



Title	Spontaneous Generation of Carrier Electrons at the Interface between Polycrystalline ZnO and Amorphous InGaZnO4
Author(s)	Fabian, Krahl; Yuzhang, Wu; Cho, Hai Jun; Karppinen, Maarit; Ohta, Hiromichi
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5 **Spontaneous Generation of Carrier Electrons at the Interface between Polycrystalline**
6 **ZnO and Amorphous InGaZnO₄**

7
8 *Fabian Krahl, Yuzhang Wu, Hai Jun Cho*, Maarit Karppinen, and Hiromichi Ohta**

9
10 F. Krahl, Prof. M. Karppinen

11 Department of Chemistry and Materials Science, Aalto University, FI-00076 Aalto, Finland

12 Prof. H.J. Cho, Y. Wu, Prof. H. Ohta

13 Graduate School of Information Science and Technology, Hokkaido University, N14W9,

14 Kita, Sapporo 060-0814, Japan

15 Prof. H.J. Cho, Prof. H. Ohta

16 Research Institute for Electronic Science, Hokkaido University, N20W10, Kita, Sapporo

17 001-0020, Japan

18 E-mail: joon@es.hokudai.ac.jp, hiromichi.ohta@es.hokudai.ac.jp

19
20 Keywords: ZnO, InGaZnO₄, interface, multi-layered film

21
22 **The interface between two materials can be expected to show exotic optical, electrical,**
23 **and thermal transport properties due to the difference in chemical bonding and**
24 **chemical potential. However, in conventional material systems, the volume fraction of**
25 **the interface is small compared to bulk, and interfacial properties are thus difficult to**
26 **utilize. In this regard, multi-layered films are essential to increase the volume fraction of**
27 **interfaces and functionalize their properties. Here we show that carrier electrons can be**
28 **generated spontaneously at the interface between polycrystalline ZnO and amorphous**
29 **(a-) InGaZnO₄. We measured the electron transport properties of multi-layered films**
30 **composed of *c*-axis oriented polycrystalline ZnO and a-InGaZnO₄ with varying interface**
31 **density (d^{-1}). Although the carrier concentrations of both ZnO and a-InGaZnO₄ were**
32 **less than $5 \times 10^{19} \text{ cm}^{-3}$, the n increased with d^{-1} and exceeded 10^{20} cm^{-3} . The relatively**
33 **large interface thermal resistance between ZnO and a-InGaZnO₄ ($1.35 \text{ m}^2 \text{ K GW}^{-1}$)**
34 **indicated the existence of a large difference in the chemical bonding and the chemical**
35 **potential and thus conduction electrons would accumulate at the interface.**

1 The interface between two materials can be expected to show exotic optical, electrical, and
2 thermal transport properties due to the difference in chemical bonding and chemical potential.<sup>[1-
3 3]</sup> Especially in oxide systems, it is known that two-dimensional electron liquid (2DEL) can be
4 spontaneously generated at the heterointerface of two oxides such as $\text{LaAlO}_3/\text{SrTiO}_3$ ^[4,5] due to
5 their strong ionicity. Since these ABO_3 crystals can be considered as pilings of $(\text{LaO})^+(\text{AlO}_2)^-$
6 and $(\text{SrO})^0(\text{TiO}_2)^0$ layers, the heterointerface should be $(\text{AlO}_2)^-(\text{LaO})^+|(\text{TiO}_2)^0(\text{SrO})^0$. Thus, the
7 positive charge of $(\text{LaO})^+$ accumulates electrons at the heterointerface. After the pioneering
8 finding of $\text{LaAlO}_3/\text{SrTiO}_3$ ^[4], several similar systems were found one after another^[6-9]. Thus,
9 similar phenomena should occur at the oxide heterointerfaces because of the difference of
10 electronegativity and chemical potential.^[10]

11
12 However, the volume fraction of the interface is usually small compared to the bulk and thus it
13 is difficult to extract the properties only from the interface. Therefore, multi-layered films are
14 essential to increase the volume fraction of the interfaces and to exploit their properties.^[11, 12]
15 In this study, we focused on physical properties including optical, electrical, and thermal
16 transport properties of the multi-layered films composed of *c*-axis oriented polycrystalline ZnO
17 and amorphous InGaZnO_4 (a-IGZO hereafter)^[13, 14] varying the interface density (d^{-1}). We
18 chose the combination of ZnO/a-IGZO for several reasons. Firstly, both ZnO and a-IGZO are
19 known as n-type oxide semiconductors.^[15] Thus, we can measure the electron transport
20 properties without using special techniques to make the contact electrode. Second, *c*-axis
21 oriented polycrystalline ZnO films can be deposited even at room temperature^[16], whereas
22 crystallization of a-IGZO does not occur until 600 °C^[17]. The amorphous layers of IGZO should
23 help to ensure that the surface and interfaces are smooth even when fabricated at room
24 temperature. Fabrication at room temperature should also minimize doping due to diffusion
25 effects. Further, we anticipated that the trivalent In^{3+} and Ga^{3+} ions of a-IGZO accumulate
26 carrier electrons at the interface of ZnO/a-IGZO on the ZnO side.

