AFRL-AFOSR-VA-TR-2022-0010



New Superconductors near Broken Rotational Symmetry Instabilities

Chu, Jiun-Haw UNIVERSITY OF WASHINGTON 4333 BROOKLYN AVE NE SEATTLE, WA, US

10/24/2021 Final Technical Report

DISTRIBUTION A: Distribution approved for public release.

Air Force Research Laboratory
Air Force Office of Scientific Research
Arlington, Virginia 22203
Air Force Materiel Command

REPORT DOCUMENTATION PAGE

Form Approved OMB No. 0704-0188

The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing the burden, to Department of Defense, Washington Headquarters Services, Directorate for Information Operations and Reports (0704-0188), 1215 Jefferson Davis Highway, Suite 1204, Arlington,

U	U	U	UU	4	19b. TEI	LEPHONE NUMBER (Include area code)
a. REPORT	b. ABSTRACT	c. THIS PAGE	ABSTRACT	OF PAGES	KENNETH GORETTA	
	CLASSIFICATION	OF:	17. LIMITATION OF	18. NUMBER		ME OF RESPONSIBLE PERSON
instabilities in the superconducting character. In the understanding superconductive developed a neex-ray diffraction a striking transproperconductor	re superconductivity ne iron-based and comparing might be experied and the application and the application are experimental plater evealed port-structural corrects. It also uncovered on pnictides. During	opper-based materinhanced by the quath program, we have of this idea. We dische nematic fluctuatiform, the elasto x-raspondence of the ned an unusual couplir	cinity of rotational symmials. It has been argued antum critical fluctuation made significant progrecovered a new mechanions with an anisotropic ay diffraction, to study the matic state in the iron progressive to the study of the state in the period, we have total states.	that the s with a nematic ess towards the sm to control strain. We also ne electronlattice conictide	oupling of t	the broken rotational symmetry phase. The elasto
A Distribution U	TION/AVAILABILIT Inlimited: PB Public ENTARY NOTES					
AF Office of Sc	ientific Research oh St. Room 3112	AGENCY NAME(S)	AND ADDRESS(ES)		10. SPONSOR/MONITOR'S ACRONYM(S) AFRL/AFOSR RTA1 11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-AFOSR-VA-TR-2022-0010	
UNIVERSITY OF WASHINGTON 4333 BROOKLYN AVE NE SEATTLE, WA US REPORT NUMBER						
						WORK UNIT NUMBER
6. AUTHOR(S) Jiun-Haw Chu					5d. PROJECT NUMBER 5e. TASK NUMBER	
					5c. PROGRAM ELEMENT NUMBER 61102F	
New Superconductors near Broken Rotational Symmetry Instabilities					5b. GRANT NUMBER FA9550-17-1-0217	
4. TITLE AND	SUBTITLE	Fillal			5a. CONTRACT NUMBER	
PLEASE DO N	OT RETURN YOU	R FORM TO THE A	BOVE ADDRESS. PORT TYPE		3. DATES COVERED (From - To) 01 Jul 2017 - 30 Jun 2021	
			otwithstanding any othe splay a currently valid O			shall be subject to any penalty for failing to

Standard Form 298 (Rev.8/98) Prescribed by ANSI Std. Z39.18

AFOSR-YIP Final Report

Award Information

Project title: New Superconductors near Broken Rotational Symmetry Instabilities

Award number: FA9550-17-1-0217;

Report date: 09/20/2021

Period covered by the report: July 1st 2017 – July 1st 2021.

Principle Investigator: Jiun-Haw Chu (Department of Physics, University of Washington,

Seattle, WA 98195)

Email: jhchu@uw.edu; phone: 650-862-2808

Program Manager: Kenneth C. Goretta, kenneth.goretta@us.af.mil

Narrative

High temperature superconductivity emerges in the vicinity of rotational symmetry-breaking instabilities in the iron-based and copper-based materials. It has been argued that the superconducting paring might be enhanced by the quantum critical fluctuations with a nematic character. In this four-year research program, we have made significant progress towards the understanding and the application of this idea. We discovered a new mechanism to control superconductivity by suppressing the nematic fluctuations with an anisotropic strain. We also developed a new experimental platform, the elasto x-ray diffraction, to study the electron-lattice coupling of the broken rotational symmetry phase. The elasto x-ray diffraction revealed a striking transport-structural correspondence of the nematic state in the iron pnictide superconductors. It also uncovered an unusual coupling between the Eu and Fe moments in the Eu based iron pnictides.

