Large positive magnetoresistance in photocarrier-doped potassium tantalites

Rui-Shu Yang(杨睿姝), Ding-Bang Wang(王定邦), Yang Zhao(赵阳), Shuan-Hu Wang(王拴虎), and Ke-Xin Jin(金克新)[†]

Shaanxi Key Laboratory of Condensed Matter Structures and Properties and MOE Key Laboratory of Materials Physics and Chemistry under Extraordinary Conditions, School of Physical Science and Technology, Northwestern Polytechnical University, Xi'an 710072, *China*

(Received 20 February 2022; revised manuscript received 8 April 2022; accepted manuscript online 14 April 2022)

We report on the high-field magnetotransport of $KTaO_3$ single crystals, which are a promising candidate for study in the extreme quantum limit. By photocarrier doping with 360 nm light, we observe a significant positive, non-saturating, and linear magnetoresistance at low temperatures accompanied by a decreasing Hall coefficient. When cooling down to 10 K, the magnetoresistance value of $KTaO_3$ (100) reaches $\sim 433\%$ at a magnetic field of 12 T. Such behavior can be attributed to all the electrons occupying only the lowest Landau level in the extreme quantum limit. Light inhomogeneity may also contribute to large linear magnetoresistance. These results provide insights into novel magnetic devices based on complex materials and add a new family of materials with positive magnetoresistance.

Keywords: photocarriers, large positive magnetoresistance, extreme quantum limit

PACS: 73.43.Qt, 73.50.Fq, 73.40.Gk **DOI:** 10.1088/1674-1056/ac6742

1. Introduction

The magnetoresistance (MR) effect has been extensively studied since its discovery in 1856. This effect can be used in sensors, memory devices, and the emergent physics of complex interactions. Many different MRs, such as ordinary MR,[1,2] anisotropic MR,[3,4] tunnel MR,[5] giant MR,[6] and colossal MR[7,8] have been investigated in magnetic and multilayer materials. Generally, the positive and conventional MR has a magnitude of only a few percent. It is even negligible in some nonmagnetic materials due to the curving of the carrier trajectory by the Lorentz force. Nevertheless, a large and linear MR effect breaks this familiar rule. It has been observed in several non-magnetic semiconductors, such as Si,[9] Bi,[10,11] $PtBi_2$,[12] and graphene,[13] raising many lively discussions in the fields of condensed physics and functional materials. Moreover, the kind of MR in nonmagnetic materials can avoid Barkhausen noise in magnetic recording and sensor devices.

Complex oxides have extraordinary and multifunctional properties, including colossal MR in manganites, high-temperature superconductivity in cuprates, and multiferroics.[14–19] Among these materials, $KTaO_3$ (KTO) exhibits a cubic structure with a lattice constant of 0.3989 nm and a high dielectric constant (~ 4500). These characteristics make it a promising material for the extreme quantum limit (EQL) under a high magnetic field and at low temperatures.[20] In particular, such a material could integrate its functions in the emerging electronics for all-oxide devices.[21,22] Due to ionic liquid gating or Ar^+ bombardment, the surface of KTO possesses a spectrum of emergent phenomena, such as electrostatic superconductivity, the topological Hall effect (THE), and de-Haas oscillation.[23–26] Recently, KTO has been used in the engineering design of two-dimensional electronic systems. Many heterointerfaces based on KTO have been studied, involving EuO/KTO,[27,28] $LaTiO_3$/KTO,[29] $LaVO_3$/KTO,[30] amorphous-$LaAlO_3$/KTO,[31] and $LaAlO_3$/KTO.[32,33] They show intriguing properties, such as two-dimensional superconductivity, high-mobility spin-polarized electron gas, the THE, and the anomalous Hall effect. However, the MR in these systems is usually small, with a value less than 10%.[34–37] And a reliable large MR effect in complex oxides has not been found so far.

