nonlinear optical Kerr rotation and we present images revealing multiple magnetic domains in a single crystalline host for the first time. 3) We describe the layout of a time- and energy-resolved optical reflectivity setup to investigate ultrafast photo-induced changes in the optical conductivity spectrum of a correlated spin-orbit system.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
10/19/2014	1.00	Darius H. Torchinsky, Hao Chu, Tongfei Qi, Gang Cao, David Hsieh. A low temperature nonlinear optical rotational anisotropy spectrometer for the determination of crystallographic and electronic symmetries, Review of Scientific Instruments, (08 2014): 83102. doi: 10.1063/1.4891417
TOTAL:	**1**	

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received		Paper
TOTAL:		

Number of Papers published in non peer-reviewed journals:

(c) Presentations

1) Gordon Research Seminar on Ultrafast Phenomena in Cooperative Systems, Ventura CA, February 1-2 2014. "Characterization of the Structural and Magnetic Symmetries of Sr2IrO4 via Nonlinear Optical Spectroscopy". (oral)

2) Gordon Research Conference on Ultrafast Phenomena in Cooperative Systems, Ventura CA, February 2-7 2014. "Characterization of the Structural and Magnetic Symmetries of Sr2IrO4 via Nonlinear Optical Spectroscopy". (poster)

3) Gordon Research Conference on Correlated Electron Systems, South Hadley MA, June 22-27 2014. "Characterization of the Structural and Magnetic Symmetries of Sr2IrO4 via Nonlinear Optical Spectroscopy". (poster)

4) APS March Meeting, Denver CO, March 3-7 2014. "A diffractive-optic based nonlinear optical generation spectrometer for measurement of crystallographic and magnetic point group symmetries". (oral)

5) APS March Meeting, Denver CO, March 3-7 2014. "A Spatially Resolved Optical Second Harmonic Generation (SHG) Study of the Perovskite Iridate Sr2IrO4 with Bulk Sensitivity". (oral)

6) APS March Meeting, Denver CO, March 3-7 2014. "A time- and wavelength-resolved optical pump-probe reflectivity study of the Metal-to-Insulator Transition in Sr2IrO4". (oral)

7) APS March Meeting, Denver CO, March 3-7 2014. "Characterization of the Structural and Magnetic Symmetries of Sr2IrO4 via Nonlinear Optical Spectroscopy". (oral)

8) Caltech-Tsinghua Workshop on Frontiers of Science and Technology, Pasadena CA, September 13-15 2014. "Search for novel quantum phases in 5d transition metal oxides using nonlinear optics". (oral)

9) The 3rd GIST-Caltech Workshop on Innovative Research, Pasadena CA, September 17-18 2014. "Search for novel quantum phases in 5d transition metal oxides using nonlinear optics". (oral)

Number of Presentations: 9.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received	Paper	
10/19/2014	2.00	D. H. Torchinsky, H. Chu, L. Zhao, N. B. Perkins, Y. Sizyuk, T. Qi, G. Cao, D. Hsieh. A structural distortion induced magneto-elastic locking in Sr2IrO4 revealed through nonlinear optical harmonic generation, P H Y S I C A L R E V I EW L E T T E R S (07 2014)
TOTAL:	**1**	

Number of Manuscripts:

Books

Received	Book
TOTAL:	

Received	Book Chapter
TOTAL:	

Patents Submitted

A rotating scattering plane based nonlinear optical spectrometer to study the crystallographic and electronic symmetries of crystals

Patents Awarded

Awards

David Hsieh (PI)
-Caltech Earnest C. Watson Lecture (October 2014)
-Sloan Research Fellowship (2014-2016)
-Plenary Speaker, RQMP Prestigious Lecture Series on Advanced Materials (November 2013)

Tejas Deshpande (Graduate student)
-NSERC Fellowship (2014)

Hao Chu (Graduate student)
-APS FGSA Travel Award for Excellence in Graduate Research (2014)

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Hao Chu	0.25	
Tejas Deshpande	0.75	
FTE Equivalent:	**1.00**	
Total Number:	**2**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
Darius Torchinsky	0.65
FTE Equivalent:	**0.65**
Total Number:	**1**

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
David Hsieh	0.01	
FTE Equivalent:	**0.01**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

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Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

5 **Rotating Scattering Plane Based Nonlinear Optical Spectrometer to Study the Crystallographic and Electronic Symmetries**

Patent Filed in US? (5d-1) Y

Patent Filed in Foreign Countries? (5d-2) N

Was the assignment forwarded to the contracting officer? (5e) Y

Foreign Countries of application (5g-2):

5a: Darius H. Torchinsky

5f-1a: California Institute of Technology

5f-c: 1200 E. California Blvd.

Pasadena CA 91125

5a: David Hsieh

5f-1a: California Institute of Technology

5f-c: 1200 E. California Blvd.