During the four-year report period, we have total 5 publications, including one Nature Materials, one Nature Physics and one Nature Communications. Below we present a summary of research highlights.

a. Drastic suppression of superconductivity near a nematic quantum critical point (Nature Physics 2020)

Most unconventional and high temperature superconductors share a similar phase diagram. As the system is tuned by chemical doping or pressure, superconductivity emerges as a nearby symmetry breaking phase is suppressed. The empirical observation of this common phase diagram has led to the belief that the fluctuations associated with the symmetry breaking phase are beneficial, if not responsible, for the superconducting pairing. A direct test to verify this hypothesis is to observe the decrease of superconducting T_c by applying the symmetry breaking conjugate field that suppresses the dynamic fluctuations of the competing order. Nevertheless, most of the competing phases in unconventional superconductors break translational symmetry, which requires a spatially modulated conjugate field that is difficult to realize experimentally. In this work, we show that anisotropic strain, the conjugate field of nematicity, drastically suppresses the superconducting T_c of an iron pnictide superconductor. We discovered a fivefold reduction of T_c for the optimally doped samples for less than one percent of strain, whereas for the underdoped samples the suppression of T_c extends to zero temperature with the recovery of a fully metallic ground state. In

addition to providing direct evidence of the role played by the nematic fluctuations in the formation of the superconducting state, these results also demonstrate tunable mechanical control of a high temperature superconductor, an important step forward regarding the technological applications of superconductivity, as well as provide a new platform for studying a superconductor to metal

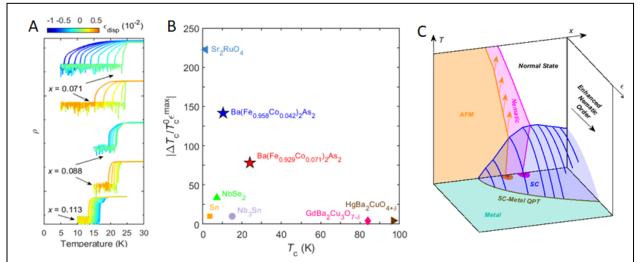


Fig. 1 Suppression of superconductivity by anisotropic strain (A) Resistivity as a function of temperature under uniaxial stress for optimally and overdoped compositions. Resistivity is on a log scale and sets of curves that overlap are offset for clarity (**B**) Sensitivity of T_c to strain for a variety of superconductors. (C) Schematic doping-strain-temperature $(x-\epsilon-T)$ phase diagram.

quantum phase transition in a three-dimensional system.

b. The transport structural correspondence across the nematic transition probed by elasto x-ray diffraction (Nature Materials 2021)

Electronic nematicity in iron pnictide materials is coupled to both the lattice and the conducting electrons, which allows both structural and transport observables to probe nematic fluctuations and the order parameter. A key question is in what regime one can establish a one-to-one correspondence between transport and structural variables. This question is especially important for 2D systems such as graphene where thermodynamic measurements are difficult. It also has important implications to the nearby superconducting phase, since the transport measurements probe the conducting electrons that form copper pars. In this work, we combine simultaneous transport and x-ray diffraction measurements with in-situ tunable strain (elasto-XRD) to measure the temperature dependence of the shear modulus and elastoresistivity above the nematic transition and the spontaneous orthorhombicity and resistivity anisotropy below the nematic transition, all within a single sample of Ba(Fe_{0.96}Co_{0.04})₂As₂. With our unprecedented multi-modal measurement, we show the four quantities perfectly follows a mean-field temperature dependence. Furthermore, the ratio of transport to structural quantities is a constant across the phase transition, suggesting that the resistivity anisotropy behaves just like a thermodynamic variable even for large values of the nematic order parameter. While the shear modulus and elastoresistivity and the spontaneous orthorhombicity and resistivity anisotropy can be well described by the Landau free energy framework, two unexpected findings stand out. First, using the shear modulus and

