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EUO--BASED MULTIFUCTIONAL HETEROSTRUCTURES

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Using molecular-beam epitaxy heterostructures were grown in which EuO was controllably doped, strained, patterned, and integrated with other materials to enable novel effects and emerging device concepts to be explored. These included (1) achieving a drastic increase of the magnetoresistance and the metal-insulator transition resistance ratios of doped EuO by interfacing this semiconductor with niobium; the observed effect is general and may be applied to any metal/semiconductor interface where the semiconductor shows large Zeeman splitting under magnetic field, (2) understanding the changes in electronic structure and Fermi-surface reconstruction that occur as doped EuO progresses through the ferromagnetic metal-insulator transition; a lack of carrier activation arises from defect states near the Γ point, (3) demonstrating that ultrafast optical pulses can be used to strengthen or weaken the magnetic order of doped EuO by ~10% in <3 ps, (4) establishing a new method for making EuO films by high-vacuum techniques with perfection and properties previously accessible only by ultra-high vacuum methods, and (5) straining EuO by over 6% to achieve the simplest and highest temperature strong multiferroic known (simultaneously ferromagnetic and ferroelectric).

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ABSTRACT

Using molecular-beam epitaxy heterostructures were grown in which EuO was controllably doped, strained, patterned, and integrated with other materials to enable novel effects and emerging device concepts to be explored. These included (1) achieving a drastic increase of the magnetoresistance and the metal-insulator transition resistance ratios of doped EuO by interfacing this semiconductor with niobium; the observed effect is general and may be applied to any metal/semiconductor interface where the semiconductor shows large Zeeman splitting under magnetic field, (2) understanding the changes in electronic structure and Fermi-surface reconstruction that occur as doped EuO progresses through the ferromagnetic metal-insulator transition; a lack of carrier activation arises from defect states near the Γ point, (3) demonstrating that ultrafast optical pulses can be used to strengthen or weaken the magnetic order of doped EuO by ~10% in <3 ps, (4) establishing a new method for making EuO films by high-vacuum techniques with perfection and properties previously accessible only by ultra-high vacuum methods, and (5) straining EuO by over 6% to achieve the simplest and highest temperature strong multiferroic known (simultaneously ferromagnetic and ferroelectric).

INTRODUCTION

EuO has long been realized to have exceptional optical, magnetic, and electronic properties as well as coupled charge-spin-lattice properties that are relevant to optical switching, spintronics, and emerging devices.[1] For example, EuO has (1) a metal–insulator transition with the largest change in resistance ($\Delta R/R > 10^{13}$) of any material,[2,3] (2) the largest Verdet constant (ν=4×10^5 ° (T·cm)$^{-1}$)—a measure of the strength of the Faraday effect,[4] and (3) the second highest spin polarization (>90%, which we updated to >96% with the support of this AFOSR program[5]) of any known material.[6] Although its ferromagnetic transition temperature is relatively low (69 K),[7] because it exhibits the highest known coefficients for many properties, its development in semiconductor-quality form will enable it to be used as a model system to allow novel effects and emerging device concepts to be explored. In this program we have perfected the growth of EuO using molecular-beam epitaxy (MBE) and developed semiconductor-grade heterostructures in which EuO can be controllably doped, strained, patterned, and epitaxially integrated with other metals, semiconductors, and insulators to enable bandgap engineering concepts to be applied to this unusual II-VI semiconductor. As we have developed a materials technology base for EuO, we have supplied appropriate samples to numerous collaborators around the world to enable novel effects and emerging device concepts to be explored.

