Cation Disorder Limited IGZO Mobility Calculation Based on the Density Functional Theory

Seung Hyo Han[1†], Deokhwa Seo[1†], Jun-Hwe Cha[2], Seiyon Kim[2] and Mincheol Shin[1]
[1]School of Electrical Engineering, Korea Advanced Institute of Science and Technology, Daejeon 34141, Rep. of Korea
[2]R&D Division, SK hynix Inc., Icheon, Rep. of Korea
[†]Equal contribution. [*]E-mail: mshin@kaist.ac.kr

2024 International Conference on Simulation of Semiconductor Processes and Devices (SISPAD) | 979-8-3315-1635-2/24/$31.00 ©2024 IEEE | DOI: 10.1109/SISPAD62626.2024.10732931

Abstract—**In this work, we present a density functional theory (DFT) based method to calculate the electrical properties of crystalline IGZO channel material. The effects of cation disorder are captured by atomic-level random cation pair switching within the DFT-generated structures. The cation disorder limited mobilities are calculated without resorting to any parameters, i.e., by directly utilizing DFT-generated samples and their Hamiltonians, yet the calculation results show remarkable agreement with experimental data. Our method enables rigorous and efficient evaluation of electrical properties for IGZO of various compositions, covering both Zn-rich and In-rich cases.**

Keywords—density functional theory, crystalline IGZO, cation disorder-limited mobility

I. INTRODUCTION

Indium gallium zinc oxide (IGZO) has emerged as a promising candidate for future semiconductor devices owing to its high mobility, extremely low off-current and feasibility for 3D stackable DRAM configurations [1, 2]. In particular, crystalline IGZO (c-IGZO), which is crystalline in the c-axis but has randomly positioned cations in the perpendicular a-b planes, exhibits a unique electrical characteristic of mobility enhancement with the increase in carrier density [3]. Previous studies have attempted to model this unusual electrical property focusing on the cation disorder as the dominant scattering mechanism, but they were limited to the In:Ga:Zn = 1:1:1 structure considering only Zn and Ga atoms, or showed an order of magnitude difference in mobility compared to experimental data [4, 5]. Moreover, these studies applied the semiclassical Fermi's golden rule to a single disorder without disorder-to-disorder interaction, whereas in actual c-IGZO structures, the cation disorder scattering rate depends on the complex interplay between the randomly distributed cation disorders. In this work, we utilize a density functional theory (DFT) based method to calculate the cation disorder limited mobility for various c-IGZO compositions. The distribution of cation disorders is captured by atomic-level random cation pair switching within the DFT-generated structure. Multiple cation configurations are sampled to obtain the statistically averaged scattering effect for a given disorder density. Using DFT, IGZO structures with composition ratios of In: Ga: Zn = 1:1:1, 1:1:2, 7:5:6, and 4:2:3 are generated, and the corresponding cation disorder limited mobilities are calculated.

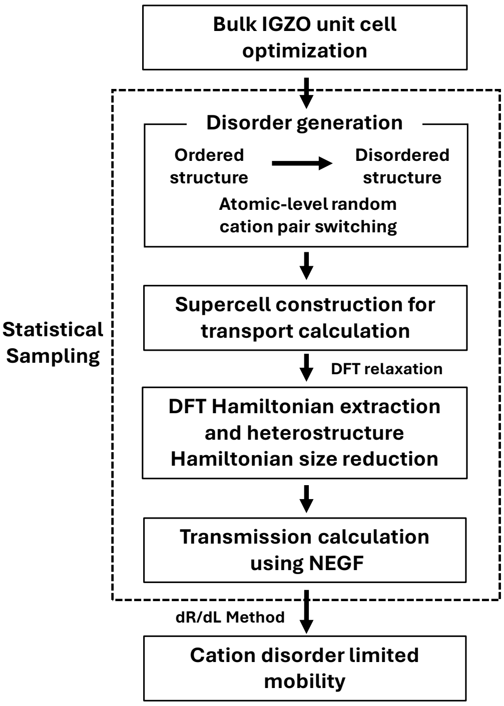

Fig. 1. Schematic of the simulation flow for calculating the cation disorder limited mobility of c-IGZO.

