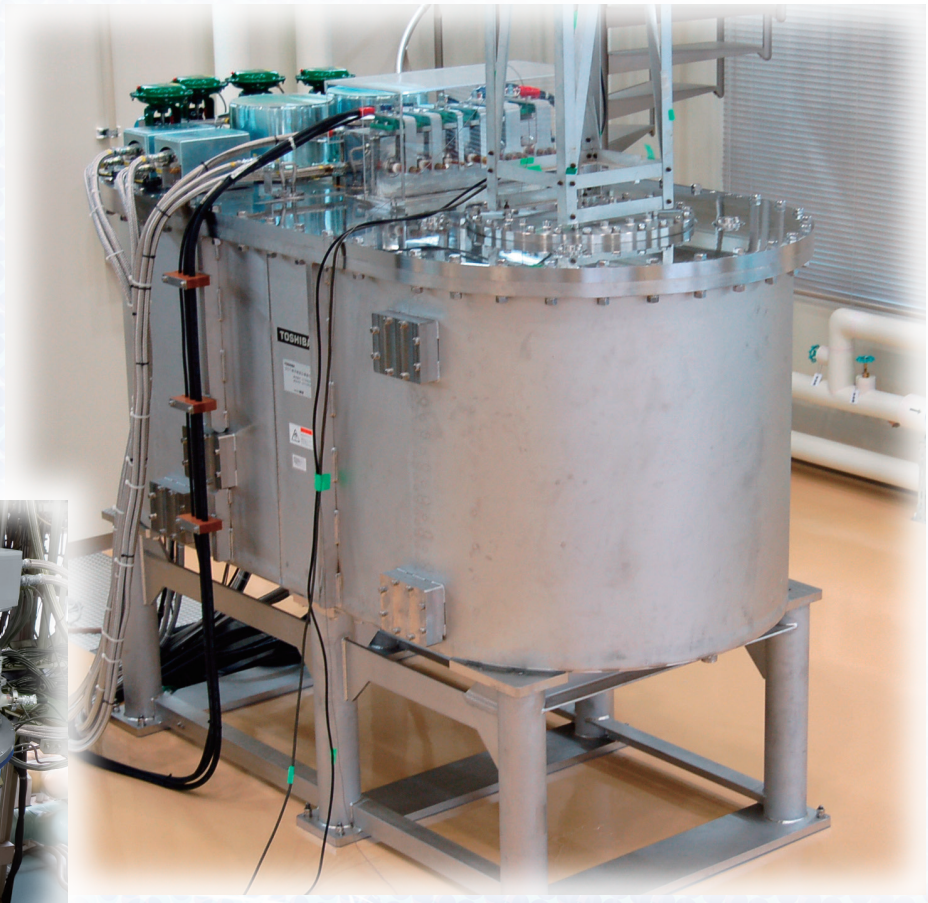
Selected Topics in 2019
Research Highlight at HFLSM

Research



28T-CHM

25T-CSM

HFLSM

High Field Laboratory for Superconducting Materials,
Institute for Materials Reserch, Tohoku University

< FRONT COVER EXPOSITION>

Cryogen-Free 28 T Hybrid Magnet and

25 T Cryogen-Free Superconducting Magnet

The world's first 28 T cryogen-free hybrid magnet developed by the HFLSM. The inner double Bitter type water-cooled resistive magnet generates 19.0 T in a room temperature experimental bore of 32 mm with the electric power of 7.2 MW. The outer cryogen-free superconducting magnet generates 9.0 T in a 360 mm room temperature bore. The cryogen-free superconducting magnet generates 24.6 T in 52 mm room temperature bore by the combination of metallic and high-T_c superconductors.

◇◆◇ Preface ◇◆◇

This booklet reports the highlights of researches and the new improvements during FY2019 performed in the HFLSM: High Field Laboratory for Superconducting Materials at Sendai. The HFLSM has been developed numbers of new magnets technologies including the cryogen-free hybrid magnet and cryogen-free superconducting magnets generating magnetic fields above 20 T. At the HFLSM, the unique 25 T cryogen-free superconducting magnet has been operational for user program and has attracted many domestic and overseas users. Such cryogen-free superconducting magnets surely offer long-term stable and high-quality steady fields. The HFLSM offers varieties of hybrid and superconducting magnets for researches in materials science, physics, applied superconductivity, chemistry and other pure and inter-disciplinary sciences performed in steady magnetic fields. It should be noted that the HFLSM is now preparing for 30 T class superconducting magnet based on our original technologies. This project is one of the main objectives in the Japan High Magnetic Field Collaboratory started operation in April 2020. It has been also accepted in the load map 2020 plan for large scale facilities form MEXT. Under difficulties in Covid-19 pandemic, the HFLSM is in operational with strict anti-infection majors by the strong supports and collaborations of users.

Form 2018, IMR has recognized as one of the six international collaboration centers and has launched the new international user program named Global Institute for Materials Science Tohoku (GIMRT). In this program, a collaboration among multiple institutions including IMR can be conducted in the “Bridge type” scheme. These new programs will enhance the diverse and strong collaboration in a global framework. Besides international collaborations with remote technologies under the pandemic, we hope that we can welcome users from abroad in 2021 for onsite experiments.