1

2 Here we report that carrier electrons indeed can be generated spontaneously at the interface
3 between polycrystalline ZnO and amorphous InGaZnO₄. We measured the electron transport
4 properties of multi-layered ~100 nm thick films composed of *c*-axis oriented polycrystalline
5 ZnO and a-InGaZnO₄ with varying interface density (d^{-1}). Although the carrier concentrations
6 (n) of both ZnO and a-InGaZnO₄ were less than $5 \times 10^{19} \text{ cm}^{-3}$, the n increased with increasing
7 interface density d^{-1} and exceeded 10^{20} cm^{-3} , indicating carrier electron generation occurred at
8 the heterointerface. Relatively large interface thermal resistance between ZnO and a-InGaZnO₄
9 ($1.35 \text{ m}^2 \text{ K GW}^{-1}$) indicated the existence of large difference in the chemical bonding and that
10 of the chemical potential. This implies that conduction electrons could accumulate at the
11 interface.

12

13 The ZnO/a-IGZO multi-layered films were deposited on non-alkali glass substrates at room
14 temperature by pulsed laser deposition (PLD) technique (see Experimental section for details)
15 as schematically shown in **Figure 1**. In order to verify the multi-layered films, X-ray diffraction
16 (XRD) and reflectivity (XRR) analyses were performed. **Figure 2** shows (a) XRR patterns, and
17 (b) out-of-plane XRD Bragg diffraction patterns of the films. The total film thickness was
18 calculated using the Kiessig fringes (**Figure 2a**); the slow decay of these fringes even at high
19 scattering vectors shows that the films are very smooth as diffuse scattering is very low. In
20 addition to the Kiessig fringes, several intense reflection peaks are seen and indicated with
21 arrows; these confirm the superlattice structure. The period of the multi-layered films was
22 calculated from these peaks. Intense diffraction peaks of the 0002 Bragg diffraction in ZnO are
23 seen in the case of ZnO/a-IGZO 1/1, 2/1, 6/1, 10/1, 19/1, and 115/0 (data not shown), whereas
24 only halo patterns are seen in the case of ZnO/a-IGZO 1/2, 1/6, 1/10, 1/19, and 0/118 (data not
25 shown). In addition to the 0002 Bragg peaks, satellite peaks are periodically seen in **Figure 2c**,
26 providing further confirmation on our superlattice structure. Thus, we could conclude that

1 periodical multi-layered films composed of *c*-axis oriented ZnO and amorphous InGaZnO₄
2 were realized with ZnO \geq 1 nm and a-IGZO layers = 1 nm. (cf. **Figure 2b**).

3

4 We measured the electron transport properties of the multi-layered films at room temperature
5 in air. **Figure 3** shows (a) electrical conductivity (σ), (b) Hall mobility (μ_{Hall}), (c) carrier
6 concentration (n) and (d) thermopower (S) of x -nm-thick ZnO/ y -nm-thick a-IGZO multi-
7 layered films as a function of the interface density (d^{-1}), i.e. the number of interfaces divided
8 by the total film thickness d . Two curves are clearly distinguished in all cases. The σ increases
9 with increasing d^{-1} except for ZnO/a-IGZO 1/1. In the case of ZnO-rich multi-layered films,
10 μ_{Hall} decreases with increasing d^{-1} , whereas it increases with increasing d^{-1} in the a-IGZO-rich
11 samples. The μ_{Hall} of ZnO and a-IGZO are $\sim 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $\sim 0.8 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively.
12 Since the observable μ_{Hall} is the weighted average of ZnO and a-IGZO against their sheet
13 conductance, it decreases with increasing a-IGZO contribution. It should be noted that the
14 carrier concentration increases with interface density and exceeds 10^{20} cm^{-3} , while n for ZnO
15 and a-InGaZnO₄ is less than $5 \times 10^{19} \text{ cm}^{-3}$. This behaviour of n has been confirmed by
16 measuring the S ; the absolute value of S gradually decreases with d^{-1} , consistent with n . These
17 results clearly indicate that carrier generation occurs at the heterointerfaces.