elastoresistivity data from the previous studies, we discovered a strong doping dependence of the ratio between transport and structural quantities, increasing by more than fivefold towards optimal doping. Second, when driving the system deep into the non-linear regime with large uniaxial stress, we found that the resistivity anisotropy shows a non-saturating behavior that is drastically different from the dampened response of the lattice, which may reveal the intertwined nature of nematicity.

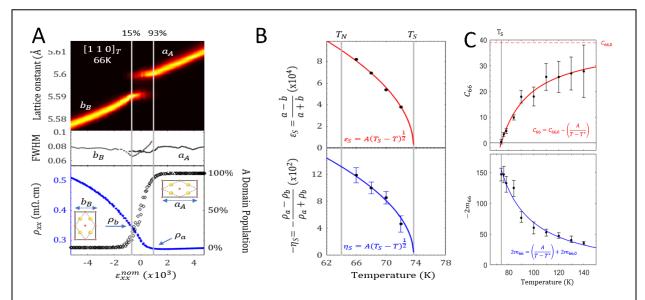


Fig. 2 Elasto X-ray diffraction measurement (A) Simultaneous x-ray diffraction (upper) and resistivity (lower, blue curve) measurement as a function of strain. These measurements allow us to extract spontaneous lattice distortion (B, upper), resistivity anisotropy (B, lower), shear modulus (C, upper) and elastoresistivity (C, lower) all in one sample.

c. Strongly anisotropic antiferromagnetic coupling in EuFe₂As₂ revealed by stress detwinning (Phys. Rev. B 2021)

Of all parent compounds of iron-based high-temperature superconductors, EuFe₂As₂ exhibits by far the largest magneto-structural coupling due to the sizable biquadratic interaction between Eu and Fe moments. While the coupling between Eu antiferromagnetic order and Fe structural/antiferromagnetic domains enables rapid field detwinning, this prevents simple magnetometry measurements from extracting the critical fields of the Eu metamagnetic transition. In this work, we measure these critical fields by combining x-ray magnetic circular dichroism spectroscopy with in-situ tunable uniaxial stress and applied magnetic field. The combination of two tuning knobs allows us to separate the stress-detwinning of structural domains from the field-induced reorientation of Eu moments. Intriguingly, we find a spin-flip transition which can only result from a strongly anisotropic interaction between Eu planes. We argue that this anisotropic exchange is a consequence of the strong anisotropy in the magnetically ordered Fe layer, which presents a new form of higher-order coupling between Eu and Fe magnetism.

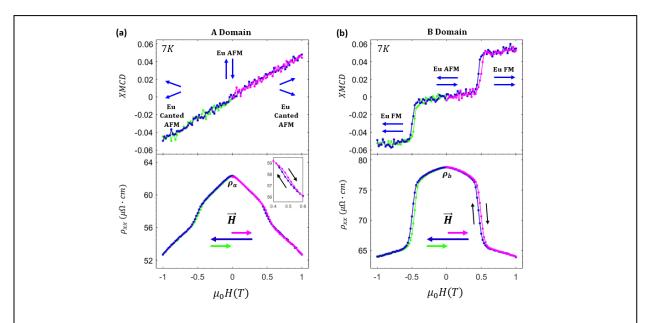


Fig. 3 Metamagnetic transition of detwinned crystals of EuFe₂As₂. (a) and (b) correspond to magnetic field perpendicular (A domain) and parallel (B domain) to the magnetic easy axes, respectively. Upper panels show the XMCD signal and lower panels show the resistivity.