KEY PERSONNEL

The numerous collaborations enabled by this AFOSR program have led to 15 published papers in refereed journals[5,8-21] including two in *Nature Communications*,[20,21] two in *Physical Review Letters*,[9,13] and one in *Advanced Materials*.[10] At Cornell the support of this AFOSR program led to the Ph.D. thesis of Alex Melville, who is now employed at MIT Lincoln Laboratory. Alex's Ph.D. thesis was entitled *Assessing the Effects of Biaxial Strain and Chemical Doping on the Magnetic and Electronic Properties of Epitaxial Europium Monoxide Films*. It also partially supported postdoc Dr. Rainer Held. Key among our collaborators was Dr. Andreas Schmehl in Jochen Mannhart's group at the University of Augsburg, who completed his Habilitation Degree entitled *Studies on Europium Monoxide—A Key Material for the Understanding of Magneto-Electronic Materials, Heterostructures and Interfaces*. Andreas had a graduate student working with him, Thomás Mairoser, who during this program completed his

Ph.D. dedicated to the electrical transport measurements (Hall and magnetization) of doped EuO films to help us establish the optimal growth conditions for the controlled growth of rare-earth-doped EuO films. Thomás' Ph.D. thesis is entitled *In Situ Neutronen-Reflektometrie und Untersuchungen von EuO-Filmen*.

RESEARCH HIGHLIGHTS

Together with the collaborators in Jochen Mannhart's group at the University of Augsburg, Germany and collaborators at Cornell we attempted to answer the key question for higher-temperature applications of EuO, "Is there an intrinsic limit to the charge carrier-induced increase of the Curie temperature of EuO?"[9] Rare earth doping is the key strategy to increase the Curie temperature (T_c) of the ferromagnetic semiconductor EuO. The interplay between doping and charge carrier density (n), and the limit of the T_c increase, however, are yet to be understood. Using a combination of Hall effect, x-ray absorption spectroscopy, and SQUID we measured n and T_c of Gd-doped EuO over a wide range of doping levels. The results show a direct correlation between n and T_c, with both exhibiting a maximum at high doping. On average, less than 35% of the dopants act as donors (with the rest being electrically inactive!), raising the question about the limit to increasing T_c.

Also in collaboration with Jochen Mannhart's group at the University of Augsburg, Germany we have worked to increase magnetoresistance using magnetic-field-tunable interface.[10] Although the T_c of EuO can be increased significantly with rare-earth doping, this increase comes at the cost of drastically reduced resistance ratios, both for the colossal magnetoresistive effect and the metal-to-insulator transition (MIT). We found a way, however, to overcome this intrinsic predicament. We achieved a drastic increase of the magnetoresistance and the MIT resistance ratios of bulk $Eu_{0.99}La_{0.01}O$ by interfacing this semiconductor with niobium. The transparency of this interface critically depends on the large Zeeman splitting of the $Eu_{0.99}La_{0.01}O$ conduction band, allowing for the temperature and magnetic field induced switching between non-linear and linear current-voltage characteristics, associated with tunneling-dominated and metallic transport across the interface.

In collaboration with Manfred Fiebig's group at the University of Bonn, Germany we have seen a large nonlinear magneto-optical effect in the centrosymmetric ferromagnetic semiconductor EuO.[8,12,14] Although EuO is known for its huge Verdet coefficient and associated magneto-optic effects, surprisingly it had *never* before been characterized by nonlinear optical techniques. Doing so on our high quality EuO films, our collaborators observed pronounced magnetic-dipole-induced second-harmonic generation (SHG). The SHG light wave emerges background-free at the Curie temperature and couples linearly to the spontaneous magnetization. The SHG spectrum is determined by spin-allowed and spin-forbidden transitions between the $4f$ ground state and the $5d(t_{2g})$ states of the Eu^{2+} ion. At the coercive field, components of the spontaneous magnetization perpendicular to the applied field are observed. Spatially resolved hysteresis measurements reveal that the ferromagnetic domains possess an average extension of $<1\ \mu$m although pinning effects can stabilize domains that are two orders of magnitude larger.[14]