II. APPROACH

A. DFT Method

As cation disorder is known to be the dominating scattering mechanism in c-IGZO [3], it is essential to compute the cation disorder limited mobility to elucidate its electrical characteristics. To uncover the cation disorder limited mobility of c-IGZO, we follow the simulation flow shown in Fig. 1. For the generation of atomic structures and cation disorders, the SIESTA package is used [6], which employs norm-conserving pseudo-potentials [7], along with the generalized gradient approximation (GGA) and the Perdew, Burke and Ernzerhof (PBE) exchange-correlation functional [8]. First, the atomic

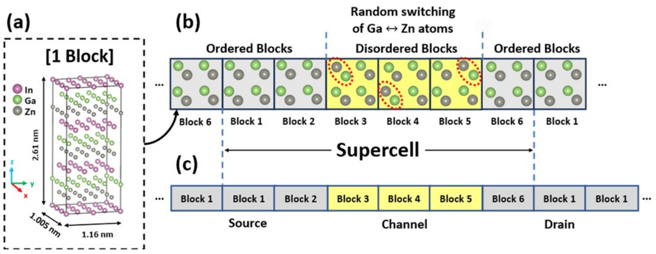

Fig. 2. Example of a DFT supercell (In:Ga:Zn = 1:1:1) with 3 nm channel length. (a) Unit cell, (b) DFT supercell, and (c) structure for transmission calculation. The ordered blocks (grey box) are used as source and drain blocks, while the disordered blocks (yellow box) are used in the channel region. Random switching of cation pairs is illustrated in the red circled region.

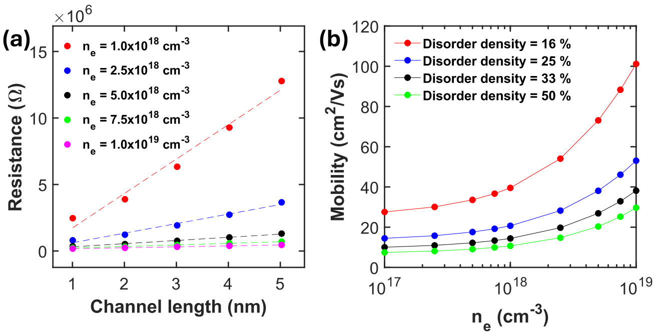

Fig. 3. DFT calculation results for the In:Ga:Zn = 1:1:1 structure. (a) Resistance vs. channel length for electron densities from 10^{18} to 10^{19} cm^{-3} (for 50 % disorder density) and (b) mobility vs. electron density for 16 to 50 % disorder densities. Mobilities are calculated using 10-20 samples per data point.

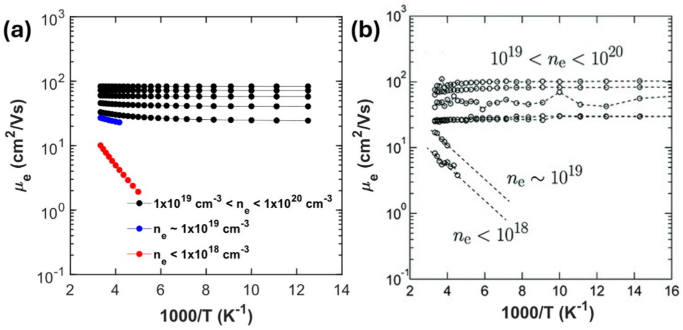

Fig. 4. The dependence of mobility on temperature and carrier density from (a) DFT calculations for 50 % disorder density and (b) experimental results (reprinted with permission from Ref. [10]).

structure of c-IGZO with the target composition is relaxed to obtain the bulk properties. Monkhorst-Pack k-grid is set to 8 × 8 × 2 for the primitive unit cell optimization, and the atomic structures are fully relaxed until the maximum force becomes less than 0.05 eV/Å. IGZO primitive unit cells with compositions of In:Ga:Zn = 1:1:1, 7:5:6, and 4:2:3 belong to the space group $R\bar{3}m$, while the composition In:Ga:Zn = 1:1:2 belongs to the space group $P6_3/mmc$.