We hope that the booklet helps you to see the overview of our activities and stimulate future research collaborations with domestic and oversea users in the HFLSM and in the High Magnetic Field Co-laboratory of Japan.

1 November 2020

Hiroyuki Nojiri

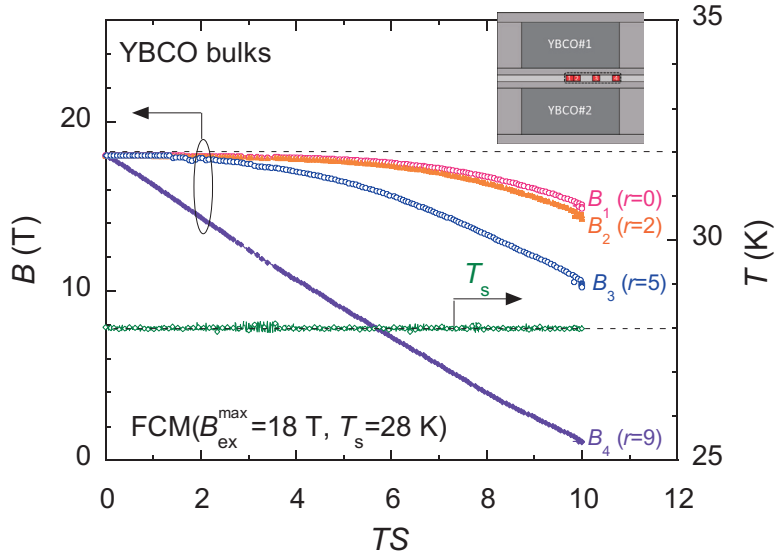
Director of HFLSM

Selected Topics in 2019 – Research Highlight at HFLSM

Selected Topics in 2019 – Research Highlight at HFLSM

Magnetization of YBCO Bulks under High Magnetic Fields of up to 22 T

YBCO バルクの最大 22 T の強磁場下における着磁現象



The stacking of YBCO superconducting bulk pair reinforced by the stainless steel (SUS) container was magnetized by the field cooled magnetization (FCM) under magnetic fields of up to 22 T using the 28 T hybrid magnet (28T-HM). The structure of the SUS container was optimized against the electromagnetic hoop stress of about several hundred MPa by the numerical simulations. We obtained the trapped field of 15.1 T by FCM from 18 T at 28 K at the center of the YBCO pair, however, the cracking occurred partially at the periphery of both YBCO bulks during FCM from 22 T at 23 K. The bulk fracture should originate from the thermal shock by the flux jumps.

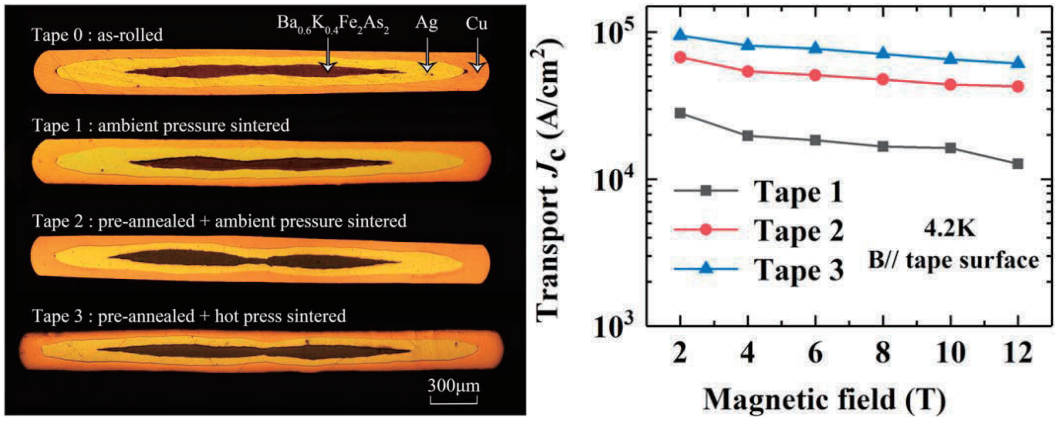
T. Naito[1], H. Fujishiro[1], S. Awaji[2]
[1] Fac. of Sci. Eng., Iwate Univ., [2] IMR, Tohoku Univ.

Reference: T. Naito *et al.*, “Field-cooled magnetization of Y-Ba-Cu-O superconducting bulk pair reinforced by full metal encapsulation under high magnetic fields up to 22 T”, J. Appl. Phys. **126** (2019) 243901.