Also in collaboration with Manfred Fiebig's group (which during this project moved to the Swiss Federal Institute of Technology in Zurich, Switzerland) we have tuned ferromagnetism optically.[20] Interest in manipulating the magnetic order by ultrashort laser pulses has thrived since it was observed that such pulses can be used to alter the magnetization on a sub-picosecond timescale. Usually this involves demagnetization by laser heating or, in rare cases, a transient increase of magnetization. What we demonstrated is a mechanism that allows the magnetic order of a material to be enhanced or attenuated at will. This is possible in systems simultaneously

possessing a low, tunable density of conduction band carriers and a high density of magnetic moments. In such systems, the thermalization time can be set such that adiabatic processes dominate the photoinduced change of the magnetic order—the three-temperature model for interacting thermalized electron, spin and lattice reservoirs is bypassed. In ferromagnetic $Eu_{1-x}Gd_xO$, we demonstrated the strengthening as well as the weakening of the magnetic order by ~10% and within <3 ps by optically controlling the magnetic exchange interaction.[20]

In collaboration with Jochen Mannhart's group we found a way to extend the large metal-to-insulator transition (MIT) of EuO to higher temperatures.[8] Using rare-earth doping, the Curie temperature (T_C) of EuO can be increased from 69 K to ~170 K.[22,23] This increase, however, comes at the cost of drastically reduced resistance ratios, both for the CMR effect and the MIT.[6] We found a way to overcome this intrinsic predicament. We were able to achieve a drastic increase of the magnetoresistance and the MIT resistance ratios of bulk $Eu_{0.99}La_{0.01}O$ by interfacing this semiconductor with niobium.[8] The transparency of this interface critically depends on the large Zeeman splitting of the $Eu_{0.99}La_{0.01}O$ conduction band,[24,25] allowing for the temperature and magnetic field induced switching between non-linear and linear current-voltage characteristics, associated with tunneling-dominated and metallic transport across the interface.[8] The result is an enhanced magnetoresistance and metal-to-insulator transition in highly electron-doped europium monoxide by interfacing it with niobium. By optimizing the rare-earth doping level, the interface properties are tuned to the point, at which changes of the band structure and of the charge carrier density cause a transition of the interface between tunneling-dominated and metallic transport. The doping levels necessary for achieving these improved properties are high enough to benefit from the carrier-mediated increase in Curie temperature. This same principle is applicable to any metal/semiconductor interface where the semiconductor shows large Zeeman splitting under magnetic field, e.g., dilute magnetic semiconductors.

Also in collaboration with Jochen Mannhart's group we studied the influence of the substrate temperature on the Curie temperature and charge carrier density of epitaxial Gd-doped EuO films.[11] Rare earth doping is a standard, yet experimentally poorly understood method to increase the Curie temperature (T_C) of the ferromagnetic semiconductor EuO. We investigated the dependence of the charge carrier density (n) and the T_C of 4.2 at. % Gd-doped EuO films grown by MBE on (110)-oriented $YAlO_3$ on substrate temperature (T_{sub}). Increasing T_{sub} leads to a decrease in n and T_C. For high substrate temperatures the Gd-doping is rendered completely inactive: n and T_C drop to the values of undoped EuO.

In collaboration with Kyle Shen's group at Cornell University, who has connected an angle-resolved photoemission spectroscopy (ARPES) system to our oxide MBE, we have determined the temperature dependence of the electronic structure and Fermi-surface reconstruction of $Eu_{1-x}Gd_xO$ through the ferromagnetic metal-insulator transition.[13] This capability enabled us to measure the ARPES spectra of $Eu_{1-x}Gd_xO$ through the ferromagnetic metal-insulator transition. In the ferromagnetic phase, we observed Fermi surface pockets at the Brillouin zone boundary, consistent with density functional theory, which predicts a half metal. Upon warming into the paramagnetic state, our results reveal a strong momentum-dependent evolution of the electronic structure, where the metallic states at the zone boundary are replaced by pseudogapped states at the Brillouin zone center due to the absence of magnetic long-range order of the Eu 4*f* moments.[13]