After optimization, a unit cell is defined which would later serve as the base building block for the source, channel, and drain regions. This unit cell consists of 3 × 4 × 1 primitive unit cells as illustrated in Fig. 2(a), which is sufficiently large to accommodate various random configurations of cations (In, Ga, and Zn) at the cation sites between the indium layers. The ordered unit cell is defined as the one made from the primitive unit cells with the lowest energy. To generate cation disorder, we employ two different methods: the nearest neighbor switching and complete random distribution method. Starting with the ordered unit cell, disorder unit cells according to the nearest neighbor switching method are generated by switching adjacent cation positions of Zn and Ga atoms until the desired disorder density is reached. For the In rich case, switching between Zn, Ga, and In atoms is also possible. The complete random distribution method involves randomly distributing cations at the cation sites between the indium layers. This method is analogous to the nearest neighbor switching method when the switching rate reaches 50%.

After preparing the ordered and disordered unit cells, the DFT supercell is formed by relaxing a structure that connects ordered unit cells to disordered unit cells as illustrated in Fig. 2(b). For the supercell relaxation, Monkhorst-Pack k-grid is set to be 1 × 4 × 1 or 1 × 4 × 2, and the atomic structures are fully relaxed until the maximum force becomes less than 0.07 ~ 0.12 eV/Å, depending on the supercell size.

B. *Mobility calculation using the DFT Hamiltonians*

The full DFT Hamiltonian of the supercell is extracted and used directly in calculating the transmission using the non-equilibrium Green's function (NEGF) formalism. For the transmission calculation, the DFT supercell is utilized to form a structure with source, channel and drain regions by sequentially linking ordered and disordered blocks up to a total of 8 blocks. The source and drain regions are constructed by repeating Block 1 of the DFT supercell, which is essentially the same as the ordered unit cell, while the channel region is constructed using the disordered blocks as illustrated in Fig. 2(c). This device structure enables the observation of disorder-induced scattering within the channel regions, and the length of the scattering region can be varied by the channel length. Since each block consists of 252 atoms with a Hamiltonian size of 3852 × 3852, the total Hamiltonian size of the device structure can reach up to ~30000×30000. (For the In:Ga:Zn = 1:1:2 case, each block consists of 216 atoms with a Hamiltonian size of 3336 × 3336.) Such massive Hamiltonian sizes require extensive computation time; the transmission calculation would take a minimum of a week per sample. In this work, using the novel heterostructure Hamiltonian reduction method in Ref. [9], the DFT Hamiltonian size is reduced to less than 5 % of its original size without losing accuracy. This approach reduces the NEGF computation time from days to a few minutes, thereby enabling a framework for the statistical analysis of random c-IGZO samples (400 in total). The aforementioned steps are iterated across randomly sampled disordered structures to obtain the average transmission for the

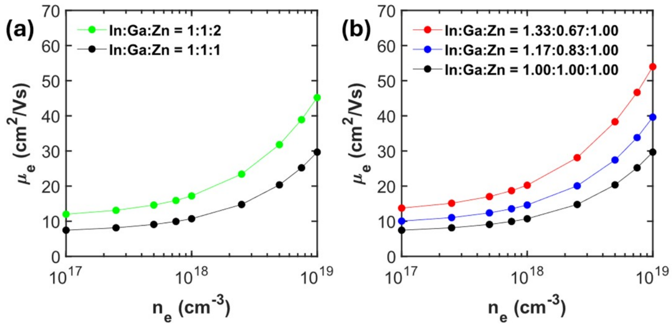

Fig. 5. Calculated DFT mobility values for IGZO with different composition ratios (a) Zn-rich case and (b) In-rich cases.

