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Highly-Efficient Advanced Thermoelectric Devices from Different Multilayer Thin Films

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Abstract: The efficiency of the thermoelectric materials and devices is shown by the dimensionless Figure of merit, ZT. ZT is calculated by multiplying the Seebeck coefficient with square of the electrical conductivity and absolute temperature and dividing it all by the thermal conductivity. Thermoelectric devices were prepared using different multilayered thin film structures in the order of SiO_2/SiO_2 + Ge/Ge/Sb + Ge/Si/Si + Ge/Ge/Ge + Si by DC/RF Magnetron Sputtering. The prepared thermoelectric devices have been tailored with 5 MeV Si ions bombardment at the different fluences (doses) to form quantum structures in the multilayer thin films to improve the efficiency of the thermoelectric devices. Seebeck coefficients, van der Pauw-four probe resistivity, Hall Effect coefficient, density and mobility have been measured. After the samples were prepared, SEM/EDS data were collected. FIB/SEM images were provided to figure out the cross-section of the fabricated devices. Seebeck coefficients and electrical resistivity results were affected positively if the appropriate ion beam dose was selected.

Keywords: Thermoelectric Materials, Figure of Merit, Seebeck Coefficient, High Energy Ion Beam Bombardment

Introduction

Thermoelectric materials could convert the heat energy to electrical energy directly (Li *et al*., 2016). Thermoelectric devices took an important role due to their applications in our life in many areas. Thermoelectric materials were founded in the 19th century. In 1821, Thomas Seebeck performed the first discovery on the thermoelectric materials while he was watching the deviation of the needle placed in the circuit formed by two unlike metal conductors. Seebeck found that the deflection of the needle was proportionally affected by the change of the temperature. He found that the temperature change across the unlike metals produced an electric current.

In 1834, Jean Peltier found that the current flow resulted from the heat flow through the unlike metals caused the absorbed or discharged at the closed electrical circuit. The foundations related to thermoelectrics are big advancements for today's life. The effect through junction between unlike metals due to heat flow is called as Seebeck coefficient (Graham and Lindsay, 2014; Ratliff and Bronwyn, 2014). In other words, Seebeck coefficient could be defined as the induced thermoelectric voltage for the response to the temperature change across the studied materials (Ratliff and Bronwyn, 2014).

Until 20th century, there were not many researches performed on the thermoelectrics. Edmund Altenkirch described the fundamentals of the materials to make practical thermoelectric devices and carry out the efficiency of the thermoelectric generators (Budak *et al*., 2015a; 2013). He found that the high quality thermoelectrics devices should have high Seebeck coefficient and high electrical conductivity and low thermal conductivity.

Thermoelectric materials and devices have the dimensionless efficiency described as Figure of merit, ZT. ZT could be calculated by $ZT = S^2\sigma T/\kappa$, where S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature in Kelvin (K) and κ is the thermal conductivity (Budak *et al*., 2009; Riffat and Ma, 2003; Meroz *et al*., 2016). In this study, the progress on the fabrication of the thin films from the different multilayers and the effects of the high energy ion beam bombardments on the thermoelectrical properties of the thin films were reported.

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Experimental Program

Figure 1 shows the used geometry during the deposition of the different multilayer thin films from SiO_2/SiO_2 + Ge, Ge, Sb + Ge, Si, Si + Ge, Ge/Ge + Si materials. To perform the fabrication of the thermoelectric devices, the SiO_2 substrates were cut into smaller pieces. Then the cut-pieces were put on the substrate holder, attaching them with double-side-carbon tape. Figure 2 shows the prepared samples on the sputtering machine's substrate holder before multilayer deposition were performed.

