Electronic Supplementary Item for: First Principle Study of the optical and thermoelectric properties of tetragonal-silicene

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Part-I

1   Validation of DZP basis

We have made a detailed analysis for the applicability of different functional and basis sets for our study by applying our method to known materials. The application of different basis sets had already been applied to bulk-silicon to calculate its lattice parameter, Bulk modulus, cohesive energy and compared with experimental values [1,2]. Results suggest that the DZP basis set provides a good balance between converged high-quality results and computational cost for silicon. In the next two subsections, we provide a comparison of the optical properties of bulk-silicon and silicene with the previous experimental and theoretical findings.

1.1   Bulk-silicon

We have calculated the optical properties like absorption coefficient (α), reflectance ($R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}$), extinction coefficient (k) of bulk silicon (figure 1, 2 and 3 respectively) and demonstrated the accuracy of our theoretical approach by comparing it with the experimental findings. We have considered single ζ- polarized (SZP), triple ζ- polarized (TZP), double ζ- polarized (DZP) basis set with exchange-correlation functional of Perdew-Burke-Ernzerhof (PBE) of GGA along with Perdew-Zunger (PZ)/DZP level of theory. We have also plotted the experimental results of the optical properties of bulk silicon at 300K [3,4]. The results indicate that optical properties by PBE/DZP and PBE/TZP completely match each other and are in good agreement with the experiments in the energy range 1.5 $-$ 4.5 eV. Experimental data above 4.5 eV is not available in the literature. The reflectance and absorption of bulk silicon are also at par with the previous theoretical prediction [4]. Thus for bulk silicon, the choice of PBE/DZP level of theory is good enough and provides consistent results.
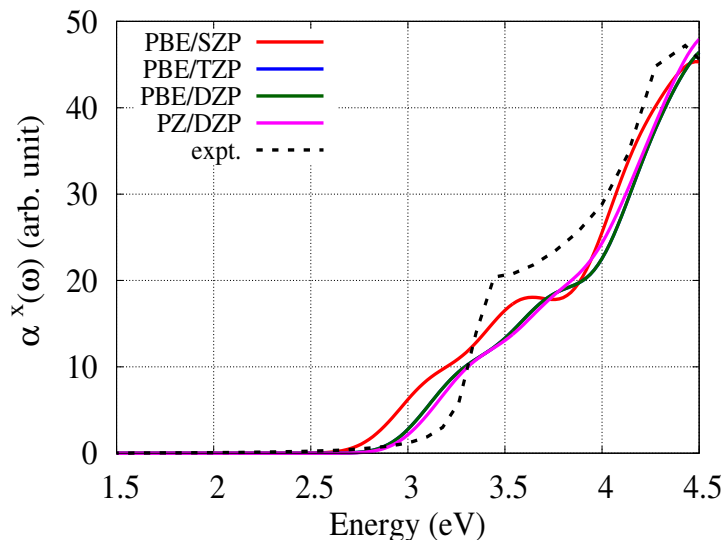

Figure 1: Absorption Coefficient α calculated for polarization along $X-$ axis for bulk-silicon by DFT method and compared with the experimental results [3].