Papers published and submitted

- 1. Junyi Yang, Lin Hao, Peyton Nanney, Kyle Noordhoek, Derek Meyers, Lukas Horak, Joshua Sanchez, Jiun-Haw Chu, Christie Nelson, Mark PM Dean, Jian Liu, "Epitaxial stabilization of Sr₃Ir₂O₇ thin films" *Applied Physics Letters* 114 (18), 182401 (2019)
- 2. Lin Hao, Zhentao Wang, Junyi Yang, D. Meyers, Joshua Sanchez, Gilberto Fabbris, Yongseong Choi, Jong-Woo Kim, Daniel Haskel, Philip J. Ryan, Kipton Barros, Jiun-Haw Chu, M. P. M. Dean, Cristian D. Batista, Jian Liu, "Anomalous Magnetoresistance due to Longitudinal Spin Fluctuations in a Jeff = 1/2 Mott Semiconductor", *Nature Communications* 10, 5301 (2019)
- **3.** Paul Malinowski, Qianni Jiang, Joshua Sanchez, Zhaoyu Liu, Joshua Mutch, Preston Went, Jian Liu, Philip Ryan, Jong-Woo Kim, Jiun-Haw Chu, "Suppression of superconductivity by anisotropic strain near a nematic quantum critical point", *Nature Physics* 16 (12), 1189-1193, (2020)
- **4.** Joshua J Sanchez, Paul Malinowski, Joshua Mutch, Jian Liu, J-W. Kim, Philip J Ryan, Jiun-Haw Chu, "The transport-structural correspondence across the nematic phase transition probed by elasto-x-ray diffraction", *Nature Materials* (2021). https://doi.org/10.1038/s41563-021-01082-
- 5. Joshua J Sanchez, Gilberto Fabbris, Yongseong Choi, Yue Shi, Paul Malinowski, Shashi Pandey, Jian Liu, II Mazin, Jong-Woo Kim, Philip Ryan, Jiun-Haw Chu, "Strongly anisotropic antiferromagnetic coupling in EuFe2As2 revealed by stress detwinning", **Phys. Rev. B** 104, 104413, (2021)
- 6. Qianni Jiang, Yue Shi, Morten H. Christensen, Joshua Sanchez, Bevin Huang, Zhong Lin, Zhaoyu Liu, Paul Malinowski, Xiaodong Xu, Rafael M. Fernandes, Jiun-Haw Chu, "Nematic Fluctuations in an Orbital Selective Superconductor Fe1+yTe1-xSex", arXiv:2006.15887 (submitted)

Invited talks and seminars

Jiun-Haw Chu

- 1. "Nematic quantum criticality in iron-based superconductors" International Conference on Quantum Liquid Crystals 2021 (QLC 2021) (online), May 2021
- 2. "Nematic Fluctuations in an Orbital Selective Superconductor Fe1+yTe1-xSex", APS March Meeting, Mar 2021
- 3. "Nematic quantum criticality in iron-based superconductors", LASSP Seminar, Cornell University, Oct 2020
- 4. Nematic quantum criticality in iron-based superconductors", Condensed Matter Seminar, University of Florida, Oct 2020
- 5. "Nematic quantum criticality in iron-based superconductors", Physics Colloquium, University of Washington, Sep 2020
- 6. "Nematic quantum criticality in iron-based superconductors", Condensed Matter Seminar, Rice University, Sep 2020
- 7. "Nematic quantum criticality in iron-based superconductors" Online Summer Seminars for Correlated Electrons and Frustrated Magnets, Sep 2020
- 8. "Nematic quantum criticality in iron-based superconductors" Workshop on Multi-Modal X-Ray Techniques for Emergent Quantum Materials, Aug 2020

Joshua Sanchez

- 9. "The transport-structural correspondence across the nematic phase transition probed by elasto-x-ray diffraction", APS March Meeting, Mar 2022
- 10. "Tuning electronic and magnetic orders with strain and magnetic field" Workshop on Multi-Modal X-Ray Techniques for Emergent Quantum Materials, Aug 2020