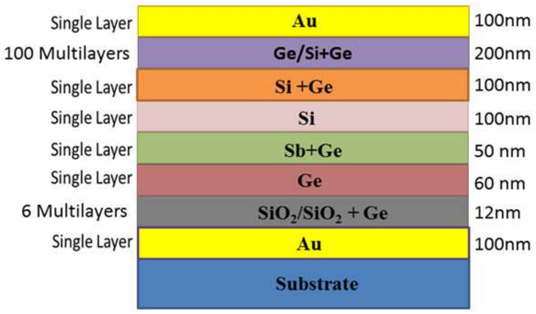

Fig. 1. Used geometry for the multilayer deposition

Fig. 2. Samples on the substrate holder of the sputtering machine for deposition

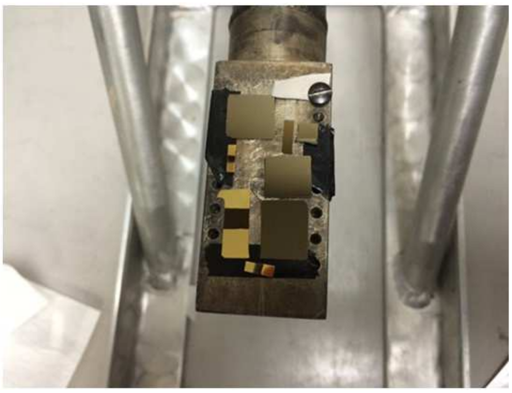

Fig. 3. Samples on the ion bombardment stage

Then, the samples were loaded into the DC/RF Magnetron sputtering machine for the beginning of the multilayer thin film deposition. The multilayer thermoelectric layers are ordered as SiO_2/SiO_2 + Ge, Ge, Sb + Ge, Si, Si + Ge, Ge/Ge + Si as seen from Fig. 1 for thermoelectric device fabrication.

The fabricated thin film thermoelectric devices have been bombarded with 5 MeV Si ions at the different fluences (doses) to form quantum structures in the multilayers to improve the efficiency of the fabricated thermoelectric devices by Pelletron Accelerator. Figure 3 shows the loading of the samples on the bombardment stage.

Results

The Seebeck coefficients have been measured for the prepared devices from the different multilayer thin films. Figure 4 shows the Seebeck coefficients of the thin films at the different fluences under the different temperatures.

After obtaining Seebeck measurements, van der Pauw-four probe resistivity, Hall Effect coefficient, density and mobility measurements have been carried out.

After the thermoelectric devices were prepared, SEM+EDS results were collected. Scanning Electron Microscopy (SEM) scans a focused electron beam on the surface to produce an image. It interacts with the materials where it produces various signals. Energy-Dispersive X-ray Spectroscopy (EDS) when combined with SEM, it produces X-ray for the analysis of the elements present on the fabricated thin film samples. EDS analysis could be used both to figure out the elemental composition of individual points and to map the lateral distribution of elements.

Discussion

As seen from Fig. 4, appropriate ion beam bombardment could cause an increment in Seebeck coefficients. This might come from the increase in the charge carrier concentrations in the atomic level. High Seebeck coefficient is one of the expected values from the high-efficient thermoelectric devices. Highly-efficient thermoelectric devices should have high Seebeck coefficients. As seen from Fig. 5 and 6, the resistivity values decreases at the appropriate used ion beam fluences (doses) during the ion bombardment. The decrease in the resistivity means increase in the electrical conductivity. High electrical conductivity is one of the important expected values of the highly-efficient thermoelectric devices. Figure 7 shows van der Pauw density measurement of multilayered thin films. As seen from Fig. 7, the density values take the values between negative and positive values depending on the applied ion beam bombardment. The increase in the charge carrier concentration could cause the material systems to behave like n-type or p-type semiconductor materials. Figure 8 shows van der Pauw Hall coefficient measurements of the multilayered thin films. Figure 9 shows van der Pauw mobility measurements of the multilayered thin films. Depending on the applied ion beam bombardment, both Hall Effect coefficients and mobility value could change since charge carrier concentration changes as discussed in density. Figure 10-12 show the SEM images with EDS and elemental mapping on the used multilayered thin film system when it is not bombarded. Figure 13-16 show Focus Ion Beam milled thin film systems with SEM micrograph from the cross-sections on the unbombarded and bombarded at the fluence of 5×10^{12} ions/cm^2. Figure 17 shows the ion beam bombardment modeling using SRIM (Ziegler *et al*., 1985) simulation before the real ion beam bombardment on the thermoelectric thin films. SRIM simulation aids in determining how much ion beam bombardment should be used for the certain type of the materials at the deposited thickness.