Part-II

1.2   Silicene

We have now extended the calculation of optical properties using the same level of theory for silicene. Here we have calculated the real and imaginary part of dielectric function (ϵ_1 and ϵ_2, respectively), real part of optical conductivity (σ_1), absorption coefficient (α) (figure 4a,4b, 4c, 4d respectively) of free-standing silicene, and compared the results with the available literature. Though the direct comparison of any properties of free-standing silicene cannot be carried out with the experiments, as silicene has only been synthesized in substrates that modify its properties
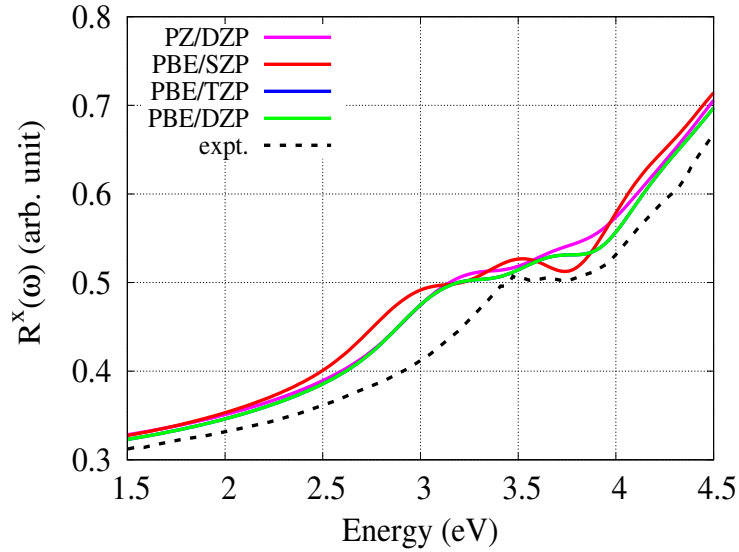
Figure 2: Reflectance R calculated for polarization along $X-$ axis for bulk-silicon by DFT method and compared with the experimental results [3, 4].
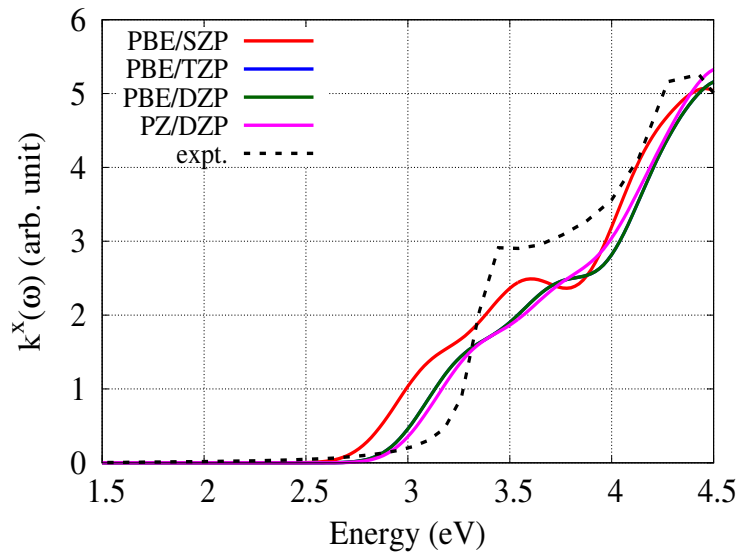

Figure 3: Extinction Coefficient k calculated for polarization along $X-$ axis for bulk-silicon by DFT method and compared with the experimental results [3].
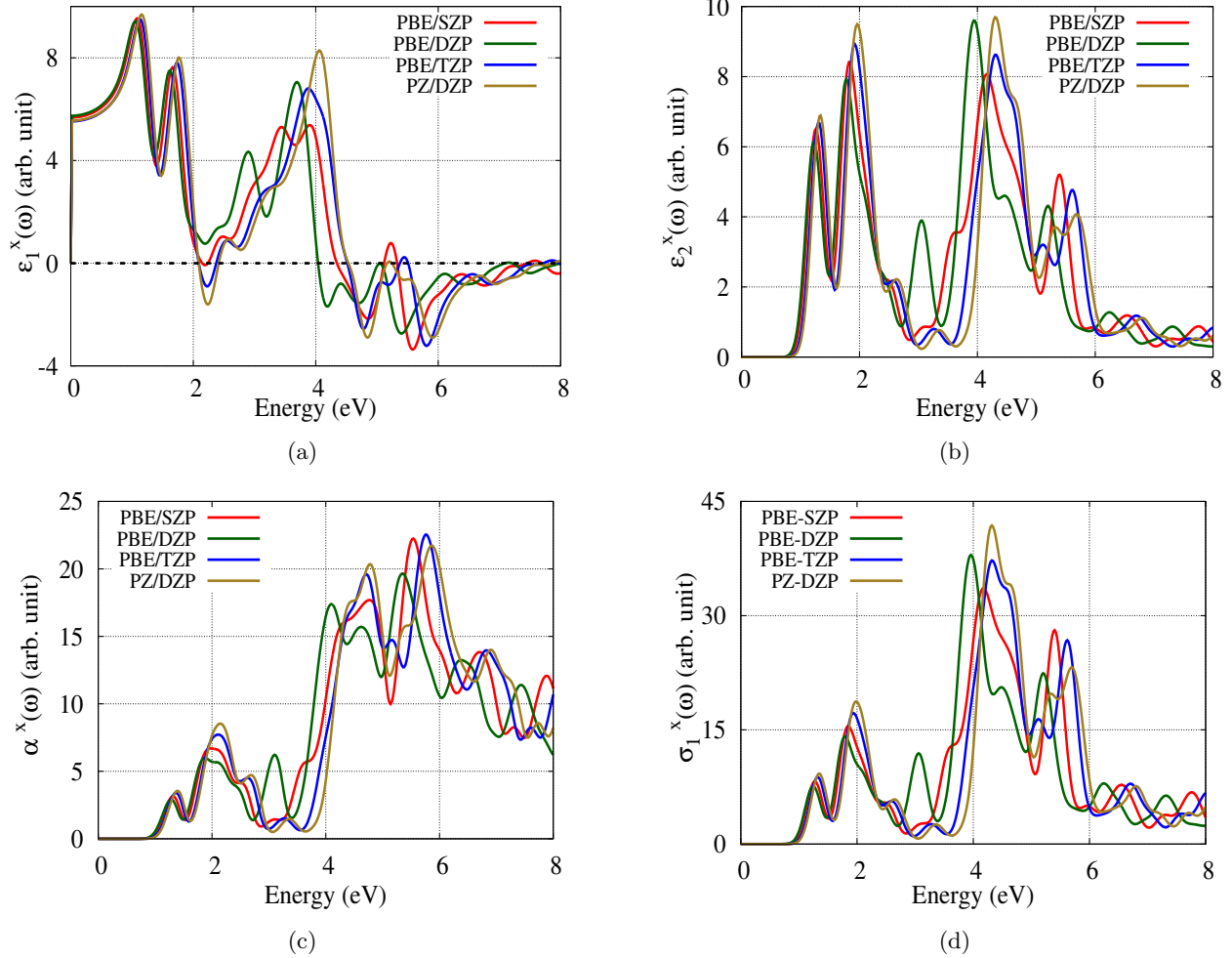
Figure 4: (a)Real part of dielectric function (b) Imaginary part of dielectric function (c) absorption coefficient (d) Real part of optical conductivity for silicene calculated by different level of theory for parallel polarization.