In this work, we find, for the first time, a significant positive MR effect in KTO single crystals with photocarrier doping under high magnetic fields (about 12 T). Under 360 nm illumination with different light intensities at different temperatures, KTOs come into the EQL state, where the electrons are confined in the lowest Landau levels. In the temperature range of 2 K–50 K, KTOs exhibit metallic behavior and their Hall coefficient vanishes as the magnetic field increases.[38]

2. Experiment details

KTO (100) and (111) single crystals (3 mm × 3 mm) with different thicknesses are commercially available. The electrical connections were realized at their surface using the ultrasonically wire-bonded aluminum wires. Both the resistivity and the Hall effect were measured with a van de Pauw geometry under 360 nm illumination with different light intensities. We used a physical property measurement system (CFMS-14T) that was equipped with an optical fiber. This apparatus is shown in the inset of Fig. 1(a). The bandgap (E_g) of KTO

[†]Corresponding author. E-mail: jinkx@nwpu.edu.cn

© 2022 Chinese Physical Society and IOP Publishing Ltd

is 3.4 eV, and the wavelength of light is 360 nm (3.44 eV). Thus, the electrons can be promoted from the valence band to the conduction band under 360 nm light. Although the light illumination generates non-equilibrium carriers, a steady-state quasi-equilibrium can be achieved because the carrier lifetime is close to 0.15 ms.[39] This lifetime is much larger than thermalization. The holes are straightforward to trap, and thus the transport is dominated by electrons.[20,39,40] Furthermore, Hall measurements confirm that electrons mainly determine the conduction. More importantly, the interaction between light and matter depends on the optical properties of matter and the wavelength of light. The optical penetration depth is deduced from the absorption coefficient (α) of KTO, as shown in the supplementary material in Fig. S1(a), which is taken from Ref. [39], from which the depth profile of the electronic distribution can be estimated. From the absorption coefficient, we can obtain that the thickness of the conductive layer is larger than 1 mm at $\lambda = 360$ nm. At this wavelength, the carrier density distribution, which is perpendicular to the surface of the samples, is uniform. Here, we define MR $= [\rho_{xx}(H) - \rho_{xx}(H=0)] \times 100\%/\rho_{xx}(H=0)$. The supplementary material in Fig. S1(b) shows the relationship between MR and H when the magnetic field is perpendicular and parallel to the current at 20 K, indicating that the samples exhibit isotropic properties. Considering the thickness of the conductive layer, the electric conduction of KTO under 360 nm light is quasi-three-dimensional. Additionally, $LaAlO_3$/KTO heterointerfaces were prepared using pulsed laser deposition at 800 °C and 1×10^{-3} Pa of O_2 with a KrF excimer laser ($\lambda = 248$ nm) that operated at 1 Hz.

3. Results and discussion

The temperature dependences of the electrical resistivity of KTO (100) and (111) are shown in Fig. 1(a). The inset shows a schematic diagram of the measured resistivity and Hall resistivity. We observe that the KTOs show metallic behavior at low temperatures. The temperature dependence of the electrical resistivity can typically be described by a power law, $\rho = \rho_0 + AT^n$.[41] When $n = 2$, the expected behavior is a Fermi liquid, while a 'non-Fermi liquid' is identified with $1 < n < 2$. The fitted values for n are 1.62 for KTO (100) and 1.64 for (111) at low temperatures. These values indicate that they prefer to behave as non-Fermi liquids at low temperatures under 360 nm irradiation.

To analyze the electronic transport of KTO (100) by photo doping, we measure the Hall resistivity (ρ_{xy}) under the magnetic field (H). Figure 1(b) shows this behavior at different temperatures. Strikingly, the ρ_{xy}–H is nonlinear below 20 K. The carrier density (n) and mobility (μ) can be obtained from the Hall coefficient at a zero magnetic field. As shown in Fig. 1(c), the carrier density slightly decreases as the temperature decreases, while the mobility increases and reaches 1200 $cm^2/V{\cdot}s$ at 2 K. More importantly, the KTOs still exhibit metallic behavior, even at very low electron density $n = 1.4 \times 10^{12}$ cm^{-3}. We also measured the carrier density and mobility of KTO single crystals with various light intensities at 2 K. Figure S2 demonstrates that the carrier density increases with the light intensity, which is consistent with previous research of STO single crystals.[42]