SUS 製容器で補強された YBCO 超伝導バルク対を最大 22 T の磁場下において磁場中冷却法で着磁した。SUS 容器は数百 MPa の電磁的フープ応力に耐える強度を YBCO バルク対に与えるよう有限要素法によるシミュレーション解析から最適設計した。その結果，YBCO バルク対に 28 K において 15.1 T の磁場を捕捉させることに成功した。しかし，23 K の着磁実験ではフラックスジャンプが発生し 2 個のバルクともに端部に割れが生じた。フラックスジャンプ発生時の急激な温度上昇による熱衝撃が破壊の起源の可能性がある。

内藤 智之[1], 藤代 博之[1], 淡路 智[2]
[1] 岩手大理工, [2] 東北大金研

Enhancement of the Critical Current Density in Cu/Ag Composite Sheathed (Ba, K)Fe_2As_2 Tapes by Pre-Annealing Process



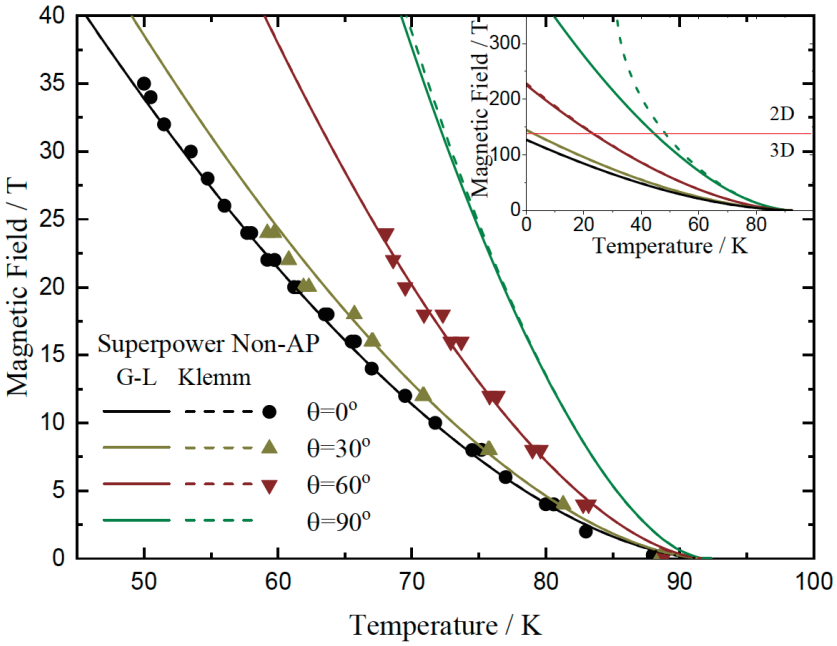
Iron-based superconductors, a promising candidate for high-field applications, have aroused rising attentions. For 122-type IBS tapes fabricated using pure silver sheath, transport J_c can achieve 1.5×10^5 A cm^{-2} (4.2 K, 10 T). To reduce the usage of pure silver, Cu/Ag composite sheath is attempted in fabricating 122 tapes. However, the melting point of Cu/Ag composite sheath is much lower than that of pure silver. The tapes will be melted and destroyed once the sintering temperature is higher than the melting point of the sheath. The commonly used temperature during the final sintering process for pure silver sheathed Ba-122 tapes is higher than 850°C. In contrast, the sintering temperature for Cu/Ag composite sheathed Ba-122 superconducting tapes is lower than 740°C. Compared with the silver sheathed tapes, the Cu/Ag composite sheathed IBS tapes exhibit low J_c as being sintered at lower temperatures. In general, voids and residual cracks formed during cold deformation will be remained when the tapes are sintered at low sintering temperature. Accordingly, the grains are not well connected in the Cu/Ag composite sheathed IBS tapes.

In this study, a pre-annealing method was used to promote the transport critical current density (J_c) in Cu/Ag composite sheathed (Ba,K)Fe_2As_2 tapes. It was found that tapes sintered at 740°C for 3h under ambient pressure showed a J_c up to 4.9×10^4 A cm^{-2} (4.2 K, 10 T). Also, a transport J_c of 6.5×10^4 A cm^{-2} (4.2 K, 10 T) was achieved using a hot-press technique. The factors affecting the J_c of tapes were systematically analyzed. The well-constructed crystallization and grain connectivity obtained in pre-annealed tapes were considered as the major causes of the high J_c. These results highlight the great potential to increase the J_c in multiple metal composite sheathed Ba-122 tapes.

L. Li[1], X. Zhang[1], C. Yao[1], S. Liu[1], H. Huang[1], C. Dong[1], Z. Cheng[1], S. Awaji[2], Y. Ma[1]
[1] IEE, CAS, PRC, [2] IMR, Tohoku Univ.

Reference: L. Li *et al.*, "Large critical current density in Cu/Ag composite sheathed (Ba, K)Fe_2As_2 tapes fabricated under ambient pressure", Supercond. Sci. Technol. **32** (2019) 065008.

3-D Properties in (RE)BCO Tapes Measured in Fields up to 35 T



We have measured the temperature and angular dependence of the upper critical field (B_{C2}) for three HTS coated conductors using AC susceptibility measurements. We have obtained fits to our B_{C2} (T, θ) data using both the anisotropic Ginzburg–Landau (G–L) and Klemm's models for layered superconductors. Our calculations suggest that these tapes are three-dimensional (3-D) at all temperatures in zero field but become 2-D in magnetic fields above a crossover field of 457/ γ T, where $\gamma = \sqrt{(m_c/m_{ab})}$ is the G - L anisotropy parameter. We expect the 3-D–2-D crossover fields in these tapes are at least 130 T.