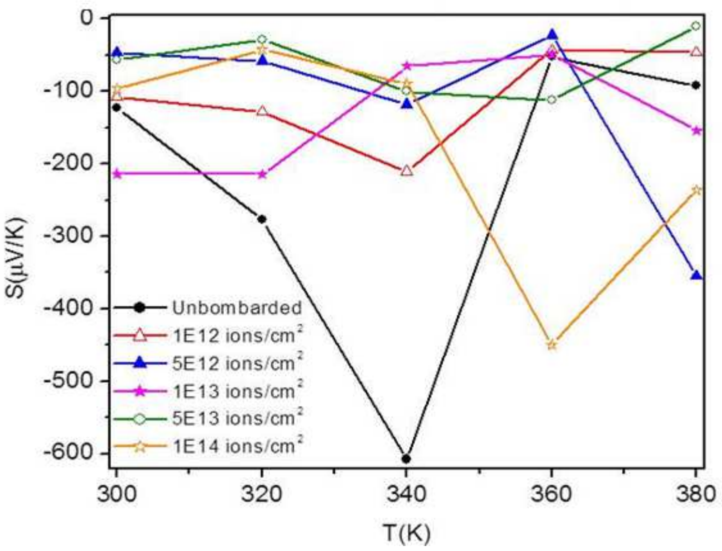

Fig. 4. Seebeck coefficients of multilayered thin films

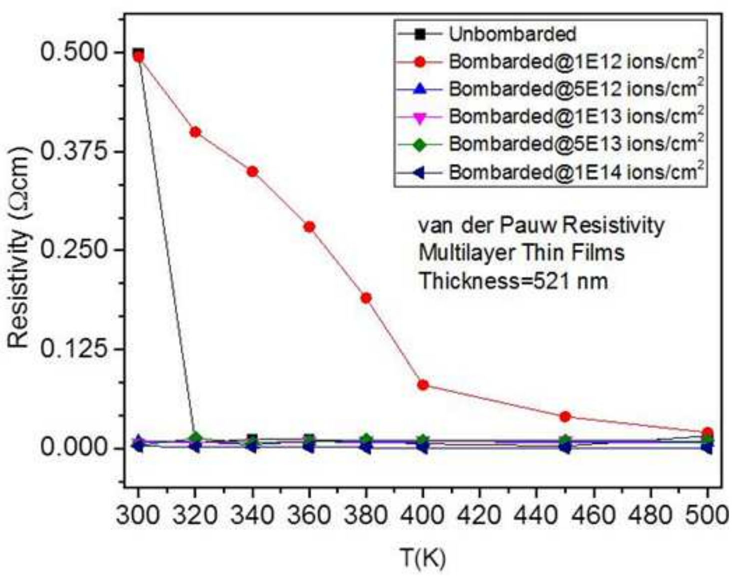

Fig. 5. Temperature dependence of van der Pauw resistivity measurements of multilayered thin films bombarded at different fluencies

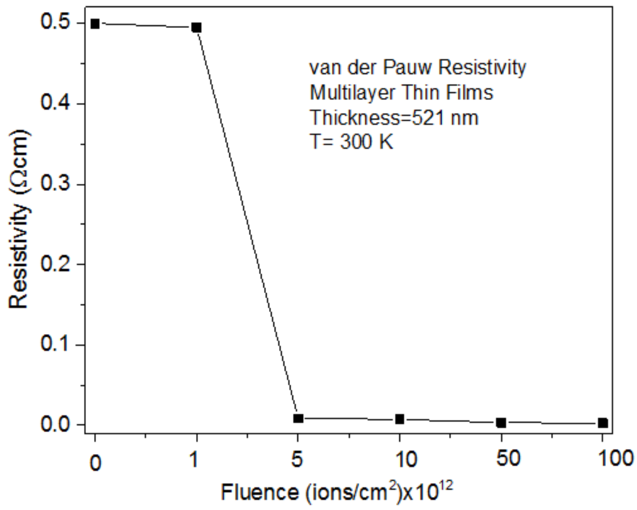

Fig. 6. Fluence dependence of van der Pauw resistivity measurements of multilayered thin films at room temperature