18

19 In order to clarify the photon and phonon transport properties across the ZnO/a-InGaZnO₄
20 interfaces, we measured optical absorption spectra and thermal conductivities (κ) of the multi-
21 layered films. The absorption spectra are shown in **Figure 4**. The absorption edge of a pure
22 ZnO film (115 nm/0 nm) looks abrupt due to the free exciton absorption at $\sim 3.3 \text{ eV}$. The
23 absorption coefficient (α) at 3.0 eV decreases with decreasing ZnO content, as seen from **Figure**
24 **4b** where α is plotted against d^{-1} (as an indirect but convenient measure of the a-IGZO content).
25 In addition, the slope of the cusp flattens with increasing a-IGZO content, and the absorption
26 edge gradually changes from a sharp cusp to an Urbach tail, which is commonly observed from

1 amorphous materials. This shows that the dominating electronic structure in the multi-layered
2 films changes from polycrystalline ZnO to a-IGZO, which agrees with the XRD
3 characterization results. These results suggest that the interfaces of ZnO/a-IGZO are optically
4 smooth and the bandgap of both layers did not change upon forming the heterointerface.
5

6 **Figure 5** summarizes the cross-plane κ of the multi-layered films at room temperature. If a-
7 IGZO dominates the multilayered films ($V_{\text{a-IGZO}} > 50\%$), the κ exhibits a plateau at $\sim 0.8 \text{ W m}^{-1}$
8 K^{-1} from $d^{-1} = 0.05 \text{ nm}^{-1}$ to 0.17 nm^{-1} , then slowly increases up to $\sim 1 \text{ W m}^{-1} \text{ K}^{-1}$ ($d^{-1} = 0.5$
9 nm^{-1}), which are all consistent with previously reported values for a-IGZO. When the a-IGZO
10 phase is dominant in the multi-layered films, the κ values are mainly affected by the volume
11 fraction of the a-IGZO layers. On the other hand, the κ value monotonically decreases with
12 increasing d^{-1} if $V_{\text{ZnO}} > 50\%$, which is a typical consequence from phonon scatterings at
13 interfaces in multi-layered films. The Kapitza resistance (R_K) of the a-IGZO layer in
14 polycrystalline ZnO, which can be obtained from the slope of the thermal resistivity (κ^{-1}) vs
15 d^{-1} , was calculated to be $1.35 \text{ m}^2 \text{ K GW}^{-1}$ (**Figure 5b**). This value is considerably higher than
16 the value ($0.5 \text{ m}^2 \text{ K GW}^{-1}$) found in ZnO/In₂O₃ superlattices (please add
17 <https://doi.org/10.1063/1.4809784>) and rather in line with the atomically different interfaces
18 such as TiN/MgO (<https://doi.org/10.1103/PhysRevB.67.054302>). Most importantly, the
19 relatively large R_K between ZnO and a-InGaZnO₄ is a strong implication of the anticipated
20 difference in the chemical bonding and the chemical potential between the crystalline ZnO layer
21 and the amorphous InGaZnO layer. Another strong argument pointing towards this conclusion
22 is the observed accumulation of conduction electrons at the interface. In principle, the observed
23 increase of charge carrier density could also be due to doping effects, but the high Kapitza
24 resistance and the XRR measurements confirm a clear separation of the ZnO and a-IGZO layers
25 in the films and together with the fact that the films were deposited at room temperature we
26 believe doping is not the primary reason for the increase in charge carriers.

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In summary, we measured the electron transport properties of multi-layered films composed of *c*-axis oriented polycrystalline ZnO and a-InGaZnO₄ with varying interface density. With increasing d^{-1} , the carrier concentration was found to increase by nearly an order of magnitude compared to the n values less than $5 \times 10^{19} \text{ cm}^{-3}$ for both ZnO and a-InGaZnO₄, exceeding 10^{20} cm^{-3} for the multi-layered films. The thermal conductivity decreased with the d^{-1} when the volume fraction of ZnO was larger than a-InGaZnO₄. Relatively large interface thermal resistance between ZnO and a-InGaZnO₄ ($1.35 \text{ m}^2 \text{ K GW}^{-1}$) indicated the existence of large difference in the chemical bonding and of the chemical potential, and thereby the accumulation of conduction electrons at the interfaces.

Experimental Section

Preparation of the multi-layered films: The multi-layered films composed of alternately stacks of polycrystalline ZnO and amorphous InGaZnO₄ were deposited on non-alkali glass substrates (Corning® EAGLE XG®) by pulsed laser deposition (PLD, KrF excimer laser, $\sim 1.5 \text{ J cm}^2 \text{ pulse}^{-1}$, 5 Hz) under an oxygen atmosphere (1 Pa) at room temperature. The total film thickness was kept around 100 nm. Prior to the multi-layered film deposition, we measured the deposition rate of polycrystalline ZnO and amorphous InGaZnO₄. We precisely controlled the thickness of the ZnO and InGaZnO₄ films by the number of the KrF excimer laser pulses using the deposition rate.^[12] The crystallographic orientation, surface roughness, and thickness of the resultant films were evaluated by X-ray diffraction (XRD) and X-ray reflectivity (XRR) measurements with monochromatic Cu K α radiation (ATX-G, Rigaku Co.).

