Materials Research Express

**PAPER**

# The effect of fabrication conditions on 2DEGs transport characteristics at amorphous-$LaAlO_3$/$KTaO_3$ interfaces

To cite this article: Hui Zhang *et al* 2019 *Mater. Res. Express* **6** 086448

IOP Publishing Mater. Res. Express 6 (2019) 086448 https://doi.org/10.1088/2053-1591/ab240a

Materials Research Express

CrossMark

RECEIVED
27 March 2019

REVISED
21 April 2019

ACCEPTED FOR PUBLICATION
23 May 2019

PUBLISHED
31 May 2019

PAPER

# The effect of fabrication conditions on 2DEGs transport characteristics at amorphous-$LaAlO_3$/$KTaO_3$ interfaces

Hui Zhang[1,2], Xi Yan[1,2], Jing Zhang[1,2], Jine Zhang[1,2], Furong Han[1,2], Hailin Huang[1,2], Shaojin Qi[1,2], Wenxiao Shi[1,2], Baogen Shen[1,2] and Jirong Sun[1,2]

[1] Beijing National Laboratory for Condensed Matter physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, Peoples' Republic of China

[2] School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, Peoples' Republic of China

E-mail: jrsun@iphy.ac.cn

**Keywords:** oxide film, two-dimensional electron gas, fabrication condition, 5d electrons, hall mobility, two-band model

## Abstract

Two-dimensional electron gases (2DEGs) formed at the interface between two oxide insulators present a promising platform for the exploration of emergent phenomena. While most of the previous works focused on $SrTiO_3$-based 2DEGs, here we report on a systematic investigation of the 2DEGs at amorphous-$LaAlO_3$/$KTaO_3$ (a-LAO/KTO) interfaces, focusing on the effect of fabrication conditions on 2DEGs. We found that 2DEGs can be formed in a wide temperature range from room temperature to 750° under the oxygen pressure $1 \times 10^{-4}$ Pa. Unexpectedly, its performance shows a unusual strong dependence on fabrication temperature: the Hall mobility increases rapidly with the decrease of substrate temperature. The highest extracted mobility of charge carriers coming from $d_{XZ}/d_{YZ}$ subband is ~$6.6 \times 10^3$ $cm^2\,V^{-1}s^{-1}$, achieved under the condition of $T_S = 100$ °C and $P_{O_2} = 3 \times 10^{-5}$ Pa. This value is higher than that of the 2DEGs of a-LAO/$SrTiO_3$ by a factor of 30, which reveals the unique character of the 2DEGs formed by 5*d* electrons. Two-band model is applied for the analysis of the transport behavior, from which information on carrier density and Hall mobility and their dependence on fabrication conditions are determined.

## 1. Introduction

Two-dimensional electron gas (2DEG) at $LaAlO_3$/$SrTiO_3$ (LAO/STO) interface has attracted extensive attentions because of its great potential for the exploration of emergent phenomena [1–14]. After intensive investigations, a lot of exotic properties are observed, such as two-dimensional superconductivity [2–4], interfacial magnetism [5–8], strong electrical field effect manifesting as gate-tunable metal-to-insulator transition [9], quantum Hall effect [12], and efficient spin-to-charge conversion [13, 14]. So far, most of the previous investigations focused on STO-based 2DEGs. In fact, due to the different characters of the 5*d* and 3*d* electrons, 2DEGs residing in $KTaO_3$ (KTO) could be completely different, thus deserve special attention. However, works on this kind of 2DEGs are very limited, in sharp contrast to the 2DEG of LAO/STO. In addition to a few examples of 2DEGs created on the surface of KTO by electric gating, ionic or UV light irradiation [15–19], the only reported 2DEGs at bilayer interfaces are $LaTiO_3$/KTO [20], amorphous-LAO/KTO [21, 22], and EuO/KTO [23]. Effects associated with the distinct characters of KTO are far from being fully explored.

