Final Close-Out Report for DE-FG02-00ER45818

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Principal Investigator:
Sashi Satpathy

Department of Physics and Astronomy,
University of Missouri, Columbia, MO 65211
Email: satpathys@missouri.edu

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Program Manager:
Dr. Matthias Graf
Basic Energy Sciences
U.S. Department of Energy
1000 Independence Avenue, SW.
Washington, DC 20585-1290

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Abstract

This is a final close-out report for DOE contract DE-FG02-00ER45818 entitled “Fundamental Studies of Complex Oxides and their Interfaces” at the University of Missouri, Columbia, Missouri. It funded research in Condensed Matter Physics Basic Energy Science from July 1, 2000 through Nov 30, 2022. Presented below are: (i) An overview of the research accomplishments during the entire grant period with highlights of research results from the last funding period, (ii) A list of postdocs and graduate students that received support from this grant, and (iii) A list of original papers published through the duration of the grant with impact factor metrics.

Overview

The DOE Basic Energy Sciences grant “Fundamental Studies of Complex Oxides and their Interfaces” supported the research of Professor Sashi Satpathy at the University of Missouri, Columbia, Missouri. The objective of the project was to develop the basic theory and understanding of the electronic and magnetic properties of complex oxides and their interfaces. The theoretical studies were based on density-functional calculations, from which model Hamiltonians were developed and solved using mean-field methods, exact diagonalization, and Lang-Firsov approach for the electron-phonon coupling. Over the years, the research topics evolved to take into account the emerging frontier of research in the area of complex oxide materials. Towards the end of the project period, insights developed from the complex oxide physics was applied to other 2D systems, such as the Rashba effect and the magnetic interactions including the RKKY and the Dzyaloshinskii-Moriya interactions. Seventy-six publications resulted from this research, which received 2822 citations as of May 8, 2022, or 37 cites per paper on average.

We summarize below very broadly the main accomplishments of this grant, and then in more detail the result of the last funding period. We divide our developments into three broad themes of research work.

Physics of CMR Manganites and Other Oxide Bulk Crystals [2000-2021]: This topic occupied us exclusively for the first five years, but continued off and on until the end of the grant period. The grant started out with primary focus on the physics of the bulk oxides, especially the colossal magneto-resistance (CMR) phenomena in the manganites compounds typified by $La_{1-x}Ca_xMnO_3$ and related materials. The physics of the CMR manganites is controlled by a score of competing interactions including electronic correlation effects, spin-orbit coupling, double and super exchange interaction, and the static and dynamic Jahn-Teller effects. A number of significant results was obtained to elucidate the physics of the manganites including studies of the electronic structure, theory of the exchange interaction [1], formation of the self-trapped magnetic polarons [7, 11], and charge-orbital order in the half-doped manganites [2], among others.

Oxide Interfaces [2005 – 2021]: Our expertise on the manganite physics naturally evolved into our work on the complex oxide interfaces. Though this work was done primarily between 2005-2014, work continued to some degree until 2021, the end of the grant period. Beginning in 2004, experimenters were able to grow atomically sharp, lattice matched oxide interfaces, which led to considerable excitement in the field, leading to the discovery of many phenomena involving two-dimensional electron gases (2DEG) at the interfaces and surfaces. Soon after the experimenters successfully grew the interfaces, we performed the very first density-functional study of the oxide interfaces [13], where we proposed the Airy-function localization of the electron states at the SrTiO3/LaTiO3 interface. This was followed by a series of work on the subject, notably on the: Double-exchange ferromagnetism [20], Origin of the 2DEG at the $LaAlO_3$ on $SrTiO_3$ Interface and the formation of the interface sub-bands [24], Prediction of the spin-polarized electron gas in the layer-doped SrMnO3 compound [28], Effect of strain [27], Electric field tuning of the Rashba effect [47], Electric field control of magnetism at the $CaMnO_3$/$CaRuO_3$ interface [76], and others. Many of these results were subsequently observed in a large number of experiments; Especially our work on the $LaAlO_3$ on $SrTiO_3$ Interface [24], which predicted the existence of the interface sub-bands, had a large impact in the field. Several works were performed during the years 2018-2021 on the iridate interfaces, where we predicted a large anomalous Hall effect [66, 69] and the existence of a spin-orbital entangled 2DEG [64].