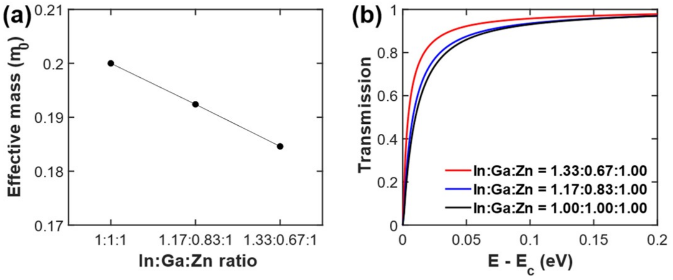

Fig. 6. Bulk properties of In-rich IGZO with increasing indium composition: (a) effective mass and (b) transmission function.

desired disorder density at a specific channel length. Subsequently, the average transmission is calculated for different channel lengths, allowing the calculation of the corresponding resistance R using the Landauer formula:

$$\frac{1}{R} = G = \frac{2e^2}{h}\int < T_L(E) > \left(-\frac{\partial f(E,T)}{\partial E}\right)dE \tag{1}$$

where G is the conductance, E is the energy, T is the temperature, e is the elementary charge, h is the Plank constant, f is the Fermi-Dirac distribution function, and $< T_L(E) >$ is the average transmission for a given channel length L [11]. The cation disorder-limited mobility in then calculated by considering the variation in resistance with respect to channel length using the dR/dL method:

$$\mu = \frac{1}{en\rho} = \left(\frac{dR}{dL}\right)^{-1}\frac{1}{enA} \tag{2}$$

where n is the carrier density, ρ is the bulk resistivity, and A is the channel cross-sectional area [12]. The linear relationship between R and L is crucial to ensure the validity of Eq. 2.

III. Results

We have conducted DFT calculations for the In:Ga:Zn = 1:1:1 ratio IGZO structure for a total of 220 samples; 20 samples each for channel lengths of 1, 2, 3, 4 and 5 nm with cation disorder generated by the complete random distribution method (50 % disorder density) and 10-15 samples each for channel lengths of 1, 2, and 3 nm for 16 %, 25 %, and 33 % disorder densities. Upon performing the sample average, we have examined the linear relationship between the resistance and channel length. IGZO supercell samples with 50 % disorder density and channel lengths ranging from 1 nm to 5 nm are used to calculate the channel resistance. A linear relationship is observed across various carrier densities as shown in Fig. 3(a), confirming the validity of our mobility calculation using the dR/dL method. Then, mobility calculations are performed for disorder densities ranging from 16 to 50 %. The unique characteristic of mobility enhancement with increasing carrier density is successfully captured as shown in Fig. 3(b). Furthermore, the calculated mobilities depending on temperature and carrier density for 50 % disorder density samples are in well agreement with high-quality bulk IGZO single crystal experimental data, showing similar magnitudes as shown between Fig. 4 (a) and (b) [10]. Additionally, a comparison of the total energy of the IGZO samples reveals that those with higher disorder densities exhibit lower energies. This observation indicates that cations between the indium layers favor disorder formation, resulting in a more energetically stable configuration.

Similar DFT calculations have been performed for the Zn-rich and In-rich IGZO structures with cation disorder generated by the complete random distribution method (50 % disorder density), including a total of 60 samples for each case. In the Zn-rich case (In:Ga:Zn = 1:1:2), a slight increase in mobility is observed compared to the In:Ga:Zn = 1:1:1 ratio case as shown in Fig. 5 (a). This suggests reduced Ga-Zn scattering as an additional zinc oxide layer is added to the IGZO structure. For the In-rich cases (In:Ga:Zn = 7:5:6 and 4:2:3), the effective mass decreases and transmission rate increases with the increase in the In ratio as shown in Fig. 6 (a) and (b). This tendency suggests that the increased overlap of In 5s orbitals reduces the effective mass, and the scattering rate between Ga-Zn atoms is higher than that between In-Zn and In-Ga atoms. The reduced scattering further enhances IGZO mobility, resulting in In-rich (In:Ga:Zn = 4:2:3) IGZO having the highest mobility among the studied cases as illustrated in Fig. 5 (b).

IV. Conclusion

In summary, we have evaluated the electrical properties of crystalline IGZO with various composition ratios using the DFT method. We were able to observe the mobility enhancement with increasing temperature and carrier density which are in well agreement with experimental results. This suggests that the proposed DFT-based mobility calculation method, which does not require any fitting parameters, successfully captures the cation disorder scattering effect in IGZO. Furthermore, we have found that the effective mass decreases and transmission probability increases under In-rich conditions. Mobility is also improved when additional zinc oxide layers are added to the IGZO structure. Our methodology can further be expanded to identify optimal IGZO compositions with superior electrical properties, and it can also be readily applied to simulate IGZO channel transistors.