Pasadena CA 91125

Scientific Progress

See Attachment

Technology Transfer

I. Statement of the problem studied

The goal of this research project is to experimentally identify and to understand new electronic phases of matter in solids that emerge from the interplay of strong electron-electron correlations and strong spin-orbit coupling. Recently it has been found that 5*d* transition metal oxide compounds can realize this unique parameter regime. This has stimulated the discovery and synthesis of many new families of 5*d* transition metal oxide materials as well as rapid improvements in the quality of their single crystals. However many of the exotic quantum phases predicted to exist in these crystals are challenging to probe experimentally for two reasons. First, the crystals are typically small in size and the 5*d* transition metal ions have a large cross section for neutron absorption, which makes crystallographic and magnetic structure determination difficult to perform with neutron diffraction. Second, the experimental signatures of many of the predicted quantum phases cannot be revealed through conventional probes, thus necessitating the development of new experimental techniques.

This DURIP award has supported the development of three new femtosecond pulsed laser-based optical spectroscopy techniques that are well suited to studying the electronic properties of 5*d* transition metal oxides. The three techniques are: 1) Low temperature nonlinear optical rotational anisotropy to probe crystallographic and electronic symmetry breaking phase transitions; 2) Low temperature nonlinear optical and Kerr microscopy to probe the spatial distribution of time-reversal symmetry broken phases; and 3) time- and energy-resolved optical spectroscopy to probe the nature of unconventional metal-to-insulator transitions.

II. Summary of most important results

In this Final Progress Report we will focus on a technical description of the three new instruments developed through this DURIP award. The scientific findings are discussed in depth in our Interim Progress Report submitted for ARO award W911NF-13-1-0059, therefore we will only briefly summarize them here. 1) We have performed a detailed study of the crystallographic structure of the 5*d* transition metal oxide Sr_2IrO_4 using room temperature optical second and third harmonic generation rotational anisotropy. Our study reveals a lowered symmetry compared to existing neutron and x-ray diffraction results. This resolves the origin of several forbidden Bragg peaks observed in diffraction studies and reconciles the observations of a perfect magneto-elastic locking with a tetragonal distortion of the oxygen octahedra in Sr_2IrO_4. 2) We have investigated the magnetic structure of Sr_2IrO_4 using a combination of low temperature optical second harmonic generation rotational anisotropy and microscopy. We discovered a hidden magnetic order parameter below the Neel temperature and have identified ordered regions tens of microns in size that may explain its unusual bulk magneto-electric properties. 3) We have performed time- and energy-resolved pump-probe optical reflectivity

measurements on Sr_2IrO_4. We observe no dependence of the relaxation dynamics as a function of probe wavelengths spanning all transition energies within the t_{2g} complex of Ir 5*d* states, which suggests that there is significant overlap in energy between these bands. No evidence for a photo-induced insulator-to-metal transition has been observed so far at the pump fluences used.

III. Scientific instrumentation developed through this award

III.A.1 *Low temperature nonlinear optical rotational anisotropy*

We proposed to use second harmonic generation rotational anisotropy (SHG-RA) to resolve the low temperature long-range ordered magnetic structures of 5*d* transition metal oxides, with particular emphasis on the iridate family. In a conventional SHG-RA experiment, light of frequency ω is impinged onto the surface of a crystal and the intensity of light transmitted or reflected at twice the incident frequency 2ω is collected as the crystal is rotated about its surface normal axis (Fig.1). The angular dependence of the SHG intensity, in principle, provides information about the symmetry properties of both the crystal lattice [1] as well as the underlying magnetic lattice [2]. This technique is therefore a powerful complement to neutron and x-ray diffraction based structure refinement techniques.

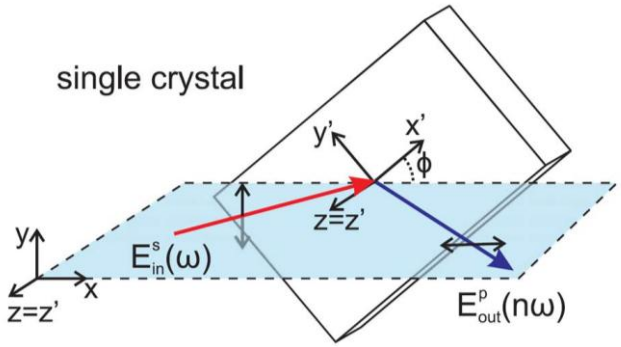


FIG. 1. Schematic of an SHG-RA experiment. The scattering plane (light blue) is defined by the incident (red arrow) and radiated (dark blue arrow) beams. S(P)-polarization denotes an electric field (E) pointing perpendicular (parallel) to the scattering plane. ϕ is the angle that the crystalline axis *x*' makes with respect to the scattering plane axis *x* upon rotation about the surface normal *z* = *z*' axis.