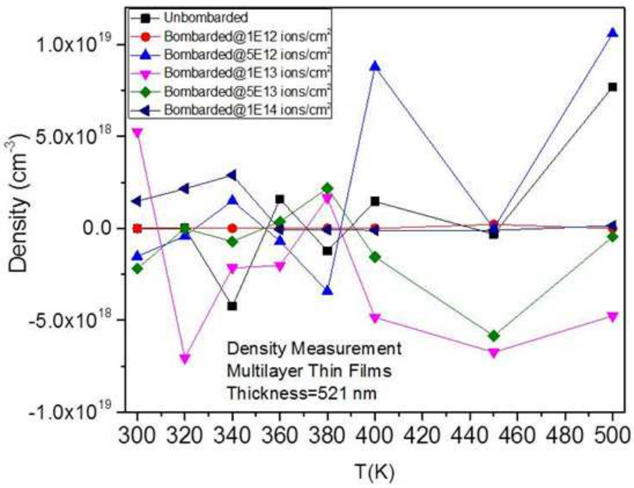

Fig. 7. Van der Pauw density measurement of multilayered thin films

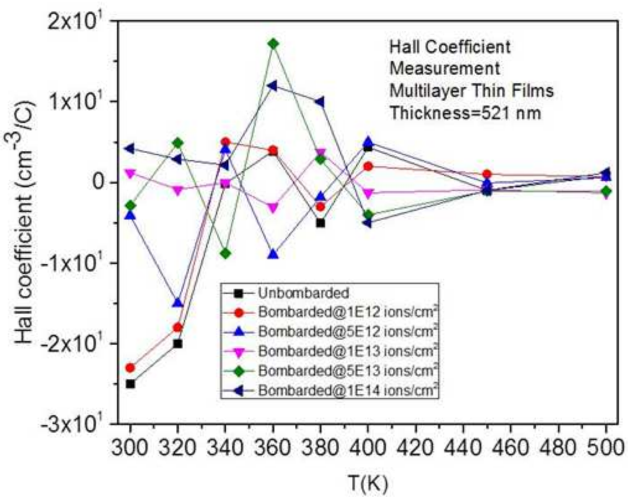

Fig. 8. Van der Pauw Hall coefficient measurement of multilayered thin films

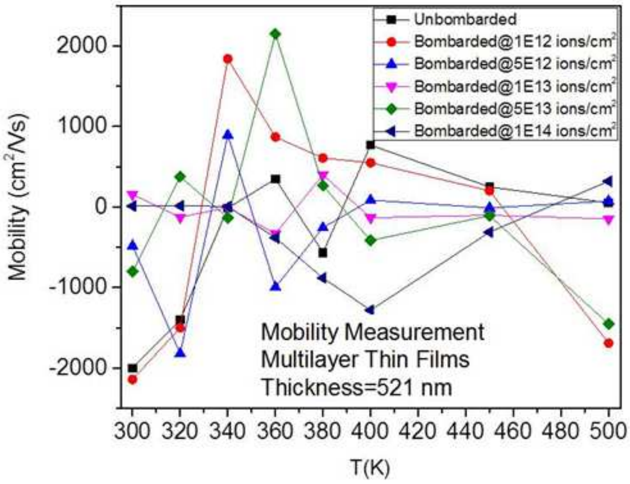

Fig. 9. Van der Pauw mobility measurements of multilayered thin films

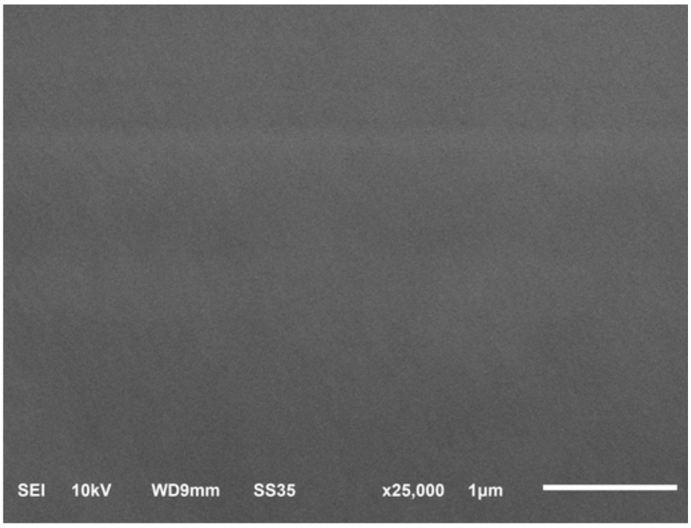

Fig. 10. SEM micrograph of the fabricated device

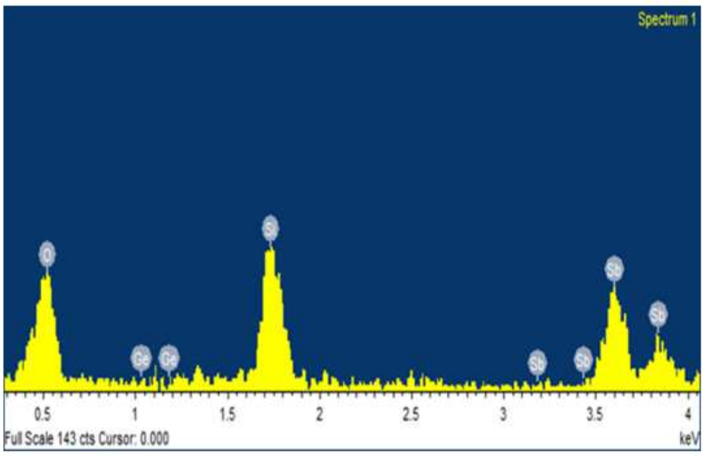