Measurement of optical, electrical, and thermal properties: Optical transmission and reflection spectra of the films were measured by using a UV-Vis spectrometer (Solidspec-3700, Shimadzu Co.) at room temperature in air. Electrical conductivities were measured by conventional d.c.

1 four-point probe method with the van der Pauw electrode configuration at room temperature in
2 air. Hall effect measurements were also performed. Thermopower of the films was measured
3 by the standard steady-state method.^[18-20] The film sample was placed on the gap (~5 mm)
4 between two Peltier devices. By applying the forward/reverse current to each Peltier device, the
5 temperature difference was generated in the sample. We measured the temperature difference
6 (ΔT) and the thermo-electromotive force (ΔV) simultaneously at room temperature. The
7 thermopower was calculated as the linear slope of the $\Delta T - \Delta V$ plot. Cross-plane thermal
8 conductivity (κ) of the resultant films was measured by the time domain thermo-reflectance
9 (TDTR, PicoTR, PicoTherm Co.). The 116-nm-thick Mo was deposited on the
10 ZnO/amorphous InGaZnO₄ films as a transducer by dc sputtering. The decay curves of the
11 TDTR signals were simulated using the software provided by the manufacturer.

12

13 **Supporting Information**

14 Supporting Information is available from the Wiley Online Library or from the author.

15

16

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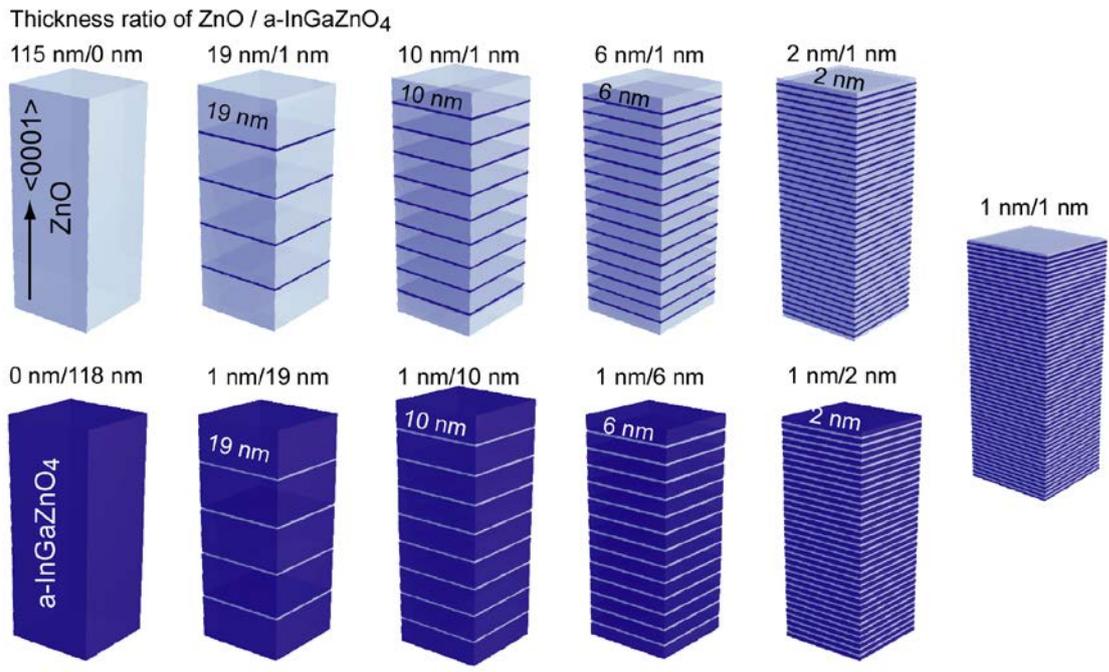
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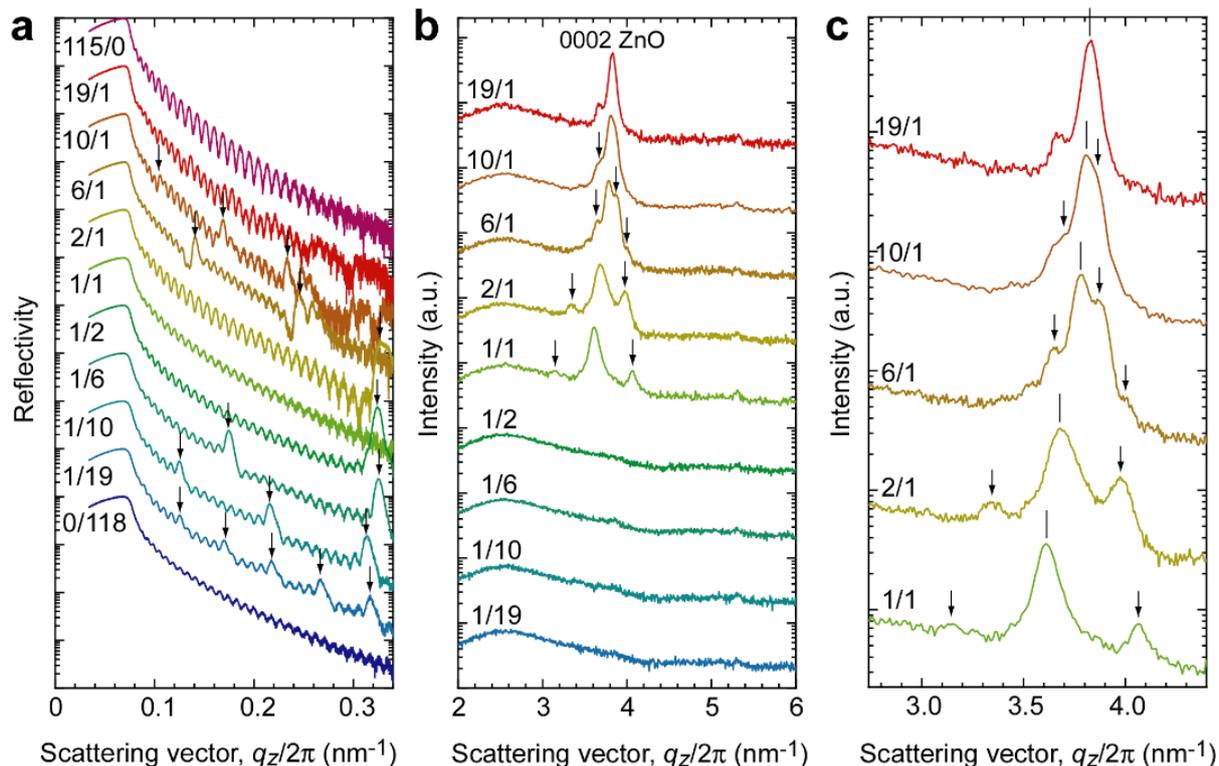
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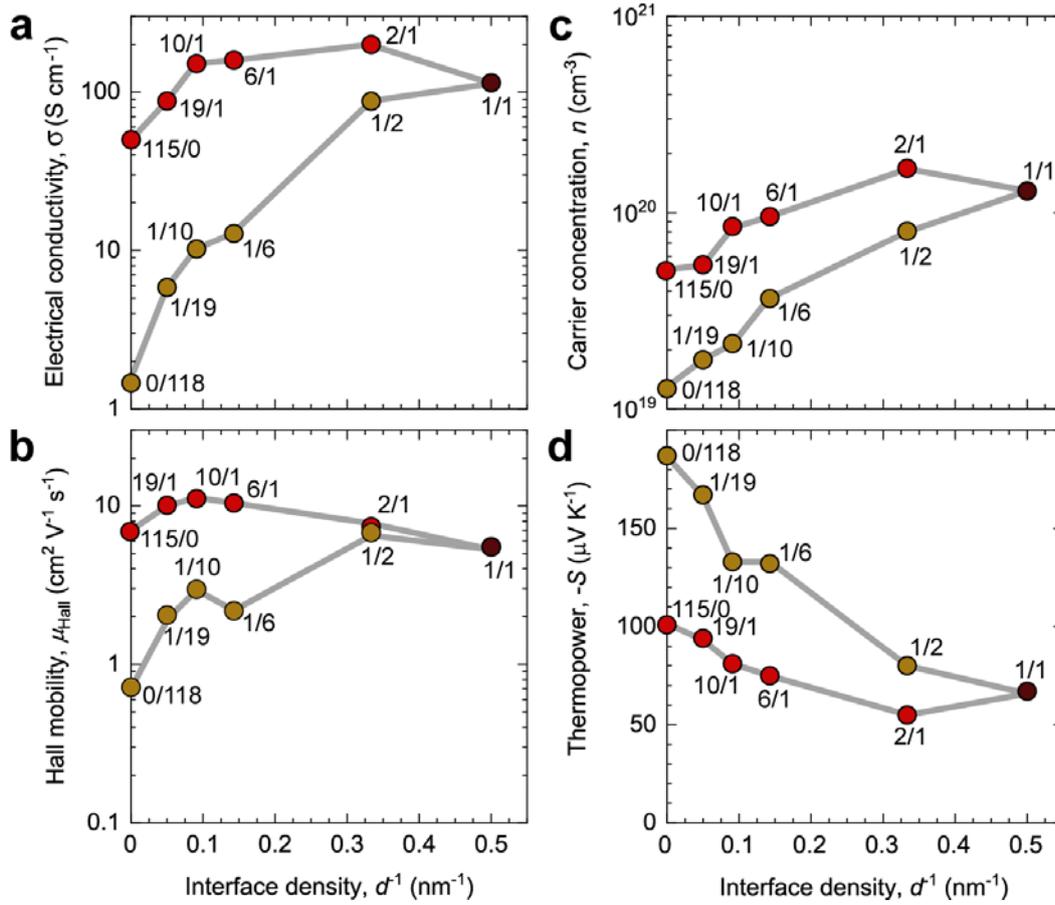