Although this technique has been successfully used to study the lattice and magnetic structures of systems such as semiconductor surfaces, multiferroic crystals, magnetic thin films and multilayers, challenging technical requirements have prevented its application to the plethora of complex electronic phases found in strongly correlated electron systems including 5*d* transition metal oxides. These requirements include an ability to probe small bulk single crystals at the μm length scale, a need for sensitivity to the entire nonlinear optical susceptibility tensor, oblique light incidence reflection geometry and incident light frequency tunability. A further challenge is presented by the need for extreme sample environments such as ultra low temperatures, high magnetic fields, or high pressures.

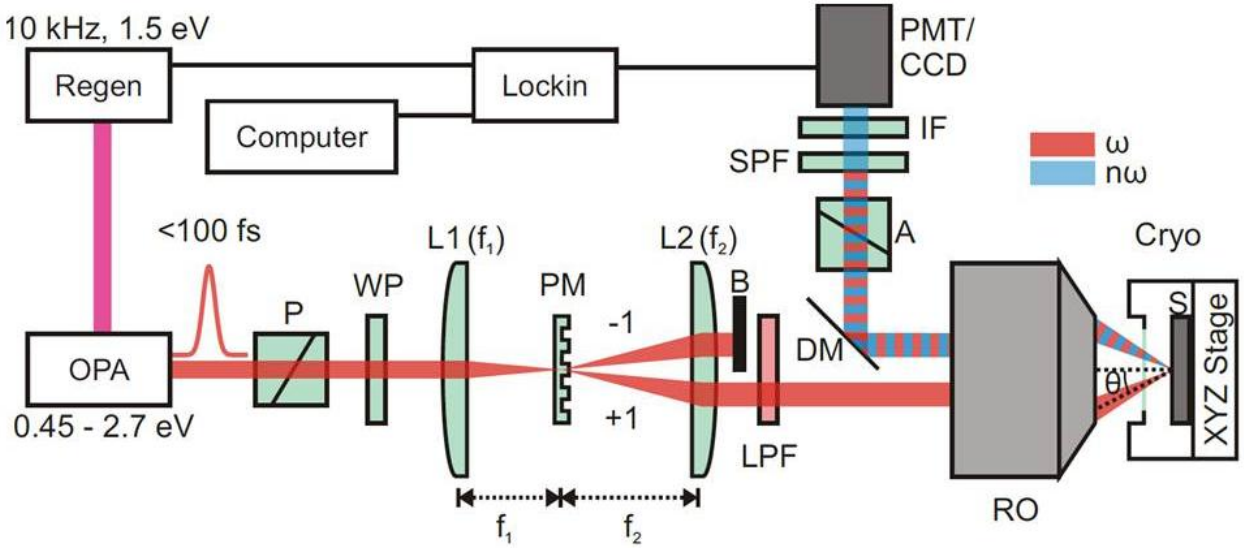


Fig. 2 Layout of the SHG-RA experiment. A pulsed laser beam from an optical parametric amplifier (OPA), which is seeded by a Ti:Sapphire regenerative amplifier, passes through a polarizer (P) and waveplate (WP) and is focused by the first lens (L1) onto a phase mask (PM). A 1st order diffracted beam is collimated by a second lens (L2), sent through a long-pass filter (LPF), and then focused using a reflective objective (RO) onto a sample (S) in the cryostat. The reflected beam passes back through the RO and is picked off by a d-cut mirror (DM). An analyzer (A), short-pass filter (SPF) and interference filter (IF) select a polarization component of the n^{th} harmonic, which is measured using either a photomultiplier tube (PMT) or cooled CCD camera. The scattering plane is rotated by placing the optics P, WP, PM, DM, A, SPF, IF and PMT on rotation stages.

Through DURIP support, we have devised and constructed a novel scheme for performing wavelength tunable SHG-RA measurements under oblique incidence geometry that overcomes all of the previous technical obstacles for the first time. Moreover it can be used to measure any reflected harmonic of the incident beam and not only the second harmonic. Our scheme, which has been published in the Review of Scientific Instruments [3], works by rotating the light scattering plane while keeping the crystal stationary (Fig. 2). By removing the need to rotate the crystal one removes the need for any mechanical degrees of freedom on the cryostat cold finger, which allows samples to be cooled to significantly lower temperatures. Moreover, this allows magnetic field and pressure dependent experiments to be performed in standard magnet cryostats and pressure cells where the sample environment remains stationary. By using a novel alignment technique that uses a rotating optical grating generated by a binary phase mask (Fig. 2), we have achieved a very high degree of alignment accuracy including both negligible beam walk on the crystal (≤ 1 μm) and negligible deviation ($\leq 0.2°$) of the crystal surface normal away from the rotation axis over the entire 360° angular ϕ range of a measurement.