## 2. Experimental methods

Herein we report on a systematic investigation on the transport characteristics of the 2DEGs at the amorphous-$LaAlO_3$/$KTaO_3$ (a-LAO/KTO) interfaces prepared with different conditions. Amorphous $LaAlO_3$ thin films with a thickness of 10 nm were grown by the technique of pulsed laser deposition (PLD) on (001)-oriented KTO substrates ($3 \times 3 \times 0.5$ $mm^3$). The substrate temperature ($T_S$) and the oxygen pressure ($P_{O_2}$)

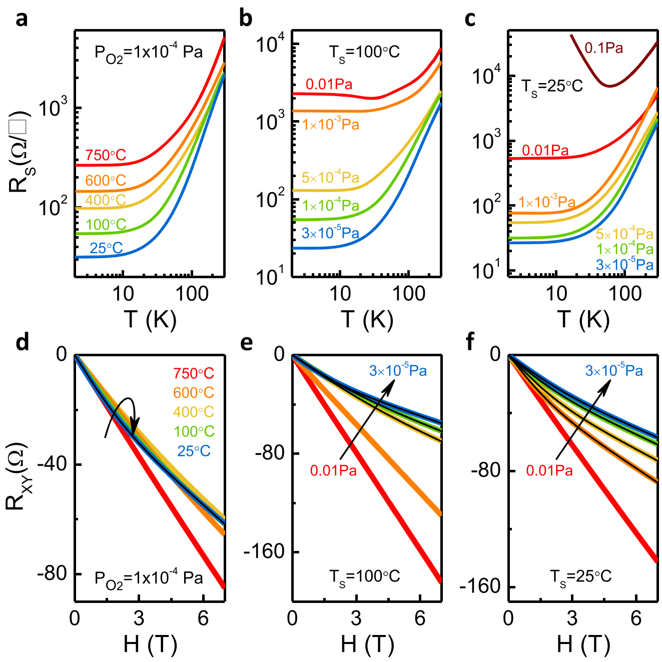


**Figure 1.** (a)–(c) Temperature dependence of sheet resistance for the a-LAO/KTO interfaces fabricated under different substrate temperatures or oxygen pressures. (d)–(f) Corresponding Hall resistance as a function of magnetic field, recorded at 2 K. Thick solid lines are experimental results, and thin lines are results of curve fitting to two-band model.

were varied, while the repetition rate (2 Hz) and fluence ($\sim$2 J cm$^{-2}$) of the KrF excimer laser ($\lambda$ = 248 nm) were held constant. The transport characteristics were measured by a Quantum-designed physical property measurement system (PPMS) adopting Van der Pauw geometry.

## 3. Results and discussion

We prepared three sets of samples to investigate the effect of fabrication conditions. The first set samples were fabricated under different $T_S$ from 750 °C down to room temperature (25 °C), with a fixed oxygen pressure of $P_{O_2} = 1 \times 10^{-4}$ Pa. As evidenced by the previous work, the LAO film presents in the form of an amorphous phase even it was grown at 750 °C [21]. The second and the third sets samples were prepared by setting $T_S$ to 100 °C and 25 °C, respectively, but varying $P_{O_2}$ from 0.1 to $3 \times 10^{-5}$ Pa. Figure 1(a) shows the temperature dependence of the sheet resistance ($R_S$) for the first set samples. An obvious feature is that all 2DEGs are well metallic throughout the temperature range investigated and the metallicity enhances as fabrication temperature decreases, as indicated by the growth of the $R_S$(300 K)/$R_S$(2 K) ratio. A direct calculation shows that $R_S$(300 K)/$R_S$(2 K) is $\sim$19 when $T_S$ is 750°C and $\sim$68 when $T_S$ is 25°C. The greatly increase indicates an enhanced Hall mobility, as will be seen later. This is in sharp contrast to the 2DEGs in the LAO/STO system, where poor conduction was observed at amorphous-LAO/STO interface, except for introducing the $La_{1-x}Sr_xMnO_3$ buffer layer [24]. Figures 1(b) and (c) show $R_S$ versus temperature for the second and the third sets samples, respectively. For the highest $P_{O_2}$ of 0.1 Pa, the sample prepared at 100 °C is totally insulating, while the sample grown at 25 °C exhibits a metal-to-insulator transition around 50 K. As for the remaining samples, their $R_S$-$T$ curves show a general trend: 2DEGs are well metallic over the entire temperature range investigated, and the overall $R_S$-$T$ curve shows a continuous downwards shift as $P_{O_2}$ decreases. This feature is especially obvious for the samples fabricated at $T_S$ = 100 °C. A rather similar oxygen pressure dependent metallic conductivity was observed in a-LAO/STO interfaces prepared at room temperature [25].