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Figure 1. Structure of the polycrystalline ZnO / amorphous InGaZnO₄ multi-layered films. Schematic cross-sectional views of the multi-layered films composed of x -nm-thick c -axis oriented ZnO (light blue) and y -nm-thick amorphous InGaZnO₄ (dark blue). The thickness ratio of x/y is systematically varied from 115 nm/0 nm to 0 nm/118 nm.

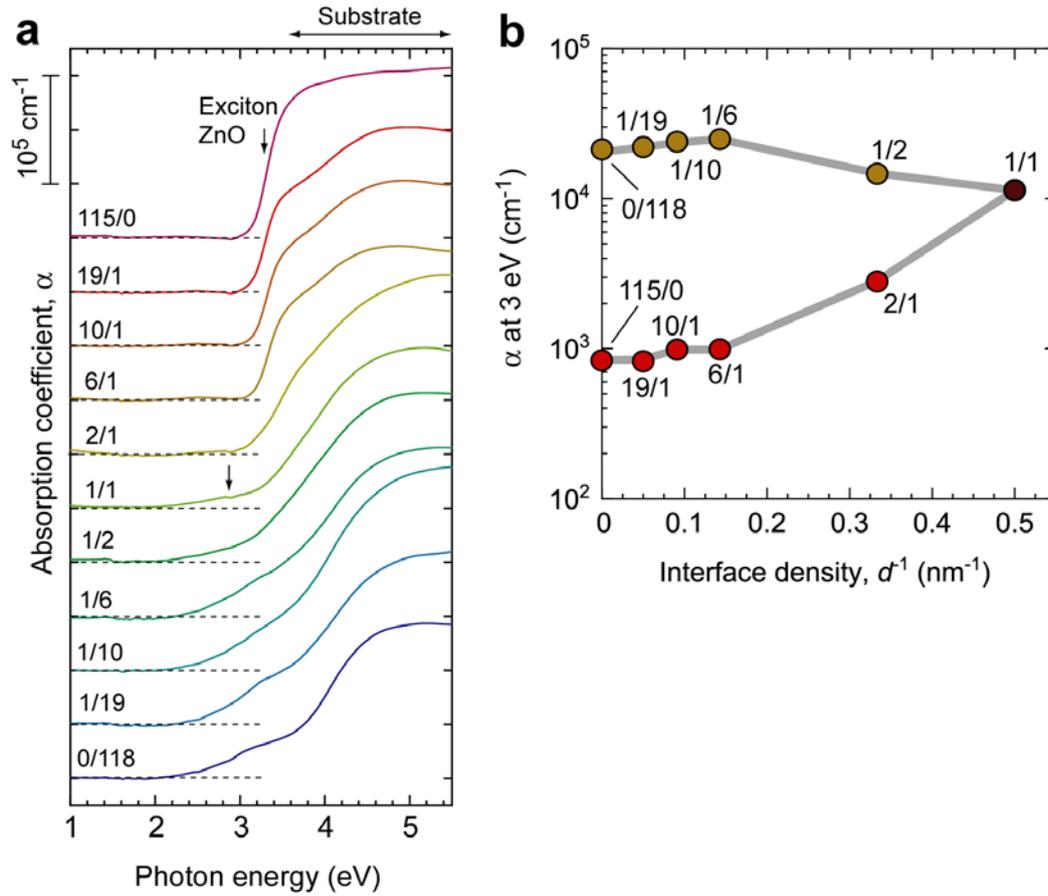


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4 **Figure 2. X-ray diffraction analyses of the multi-layered films composed of x -nm-thick c -**
5 **axis oriented ZnO and y -nm-thick amorphous InGaZnO₄.** (a) X-ray reflectivity curves, (b)
6 out-of-plane Bragg diffraction patterns, and (c) magnified patterns of (b). The total film
7 thickness of the samples was calculated using the Kiessig fringes. In addition to the Kiessig
8 fringes, several intense reflection peaks are seen and indicated with arrows. These confirm the
9 superlattice structure and the period of the multilayered film was calculated from these peaks.
10 (b) Intense diffraction peaks of 0002 ZnO are seen in the case of ZnO/a-InGaZnO₄ is 1/1, 2/1,
11 6/1, 10/1, 19/1, and 115/0 (data not shown), whereas only halo patterns are seen in the case of
12 ZnO/a-InGaZnO₄ is 1/2, 1/6, 1/10, 1/19, and 0/118 (data not shown). In addition to the 0002
13 Bragg peaks, satellite peaks are periodically seen in (c). The period of the multi-layered film
14 was confirmed using these satellite peaks. Periodical multi-layered films composed of c -axis
15 oriented ZnO and amorphous InGaZnO₄ can be fabricated when ZnO film is thicker than 1