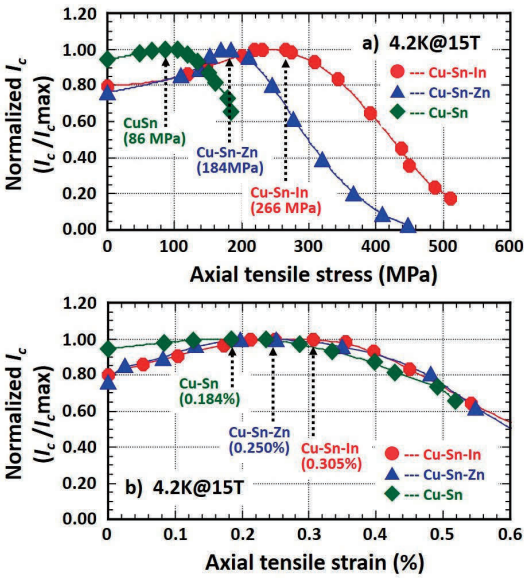
In the standard description of HTS materials, superelectrons condense in parallel 2-D Cu-O planes that are coupled via interlayer Josephson coupling. This layered structure leads to a dimensional crossover from 3-D properties close to the critical temperature (T_c) where the coherence length (ξ) is large, to 2-D properties at low temperatures where ξ is small. From a technological point of view, we have found 3-D behavior in these tapes at all measured fields, temperatures and angles up to 35 T. We conclude that concerns about using (RE)BCO tapes in high field applications because of a dimensional crossover can be set aside.

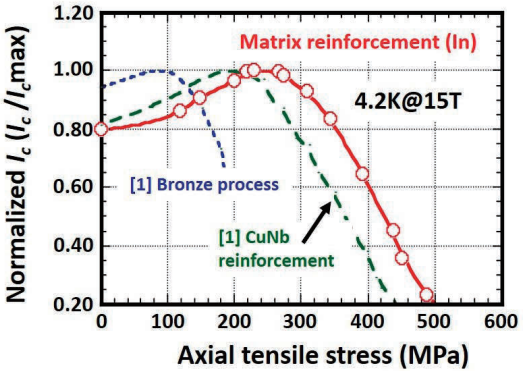
A.P. Smith[1], T. Okada[2], S. Awaji[2], E. Surrey[3] and D.P. Hampshire[1]
[1] Dep. Phys., Durham Univ., [2] IMR, Tohoku Univ., [3] Culham Centre for Fusion Energy, Oxford.

References: A.P. Smith *et al.*, "3D Properties in (RE) BCO Tapes Measured in Fields up to 35 T.", IEEE Trans. Appl. Supercond., 29 (2019) 6601005.
R. A. Klemm *et al*, "Theory of the upper critical field in layered superconductors," Physical Review B, vol. 12, no. 2, pp. 877-891, 1975.

Mechanical Strength Evaluation of the Internal Matrix Reinforced Nb_3Sn Multifilamentary Wires Using Cu-Sn-In Ternary Alloy Matrix

Cu-Sn-In 系三元系合金母材を用いた内部マトリクッス補強 Nb_3Sn 極細多芯線材の機械強度評価





[1] K. Watanabe et al., AIP Conf. Proc. 1574 (2014) pp.186-191.

Critical current (I_c) under the tensile deformation in various Nb_3Sn wires using Cu-Sn-Zn and Cu-Sn-In ternary matrices were evaluated. The tensile stress and strain obtained to the maximum peak I_c value on the Cu-Sn-In ternary alloy matrix sample was estimated to approximately 265 MPa and 0.305%, and these values were much higher than those of the conventional bronze processed and CuNb reinforcement Nb_3Sn wire samples. The In element remained homogeneously in the wire matrix after the Nb_3Sn phase synthesis. Mechanical strength improvements were caused by the phase transformation of the wire matrix into a mechanical harder (Cu, In) solid solution.

Y. Hishinuma[1,5], H. Oguro[2], H. Taniguchi[3], S. Awaji[4], A. Kikuchi[5]

[1] National Institute for Fusion Science, [2] Dept. of Mater. Sci., Tokai Univ., [3] Osaka Alloying Works Co., Ltd, [4] IMR, Tohoku Univ., [5] National Institute for Materials Science

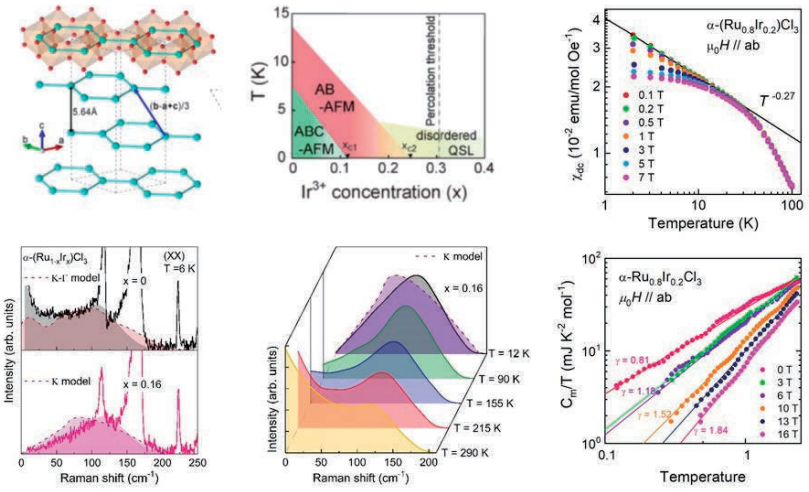
Reference: Y. Hishinuma *et al.*, "Mechanical strength evaluation of the internal matrix reinforced Nb_3Sn multifilamentary wires using Cu-Sn-In ternary alloy matrix", IEEE Trans. Appl. Supercond., **30** (2020) 6001104.