extensively. We, therefore, compared our results with the established theoretical results of free-standing silicene. Our results of optical properties on free-standing silicene are in good agreement with the earlier studies. For example, the real part of the dielectric function matches well with the results of John and Merlin [5], while the real part of optical conductivity shows similar behavior with that calculated by the DFT-independent particle method and GW-BSE method [6]. The imaginary part of the dielectric constant and absorption coefficient are in agreement with the results of Ref. [7]. We have also compared the values of the static part of the dielectric constant, plasma frequencies and peaks in absorption coefficients and $\epsilon_2(\omega)$ of silicene by different level of DFT theory with those reported earlier theoretically and is tabulated in table 1. Results indicate that the PBE/DZP level of theory provides sensible results with the optical properties of silicene.

Thus it can be concluded that DZP basis with PBE functional of GGA exchange-correlation provides a good agreement with the results of bulk-silicon and silicene, and hence can be applied to our silicon-based structure T-silicene.

Table 1: Comparison of few optical parameters for parallel polarization calculated by various level of theory by DFT with previous findings

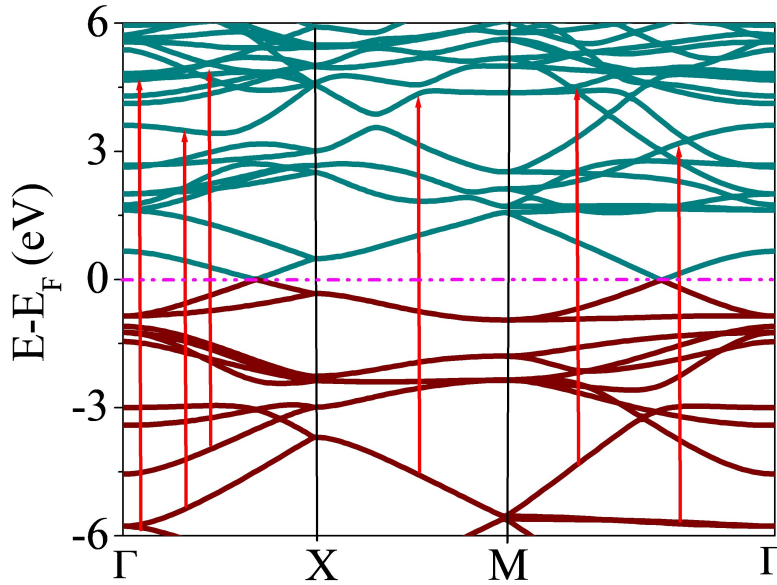| Parameters	PBE-SZP	PBE-DZP	PBE-TZP	PZ-DZP	Previous results
Static part of Dielectric function	5.6774	5.7428	5.5278	5.5524	5.43 [5], 5.41 [8]
Plasma Frequency (eV)	2.23, 5.12	5.04	2.38, 5.38	2.42, 5.18	4.86 [5]
Peaks position in $\alpha(\omega)$ (eV)	1.32, 4.76, 5.54	1.87, 4.62, 5.34	2.11, 4.72, 5.76	1.40, 4.78, 5.86	1.81, 4.35 [7]
Peaks position in $\epsilon_2(\omega)$(eV)	1.26, 1.84, 4.16	1.22, 1.78, 3.94	1.30, 1.92, 4.30	1.34,1.96, 4.30	0.41, 1.55, 3.89 [7]; 0.12, 1.60, 3.91 [9];