Paul Malinowski

- 11. "Suppression of superconductivity by anisotropic strain near a nematic quantum critical point", APS March Meeting, Virtual, March 2021
- 12. "Suppression of superconductivity by anisotropic strain near a nematic quantum critical point", RCQM Symposium on Fe-based Superconductivity, Virtual (Rice University), Jan. 2021

Award and patents

Jiun-Haw Chu

Presidential Early Career Award for Scientists and Engineers, 2019

Packard Fellowship for Science and Engineer, 2018

Sloan Research Fellow, 2018

Paul Malinowski

Dehmelt Prize, UW Department of Physics 2019

Joshua Sanchez

Dehmelt Prize, UW Department of Physics 2018

Students and postdocs supported

Joshua Sanchez (PhD student, Graduated Aug 2021, Now NSF MPS-Ascend Postdoctoral Fellow at MIT)

Paul Malinowski (PhD student)

Qianni Jiang (PhD student)

Yue Shi (PhD student)

Major collaborations

Collaboration on the elasto X-ray and iridates

Phillip Ryan, Jong Woo Kim (Advanced Photon Source, Argonne National Laboratory)

Jian Liu (University of Tennessee, Knoxville)

Collaboration on the nematicity in iron chalcogenides

Rafael Fernades (University of Minesota)

Xiaodong Xu (University of Washington)

AFOSR-YIP Final Report

Award Information

Project title: New Superconductors near Broken Rotational Symmetry Instabilities

Award number: FA9550-17-1-0217;

Report date: 09/20/2021

Period covered by the report: July 1st 2017 – July 1st 2021.

Principle Investigator: Jiun-Haw Chu (Department of Physics, University of Washington,

Seattle, WA 98195)

Email: jhchu@uw.edu; phone: 650-862-2808

Program Manager: Kenneth C. Goretta, <u>kenneth.goretta@us.af.mil</u>

Research Accomplishment

High temperature superconductivity emerges in the vicinity of rotational symmetry-breaking instabilities in the iron-based and copper-based materials. It has been argued that the superconducting paring might be enhanced by the quantum critical fluctuations with a nematic character. In this four-year research program, we have made significant progress towards the understanding and the application of this idea. We discovered a new mechanism to control superconductivity by suppressing the nematic fluctuations with an anisotropic strain. We also developed a new experimental platform, the elasto x-ray diffraction, to study the electron-lattice coupling of the broken rotational symmetry phase. The elasto x-ray diffraction revealed a striking transport-structural correspondence of the nematic state in the iron pnictide superconductors. It also uncovered an unusual coupling between the Eu and Fe moments in the Eu based iron pnictides.

During the four-year report period, we have total 5 publications, including one Nature Materials, one Nature Physics and one Nature Communications. Below we present a summary of research highlights.

a. Drastic suppression of superconductivity near a nematic quantum critical point (Nature Physics 2020)

Most unconventional and high temperature superconductors share a similar phase diagram. As the system is tuned by chemical doping or pressure, superconductivity emerges as a nearby symmetry breaking phase is suppressed. The empirical observation of this common phase diagram has led to the belief that the fluctuations associated with the symmetry breaking phase are beneficial, if not responsible, for the superconducting pairing. A direct test to verify this hypothesis is to observe the decrease of superconducting T_c by applying the symmetry breaking conjugate field that suppresses the dynamic fluctuations of the competing order. Nevertheless, most of the competing phases in unconventional superconductors break translational symmetry, which requires a spatially modulated conjugate field that is difficult to realize experimentally. In this work, we show that anisotropic strain, the conjugate field of nematicity, drastically suppresses the superconducting T_c of an iron pnictide superconductor. We discovered a fivefold reduction of T_c for the optimally doped samples for less than one percent of strain, whereas for the underdoped samples the suppression of T_c extends to zero temperature with the recovery of a fully metallic ground state. In addition to providing direct evidence of the role played by the nematic fluctuations in the formation of the superconducting state, these results also demonstrate tunable mechanical control of a high

temperature superconductor, an important step forward regarding the technological applications of superconductivity, as well as provide a new platform for studying a superconductor to metal quantum phase transition in a three-dimensional system.