To get further information on 2DEGs, Hall resistance ($R_{XY}$) is measured. Figures 1(d)–(f) display the corresponding $R_{XY}$ for three series of samples. All $R_{XY}$-H curves were recorded at 2 K. As shown in figure 1(d), $R_{XY}$ exhibits a linear variation with applied field for the 2DEG grown at $T_S$ = 750 °C. This is the typical feature of the 2DEG with only one species of charge carriers. With the decrease of $T_S$, however, $R_{XY}$-$H$ deviates from linearity, exhibiting an obvious deviation around 1.5 T. This feature enhances with the decrease of $T_S$. A similar phenomenon is also observed in figure 1(e) and (f): the $R_{XY}$-$H$ relation is well linear when $P_{O_2}$ is high and gradually deviates from linearity as $P_{O_2}$ is decreased. Meanwhile, the $R_{XY}$-$H$ slope declines considerably, implying a concomitant increase in carrier density. The non-linear $R_{XY}$-$H$ relation is an indication of either the occurrence of anomalous Hall effect or the coexistence of more than two species of charge carriers. We tried to fit

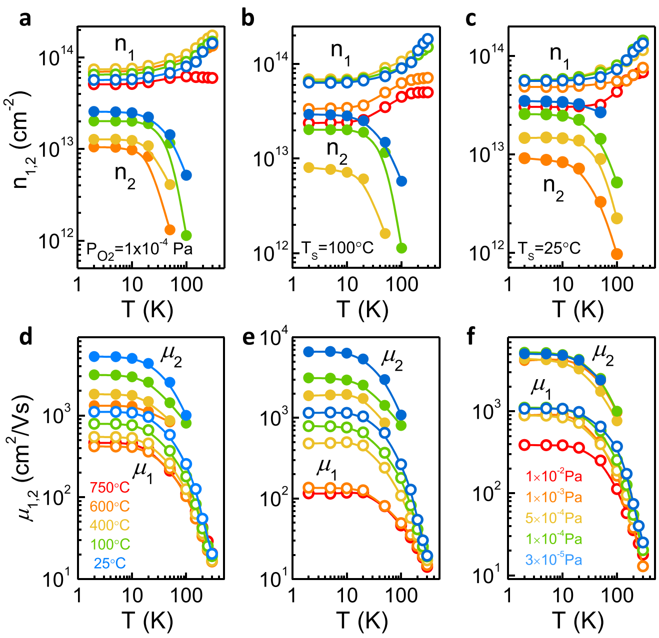


**Figure 2.** Temperature dependence of the carrier density (a)–(c) and Hall mobility (d)–(f) for three series of samples. The corresponding temperatures are denoted in (d) for the first set of samples. The oxygen pressures adopted by the second and third sets of samples are given in (f). The solid lines are guides to the eye.