Figure 5: Few probable transition for the peak at ω=8.88eV in imaginary part of dielectric function $\epsilon_2(\omega)$ for perpendicular polarization.
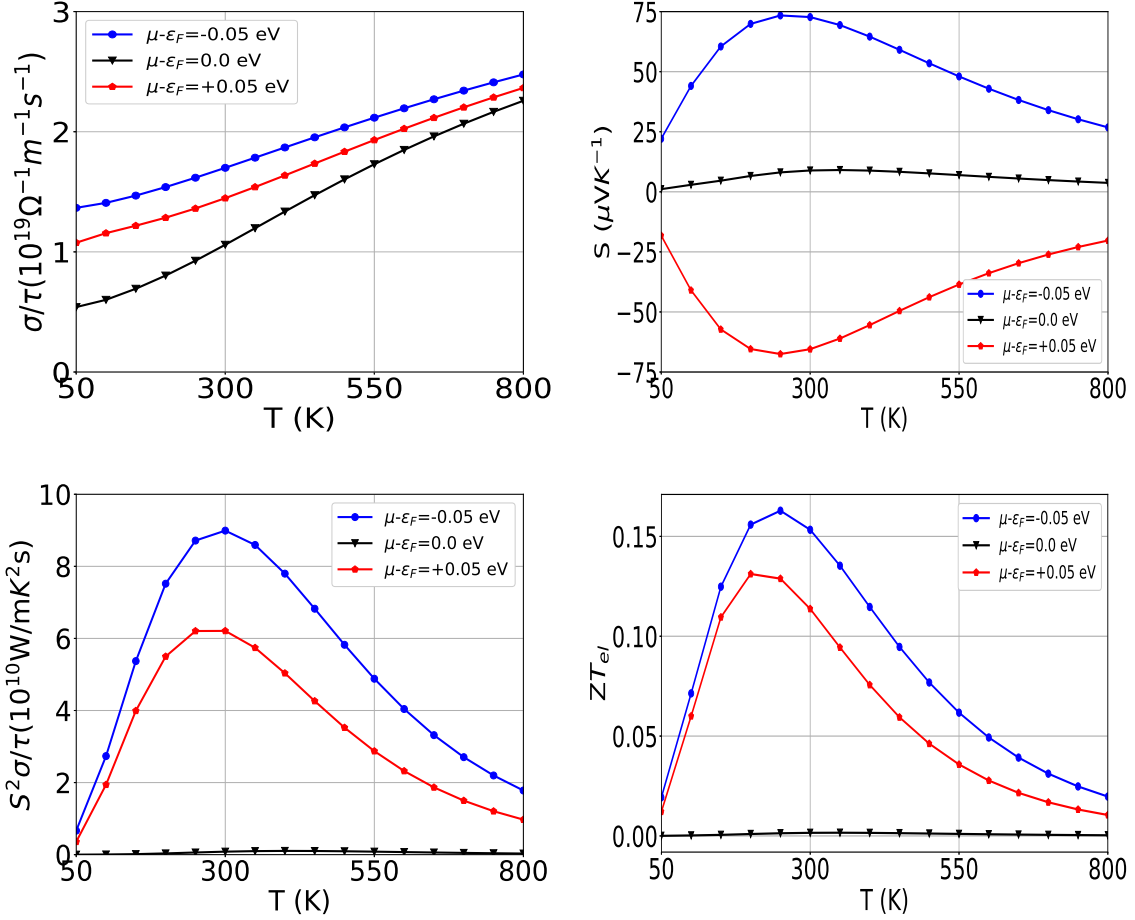
Figure 6: Variation of the (a) electrical conductivity, (b) Seebeck coefficient, (c) Power factor, (d) Electronic figure of merit