三元系合金母材を用いた Nb_3Sn 極細多芯線材の引張変形に伴う臨界電流(I_c)を評価した結果、最大の I_c 値を示すピーク引張応力及び引張ひずみの大幅な改善(高応力・高ひずみ側にシフト)が Cu-Sn-In 合金線材に確認され、更に CuNb 補強線材と比較しても高い機械強度を示した。機械強度の改善は、Nb_3Sn 相の生成後の母材が残存した In 元素と反応して機械的に硬い(Cu, In)固溶体に相変態したためである。

菱沼　良光[1,5], 小黒　英俊[2], 谷口　博康[3], 淡路　智[4], 菊池　章弘[5]

[1] 核融合研, [2] 東海大工, [3] 大阪合金, [4] 東北大金研, [5] 物質材料研究機構

Randomly Hopping Majorana Fermions in the Diluted Kitaev System



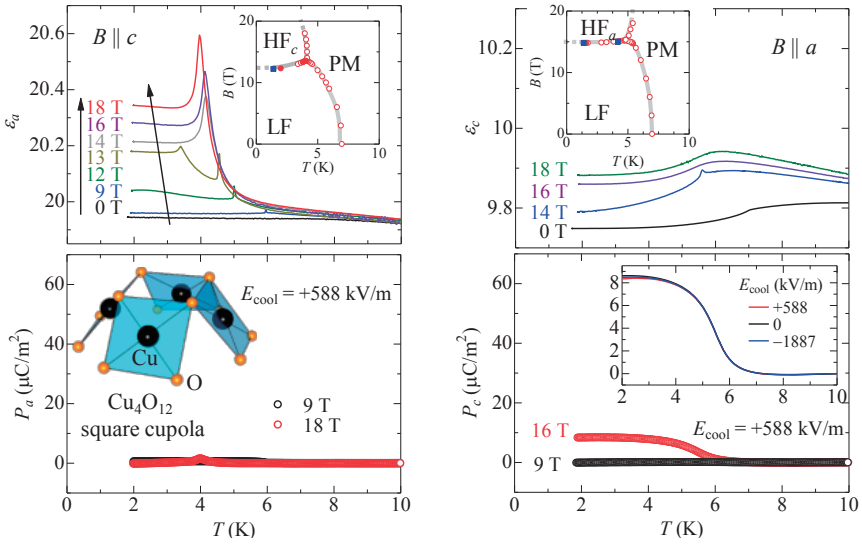
Combined magnetic susceptibility, specific heat, and Raman scattering experiments show evidence consistent with a disordered Kitaev spin liquid in α-$Ru_{1-x}Ir_xCl_3$ ($x \approx 0.2$). At ambient pressure and zero field, the Kitaev candidate material α-$RuCl_3$ in its pristine form harbors a zigzag antiferromagnetic order below 6.5 K. On introducing nonmagnetic impurities ($x \geq 0.2$), at intermediate energies above 3 meV, Raman spectroscopy gives signatures of Majorana-like excitations: asymptotic ω-linear spectral form and the Fermi statistics. At low energies below 3 meV, the magnetic specific heat shows a power-law behavior $C_m \sim T^{2/(1+\Delta)}$ with the disorder strength $\Delta \approx 0.1$. In addition, the magnetic susceptibility and magnetization obey a scaling relation of $\chi[H,T]$ and $M[H,T]$ in H/T with the scaling exponent $\alpha = 0.27$. This scaling phenomenology implies the presence of a weakly divergent low-energy density of states, characteristic for a bond-disordered Kitaev model. Our results demonstrate that the random hoppings of Majorana fermions feature low-lying excitations in a Kitaev honeycomb system subject to spin vacancies and bond randomness.

S. Do[1], C. H. Lee[1], Y. S. Choi[1], K.-Y. Choi[1], T. Kihara[2], H. Nojiri[2]
[1] Dept. of Physics, Chung-Ang Univ., [2] IMR, Tohoku Univ.,

Reference: Seung-Hwan Do *et al.*, "Randomly Hopping Majorana Fermions in the Diluted Kitaev System α-$Ru_{0.8}Ir_{0.2}Cl_3$", Phys. Rev. Lett. **124** (2020) 047204.

Magnetoelectric Behaviors in Magnetic-Field-Induced Phases of Square-Cupola-Based Antiferromagnet $Pb(TiO)Cu_4(PO_4)_4$

正四角台塔反強磁性体 $Pb(TiO)Cu_4(PO_4)_4$ の磁場誘起相での電気磁気応答



Recently, $Pb(TiO)Cu_4(PO_4)_4$ has gained interest because of its unique magnetoelectric (ME) responses due to a peculiar geometry of magnetic Cu_4O_{12} square-cupola clusters in this material. Here we study ME properties in high-magnetic-field (B) phases of $Pb(TiO)Cu_4(PO_4)_4$ for B applied along a (B_a) and c axes (B_c). For B_c, a dielectric constant shows a peak at a paraelectric to high-field phase transition, indicative of an antiferroelectric state. For B_a, by contrast, an electric polarization is observed in a high-field phase, whose sign is, however, independent of the electric field cooling procedure. These behaviors are distinct from a previously-reported ferroelectricity in a high field phase for B ∥ [110]. Thus, magnetic square-cupolas are promising units leading to rich ME phenomena.