the $R_{XY}$-$H$ relation to the two-band model [26–28],

$$R_{XY}(H) = -\frac{1}{e}\frac{\left(\frac{n_1\mu_1{}^2}{1+\mu_1{}^2H^2}+\frac{n_2\mu_2{}^2}{1+\mu_2{}^2H^2}\right)H}{\left(\frac{n_1\mu_1}{1+\mu_1{}^2H^2}+\frac{n_2\mu_2}{1+\mu_2{}^2H^2}\right)^2+\left(\frac{n_1\mu_1{}^2}{1+\mu_1{}^2H^2}+\frac{n_2\mu_2{}^2}{1+\mu_2{}^2H^2}\right)^2H^2} \tag{1}$$

with the constraint

$$R_S(0) = \frac{1}{e(n_1\mu_1 + n_2\mu_2)}. \tag{2}$$

where $n_1$ and $n_2$ denote, respectively, the density of the two species of charge carriers, $\mu_1$ and $\mu_2$ are the corresponding Hall mobility. We obtained a very good description of the non-linear $R_{XY}$-$H$ relations in figures 1(d)–(e) (thin solid curves). Therefore, the appearance of the second species of charge carriers is responsible for the curvature of the Hall resistance.

We also measured the $R_{XY}$-$H$ curves at various temperatures for all 2DEGs (not shown). Based on the two-band-model curve fitting, the carrier density and corresponding Hall mobility can be deduced, as shown in figure 2. The 5$d$ subband structure populated by 2DEG at the (100) surface of $KTaO_3$ has been directly measured by angle-resolved photoemission (ARPES) [16, 19] and the band structure calculation is in agreement with ARPES measurement. The results show that quantum confinement lifts the orbital degeneracy of the bulk band structure and leads to a 2DEG composed of ladders of subband states of both light and heavy carriers. Considering the hierarchic band structure of 2DEG at the a-LAO/KTO interface, the carriers originate from the 5$d$ electronic shell of the tantalum (Ta) ions, and the light and heavy charge carriers may come from $d_{XY}$ and $d_{XZ}/d_{YZ}$ orbital states, respectively. The $d_{XY}$ electrons ($n_1$, $\mu_1$) occupy the lowest energy states of the 2DEG and locate very close to interface. They will suffer strong interface scattering thus exhibit low Hall mobility. In contrast, $d_{XZ}/d_{YZ}$ electrons ($n_2$, $\mu_2$) occupy further higher energy states and amount for only a small fraction of the total carrier density. Moreover, these carriers populate deeper KTO layers. Consequently, they are less scattered by interfacial defects thus exhibit high mobility. Different from $n_1$, $n_2$ exhibits a strong temperature dependence. It appears to be very small around ~100 K (~$10^{12}$ cm$^{-2}$) and grows by an order of magnitude when cooled from 100 K to 2 K. However, the temperature dependence of $\mu_2$ is similar to $\mu_1$, first growing and then saturating upon cooling.

Figures 3(a) and (d) show carrier density and Hall mobility, respectively, as a function of the $T_S$. The total carrier density ($n_{total} = n_1 + n_2$) slightly increases as $T_S$ varies from 25 °C to 400 °C and rapidly decreases when $T_S$ exceeds 400 °C. Notably, above 600 °C, only one species of charge carrier survives. The meanings of these