ACKNOWLEDGMENT

This work was a result of the research project supported by SK hynix Inc.

REFERENCES

[1] M. Oota, Y. Ando, K. Tsuda, T. Koshida, S. Oshita, A. Suzuki, K. Fukushima, S. Nagatsuka, T. Onuki, R. Hodo, T. Ikeda, and S. Yamazaki, "3D-Stacked CAAC-In-Ga-Zn Oxide FETs with Gate Length of 72nm," 2019 IEEE International Electron Devices Meeting (IEDM), San Francisco, CA, USA, 2019, pp. 3.2.1-3.2.4,

[2] A. Belmonte, H. Oh, N. Rassoul, G. L. Donadio, J. Mitard, H. Dekkers, R. Delhougne, S. Subhechha, A. Chasin, M. J. van Setten, L. Kljucar, M. Mao, H. Puliyalil, M. Pak, L. Teugels, D. Tsvetanova, K. Banerjee, L. Souriau, Z. Tokei, L. Goux, and G. S. Kar, "Capacitor-less, Long-Retention (>400s) DRAM Cell Paving the Way towards Low-Power and High-Density Monolithic 3D DRAM," 2020 IEEE International Electron Devices Meeting (IEDM), San Francisco, CA, USA, 2020, pp. 28.2.1-28.2.4

[3] N. Kimizuka and S. Yamazaki, Eds., Physics and Technology of Crystalline Oxide Semiconductor CAAC-IGZO: Fundamentals, Chapter 3, pp. 190-200. Hoboken, NJ, USA: John Wiley & Sons, 2017

[4] Y. Jegal, A. H. Kulahlioglu, C. -K. Baek and B. D. Kong, "Disorder Originated Unusual Mobility in Crystalline InGaZnO4," in IEEE Electron Device Letters, vol. 41, no. 6, pp. 872- 875, 2020

[5] Y. Kang, Y. Cho, and S. Han, "Cation disorder as the major electron scattering source in crystalline InGaZnO," Applied Physics Letters, vol. 102, no. 15, pp. 152104, 2013.

[6] J. M. Soler, E. Artacho, J. D. Gale, A. García, J. Junquera, P. Ordejón, and D. Sánchez-Portal, "The SIESTA method for ab initio order-N materials simulation," Journal of Physics-Condensed Matter, vol. 14, no. 11, pp. 2745-2779, 2002.

[7] A. García, M. J. Verstraete, Y. Pouillon, and J. Junquera, "The psml format and library for norm-conserving pseudopotential data curation and interoperability," Computer Physics Communications, vol. 227, pp. 51-71, 2018.

[8] J. P. Perdew, K. Burke, and M. Ernzerhof, "Generalized Gradient Approximation Made Simple," Physical Review Letters, vol. 77, no. 19, pp. 3865-3868, 1996.

[9] M. Shin, W. J. Jeong, and J. Lee, "Density functional theory based simulations of silicon nanowire field effect transistors," Journal of Applied Physics, vol. 119, no. 15, pp. 154505, 2016.

[10] Y. Tanaka, K. Wada, Y. Kobayashi, T. Fujii, S. J. Denholme, R. Sekine, N. Kase, N. Kimizuka, and N. Miyakawa, "Single crystal growth of bulk InGaZnO4 and analysis of its intrinsic transport properties," CrystEngComm, vol. 21, no. 19, pp. 2985-2993, 2019.

[11] Markussen, T., Palsgaard, M., Stradi, D., Gunst, T., Brandbyge, M., and Stokbro, K., “Electron-phonon scattering from Green’s function transport combined with molecular dynamics: Applications to mobility predictions,” Physical Review B, vol. 95, 245210, 2017

[12] K. Rim, S. Narasimha, M. Longstreet, A. Mocuta and J. Cai, "Low field mobility characteristics of sub-100 nm unstrained and strained Si MOSFETs," Digest. International Electron Devices Meeting, San Francisco, CA, USA, 2002, pp. 43-46