Fig. 11. Energy-dispersive X-ray spectroscopy (EDS)

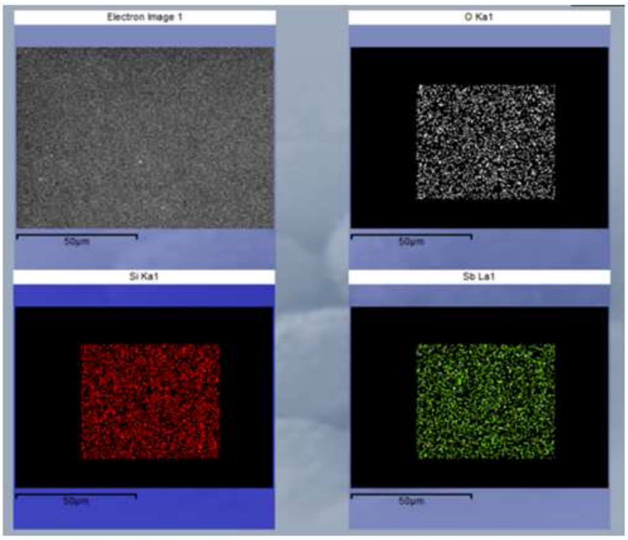

Fig. 12. Elemental mapping of used materials at the surface of the fabricated devices

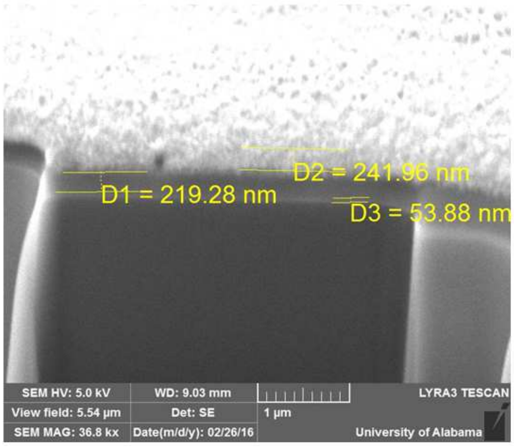

Fig. 13. FIB milled SEM cross-section image when geometric transformations is on by UA support on unbombarded multilayered thin film

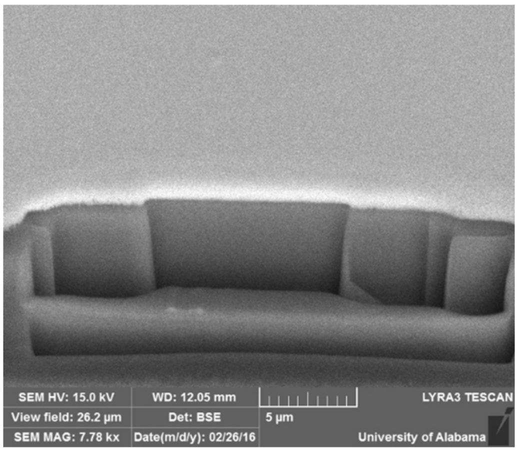

Fig. 14. FIB milled SEM cross-section image by UA support on unbombarded multilayered thin film