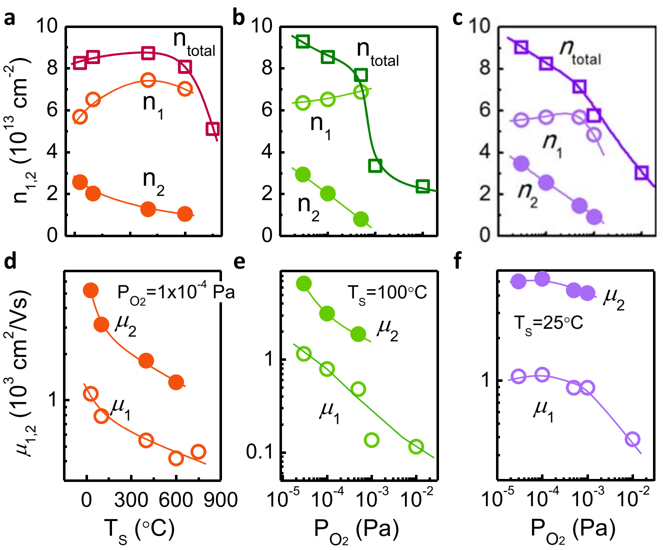


**Figure 3.** Evolution of carrier density (a)–(c) and Hall mobility (d)–(f) with substrate temperature or oxygen pressure, obtained at 2 K. Solid lines are guides to the eye. The highest mobility is $6.6 \times 10^3$ cm$^2$ V$^{-1}$s$^{-1}$, gained under the condition of $T_S = 100$ °C and $P_{O_2} = 3 \times 10^{-5}$ Pa.

results are two-fold. At first, a high growth temperature disfavors high carrier density. Probably interlayer mixing takes place at high temperatures, causing a reduction in carrier density. Secondly, second species of charge carriers appear only when the total carrier density is high enough; it seems that the charge carriers in $d_{XY}$ states spill over, filling into the $d_{XZ}/d_{YZ}$ states.

It is interesting that $\mu_1$ and $\mu_2$ exhibit a similar dependence on $T_S$ though the latter is greater than the former by a factor of ~5, rapidly decreasing with the increase of $T_S$. Take $\mu_2$ as an example. It is ~5233 cm$^2$ V$^{-1}$s$^{-1}$ for $T_S = 25$ °C and ~1312 cm$^2$ V$^{-1}$s$^{-1}$ for $T_S = 600$ °C. In general, Hall mobility contains the information about the defects of the sample. For KTO, a possible process at high temperature could be the evaporation of K atoms due to the sputtering of the plasma jet during sample preparation. As mentioned above $d_{XZ}/d_{YZ}$ electrons are more extended into the interior of KTO, therefore we propose that the volatilization of K which far from the interface in the KTO substrate becomes more serious for the higher $T_S$, which will introduce the more defect and consequently lead to the stronger scattering. The K volatilization could be unobvious at low temperatures. As a result, the interface fabricated at room temperature exhibits a much better performance as characterized by high mobility. This is different from the 2DEG at a-LAO/STO, which usually shows a mobility of 1–200 cm$^2$ V$^{-1}$s$^{-1}$ when prepared at room temperature [29–31].

The carrier density and Hall mobility, corresponding to the fabrication temperatures of 100 °C and 25 °C, as functions of $P_{O_2}$ are also shown in figure 3. $n_{total}$ increases as $P_{O_2}$ decreases, indicating that the electron doping by oxygen vacancies plays an important role in forming the 2DEG. Obviously, the lower the $P_{O_2}$ is the higher the $n_{total}$ will be. Remarkably, above a threshold $P_{O_2}$ ($5 \times 10^{-4}$ Pa for $T_S = 100$ °C and $1 \times 10^{-3}$ Pa for $T_S = 25$ °C), the second species of charge carriers disappear. Accordingly, $n_{total}$ undergoes a sudden drop for $T_S = 100$ °C in figure 3(b). It is possible that the interlayer mixing is severe when $T_S$ is high, strongly counteracting the effect of outward oxygen diffusion from KTO to LAO when $P_{O_2}$ exceeds $5 \times 10^{-4}$ Pa. In contrast, the competition of interlayer mixing and oxygen outwards diffusion is not strong for $T_S = 25$ °C. As a result, the $n_{total}$-$P_{O_2}$ dependence is smooth in figure 3(c). In addition, as shown in figures 3(e) and (f), $\mu_2$ is almost independent of oxygen pressure when $T_S = 25$ °C and rapidly decreases with $P_{O_2}$ when $T_S = 100$ °C. Presumably, the plasma jet is unable to yield considerable K volatilization without the help of temperature. As a result, the ionic scattering is weak and the mobility is only weakly $P_{O_2}$-dependent. When $T_S = 100$ °C, however, the content of ionic defects could be obviously $P_{O_2}$-dependent since Hall mobility rapidly reduces with $P_{O_2}$. We postulate that some of the surface K atoms are oxidized by oxygen atmospheres, and are peeled by the plasma jet in the growth process of a-LAO. In this picture, the lower the $P_{O_2}$ is, the higher the mobility will be. The highest mobility is $6.6 \times 10^3$ cm$^2$ V$^{-1}$s$^{-1}$, gained under the condition of $T_S = 100$ °C and $P_{O_2} = 3 \times 10^{-5}$ Pa. Notably, the highest mobility of a-LAO/KTO interface is also much higher than that of the 2DEGs at the a-LAO/STO interface [27–29]. We attribute the higher mobility of the 5$d$ 2DEGs to a reason that 5$d$ electrons are less tightly bound to the nucleus thus have light effective mass.