In collaboration with Jochen Mannhart's group at the University of Augsburg we studied the effect of lutetium doping on the structural, electronic, and magnetic properties of epitaxial EuO thin films grown by reactive molecular-beam epitaxy.[5] The behavior of Lu-doped EuO was

contrasted with doping by lanthanum and gadolinium. All three dopants were found to behave similarly despite differences in electronic configuration and ionic size. Andreev reflection measurements on Lu-doped EuO revealed a spin-polarization of 96% in the conduction band, despite non-magnetic carriers introduced by 5% lutetium doping.[5] This high spin polarization, second only to CrO_2 with 98% spin polarization,[26] is in agreement with previous reports of the near-complete spin-polarization of EuO.[6] The high spin-polarization despite 5% doping of nonmagnetic atoms is critically important, as it shows that EuO retains its high spin-polarization in the presence of a dopant which significantly boosts its T_C. This is not so surprising given that doped EuO is a half-metal due to the spin-splitting of the conduction band,[25,27] so the carriers are spin-polarized anyway despite the nature of the dopant.

We further showed how the transition temperature of EuO could be shifted by biaxial strain in epitaxial films,[16] epitaxially integrated EuO with diamond,[18] and in collaboration with David Muller's group at Cornell succeeded in imaging EuO for the first time by transmission electron microscopy.[19] Spin injection is greatly affected by the quality of the interface between the injector and silicon. Using atomic-resolution scanning transmission electron microscopy in conjunction with electron energy loss spectroscopy we directly imaged and chemically characterized a series of EuO/Si and EuO/$YAlO_3$ interfaces fabricated using different growth conditions.[19] We identified the presence of europium silicides and regions of disorder at the EuO/Si interfaces, imperfections that could significantly reduce spin injection efficiencies via spin-flip scattering.

Finally, in collaboration with Andreas Schmehl and Thomás Mairoser at the University of Augsburg in Germany, we discovered a way to make high quality epitaxial EuO films via a topotactic transformation of epitaxial Eu_2O_3 films.[21] Epitaxy is widely employed to create highly oriented crystalline films. A less appreciated, but nonetheless powerful means of creating such films is via topotactic transformation, in which a chemical reaction transforms a single crystal of one phase into a single crystal of a different phase, which inherits its orientation from the original crystal. Topotactic reactions may be applied to epitactic films to substitute, add, or remove ions to yield epitactic films of different phases. We exploited a topotactic reduction reaction to provide a non-ultra-high vacuum (non-UHV) means of growing highly oriented single crystalline thin films of the easily over-oxidized half-metallic semiconductor EuO with a perfection rivalling that of the best films of the same material grown by molecular-beam epitaxy or UHV pulsed-laser deposition. As the technique only requires high-vacuum deposition equipment, it has the potential to drastically improve the accessibility of high-quality single crystalline films of EuO as well as other difficult to synthesize compounds.[21]

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1. M. Matsubara, A. Schmehl, J. Mannhart, D.G. Schlom, and M. Fiebig, “Large Nonlinear Magneto-Optical Effect in the Centrosymmetric Ferromagnetic Semiconductor EuO,” Physical Review B 81 (2010) 214447.

2. T. Mairoser, A. Schmehl, A. Melville, T. Heeg, L. Canella, P. Böni, W. Zander, J. Schubert, D.E. Shai, E.J. Monkman, K.M. Shen, D.G. Schlom, and J. Mannhart, “Is there an Intrinsic Limit to the Charge-Carrier-Induced Increase of the Curie Temperature of EuO?,” Physical Review Letters 105 (2010) 257206.

3. A. Schmehl, D.G. Schlom, and J. Mannhart, “Increasing Magnetoresistance Using Magnetic-Field-Tunable Interfaces,” Advanced Materials 23 (2011) 1242-1245.

4. T. Mairoser, A. Schmehl, A. Melville, T. Heeg, W. Zander, J. Schubert, D.E. Shai, E.J. Monkman, K.M. Shen, T.Z. Regier, D.G. Schlom, and J. Mannhart, “Influence of the Substrate Temperature on the Curie Temperature and Charge Carrier Density of Epitaxial Gd-Doped EuO Films,” Applied Physics Letters 98 (2011) 102110.