K. Kimura[1], S. Kimura[2], T. Kimura[1]

[1] Dept. of Adv. Mater. Univ. of Tokyo, [2] IMR, Tohoku Univ.

Reference: K. Kimura *et al.*, "Magnetoelectric Behaviors in Magnetic-Field-Induced Phases of $Pb(TiO)Cu_4(PO_4)_4$", J. Phys. Soc. Jpn. **88** (2019) 093707.

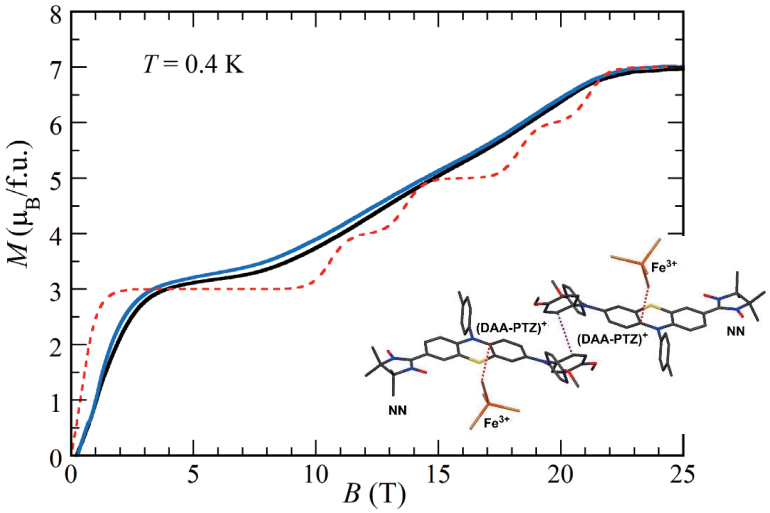
正方晶反強磁性体 $Pb(TiO)Cu_4(PO_4)_4$ は、その磁性を支配している正四角台塔クラスターCu_4O_{12} の幾何学的形状に起因した特異な電気磁気応答に興味がもたれる。本研究では、a 軸(B_a)および c 軸方向の磁場(B_c)中で現れる高磁場相の電気磁気応答を調べた。B_a 誘起相では、電気分極が現れない一方で誘電率が相転移に起因してピークを示すことから、反強誘電状態の形成が示唆される。一方、B_c 誘起相では、有意な分極は観測されたが、その符号は冷却電場の符号に依存しないことが分かった。これらの振る舞いは、[110]磁場誘起相で過去に報告された強誘電性とは全く異なる。本研究により、正四角台塔クラスターが磁場中で多彩な電気磁気応答を引き起こすことが実証された。

木村　健太[1], 木村　尚次郎[2], 木村　剛[1]

[1] 東大新領域, [2] 東北大金研

Development of a New Biradical Cation Forming $S = 1$ and Magnetic Properties of its Salts

有機ビラジカルカチオンが形成する $S = 1$ 種の開発とその塩の磁気的性質



We have designed and synthesized a novel $S = 1/2$ organic radical DAA-PTZ-NN and its one-electron oxidized species (DAA-PTZ)$^+$-NN, which is a biradical system since the (DAA-PTZ)$^+$ moiety carries an $S = 1/2$. This biradical cation is so stable that single crystals of its salts (DAA-PTZ)$^+$-NN •MBr_4^- (M = Ga, Fe) were successfully obtained. The magnetic measurements of Ga salt have revealed that (DAA-PTZ)$^+$-NN is a good $S = 1$ species with strong ferromagnetic intramolecular interaction $2J/k_B$ = +640 K. For the crystals of Fe salt, the stationary behavior of magnetization at 3μ_B/f.u. reveals the formation of the 6-spin cluster. The monotonous increase of magnetization above 3 T means the existence of magnetic interaction between the 6-spin cluster.

T. Tahara[1], S. Suzuki[1,2], M. Kozaki[1], D. Shiomi[1], K. Sugisaki[1], K. Sato[1], T. Takui[1], Y. Miyake[3], Y. Hosokoshi[3], H. Nojiri[4], and K. Okada[1]

[1] Grad. Sch. of Sci., Osaka City Univ., [2] Grad. Sch. of Eng. Sci., Osaka Univ., [3] Grad. Sch. of Sci., Osaka Pref. Univ., [4] IMR, Tohoku Univ.