of TS nanosheet with respect to temperature. We have taken the pristine system $\mu - \varepsilon_f = 0$ and two arbitrary doping $\mu - \varepsilon_f = \pm 0.05$
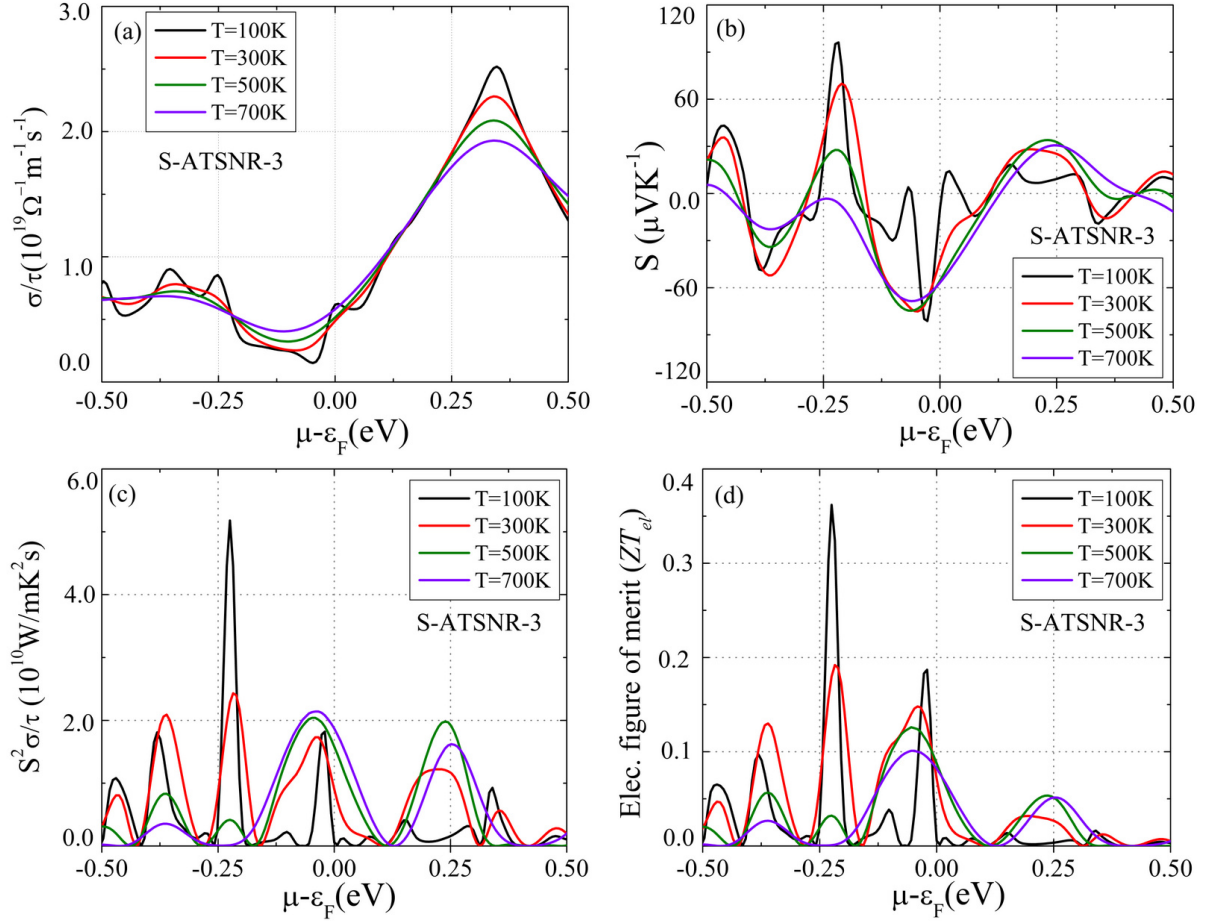
Figure 7: Variation of the (a) electrical conductivity, (b) Seebeck coefficient, (c) Power factor, (d) Electronic figure of