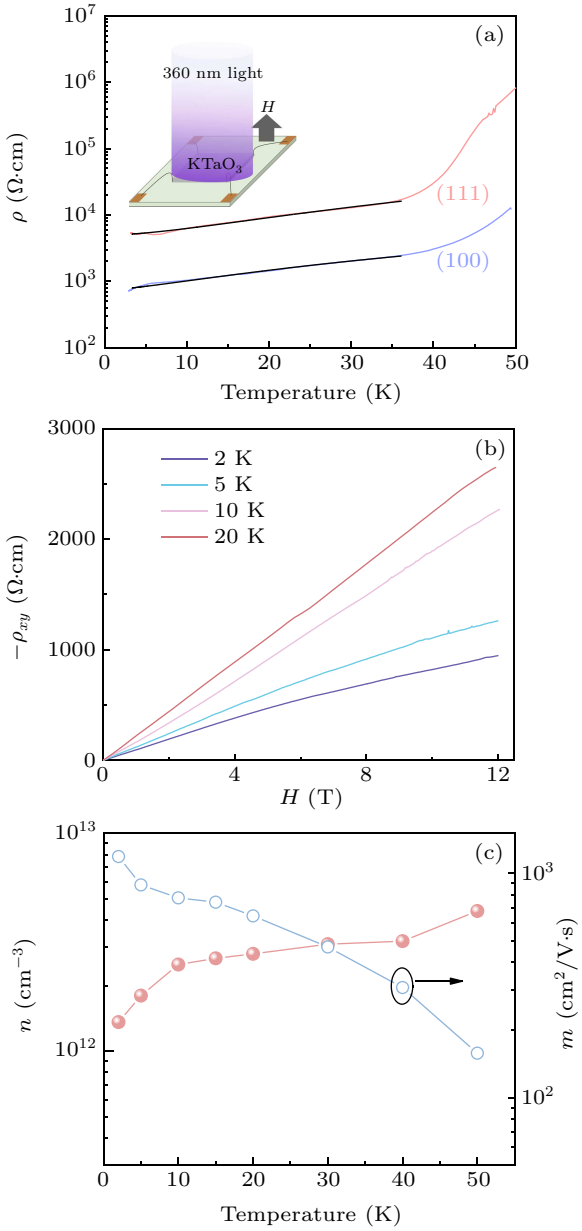


Fig. 1. (a) The temperature dependence of the resistivity of KTO (100) and (111) under illumination at $\lambda = 360$ nm and 33 mW/cm^2. The inset of (a) shows a sketch for the measurements of resistivity and Hall resistivity. (b) Hall resistivity (ρ_{xy}) of KTO (100) under the illumination as a function of the magnetic field at different temperatures. (c) Carrier density (n) and mobility (μ) of KTO (100) as a function of temperature.

The Hall resistivity of KTO (111) as a function of the magnetic field in the temperature range of 2 K–20 K is shown in Fig. 2(a). The Hall resistivity (ρ_{xy}) favors a nonlinear dependence at low temperatures. The mobilities of KTO (111)

as a function of thicknesses and temperatures are shown in Figs. 2(b) and 2(c), respectively. The mobility is enhanced as the temperature decreases and the thicknesses increase. Generally, the nonlinear Hall effect has two origins: one is the abnormal Hall effect caused by magnetism, and the other is caused by the coexistence of two or more types of carriers. The abnormal Hall effect can be determined by the derivative relationship between ρ_{xy} and H if the derivation curve has a peak near 0 T.[43] We exclude the abnormal Hall effect according to the curves of $\mathrm{d}\rho_{xy}/\mathrm{d}H$ (Fig. S3). To further investigate the nonlinear Hall effect, the two-channel conduction from the electronic bands is taken into account. It is found that the fitting result cannot explain the nonlinear Hall effect (Fig. S4 and Table S1). Thereby, the relationship between the magnetic field and the normalized Hall coefficient is analyzed, as shown in Fig. 2(d). The normalized Hall coefficient decreases with the increasing magnetic field, which is consistent with reports by Kozuka *et al.*[38] The decreasing Hall coefficient is probably caused by the low level of scattering under a high magnetic field in the EQL.