Figure 4 presents the relation between carrier density and Fermi energy ($E_F$), obtained at 2 K for all samples. $E_F$ is estimated from the formula $E_F = n_{total}h^2/4\pi m^*$ to be 158 ~ 618 meV above the conduction band bottom,

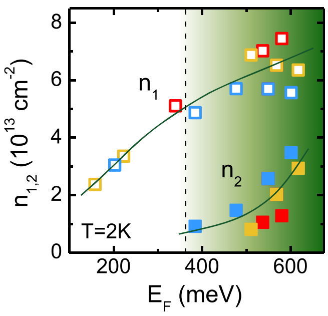


**Figure 4.** Variation of carrier density with Fermi energy, obtained at 2 K. Symbols with different colors correspond to different samples. Dashed line marks the band filling state above which a second species of charge carriers generates. Solid lines are guides for the eye.

where $m^* \approx 0.36m_0^{20}$ with $m_0$ being the free electron mass, and $h$ is the Planck constant. The red, yellow and blue symbols represent the results of three sets of samples, respectively. When the Fermi energy is low there is only one type of charge carriers, populated at the lowest $d_{XY}$ bands. As the Fermi energy rises and passes the bottom of the $d_{XZ}/d_{YZ}$ bands, the 2DEG undergoes a crossover from one to two species of charge carriers. The threshold Fermi level is $E_F \sim$360 meV for this transition (dashed line in figure 4).

## 4. Conclusions

In summary, we systematically investigated the effect of fabrication conditions on 2DEGs transport characteristics at a-LAO/KTO interfaces, by varying the substrate temperature and the oxygen pressure. We found that the interface fabricated at low temperature and high vacuum exhibits a much better metallic character as characterized by high Hall mobility though LAO is amorphous. The highest mobility is $\sim 6.6 \times 10^3$ cm$^2$ V$^{-1}$s$^{-1}$, which is higher by a factor of 30 than that of the 2DEGs at the a-LAO/STO interface. The threshold Fermi energy above which two species of charge carriers coexist is determined. The low growth temperature and high Hall mobility assign 5$d$ 2DEGs great potential for the investigation of quantum phenomenon.


## Acknowledgments

This work has been supported by the National Basic Research of China (Nos. 2016YFA0300701, 2017YFA0206300, 2017YFA0303601 and 2018YFA0305704), the National Natural Science Foundation of China (Nos. 11520101002, 51590880, and 11674378), and the Key Program of the Chinese Academy of Sciences.