merit of S-ATSNR (N=3) with respect to the chemical potential.
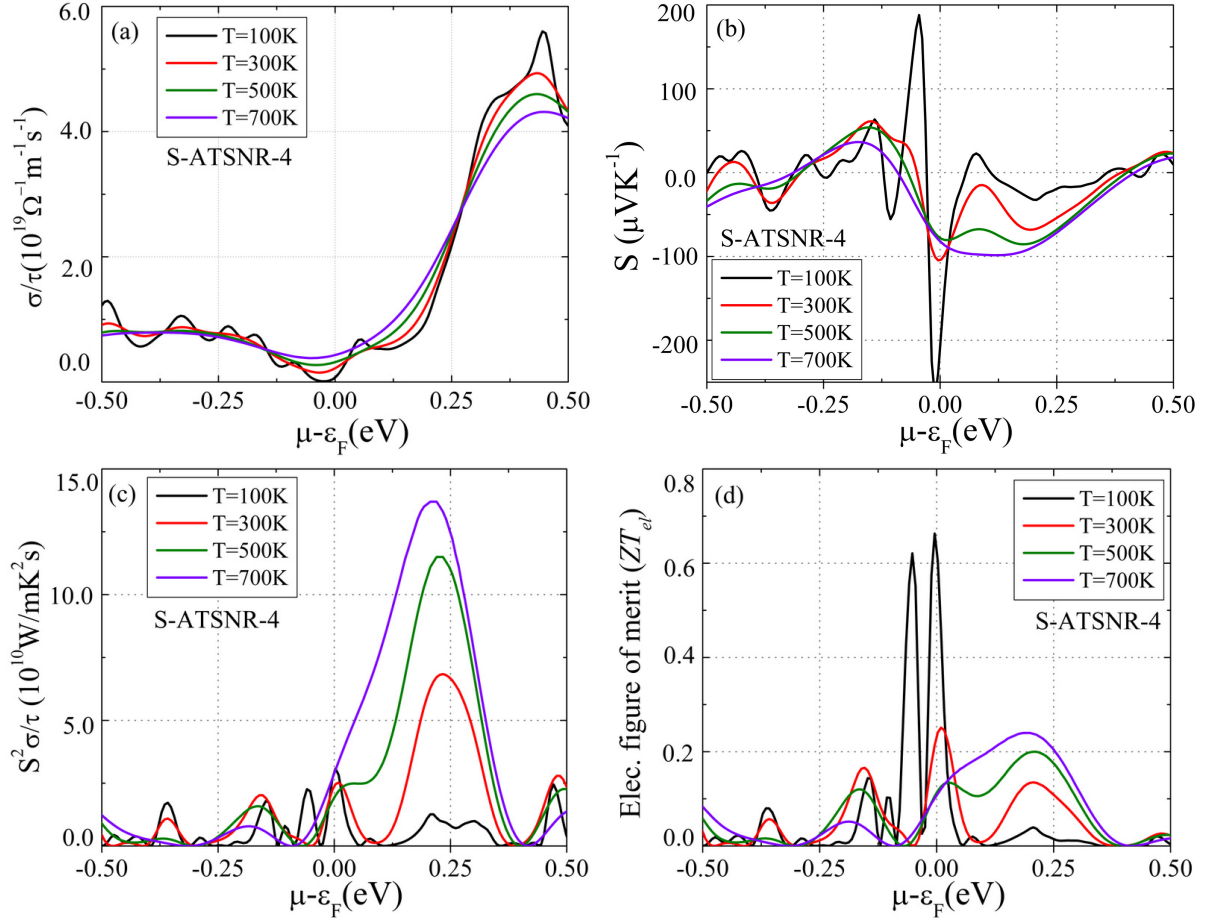
Figure 8: Variation of the (a) electrical conductivity, (b) Seebeck coefficient, (c) Power factor, (d) Electronic figure of merit of S-ATSNR (N=4) with respect to the chemical potential.
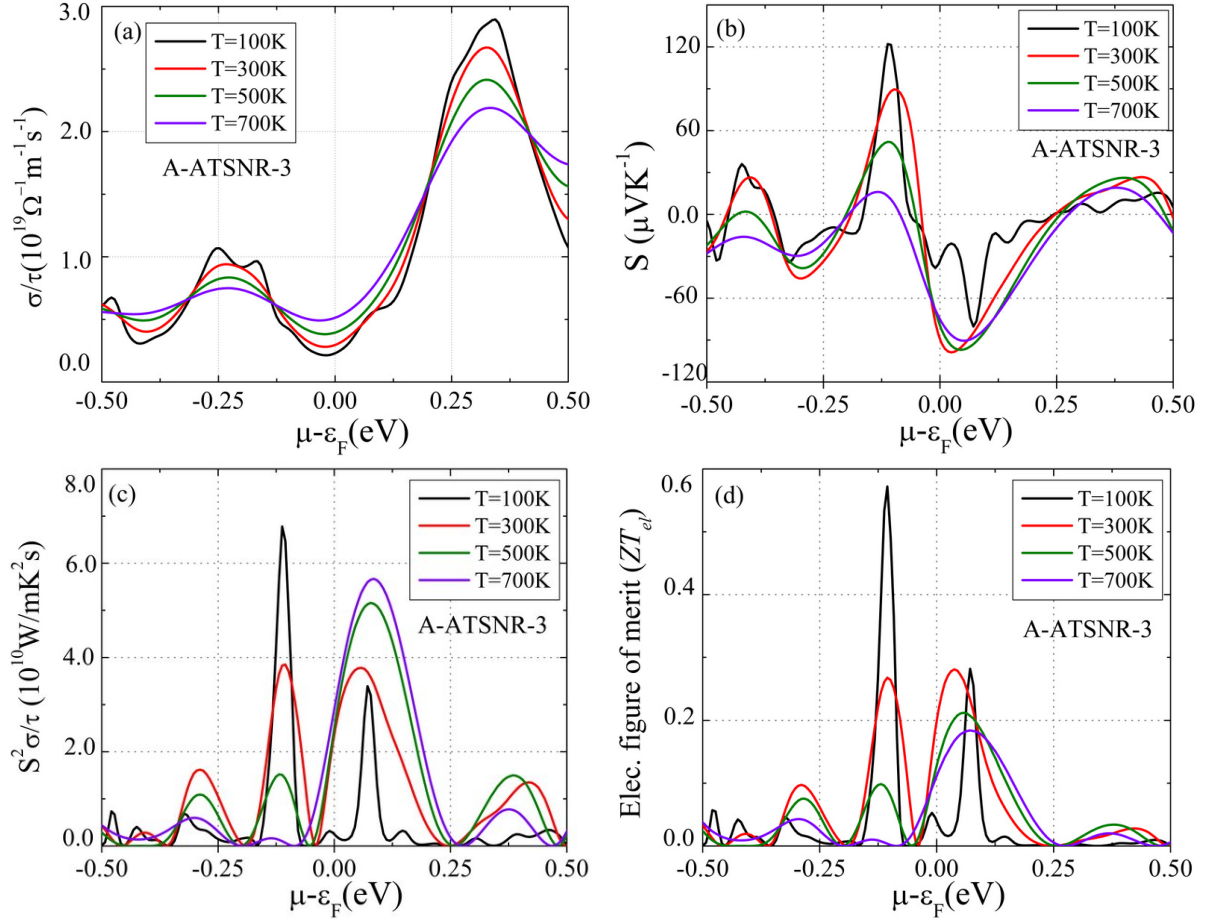