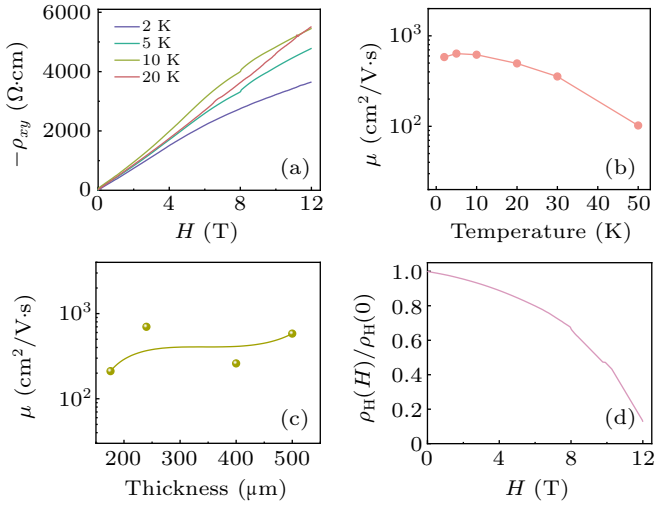


Fig. 2. (a) Hall resistivity (ρ_{xy}) of KTO (111) under the illumination as a function of the magnetic field in the temperature range of 2 K–20 K. (b) and (c) The mobility of KTO (111) as a function of temperatures and thicknesses at 0 T under the illumination with $\lambda = 360$ nm and 33 mW/cm^2, respectively. (d) The normalized Hall coefficient of KTO under illumination as a function of the magnetic field at 2 K.

The requirements to realize the EQL state are $\omega_c\tau > 1$ and $\hbar\omega_c > k_BT$, E_F, where $\omega_c = eH/m^*$ is the cyclotron frequency (e is the elementary charge, H is the magnetic field, and m^* is the electron effective mass), τ is the carrier relaxation time, k_BT is the thermal energy, and E_F is the Fermi energy. In this work, the dielectric constant (ε_r) of KTO is ~ 4500 (2 K)[20] and m^* is $0.8m_0$.[20,40] Thus, the $\hbar\omega_c$ of KTO is estimated about 2.78 meV, which is far larger than k_BT (2 K) (0.18 meV). The E_F of KTO is about 5.4×10^{-4} meV according to $\hbar^2k_F^2/2m^*$. In addition, if we apply the Mott criterion to $\hbar\omega_c > k_BT$, E_F, the EQL state can be realized by satisfying the following conditions: $\varepsilon_r/m_r > 1.3 \times 10^3$, where $\varepsilon_r = \varepsilon/\varepsilon_0$ (ε_0 is the vacuum permittivity) and $m_r = m^*/m_0$ (m_0 is the bare electron mass). For the KTOs, the value of $\omega_c\tau$ is ~ 1.44 at a high magnetic field because of the high mobility ($> 10^3$ cm^2/V·s). Further, the ε_r/m_r is about 5.6×10^3, which is larger than 1.3×10^3. Therefore, KTO is likely to come into an EQL state. Electronic wave functions are highly localized and the small gyration radius of electrons in the perpendicular direction reduces the probability of scattering between electrons. Hence, the Hall coefficient decreases as the magnetic field increases.

Figure 3(a) shows the relationship between the MR of KTO (100) and the magnetic field. The results for KTO (111) are shown in Fig. S5. The MR displays giant positive linear non-saturating features. As shown in Fig. 3(b), the MR increases nonlinearly when decreasing the temperature. The MR values with a thickness of 500 μm under the magnetic field of 12 T are 256%, 340%, 433%, 367%, 263%, and 119% at 2 K, 5 K, 10 K, 20 K, 30 K, and 50 K, respectively. These values are far greater than that of the ordinary MR effect. The reduction of MR below 10 K might be attributed to the localization of electrons due to the ferroelectric phase of KTO.[44] This phenomenon is consistent with the drop in carrier density. Furthermore, we measured the MR of KTOs with different thicknesses and light intensities at 2 K. Figure 3(c) shows that the MR of KTO (111) reaches $\sim 200\%$ (12 T) with different thicknesses and $\sim 20\%$ (2 T) at various light intensities (25 mW/cm^2–35 mW/cm^2). These values are far greater than those of oxide interfaces.[24,29]