Data obtained from a μm scale facet of the (001) cleaved surface of Sr_2IrO_4 are shown in Fig. 3. SHG-RA patterns were taken both above and below the antiferromagnetic ordering Neel temperature T_N = 240 K using a liquid nitrogen cooled optical cryostat with the sample

maintained at a pressure better than 1×10^{-6} torr in order to preserve a clean surface. The SHG-RA pattern at a temperature $T > T_N$ is very well accounted for exclusively by a bulk electric quadrupole SHG radiation process (red lines) that is representative of the bulk crystal symmetry of Sr_2IrO_4, with no evidence for surface adsorbate contamination. Therefore despite SHG conventionally being used as a surface sensitive probe, our measurements show that it can be used as a bulk sensitive probe of iridates. These results have been submitted to Physical Review Letters and is currently under review. The data for $T < T_N$ shows a clear change in symmetry that is representative of the low temperature antiferromagnetic structure of Sr_2IrO_4, which demonstrates that our SHG-RA scheme can effectively be used to probe the symmetry of bulk magnetic structures. We have recently also shown that our technique can similarly be applied to Na_2IrO_3, a compound which has been predicted to be the host for an exotic topological spin liquid, and we are currently in the process of analyzing this data.

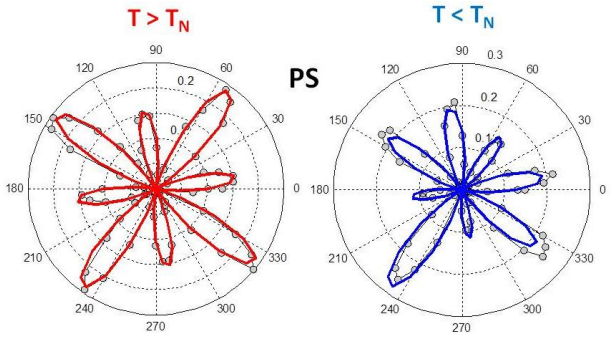


FIG. 3. SHG-RA data obtained from the (001) surface of Sr_2IrO_4 under P-input and S-output geometry both above (left) and below (right) the Neel temperature T_N = 240 K. The SHG intensity is plotted radially as a function of ϕ. The red line is a fit to a bulk crystallographic electric quadrupole radiation process while the blue line includes an additional bulk magnetism induced electric dipole radiation process.

III.A.2 *Second generation low temperature nonlinear optical rotational anisotropy*

The scheme outlined in Fig. 2 takes data like those shown in Fig. 3 by incrementally stepping ϕ and averaging the signal over a specified time at each value of ϕ. Typically each data point requires ~1 min of averaging time. Owing to the considerable weight of the optics and detector that need to be rotated as ϕ is stepped, a large and heavy rotation stage is used, which means that the rotation speed is rather slow and adds time between each measurement point. Altogether, a single data set where ϕ spans 360° takes roughly 1 hour.

A drawback of this long data acquisition time is the susceptibility to sources of low frequency noise, the most serious being laser power fluctuations, which are known to be especially large for Ti:Sapphire regenerative amplifiers. In effort to acquire our SHG-RA data at much higher frequency and thus improve our signal-to-noise ratio, we have devised an upgraded 2nd generation setup that eliminates the need for a rotating detector and reduces the number of optics that need to be rotated, thus enabling us to take 360° datasets at about 3000 times higher frequency. The working principle of this 2nd generation setup is illustrated in Fig. 4. The pulsed laser beam is circularly polarized by a quarter waveplate and a rotating linear polarization is produced by a rotating linear polarizer. Similar to our 1st generation setup (Fig. 2), a rotating