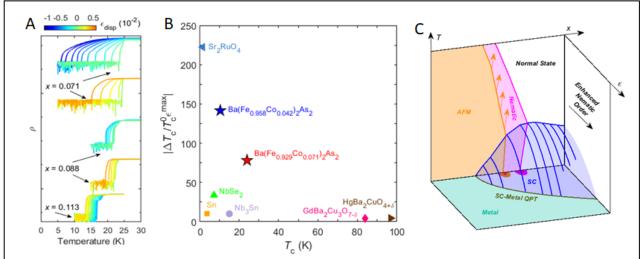


Fig. 1 Suppression of superconductivity by anisotropic strain (A) Resistivity as a function of temperature under uniaxial stress for optimally and overdoped compositions. Resistivity is on a log scale and sets of curves that overlap are offset for clarity (B) Sensitivity of T_c to strain for a variety of superconductors. (C) Schematic doping-strain-temperature $(x-\epsilon-T)$ phase diagram.

b. The transport structural correspondence across the nematic transition probed by elasto x-ray diffraction (Nature Materials 2021)

Electronic nematicity in iron pnictide materials is coupled to both the lattice and the conducting electrons, which allows both structural and transport observables to probe nematic fluctuations and the order parameter. A key question is in what regime one can establish a one-toone correspondence between transport and structural variables. This question is especially important for 2D systems such as graphene where thermodynamic measurements are difficult. It also has important implications to the nearby superconducting phase, since the transport measurements probe the conducting electrons that form copper pars. In this work, we combine simultaneous transport and x-ray diffraction measurements with in-situ tunable strain (elasto-XRD) to measure the temperature dependence of the shear modulus and elastoresistivity above the nematic transition and the spontaneous orthorhombicity and resistivity anisotropy below the nematic transition, all within a single sample of Ba(Fe_{0.96}Co_{0.04})₂As₂. With our unprecedented multi-modal measurement, we show the four quantities perfectly follows a mean-field temperature dependence. Furthermore, the ratio of transport to structural quantities is a constant across the phase transition, suggesting that the resistivity anisotropy behaves just like a thermodynamic variable even for large values of the nematic order parameter. While the shear modulus and elastoresistivity and the spontaneous orthorhombicity and resistivity anisotropy can be well described by the Landau free energy framework, two unexpected findings stand out. First, using the shear modulus and elastoresistivity data from the previous studies, we discovered a strong doping dependence of the ratio between transport and structural quantities, increasing by more than fivefold towards optimal doping. Second, when driving the system deep into the non-linear

regime with large uniaxial stress, we found that the resistivity anisotropy shows a non-saturating behavior that is drastically different from the dampened response of the lattice, which may reveal the intertwined nature of nematicity.

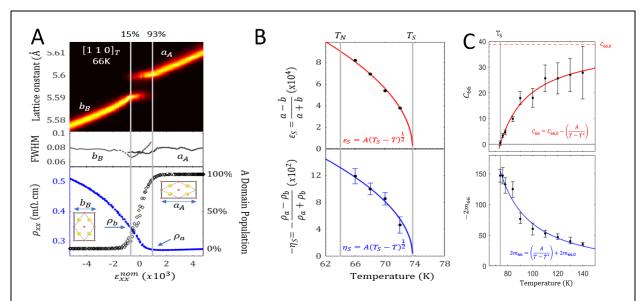


Fig. 2 Elasto X-ray diffraction measurement (A) Simultaneous x-ray diffraction (upper) and resistivity (lower, blue curve) measurement as a function of strain. These measurements allow us to extract spontaneous lattice distortion (B, upper), resistivity anisotropy (B, lower), shear modulus (C, upper) and elastoresistivity (C, lower) all in one sample.

c. Strongly anisotropic antiferromagnetic coupling in EuFe₂As₂ revealed by stress detwinning (Phys. Rev. B 2021)