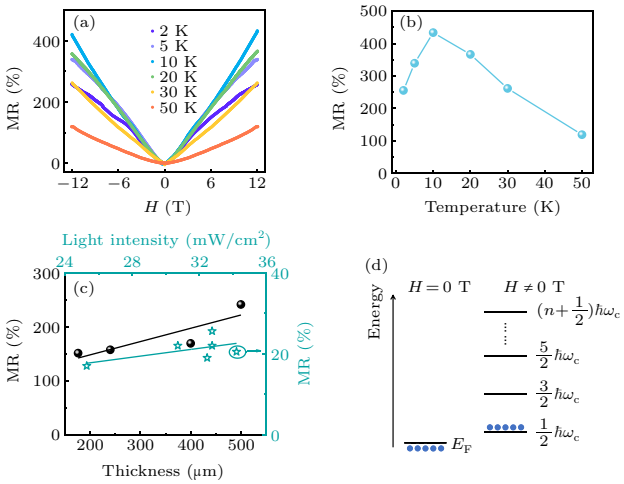


Fig. 3. (a) MR of KTO (100) as a function of the magnetic field at different temperatures. (b) MR of as a function of temperatures with KTO (100) at 12 T. (c) The MR of KTO (111) dependence of different thicknesses (12 T) and light intensities (2 T) at 2 K; the solid lines are linear fitting curves. (d) A schematic diagram of the energy level under $H = 0$ T and $H \neq 0$ T. All the electrons occupy only the lowest Landau level and other levels are empty when $\hbar\omega_c \gg E_F$.

Early in 1959, Lifshits and Peschansky[45] proposed that the linear MR effect could be caused when the Larmor radius of electrons was smaller than their mean free path for a metal in a high magnetic field. Further, Abrikosov declared[46,47]

that the system would reach the EQL state for a material with a small m^* if the magnetic field was very high. This conjecture assumes that all of the electrons would occupy only the lowest Landau energy level in a gapless semiconductor with a linear energy spectrum. This will produce a linear MR with a magnetic field, $\rho_{xx} \propto H$. Following this, various theories have been proposed to explain the positive MR in nonmagnetic materials by mechanisms, including electric field inhomogeneity,[48] density inhomogeneity,[49] density fluctuations,[50] and antiferromagnetic fluctuations.[51] In our work, although the carrier density is very low, the photocarrier-doped KTO favors metallics at low temperatures because a combination of a small m^* and a large dielectric constant can push the insulator–metal transition boundary to low densities.[9] The electrons are distributed below the Fermi surface when $H = 0$ T, as shown in Fig. 3(d), and E_F decreases as H increases. At such a low carrier density and high magnetic field, the Fermi energy of $E_F = 5.4 \times 10^{-4}$ meV is very low. Thus, the lowest Landau energy, $\hbar\omega_c = 2.78$ meV, is far larger than E_F. At this time, the electrons occupy the lowest Landau level, and all the other levels are empty. As a result, KTO shows a quantum MR effect.[13,46,52] A non-saturating MR further verifies that the system of photocarrier-doped KTO comes into the EQL state. Another interpretation is the inhomogeneity in bulk or films,[48–51] which might arise from the carrier-density inhomogeneity, mainly caused by the light distribution from an uneven surface. To further analyze our experimental results, we measured the MR of STO single crystals and $LaAlO_3$/KTO heterointerfaces under 360 nm light illumination. Figure S6 shows that single crystal STO and LAO/KTO exhibit no significant and linear MR under the same conditions. Although we cannot rule out the contribution of light inhomogeneity to the large linear MR, the experiment illustrates very interesting results for KTO under 360 nm irradiation, which further proves that KTO is a novel material with large positive MR based on complex materials.

4. Conclusion

In summary, KTO single crystals exposed to 360 nm light show metallic behavior at low temperatures with very low carrier density and high mobility. We conclude that these photo-doping KTOs reach the EQL state, which therefore provides a new oxide material with EQL. Importantly, we discover a significant positive MR effect and the decreasing of the Hall coefficient at low temperature and a high magnetic field. In particular, the MR value of KTO (100) reaches 433% at 10 K under 12 T. This is caused by all the electrons occupying the lowest Landau level in the EQL state. At the same time, we consider the contribution of light inhomogeneity to the large linear magnetoresistance. This work paves a way to the understanding of the fundamental physics of the interaction between light and complex materials.