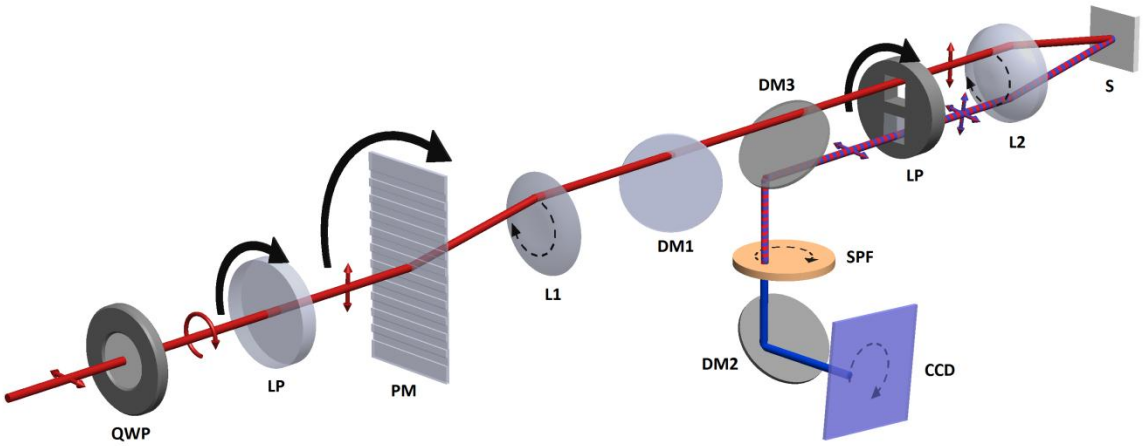


Fig. 4 Layout of the 2nd generation SHG-RA experiment. A linearly polarized pulsed laser beam from an OPA, which is seeded by a Ti:Sapphire regenerative amplifier, becomes circularly polarized by a quarter waveplate (QWP). The beam path sweeps out a cone upon passing through a co-rotating linear polarizer (LP) and phase mask (PM), with the linear polarization locked with respect to the beam deflection angle, and is then collimated by a lens (L1) so that the beam path sweeps out a cylinder. After being transmitted through a pair of SP-compensating dichroic mirrors (DM1 and DM3), the beam is focused onto the sample using an objective lens (L2). The polarization of reflected light is selected through a rotating linear polarizer (LP) and is SP-compensated by reflecting off a pair of dichroic mirrors (DM3 and DM2). The combination of dichroic mirrors and short-pass filter (SPF) serve to select the desired n^{th} harmonic. This beam draws a circle on a cooled CCD camera, from which rotational anisotropy patterns are directly obtained.

scattering plane is produced by using a phase mask that co-rotates with the linear polarizer. The beam is then re-collimated by a lens and then focused onto the sample using an objective lens. The beam reflected from the sample is polarization selected using a rotating linear polarizer and then wavelength filtered to isolate the desired harmonic. Pairs of 45° mounted dichroic mirrors are put into both the incident and reflected beam paths to compensate for the slight differences in their S and P polarized transmission and reflection coefficients as the incident and reflected beam polarizations are rotated during measurement. The key difference with this design is that rather than use a rotating detector, the signal collected at different ϕ are projected onto different spatial locations on a position sensitive CCD camera that remains stationary. The ϕ dependent SHG intensity can then be extracted by analyzing the pixel position dependent intensity on the CCD camera. Unlike the original setup shown in Fig. 2, in this setup the angle ϕ can be continuously swept rather than discretely stepped and because only three small and lightweight optics need to be rotated, the rotation speed can be several Hz. We are currently in the final testing phase of this instrument and are preparing to write an instrument review paper detailing its operation.

III.B *Low temperature nonlinear optical and Kerr microscopy*

We proposed to use low temperature nonlinear optical and Kerr microscopy to search for time-reversal symmetry broken phases in 5*d* transtion metal oxides that are likely to exhibit domain formation. Whereas domains can obscure the presence of a broken symmetry when performing a spatially averaged experiment, a microscopy experiment overcomes this problem by spatially resolving these domains. Furthermore, we proposed to use nonlinear magneto-optical Kerr rotation as opposed to linear magneto-optical Kerr rotation to probe time-reversal symmetry breaking owing to their typically larger rotation angles.

We discovered that Sr_2IrO_4 exhibits a very large (~1°) SHG magneto-optical Kerr rotation that onsets below T_N (Fig. 5). This curve was obtained by measuring both the P- and S-polarized SHG output intensity with P-polarized incident light at a particular value of ϕ using the SHG-RA setup (Fig. 2). The magneto-optical Kerr rotation angle was then obtained by studying the changes in the P- and S-polarized output intensities. We also found that by sweeping our beam spot over the surface of the sample, we could find regions that exhibit an opposite sense of magneto-optical Kerr rotation, which suggests that domains are present.

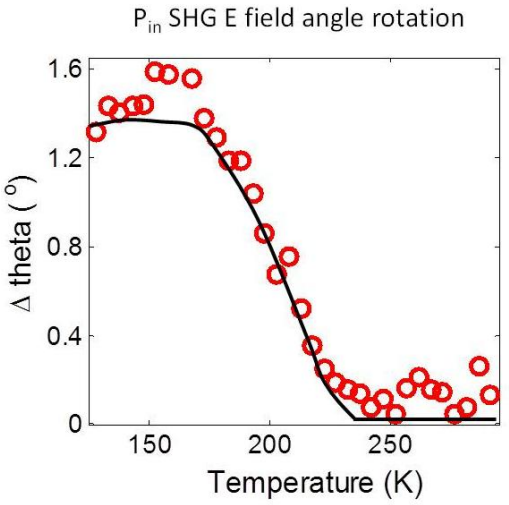


FIG. 5. SHG magneto-optical Kerr rotation obtained from the (001) surface of Sr_2IrO_4 at $\phi = 55°$ using P-polarized incident light.