2D Materials: ElecSpin/Orbital Transport, RKKY interaction [2011-2021]: The third broad research topic was the physics of the 2D materials, which was started in 2011, based on our considerable experience on the 2D-like systems from the study of the oxide interfaces and surfaces. These included the study of the Rudermann-Kittel-Kasuya-Yosida (RKKY) as well as the Dzyaloshinski-Moriya interactions in 2D electron gas and 2D materials like graphene. We studied the RKKY interaction in undoped and doped graphene [38, 39] from lattice Green's function, as well as the vacancy induced charge oscillations in graphene [42]. Rashba effect became a main topic of our research during this period. The effect requires the presence of an electric field, which is naturally present at the surfaces and interfaces due to broken symmetry. Rashba effect was studied for the polar perovskite structures, taking the example of the $LaTiO_3$ surface [47]; a model was developed for the d electron systems in general [49]; anisotropic Rashba effect was predicted for black phosphorous [51], an important 2D material; and a theory of the Dzyaloshinskii-Moriya interaction in the 2DEG in the presence of Rashba and Dresselhaus spin-orbit coupling was developed in Ref. [59]. During the last part of the project, we studied the spin and orbital Hall effects in solids. The orbital Hall effect, which involves the transport of the orbital moment, analogous to the transport of the spin moment in the spin Hall effect, is a relatively new effect, which however has not been conclusively established from experiments. As a step towards this direction, we proposed the 2D monolayer dichalcogenide materials as candidates for the observation of the orbital Hall effect [71, 73].

Below we report in more detail the highlights of work carried out in the last five-years of the funding period, during December 2016 – November 2021, which included a three-year regular grant period plus a two-year no-cost extension.

Highlights of Research (December 2015 – November 2021)

The research accomplishments in this period resulted in nineteen publications in refereed journals. We divide the resulting papers into four categories:

1. Theory of oxide interfaces
2. Theory of bulk solids including complex oxides
3. Magnetic exchange interactions
4. Orbital and Spin Hall effects

1. Theory of oxide interfaces

We continued our study of the physics of the oxide interfaces, extending our work to especially those with a large spin-orbit coupling, such as the iridate interfaces. From density-functional studies, we showed [64] that a novel type of spin-orbital entangled 2DEG forms at the $LaAlO_3$/Sr2IrO_4 polar interface between an ordinary band insulator and a spin-orbit coupled Mott insulator, aided by the combination of the spin-orbit coupling, Coulomb interaction, and polar catastrophe. Quite remarkably, the 2DEG was found to be localized to a single IrO_2 monolayer, with the electron gas occupying the upper Hubbard band in the interface layer. If successfully grown, this would be the first material to host the spin-orbital entangled 2DEG. In [66, 69], the possibility of tuning the anomalous Hall effect (AHE) at a magnetic interface by modifying the Rashba spin-orbit interaction with an electric field was demonstrated with a square lattice model as well as from explicit density-functional calculations of ($SrIrO_3$)/($SrMnO_3$), a recently grown iridate interface. The effect may be potentially useful in future spintronics applications. For the double exchange manganite interface $CaMnO_3$/$CaRuO_3$, the ferromagnetism at the interface can be similarly controlled by an electric field, but this time by controlling the amount of charge in the interfacial $CaMnO_3$ layer, which affects the double exchange interaction and therefore the magnetism [76].

2. Theory of bulk solids including complex oxides

We continued our work on the lanthanum manganites. Quite interestingly, in a theory-experiment collaboration [53, 54], using the Gutzwiller method to treat correlation effects, we showed that the pressure-induced metal-insulator transition in $LaMnO_3$ is fundamentally different from the Mott-Hubbard transition and is percolative in nature. The colossal magneto-resistance is inhomogeneity-driven, with the measured resistivity obeying the percolation scaling laws, while the temperature dependence followed the Efros-Shklovskii variable-range hopping behavior, clearly demonstrating the percolative nature of the metal-insulator transition. Along the line of our study of large spin-orbit coupled systems, in a series of papers [60, 61, 67], we studied the effect of the large spin-orbit coupling on the magnetic anisotropy and spin-wave gap in the well known osmate compound, NaOsO3, as well as the effect of strain on its optical properties. Strain effects on the prototypical iridate compound Sr_2IrO_4 was studied in [67] to understand the evolution of the electronic structure with strain including the Γ-X crossover of the valence bands. The dipole forbidden optical transitions between the Ir states, observed in the experiments, were explained in terms of the admixture of Ir p and d bands, which makes the optical matrix elements non-zero.