Acknowledgments

Project supported by the National Natural Science Foundation of China (Grant No. 51572222), Key Research Project of the Natural Science Foundation of Shaanxi Province, China (Grant Nos. 2021JZ-08 and 2020JM-088), the Natural Science Basic Research Plan in Shaanxi Province of China (Grant No. 2021JM-041), and the Fundamental Research Funds for the Central Universities (Grant Nos. 3102017OQD074 and 310201911cx044).

References

[1] Cong H V and Mesnard G 2010 *Phys. Stat. Sol.* **51** 777
[2] Honda S, Ishikawa T, Takai K, Mitarai Y and Harada H 2004 *J. Appl. Phy.* **96** 5915
[3] Mcguire T R, Hempstead R D and Krongelb S 1976 *AIP Conference Proceedings* **29** 526
[4] Moser J, Matosabiague A, Schuh D, Wegscheider W, Fabian J and Weiss D 2007 *Phys. Rev. Lett.* **99** 056601
[5] Santos T S, Lee J S, Migdal P, Lekshmi I C, Satpati B and Moodera J S 2007 *Phys. Rev. Lett.* **98** 016601
[6] Piraux L, George J M, Despres J F, Leroy C and Fert A 1994 *J. Mag. Mag. Mater.* **65** 2484
[7] Shraiman B I and Mueller R 1996 *Phys. Rev. Lett.* **77** 175
[8] Fontcuberta J, Martínez B, Seffar A, Piñol S, García-Muñoz J L and Obradors X 1996 *Phys. Rev. Lett.* **76** 1122
[9] Delmo M P, Yamamoto S, Kasai S, Ono T and Kobayashi K 2009 *Nature* **457** 1112
[10] Liu K, Chien C L, Searson P C and Yu Z K 1998 *Appl. Phys. Lett.* **73** 1436
[11] Yang F Y, Liu K, Hong K, Reich D H, Searson P C and Chien C L 1999 *Science* **284** 1335
[12] Wu B, Barrena V, Suderow H and Guillamón I 2020 *Phys. Rev. Res.* **2** 022042
[13] Friedman A L, Tedesco J L, Campbell P M, Culbertson J C, Aifer E, Perkins F K, Ward R L, Hite J K, Eddy C R, Jernigan G G and Gaskill D K 2010 *Nano Lett.* **10** 3962
[14] Haeni J H and Al E 2004 *Nature* **430** 758
[15] Hideki, Kato, Akihiko and Kudo 2002 *J. Phys. Chem. B* **106** 5029
[16] Higuchi T, Tsukamoto T, Kobayashi K, Ishiwata Y and Shin S 2000 *Phys. Rev. B* **61** 12860
[17] Li Z, He W, Xiong T, Wang Y, Zhang J, Guo C, Lan K, Shang S, Zhang Y and Li J 2020 *Bio Res.* **15** 5871
[18] William C M, Muthu S P and Ramasamy P 2020 *J. Mater. Sci. Mater. Electron.* **31** 9894
[19] Misra S, Zhang D, Lu P and Wang H 2020 *Nanoscale* **12** 23673
[20] Wemple S H 1965 *Phys. Rev.* **137** A1575
[21] Zhang H, Yu Y, Zhang X, Hui Z and Sun J 2018 *Phys. Rev. Lett.* **121** 116803
[22] Yoshikawa A, Uchida K, Koumoto K, Kato T, Ikuhara Y and Ohta H 2009 *Appl. Phys. Express* **2** 121103
[23] Nakamura H and Kimura T 2009 *Phys. Rev. B* **80** 121308
[24] Ojha S K, Gogoi S K, Patidar M M, Patel R K, Mandal P, Kumar S, Venkatesh R, Ganesan V, Jain M and Middey S 2020 *Adv. Quantum Technologies* **3** 2000021
[25] Ueno K, Nakamura S, Shimotani H, Yuan H T, Kimura N, Nojima T, Aoki H, Iwasa Y and Kawasaki M 2011 *Nat. Nanotech.* **6** 408
[26] Harashima S, Bell C, Kim M, Yajima T, Hikita Y, Hwang H Y and Bell C 2013 *Phys. Rev. B* **88** 085102
[27] Ma Y, Niu J, Xing W, Yao Y and Han W 2020 *Chin. Phys. Lett.* **37** 117401
[28] Zhang H, Ma Y, Zhang H, Chen X, Wang S, Li G, Yun Y, Yan X, Chen Y and Hu F X 2019 *Nano Lett.* **19** 1605
[29] Zou K, Ismail-Beigi S, Kisslinger K, Shen X, Su D, Walker F J and Ahn C H 2015 *APL Mater.* **3** 036104