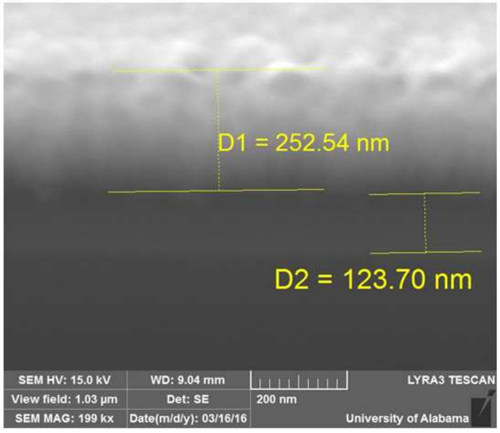

Fig. 15. FIB milled SEM cross-section image when geometric transformations is on by UA support on bombarded multilayered thin film at the fluence of 1E12 ions/cm^2

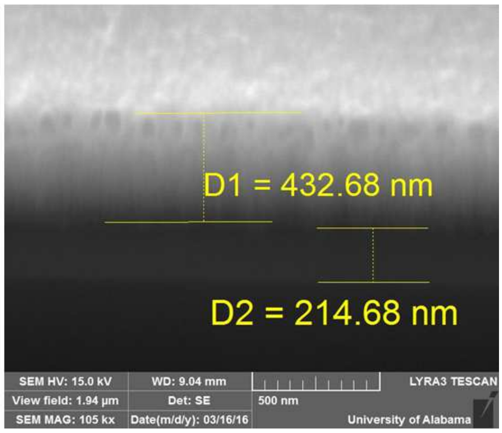

Fig. 16. FIB milled SEM cross-section image when geometric transformations is off by UA support on bombarded multilayered thin film at the fluence of 1E12 ions/cm^2

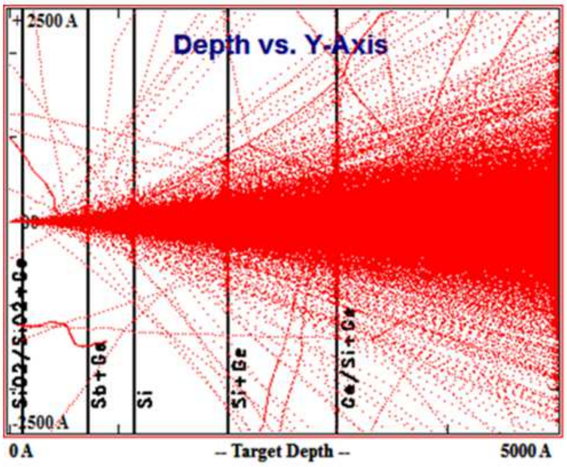

Fig. 17. Ion beam bombardment simulation by SRIM software (http://www.srim.org/)

Conclusion

The increase of the electronic density of states in the mini-band of the quantum structures due to ion beam bombardment could increase the electrical conductivity and Seebeck coefficient. When the virgin sample (unbombarded sample) is bombarded with 5-MeV Si ions at the appropriate fluence (doses), the numbers of charge carriers in both the conduction and valence bands could increase. This increase could cause a narrower energy-gap between the conduction and valence bands, which could cause the electrical conductivity to increase (Budak *et al*., 2015b). FIB milled SEM images show that the thickness measurement system used during the characterization is compatible with the result of cross-section images of FIB milled SEM images. The studied multilayered thin film systems show meaningful results from Seebeck coefficients and electrical resistivity results. Main problem for the thermoelectric devices is that excess of heat during the operation of the devices. High Seebeck coefficient shows that high amount of waste heat could be converted into electric energy. To calculate the Figure of merit for the studied thermoelectric devices, the third item needs to be studied is the thermal conductivity. For the future work, the thermal conductivity of thermoelectric material systems will be studied using Laser PIT thermal conductivity system located in the clean room.

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Author's Contributions

Each author of this manuscript made considerable contributions in conducting the experimental testing, data-analysis and contributed to the writing of this manuscript.

Ethics

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