## ORCID iDs

Jirong Sun https://orcid.org/0000-0003-1238-8770


## References


[1] Ohtomo A and Hwang H Y 2004 *Nature* **427** 423
[2] Reyren N *et al* 2007 *Science* **317** 1196
[3] Shalom M B, Sachs M, Rakhmilevitch D, Palevski A and Dagan Y 2010 *Phys. Rev. Lett.* **104** 126802
[4] Herranz G *et al* 2015 *Nat. Commun.* **6** 6028
[5] Brinkman A, Huijben M, Van Zalk M, Huijben J, Zeitler U, Maan J C, Van der Wiel W G, Rijnders G, Blank D H A and Hilgenkamp H 2007 *Nat. Mater.* **6** 493
[6] Lee J S, Xie Y W, Sato H K, Bell C, Hikita Y, Hwang H Y and Kao C C 2013 *Nat. Mater.* **12** 703
[7] Bi F, Huang M, Ryu S, Lee H, Bark C-W, Eom C-B, Irvin P and Levy J 2014 *Nat. Commun.* **5** 5019
[8] Kalisky B, Bert J A, Klopfer B B, Bell C, Sato H K, Hosoda M, Hikita Y, Hwang H Y and Moler K A 2012 *Nat. Commun.* **3** 922
[9] Thiel S, Hammerl G, Schmehl A, Schneider C W and Mannhart J 2006 *Science* **313** 1942
[10] Bell C, Harashima S, Kozuka Y, Kim M, Kim B G, Hikita Y and Hwang H Y 2009 *Phys. Rev. Lett.* **103** 226802
[11] Caviglia A D, Gariglio S, Cancellieri C, Sacepe B, Fete A, Reyren N, Gabay M, Morpurgo A F and Triscone J M 2010 *Phys. Rev. Lett.* **105** 236802
[12] Trier F, Prawiroatmodjo G E D K, Zhong Z, Christensen D V, von Soosten M, Bhowmik A, Lastra J M G, Chen Y, Jespersen T S and Pryds N 2016 *Phys. Rev. Lett.* **117** 096804
[13] Lesne E *et al* 2016 *Nat. Mater.* **15** 1261

[14] Song Q, Zhang H R, Su T, Yuan W, Chen Y Y, Xing W Y, Shi J, Sun J R and Han W 2017 *Science Advances* **3** e1602312
[15] Nakamura H and Kimura T 2009 *Phys. Rev.* B **80** 121308
[16] King P D C *et al* 2012 *Phys. Rev. Lett.* **108** 117602
[17] Harashima S, Bell C, Kim M, Yajima T, Hikita Y and Hwang H Y 2013 *Phys. Rev.* B **88** 085102
[18] Bareille C *et al* 2014 *Sci. Rep.* **4** 3586
[19] Santander-Syro A F *et al* 2012 *Phys. Rev.* B **86** 121107
[20] Zou K, Ismail-Beigi S, Kisslinger K, Shen X, Su D, Walker F J and Ahn C H 2015 *APL Mater.* **3** 036104
[21] Zhang H *et al* 2017 *ACS Appl. Mater. Interfaces* **9** 36456
[22] Zhang H *et al* 2019 *ACS Nano* **13** 609
[23] Zhang H R *et al* 2018 *Phys. Rev. Lett.* **121** 116803
[24] Chen Y Z *et al* 2015 *Nat. Mater.* **14** 801
[25] Sambri A, Cristensen D V, Trier F, Chen Y Z, Amoruso S, Pryds N, Bruzzese R and Wang X 2012 *Appl. Phys. Lett.* **100** 231605
[26] Gunkel F *et al* 2016 *Phys. Rev.* X **6** 031035
[27] Joshua A, Pecker S, Ruhman J, Altman E and Ilani S 2012 *Nat. Commun.* **3** 1129
[28] Kim J S, Seo S S A, Chisholm M F, Kremer R K, Habermeier H U, Keimer B and Lee H N 2010 *Phys. Rev.* B **82** 201407
[29] Lee S W, Liu Y, Heo J and Gordon R G 2012 *Nano Lett.* **12** 4775
[30] Chen Y Z, Pryds N, Kleibeuker J E, Koster G, Sun J R, Stamate E, Shen B G, Rijnders G and Linderoth S 2011 *Nano Lett.* **11** 3774
[31] Liu Z Q, Ariando *et al* 2013 *Phys. Rev.* X **3** 021010