16 nm and a-InGaZnO₄ is 1 nm.
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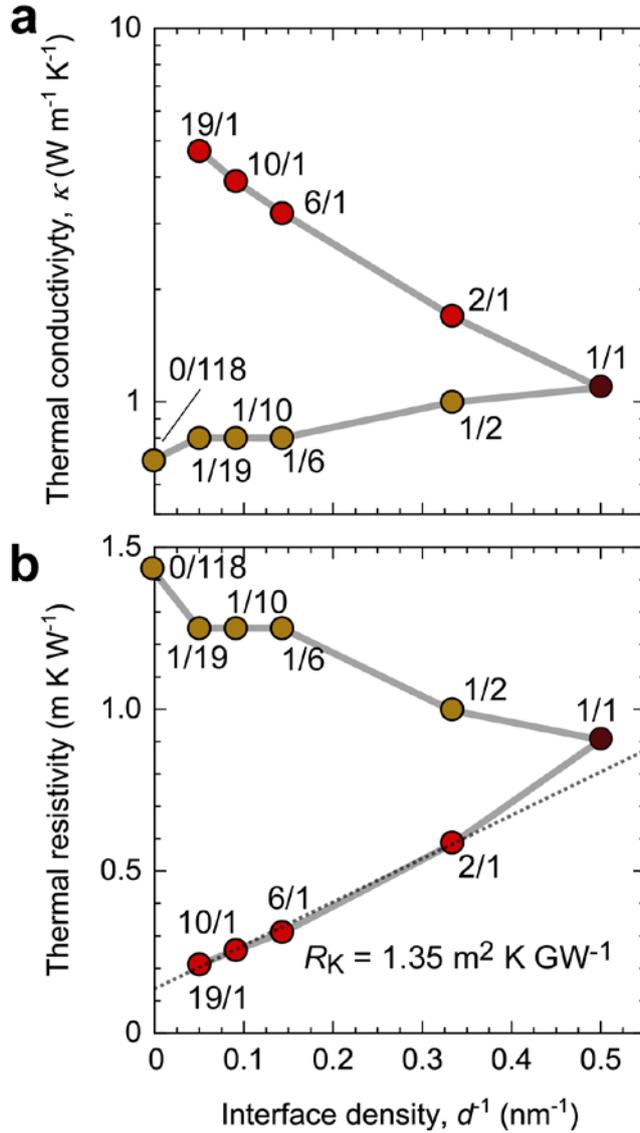
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Figure 3. In-plane electron transport properties of the multi-layered films composed of x -nm-thick c -axis oriented polycrystalline ZnO and y -nm-thick amorphous InGaZnO₄ at room temperature. (a) Electrical conductivity (σ), (b) Hall mobility (μ_{Hall}), (c) carrier concentration (n), and (d) thermopower (S) as a function of d^{-1} . Note that the carrier concentration increases with interface density and exceeds 10^{20} cm^{-3} while n of ZnO and a-InGaZnO₄ is less than $5 \times 10^{19} \text{ cm}^{-3}$. The absolute value of S gradually decreases with d^{-1} , consistent with n . These results clearly indicate that carrier generation occurs at the heterointerfaces.



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Figure 4. Optical absorption of the multi-layered films composed of x -nm-thick c -axis oriented ZnO and y -nm-thick amorphous InGaZnO₄. (a) Absorption spectra. The absorption edge of ZnO film (115 nm/0 nm) looks abrupt due to the free exciton absorption at ~ 3.3 eV. The absorption coefficient α at 3.0 eV decreases with decreasing the ZnO content. Instead, α at 2.9 eV increases with increasing a-InGaZnO₄ content. (b) Plots of the α at 3 eV as a function of interface density d^{-1} . This result clearly indicates that the optical absorption changes from excitonic absorption of c -axis oriented ZnO to Urbach tail (see arrow) of a-InGaZnO₄.