3. Magnetic exchange interactions

Chiral order in magnetic structures is currently an area of considerable interest and leads to skyrmion structures and domain walls with certain chirality. These magnetic structures are most often found in thin films and interfaces, originating from the Dzyaloshinskii-Moriya interaction, caused by broken inversion symmetry and the Rashba/Dresselhaus spin-orbit interaction, often present in the 2D structures. In [56, 59], we studied the exchange interaction between two localized magnetic moments in the 2D electron gas with both Rashba and Dresselhaus spin-orbit interaction present. Analytical expressions were found in certain limits, results that would be fundamental to discussions of the chiral structures in 2D materials. The stability of the Bloch and Néel domain walls in magnetic thin films were discussed in light of our results. Along these lines, we also studied the electronic structure and the origin of the Dzyaloshinskii-Moriya interaction in MnSi, which is a prototypical material for the experimental study of skyrmions.

4. Orbital and Spin Hall effects

Carrying information using the electronic degrees of freedom other than the electron charge such as the spin and orbital moments is an important emerging area of research, and it has led to the vision of electronics dubbed as "spintronics" and "orbitronics." While spintronics is a robust area of research, transport using orbital moments is relatively new. In fact, no definitive experiment has established the existence of the orbital current to date. In recent works [71, 73], we proposed the monolayer transition metal dichalcogenides (TMD) such as MoS_2 to be excellent candidates for the observation of orbital current, for example, as manifested in the orbital Hall effect (OHE). We showed that the already-existing opposite valley orbital moments, at different points of the Brillouin zone due to the broken inversion symmetry in TMDs, flow in opposite directions under the applied electric field due to the opposite Berry curvatures, leading to a large OHE. Simultaneously, we found the spin Hall effect in these materials to be relatively weak, which makes the TMDs to be particularly suitable for the direct observation of the OHE. A similar physics leads to the orbital magneto-electric effect in metallic TMDs such as NbSe2, when strain is present [74]. If observed, this would be the first orbital counterpart of the standard, spin-moment based magneto-electric effect.

Human Resources Development

Over the twenty-two year period, the grant supported several post-doctoral fellows and partially supported several graduate students. They continued to have careers in academia, industry, and administration (journal editor). The names and their current employment are listed below.

Post-Docs

1. Dr. Arti Kashyap (2005), Professor, L. N. Mittal Institute of Information Technology, Jaipur, India
2. Dr. Ranjit Nanda (2005 – 2010), Professor, Indian Institute of Technology, Chennai, India
3. Dr. Subodha Mishra (2006-2007), Professor, SOA University, Bhubaneswar, India (Deceased)
4. Dr. K. V. Shanavas (2011 – 2014), Data Scientist, New York
5. Dr. Mohammad Sherafati (2014), Instructor, Physics Department, University of Wisconsin-Madison, Wisconsin
6. Dr. Jamshid M. Kurdestany (2014 – 2017), Post-Doctoral Fellow, M. D. Anderson Cancer Center, Houston, Texas
7. Dr. Sayantika Bhowal (2017- 2020), Post-Doctoral Fellow, ETH, Zurich, Switzerland

Graduate Students (All partially funded by DOE)