Figure 9: Variation of the (a) electrical conductivity, (b) Seebeck coefficient, (c) Power factor, (d) Electronic figure of merit of A-ATSNR (N=3) with respect to the chemical potential.
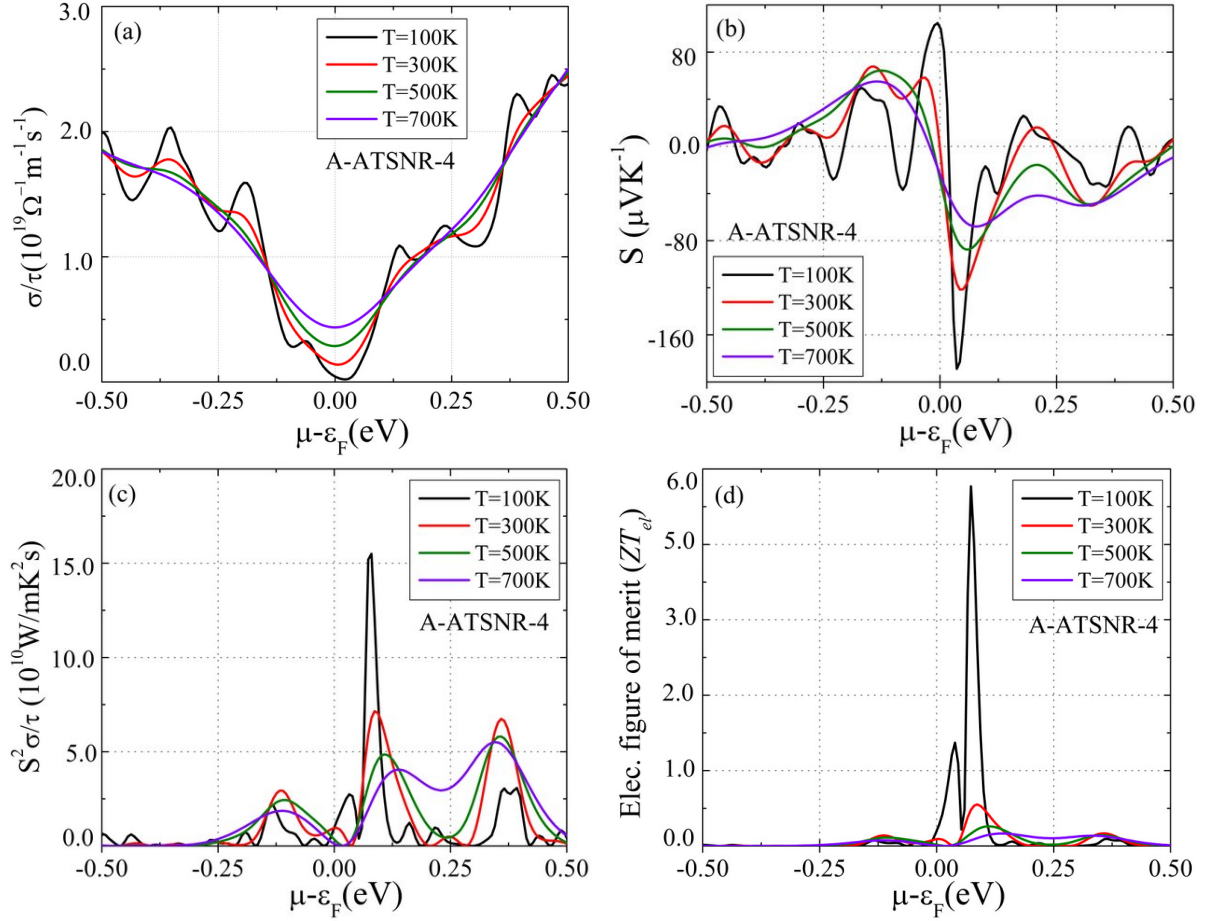
Figure 10: Variation of the (a) electrical conductivity, (b) Seebeck coefficient, (c) Power