In order to image the spatial distribution of these time-reversal symmetry broken domains in Sr_2IrO_4, through DURIP support we constructed a nonlinear optical microscope whose layout is shown in Fig. 6. A slightly oblique incidence angle geometry was used in order to distinguish between P- and S- polarized light, which does not introduce significant distortions in the image. In our current microscope setup, the light scattering plane is not rotatable. Therefore we first use the SHG-RA setup (Fig. 2) to identify the scattering plane angle ϕ where the largest intensity changes are observed below T_N, and then we mount the crystal/cryostat at a matching angle to perform the microscopy. Typical SHG microscopy images of Sr_2IrO_4 taken at the same angle ϕ used in Fig. 5 both above and below T_N are shown in Fig. 6. The images show the formation of at least two types of time-reversal symmetry broken domains below T_N with a size of order 100μm, which far exceeds our spatial resolution (~1μm). This shows that in addition to being able to resolve the symmetry of magnetic structures using SHG-RA, we also have the capability to map the spatial distribution of their domains. This work is currently in the manuscript preparation stage and we anticipate submitting the manuscript before the end of 2014.

To date we have performed such nonlinear optical microscopy studies at both liquid nitrogen and liquid helium temperatures. However the ultimate goal, as stated in our DURIP proposal, is to

reach sub-K temperatures using a custom He-3 optical cryostat. Although the magnetically shielded chamber and vacuum pumps necessary for these measurements are already available in our lab, we are still in the process of installing our He-3 cryostat and anticipate completion by early 2015.

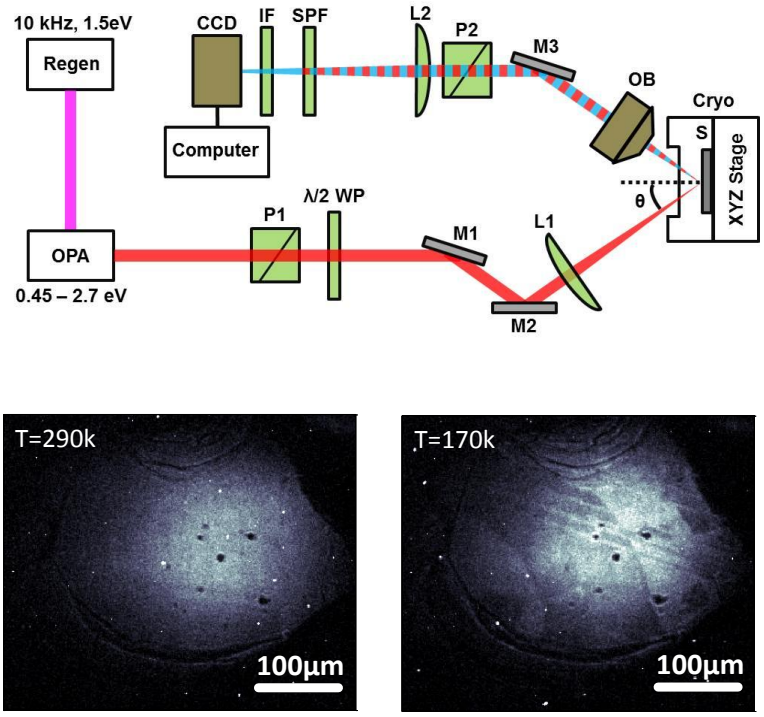


FIG. 6. (Top) Schematic layout of the SHG microscope. Laser pulses derived from an OPA are linearly polarized using a combination of a polarizing beamsplitter (P1) and half waveplate (λ/2 WP). The beam is then focused onto the sample at oblique incidence θ using a lens (L1). Mirrors are denoted M1, M2 and M3. The reflected beam is imaged onto a CCD camera via an objective lens (OB) and lens (L2). Polarization is selected via a polarizing beamsplitter (P2) and wavelength is filtered using a short-pass filter (SPF) and interference filter (IF). (Bottom) SHG images of Sr_2IrO_4 taken in PS geometry both above and below T_N. Striped domains of approximately 100μm width are clearly resolved.

III.C *Time- and energy-resolved optical spectroscopy*

We proposed to use time- and energy-resolved pump-probe optical reflectivity to understand the nature of the thermally driven spin-orbital entangled Mott insulator-to-metal phase transition in 5*d* transition metal oxides and to investigate the possibility of photo-inducing such a phase transition on ultrafast timescales. In our setup, which is realized through DURIP support and described in detail in Fig. 7, a femtosecond laser pump pulse is used to excite the system into a non-equilibrium state and a time delayed probe pulse is used to measure the instantaneous reflectivity. By collecting such data as a function of both time delay and probe wavelength the temporal evolution of spectral weight distribution can be obtained, which provides information about how the electronic structure of a material evolves following the pump excitation.