1. Dr. Shashi Pathak (Ph. D., 2001), Systems Analyst, Verisign Inc., San Jose, California
2. Dr. Hakim Meskine, Ph. D. (2005), Editor, Physica E, Elsevier, Berlin, Germany
3. Dr. Sunita Thulasi, Ph. D. (2006), System Analyst, Intel Corporation, Seattle, Washington
4. Dr. Subodha Mishra, Ph. D. (2006), Professor, SOA University, Bhubaneswar, India (Deceased)
5. Mohammad Sherafati, PhD (2013), Instructor, Physics Department, University of Wisconsin-Madison, Wisconsin
6. Masoud Valizadeh, Ph. D. (2017), Financial Engineer, City Bank, Chicago
7. Gregory S. Kissinger, Ph. D. (2018), Current position: Unknown
8. Pratik Sahu, Ph. D. (2022), Post-Doctoral Fellow, Indian Institute of Technology, Chennai, India

Publications From the DOE Grant Between Nov 1, 2000 – Oct 31, 2022 with Citation Numbers

1. H. Meskine, H. Koenig, and S. Satpathy, Orbital Ordering and Exchange Interaction in the Manganites, Phys. Rev. B 64, 094433 (2001)
   62 citations as of May 8, 2022

2. Z. Popovic and S. Satpathy, Origin of Charge-Orbital Order in the Half-Doped Manganites, Phys. Rev. Lett. 88, 197201 (2002)
   85 citations as of May 8, 2022

3. H. Meskine, Z. S. Popovic, and S. Satpathy, Electronic structure and exchange interaction in the layered perovskite $Sr_3Mn_2O_7$, Phys. Rev. B 65, 094402 (2002)
   13 citations as of May 8, 2022

4. Z. Popovic and S. Satpathy, Charge Stacking in the Half-Doped Manganites, J. Appl. Phys. 91, 8132 (2002)
   2 citations as of May 8, 2022

5. S. Mishra and S. Satpathy, One-Dimensional Photonic Crystal: The Kronig-Penney Model, Phys. Rev. B 68, 045121 (2003)
   53 citations as of May 8, 2022

6 . T. Saha-Dasgupta and S. Satpathy, Wannier-like functions and tight-binding parametrization for the manganese bands in $CaMnO_3$, J. Phys. C 15, 1685 (2003)
   7 citations as of May 8, 2022

7. H. Meskine, T. Saha-Dasgupta, and S. Satpathy, Is the self-trapped magnetic polaron energetically stable in the electron doped manganites?, Phys. Rev. Lett. 92, 056401 (2004)
   45 citations as of May 8, 2022

8. S. Satpathy, Z. S. Popovic, and W. C. Mitchel, Theoretical Study of the Composition Dependence of the Band Offset and the Sheet Carrier Density in the $GaN/Al_xGa_{1-x}N$ Heterostructure, J. Appl. Phys. 95, 5597 (2004)
   15 citations as of May 8, 2022

9. S. Thulasi and S. Satpathy, Binding Energy of the Self-Trapped Magnetic Polaron: Effect of the Intra-Sublattice Electron Hopping, Int. J. Mod. Phys. B 18, 3213 - 3225 (2004)
   2 citations as of May 8, 2022

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12. H. Meskine and S. Satpathy, Ferrodistorsive orbital ordering in the layered nickelate NaNiO2: A density-functional study, J. Appl. Phys., 97, 10A314 (2005)
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13. Z. S. Popovic and S. Satpathy, Wedge-shaped potential and Airy-function electron localization in the oxide superlattices, Phys. Rev. Lett. 94, 176805 (2005)
100 citations as of May 8, 2022

14. T. Saha-Dasgupta, Z. Popovic, and S. Satpathy, Study of the insulating ground state in $CaFeO_3$ and $La_{1/2}Sr_{2/3}FeO_3$, Phys. Rev. B 72, 045143 (2005)
36 citations as of May 8, 2022

15. S. Mishra, S. Thulasi, and S. Satpathy, Spin Polarization via Electron Tunneling through an Indirect-Gap Semiconductor Barrier, Phys. Rev. B 72, 195 347 (2005)
23 citations as of May 8, 2022

16. H. Meskine and S. Satpathy, Electronic Structure and Magnetic Interactions in the Sodium Nickelate $NaNiO_2$, Phys. Rev. B 72, 224423 (2005)
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17. S. Thulasi and S. Satpathy, Density-Functional Study of the Two-Dimensional Airy Electron Gas at the Perovskite Titanate Interfaces, Phys. Rev. B 73, 125307 (2006)
5 citations as of May 8, 2022