[30] Wadehra N, Tomar R, Varma R M, Gopal R K, Singh Y, Dattagupta S and Chakraverty S 2020 *Nat. Commun.* **11** 874

[31] Zhang H, Yan X, Zhang J, Zhang J, Han F, Huang H, Qi S, Shi W, Shen B and Sun J 2019 *Mater. Res. Express* **6** 086448

[32] Liu C, Yan X, Jin D, Ma Y, Hsiao H W, Lin Y, Bretz-Sullivan T M, Zhou X, Pearson J and Fisher B 2021 *Science* **371** 716

[33] Wang Y, Zhang Z, Cheng J, Zhang Q, Tang W and Yang K 2020 *J. Mater. Chem. C* **8** 14230

[34] Kalabukhov A, Gunnarsson R, Börjesson J, Olsson E and Winkler D 2007 *Phys. Rev. B* **75** 121404

[35] Huijben M, Brinkman A, Koster G, Rijnders G, Hilgenkamp H and Blank D 2010 *Adv. Mater.* **21** 1665

[36] Herranz G, Basletic M, Bibes M, Carretero C, Tafra E, Jacquet E, Bouzehouane K, Deranlot C, Hamzic A and Broto J M 2007 *Phys. Rev. Lett.* **98** 216803

[37] Ariando A, Wang X, Baskaran G, Liu Z Q, Huijben J, Yi J B, Annadi A, Barman A R, Rusydi A and Dhar S 2011 *Nat. Commun.* **2** 188

[38] Kozuka Y, Susaki T and Hwang H Y 2008 *Phys. Rev. Lett.* **101** 096601

[39] Frova A 1968 *Nuovo Cimento B* **55** 1

[40] Hosler W R and Frederikse H 1969 *Solid State Commun.* **7** 1443

[41] Stemmer S and Allen S J 2018 *Rep. Prog. Phys.* **81** 062502

[42] Kozuka Y, Hikita Y, Susaki T and Hwang H Y 2007 *Phys. Rev. B* **76** 085129

[43] Gan Y, Christensen D V, Zhang Y, Zhang H, Krishnan D, Zhong Z, Niu W, Carrad D J, Norrman K, von Soosten M, Jespersen T S, Shen B, Gauquelin N, Verbeeck J, Sun J, Pryds N and Chen Y 2019 *Adv. Mater.* **31** 1805970

[44] Hulm J K, Matthias B T and Long E A 1950 *Phys. Rev.* **79** 885

[45] Lifshitz I and Peschanskii V 1959 *Soviet Phys-JETP-USSR* **8** 875

[46] Abrikosov A A 1998 *Phys. Rev. B* **58** 2788

[47] Abrikosov A A 2000 *Europhys. Lett.* **49** 789

[48] Xu R, Husmann A, Rosenbaum T F, Saboungi M L and Littlewood P B 1997 *Nature* **390** 57

[49] Wang C C, Liang C T, Jiang Y T, Chen Y F, Cooper N R, Simmons M Y and Ritchie D A 2007 *Appl. Phys. Lett.* **90** 252106

[50] Khouri T, Zeitler U, Reichl C, Wegscheider W, Hussey N E, Wiedmann S and Maan J C 2016 *Phys. Rev. Lett.* **117** 256601

[51] Ji Z, Ji W J, Xu J, Geng X Y and Zhang S T 2017 *Sci. Adv.* **3** e1701473

[52] Hu J S and Rosenbaum T F 2008 *Nat. Mater.* **7** 697