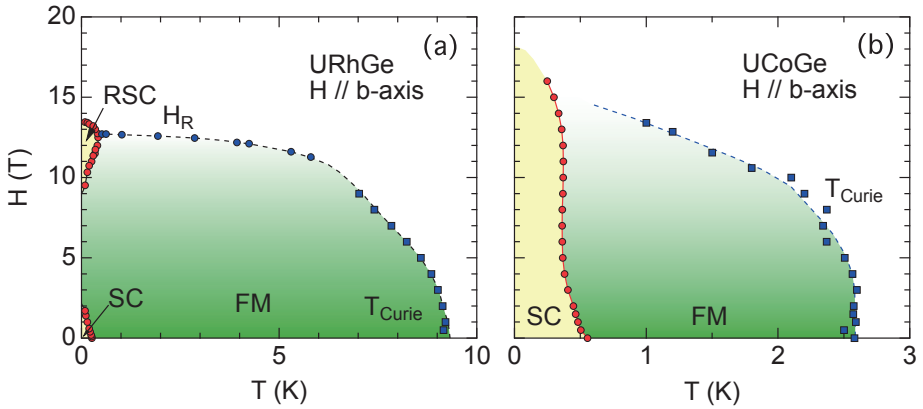
Reference: T. Tahara *et al.*, "Triplet Diradical Cation Salts Consisting of Phenothiazine Radical Cation and Nitronyl Nitroxide", Chem. Eur. J., **25** (2019) 7201-7209.

新しい有機カチオンビラジカルの開発を行った。モノラジカル DAA-PTZ-NN は、NN 部位に $S = 1/2$ が存在するが、これを一電子酸化すると(DAA-PTZ)$^+$部位がラジカカチオンとなり、(DAA-PTZ)$^+$-NN はビラジカルカチオン分子となる。この分子は安定であり、(DAA-PTZ)$^+$-NN •MBr_4^- (M = Ga, Fe)の合成と結晶化に成功した。Ga 塩 の磁気測定から(DAA-PTZ)$^+$部位と NN 部位のそれぞれ $S = 1/2$ 間には強い強磁性相互作用 $2J/k_B$ = +640 K が働き、(DAA-PTZ)$^+$-NN が良い $S = 1$ 種であることを見出した。Fe 塩について、強磁場磁化曲線が 3μ_B/f.u. で停留的な挙動を示すことから 6 スピンクラスターの形成と、3T 以上の磁化の単調増加から、6 スピン間に磁気相互作用が働くことを明らかにした。

田原　拓真[1], 鈴木　修一[1,2], 小嵜　正敏[1], 塩見　大輔[1], 杉崎　研司[1], 佐藤　和信[1], 工位　武治[1], 三宅　陽太[3], 細越　裕子[3], 野尻　浩之[4], 岡田　惠次[1]

[1] 大阪市大理, [2] 阪大基礎工, [3] 大阪府大理, [4] 東北大金研

Field-Reinforced Superconductivity in Ferromagnetic Superconductors
強磁性超伝導体の磁場強化型超伝導



The paper reviews the ferromagnetic superconductivity in UGe_2, URhGe and UCoGe. The microscopic coexistence of ferromagnetism and superconductivity is established in these systems, in which the spin-triplet state with the equal spin pairing is realized. One of the highlights is the field-reentrant (-reinforced) superconductivity when the field is applied along the hard-magnetization axis. The collapse of Curie temperature at high fields induces the strong ferromagnetic fluctuations, leading to the boost of superconductivity. The recent discovery of a new spin-triplet superconductivity in UTe_2 attracts much attention, as a family of ferromagnetic superconductivity.

D. Aoki[1], K. Ishida[2], J. Flouquet[3]
[1] IMR, Tohoku Univ., [2] Grad. Sch. of Sci., Kyoto Univ., [3] UGA/CEA-Grenoble

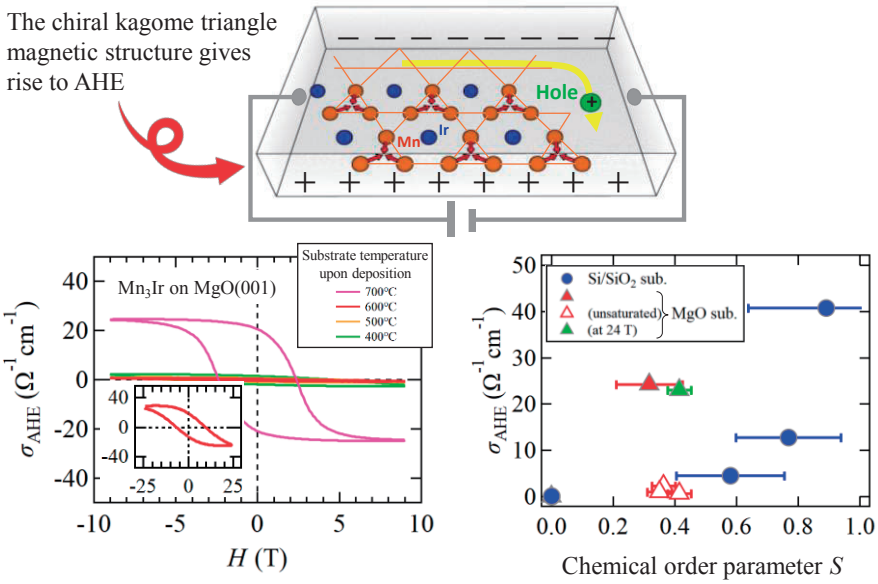
Reference: D. Aoki, K. Ishida, J. Flouquet, “Review of U-based ferromagnetic superconductors: comparison between UGe_2, URhGe, and UCoGe”, J. Phys. Soc. Jpn. **88** (2019) 022001.