18. Z. S. Popovic and S. Satpathy, Density-functional study of molybdenum purple bronze: A paradigm Luttinger liquid system, Phys. Rev. B 74, 045117 (2006)
41 citations as of May 8, 2022

19. Jun Chen, Paul Rulis, Lizhi Ouyang, S. Satpathy, and W. Y. Ching, Vacancy-enhanced ferromagnetism in Fe-doped rutile TiO_2, Phys. Rev. B 74, 235 207 (2007)
71 citations as of May 8, 2022

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66 citations as of May 8, 2022

21. P. Larson and S. Satpathy, "Effect of vacancies on ferromagnetism in GaN:Mn dilute magnetic semiconductors," Phys. Rev. B 76, 245 205 (2007)
77 citations as of May 8, 2022

22. S. Mishra and S. Satpathy, "Fiber-Mesh Photonic Molecules," Optics Comm. 281, 1077 (2008)
1 citations as of May 8, 2022

23. S. Mishra, G. S. Tripathi, and S. Satpathy, "Theory of photo-induced ferromagnetism in the dilute magnetic semiconductors," Phys. Rev. B 77, 125216 (2008)
15 citations as of May 8, 2022

24. Z. Popovic, S. Satpathy, and R. M. Martin, "Origin of the Two-Dimensional Electron Gas Carrier Density at the $LaAlO_3$ on $SrTiO_3$ Interface," Phys. Rev. Lett 101, 256801 (2008)
487 citations as of May 8, 2022

25. P. Larson, Z. Popovic, and S. Satpathy, "Lattice relaxation effects on the interface electron states in the perovskite oxide heterostructures: $LaTiO_3$ monolayer embedded in $SrTiO_3$," Phys. Rev. B 77, 245122 (2008)
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56 citations as of May 8, 2022

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67 citations as of May 8, 2022

32. S. Mishra and S. Satpathy, “Photoinduced magnetism in the ferromagnetic semiconductors,” Int. J. Mod. Phys. B 24, 359 (2010)
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33. B. R. K. Nanda and S. Satpathy, “Magnetic and Orbital Order in $LaMnO_3$ under Uniaxial Strain: A Model Study,” Phys. Rev. B 81, 174 423 (2010)
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41. M. Sherafati and S. Satpathy, “On the Ruderman-Kittel-Kasuya-Yosida interaction in graphene,” AIP Conf. Proc. 1461, 24 (2012)
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51. Z. S. Popovic, J. M. Kurdestany, and S. Satpathy, “Electronic Structure and Anisotropic Rashba Spin-Orbit Coupling in Monolayer Black Phosphorus,” Phys. Rev. B 92, 035135 (2015)
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52. M. Valizadeh and S. Satpathy, RKKY Interaction for the Spin Polarized Electron Gas, Int. J. Mod. Phys. B 29, 155 0219 (2015)

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53. M. Baldini, T. Muramatsu, M. Sherafati, H-K Mao, L. Malavasi, P. Postorino, S. Satpathy, and V. V. Struzhkin, Inhomogeneity-driven colossal magneto-resistance in compressed $LaMnO_3$, Proc. Natl. Acad. Sciences 112, 10869 (2015)
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55. K. V. Shanavas and S. Satpathy, "Electronic structure and the Dzyaloshinskii-Moriya interaction in MnSi," Phys. Rev. B 93, 195101 (2016)
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60. S. Mohapatra, C. Bhandari, S. Satpathy, and A. Singh, "Effects of the structural distortion on the electronic band structure of $NaOsO_3$ studied within density functional theory and a three-orbital model," Phys. Rev. B 97, 155 154 (2018)
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64. Churna Bhandari and S. Satpathy, "Spin-orbital entangled 2DEG at the $LaAlO_3/Sr_2IrO_4$ interface," Phys. Rev. B (RC) 98, 041303 (2018)
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4 citations as of May 8, 2022

68. Churna Bhandari and S. Satpathy, "Two dimensional electron gas in the δ-doped iridates with strong spin-orbit coupling: $La_{\delta}Sr_2IrO_4$," J. Phys.: Condens. Matter 31, 435505 (2019)
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