Using this setup shown in Fig. 7, we have performed preliminary measurements on Sr_2IrO_4 with particular focus on probe wavelengths of 800nm, 1,200nm and 2,400nm, which correspond to optical transitions from the O 2p to Ir J_{eff} = 1/2, J_{eff} = 3/2 to J_{eff} = 1/2 and J_{eff} = 1/2 to J_{eff} = 1/2 states respectively [4]. Data taken in the low pump fluence regime (Fig. 7) show no obvious

changes in temporal dynamics nor in temperature dependence across these wavelengths. We are currently in the process of 1) extending the probe wavelength range using a difference frequency generator (DFG: 2,400nm to 20,000nm) seeded by our optical parametric amplifier (OPA) and detecting using cryogenically cooled HgCdTe detectors; 2) testing electronics to perform analogue shot-to-shot normalization in effort to improve our sensitivity to fractional changes in reflectivity ($\Delta R/R$) from the current 10^{-4} regime into the 10^{-5} regime; and 3) developing a Kramers-Kronig transformation procedure [5] for converting our time- and wavelength resolved optical reflectivity data into a time-resolved optical conductivity. These improvements will enable us to perform more detailed searches for small changes in spectral weight distribution following optical excitation. We are also concurrently investigating the effect of higher pump fluences in attempt to optically induce an insulator-to-metal transition.