強磁性超伝導体 UGe_2、URhGe、UCoG のレビュー論文である。これらの系では強磁性と超伝導が微視的に共存しており、平行スピン対によるスピン三重項超伝導が実現している。この超伝導のハイライトの一つは、強磁場を磁化困難軸方向に加えたときの磁場再突入型（磁場強化型）超伝導である。強磁性キュリー温度が磁場とともにゼロに向かうところで、強い強磁性ゆらぎが発達し、その結果極めて磁場に強い超伝導が実現する。最近見つかったスピン三重項超伝導体 UTe_2 は強磁性超伝導体の仲間であり、現在大きな注目を集めている。

青木　大[1]，石田　憲二[2]，J. Flouquet[3]
[1] 東北大金研，[2] 京大院理，[3]UGA/CEA-Grenoble

Large Anomalous Hall Effect in Antiferromagnetic $L1_2$-Mn_3Ir Thin Films

反強磁性体 $L1_2$-Mn_3Ir 薄膜における巨大異常ホール効果



In this work, we explore $L1_2$-ordered Mn_3Ir thin films which are one of the non-collinear antiferromagnets predicted to exhibit the intrinsic anomalous Hall effect (AHE) due to its topologically non-trivial spin structure. The anomalous Hall conductivity as large as $\sigma_{AHE} = 40\ \Omega^{-1}cm^{-1}$ was observed at R.T. This value can be translated to the anomalous Hall conductivity per net magnetization M as $|\sigma_{AHE}/M| = 0.6\ V^{-1}$ which is much larger comparing to those for general ferromagnetic materials. We also show that σ_{AHE} depends on the crystallinity of the Mn_3Ir as well as the chemical order parameter S characterizing a content of the $L1_2$ phase. Our results experimentally verify that $L1_2$-ordered Mn_3Ir thin films exhibit the topologically-originated AHE.

H. Iwaki[1], M. Kimata[2], T. Ikebuchi[1], Y. Kobayashi[1], K. Oda[1], Y. Shiota[1], T. Ono[1], and T. Moriyama[1]

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Reference: H. Iwaki *et al.*, "Large anomalous Hall effect in $L1_2$-ordered antiferromagnetic Mn_3Ir thin films", Appl. Phys. Lett. **116** (2020) 022408. (Selected as Editor's pick)

Mn_3Ir はトポロジカルな磁気構造により、磁化がほぼゼロにも拘わらず、大きな異常ホール効果が予測されていたが、これまで実験的に実証した例はなかった。本研究では、Mn_3Ir 薄膜を作製し、その合金規則度（磁気構造の規則度に対応）が高くなるにつれて異常ホール伝導度（σ_{AHE}）が大きくなることを見出した。得られた σ_{AHE} は Mn_3Ir の磁化の大きさでは説明できない大きな値であり、トポロジカル磁気構造由来の AHE が発現することを明らかにした。

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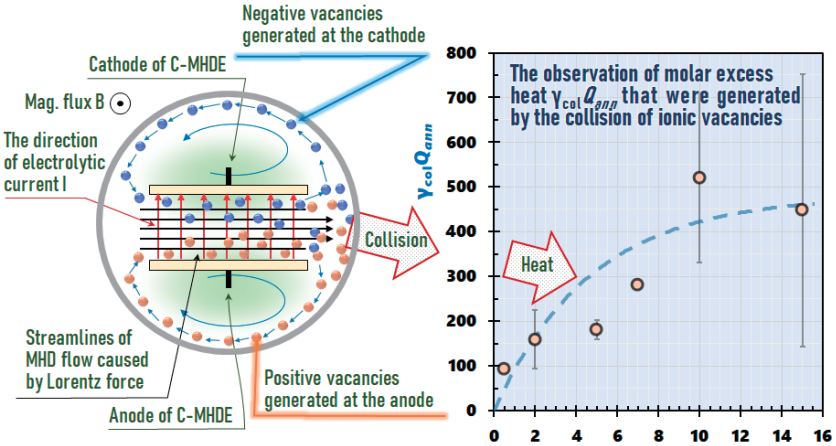
Excess Heat Production in Electrode Reaction under High Magnetic Field

高磁場下の電極反応における過剰熱生成



Schematic view of the circular-type MHD electrode (C-MHDE) in an electrolyte solution set in a magnet bore.

In electrode reaction under high magnetic field, a pair of ionic vacancies with opposite charges have been collided, and at the moment of annihilation, the stored solvation energies were liberated as excess heat. In copper redox reaction, the measured heat was 410 kJ mol^{-1} at 15 T, 1.5 times larger than the hydrogen molar combustion heat. In ferricyanide-ferrocyanide redox reaction, it was 25 kJ mol^{-1} at 15 T. Such different results come from the difference of the charge numbers of vacancies, i.e., vacancies of two and one unit-charges were created, respectively.

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電気化学の酸化・還元反応により生成するプラスとマイナスの電荷をもった二種類のイオン空孔を高磁場中で対消滅させることにより、空孔エネルギーを過剰熱として取り出すことに成功した。銅の酸化・還元反応で測定された過剰熱は 410 kJ mol^{-1} であり、フェリシアン・フェロシアンイオンの酸化・還元反応では 25 kJ mol^{-1} であった。この差は電荷の異なる二価と一価の空孔が生じることによる。

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