5. M. Matsubara, C. Becher, A. Schmehl, J. Mannhart, D.G. Schlom, and M. Fiebig, “Optical Second- and Third-Harmonic Generation on the Ferromagnetic Semiconductor Europium Oxide,” Journal of Applied Physics 109 (2011) 07C309.

6. A. Melville, T. Mairoser, A. Schmehl, D.E. Shai, E.J. Monkman, J.W. Harter, T. Heeg, B. Holländer, J. Schubert, K.M. Shen, J. Mannhart, and D.G. Schlom, “Lutetium-doped EuO Films Grown by Molecular-Beam Epitaxy,” Applied Physics Letters (2012) 222101.

7. D.E. Shai, A.J. Melville, J.W. Harter, E.J. Monkman, D.W. Shen, A. Schmehl, D.G. Schlom, and K.M. Shen, “Temperature Dependence of the Electronic Structure and Fermi-Surface Reconstruction of Eu1−xGdxO through the Ferromagnetic Metal-Insulator Transition,” Physical Review Letters 108 (2012) 267003.

8. M. Matsubara, A. Schmehl, J. Mannhart, D.G. Schlom, and M. Fiebig, “Giant Third-Order Magneto-Optical Rotation in Ferromagnetic EuO,” Physical Review B 86 (2012) 195127.

9. T. Mairoser, F. Loder, A. Melville, D.G. Schlom, and A. Schmehl, “Influence of Chemical Doping on the Magnetic Properties of EuO,” Physical Review B 87 (2013) 014416.

10. A. Melville, T. Mairoser, A. Schmehl, T. Birol, T. Heeg, B. Holländer, J. Schubert, C.J. Fennie, and D.G. Schlom, “Effect of Film Thickness and Biaxial Strain on the Curie Temperature of EuO,” Applied Physics Letters 102 (2013) 062404.

11. D.M. Kaz, C.G. Bischak, C.L. Hetherington, H.H. Howard, X. Marti, J.D. Clarkson, C. Adamo, D.G.

Schlom, R. Ramesh, S. Aloni, D.F. Ogletree, and N.S. Ginsberg, “Bright Cathodoluminescent Thin Films for Scanning Nano-Optical Excitation and Imaging,” ACS Nano 7 (2013) 10397-10404.

12. A. Melville, T. Mairoser, A. Schmehl, M. Fischer, S. Gsell, M. Schreck, D.D. Awschalom, T. Heeg, B. Holländer, J. Schubert, and D.G. Schlom, “Epitaxial Growth of Europium Monoxide on Diamond,” Applied Physics Letters 103 (2013) 222402.

13. J.A. Mundy, D. Hodash, A. Melville, R. Held, T. Mairoser, D.A. Muller, L.F. Kourkoutis, A. Schmehl, and D.G. Schlom, “Hetero-Epitaxial EuO Interfaces Studied by Analytic Electron Microscopy,” Applied Physics Letters 104 (2014) 091601.

14. M. Matsubara, A. Schroer, A. Schmehl, A. Melville, C. Becher, M.T. Martinez, D.G. Schlom, J. Mannhart, J. Kroha, and M. Fiebig, “Ultrafast Optical Tuning of Ferromagnetism via the Carrier Density,” Nature Communications 6 (2015) 6724.

15. T. Mairoser, J.A. Mundy, A. Melville, D. Hodash, P. Cueva, R. Held, A. Glavic, J. Schubert, D.A. Muller, D.G. Schlom, and A. Schmehl, “High-Quality EuO Thin Films the Easy Way via Topotactic Transformation,” Nature Communications (in press, 2015).

Changes in research objectives (if any):

none

Change in AFOSR Program Manager, if any:

Kitt Reinhardt --> James Hwang --> Kenneth C. Goretta

Extensions granted or milestones slipped, if any:

none

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, $K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
Total			

Report Document

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Appendix Documents

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