Of all parent compounds of iron-based high-temperature superconductors, EuFe₂As₂ exhibits by far the largest magneto-structural coupling due to the sizable biquadratic interaction between Eu and Fe moments. While the coupling between Eu antiferromagnetic order and Fe structural/antiferromagnetic domains enables rapid field detwinning, this prevents simple magnetometry measurements from extracting the critical fields of the Eu metamagnetic transition. In this work, we measure these critical fields by combining x-ray magnetic circular dichroism spectroscopy with in-situ tunable uniaxial stress and applied magnetic field. The combination of two tuning knobs allows us to separate the stress-detwinning of structural domains from the field-induced reorientation of Eu moments. Intriguingly, we find a spin-flip transition which can only result from a strongly anisotropic interaction between Eu planes. We argue that this anisotropic exchange is a consequence of the strong anisotropy in the magnetically ordered Fe layer, which presents a new form of higher-order coupling between Eu and Fe magnetism.

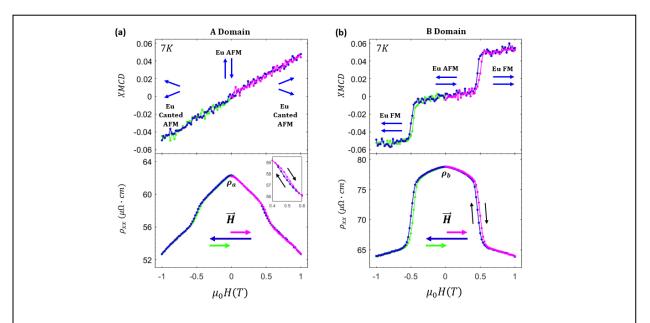


Fig. 3 Metamagnetic transition of detwinned crystals of EuFe₂As₂. (a) and (b) correspond to magnetic field perpendicular (A domain) and parallel (B domain) to the magnetic easy axes, respectively. Upper panels show the XMCD signal and lower panels show the resistivity.

Product During the Report Period

Junyi Yang, Lin Hao, Peyton Nanney, Kyle Noordhoek, Derek Meyers, Lukas Horak, Joshua Sanchez, Jiun-Haw Chu, Christie Nelson, Mark PM Dean, Jian Liu, "Epitaxial stabilization of Sr₃Ir₂O₇ thin films" *Applied Physics Letters* 114 (18), 182401 (2019)

Lin Hao, Zhentao Wang, Junyi Yang, D. Meyers, Joshua Sanchez, Gilberto Fabbris, Yongseong Choi, Jong-Woo Kim, Daniel Haskel, Philip J. Ryan, Kipton Barros, Jiun-Haw Chu, M. P. M. Dean, Cristian D. Batista, Jian Liu, "Anomalous Magnetoresistance due to Longitudinal Spin Fluctuations in a Jeff = 1/2 Mott Semiconductor", *Nature Communications* 10, 5301 (2019)

Paul Malinowski, Qianni Jiang, Joshua Sanchez, Zhaoyu Liu, Joshua Mutch, Preston Went, Jian Liu, Philip Ryan, Jong-Woo Kim, Jiun-Haw Chu, "Suppression of superconductivity by anisotropic strain near a nematic quantum critical point", *Nature Physics* 16 (12), 1189-1193, (2020)

Joshua J Sanchez, Paul Malinowski, Joshua Mutch, Jian Liu, J-W. Kim, Philip J Ryan, Jiun-Haw Chu, "The transport-structural correspondence across the nematic phase transition probed by elasto-x-ray diffraction", *Nature Materials* (2021). https://doi.org/10.1038/s41563-021-01082-

Joshua J Sanchez, Gilberto Fabbris, Yongseong Choi, Yue Shi, Paul Malinowski, Shashi Pandey, Jian Liu, II Mazin, Jong-Woo Kim, Philip Ryan, Jiun-Haw Chu, "Strongly anisotropic antiferromagnetic coupling in EuFe2As2 revealed by stress detwinning", **Phys. Rev. B** 104, 104413, (2021)