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Figure 5. Cross-plane heat transport properties of the multi-layered films composed of x -nm-thick c -axis oriented ZnO and y -nm-thick amorphous InGaZnO₄ at room temperature. (a) Thermal conductivity (κ) as a function of interface density d^{-1} . The κ decreases dramatically with increasing d^{-1} when $x/y \geq 1/1$. (b) Thermal resistivity (κ^{-1}) as a function of d^{-1} . The interface thermal resistance (Kápitza resistance, R_K) is $1.35 \text{ m}^2 \text{ K GW}^{-1}$.

1 Spontaneous carrier electron generation at the oxide semiconductor heterointerface was
2 demonstrated using multi-layered films composed of ZnO and a-InGaZnO₄ with various
3 interface densities. Relatively large interface thermal resistance ($1.35 \text{ m}^2 \text{ K GW}^{-1}$) indicated
4 the existence of large difference in the chemical bonding and that of the chemical potential
5 and thus conduction electrons would be accumulated at the heterointerface.

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7 **Keyword** ZnO, InGaZnO₄, interface, multi-layered film

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9 F. Krahl, Y. Wu, H.J. Cho*, M. Karipinen, H. Ohta*

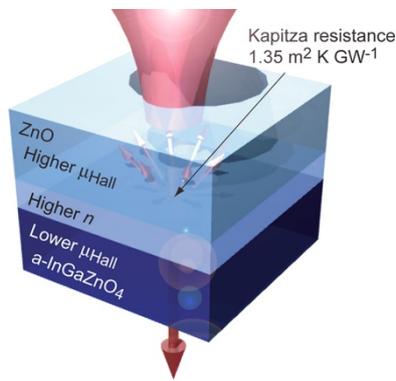
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12 **and Amorphous InGaZnO₄**

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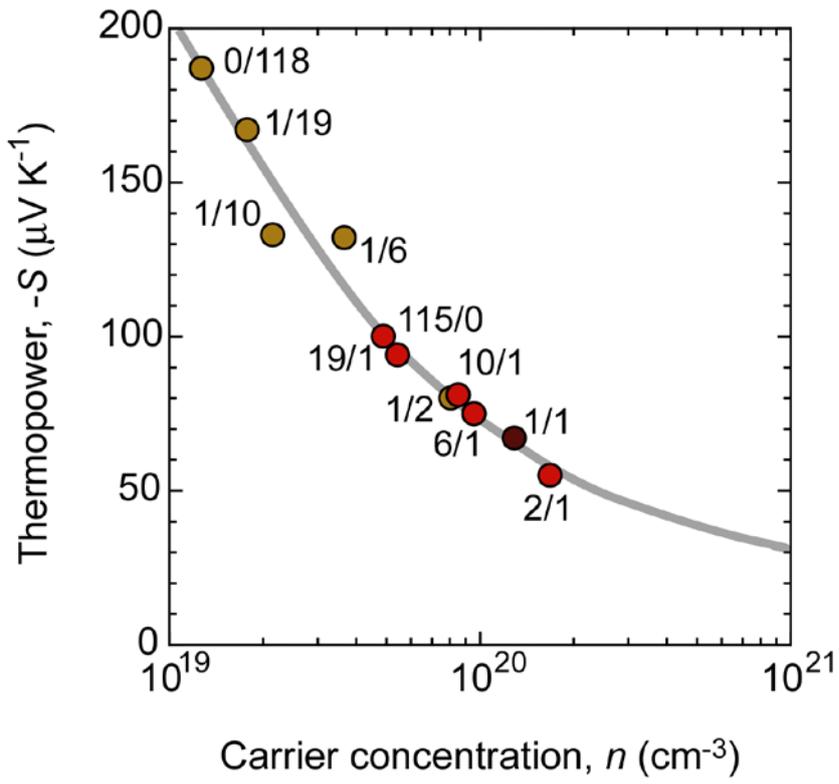
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7 **Spontaneous Carrier Electron Generation at the Interface between Polycrystalline ZnO**
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11 *Fabian Krahl, Hai Jun Cho*, Yuzhang Wu, Maarit Karppinen, and Hiromichi Ohta**

12
13 F. Krahl, Prof. M. Karppinen
14 Department of Chemistry and Materials Science, Aalto University, FI-00076 Aalto, Finland
15 Prof. H.J. Cho, Y. Wu, Prof. H. Ohta
16 Graduate School of Information Science and Technology, Hokkaido University, N14W9,
17 Kita, Sapporo 060-0814, Japan
18 Prof. H.J. Cho, Prof. H. Ohta
19 Research Institute for Electronic Science, Hokkaido University, N20W10, Kita, Sapporo
20 001-0020, Japan
21 E-mail: joon@es.hokudai.ac.jp, hiromichi.ohta@es.hokudai.ac.jp

22
23 **Keywords:** ZnO, InGaZnO₄, interface, multi-layered film

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