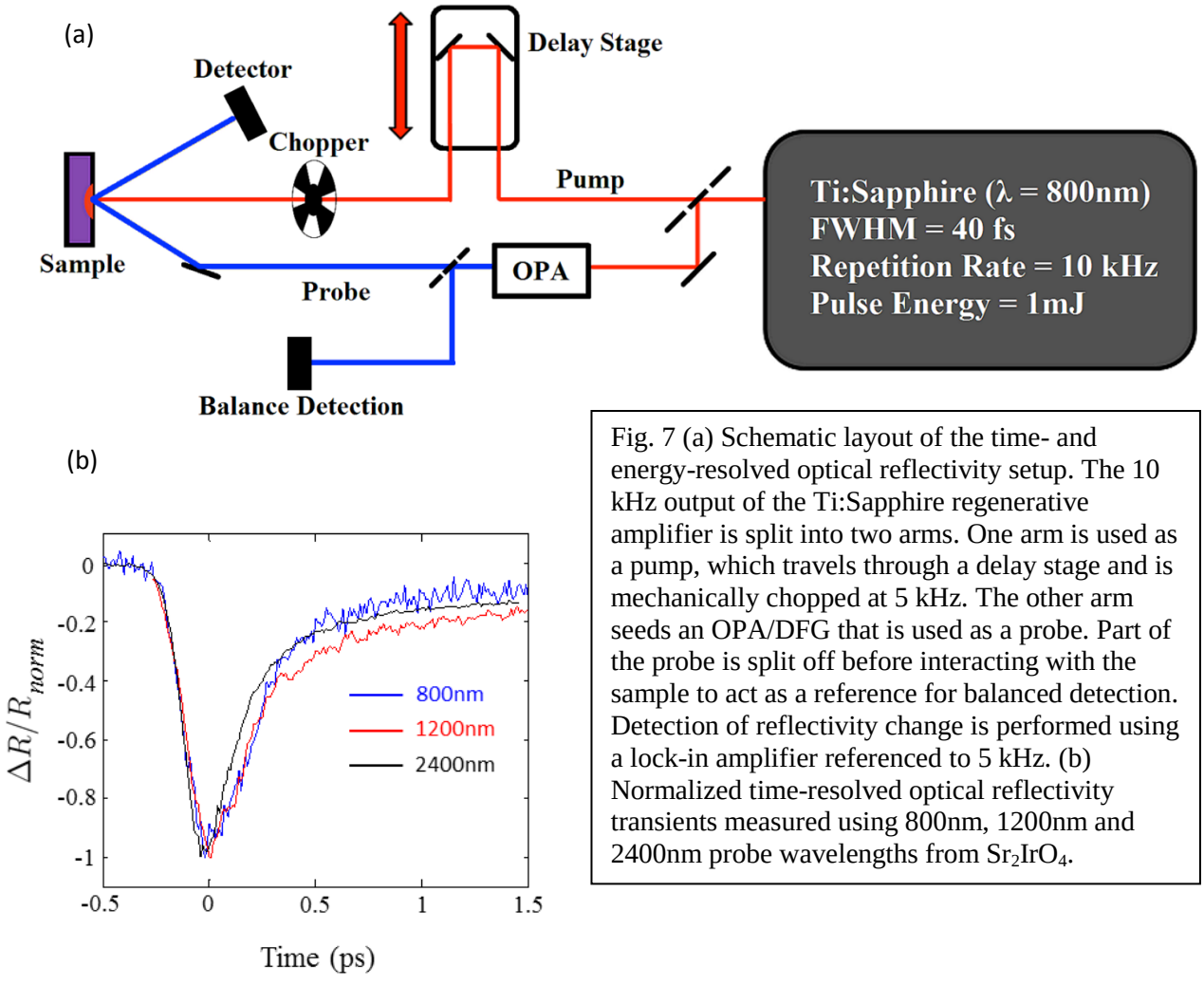
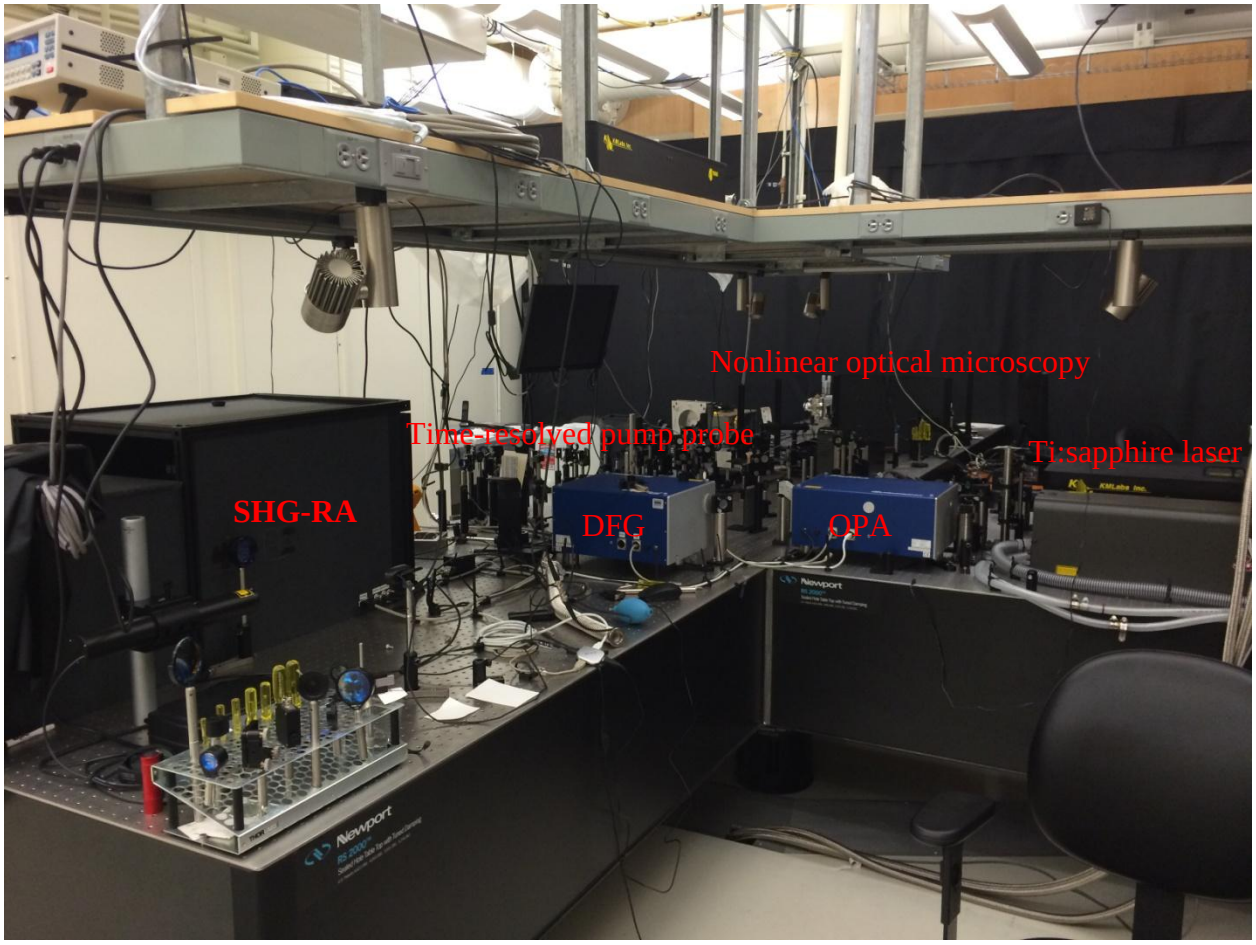


Fig. 7 (a) Schematic layout of the time- and energy-resolved optical reflectivity setup. The 10 kHz output of the Ti:Sapphire regenerative amplifier is split into two arms. One arm is used as a pump, which travels through a delay stage and is mechanically chopped at 5 kHz. The other arm seeds an OPA/DFG that is used as a probe. Part of the probe is split off before interacting with the sample to act as a reference for balanced detection. Detection of reflectivity change is performed using a lock-in amplifier referenced to 5 kHz. (b) Normalized time-resolved optical reflectivity transients measured using 800nm, 1200nm and 2400nm probe wavelengths from Sr_2IrO_4.

III.D *Physical layout of the experimental setups in Hsieh Lab*



IV. Bibliography

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