factor, (d) Electronic figure of merit of A-ATSNR (N=4) with respect to the chemical potential.
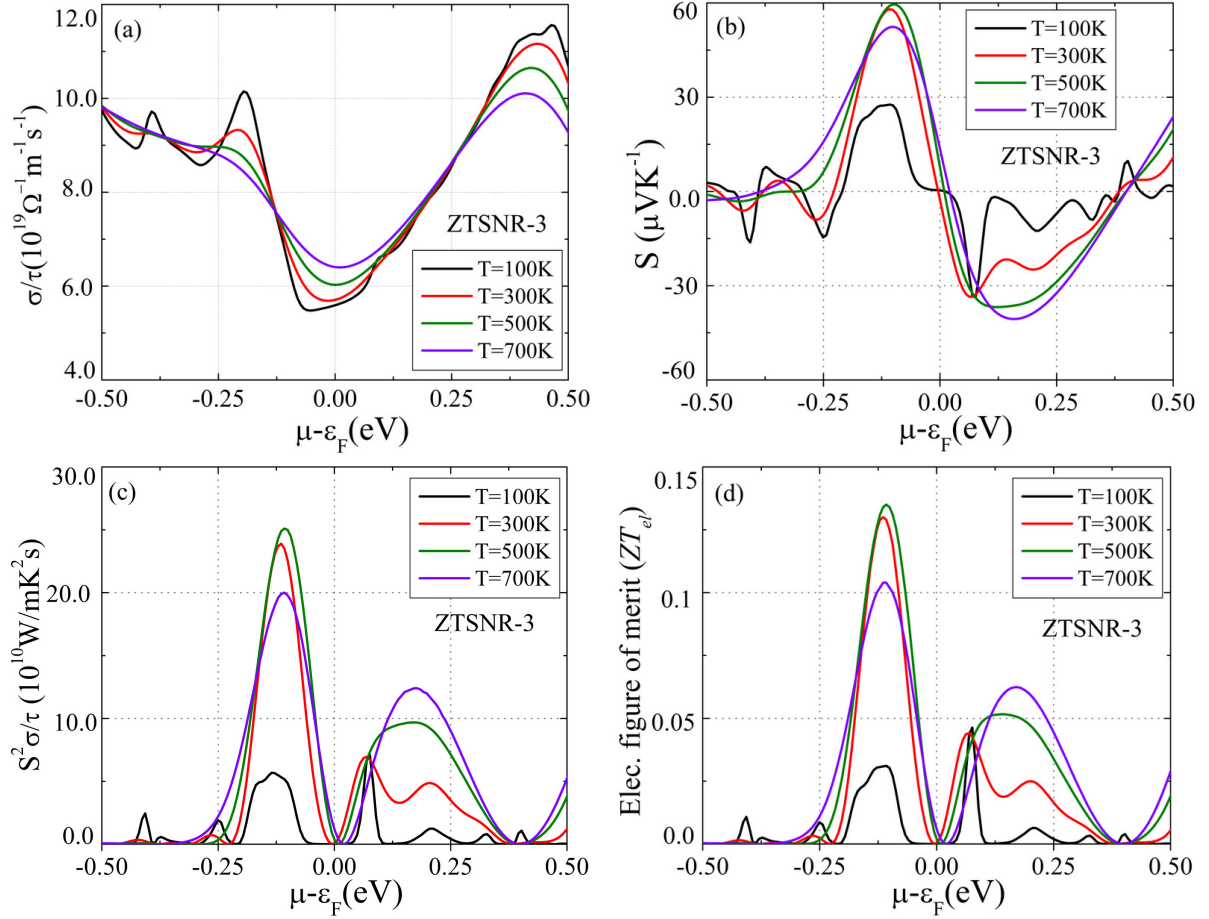
Figure 11: Variation of the (a) electrical conductivity, (b) Seebeck coefficient, (c) Power factor, (d) Electronic figure of merit of ZTSNR (N=3) with respect to the chemical potential.
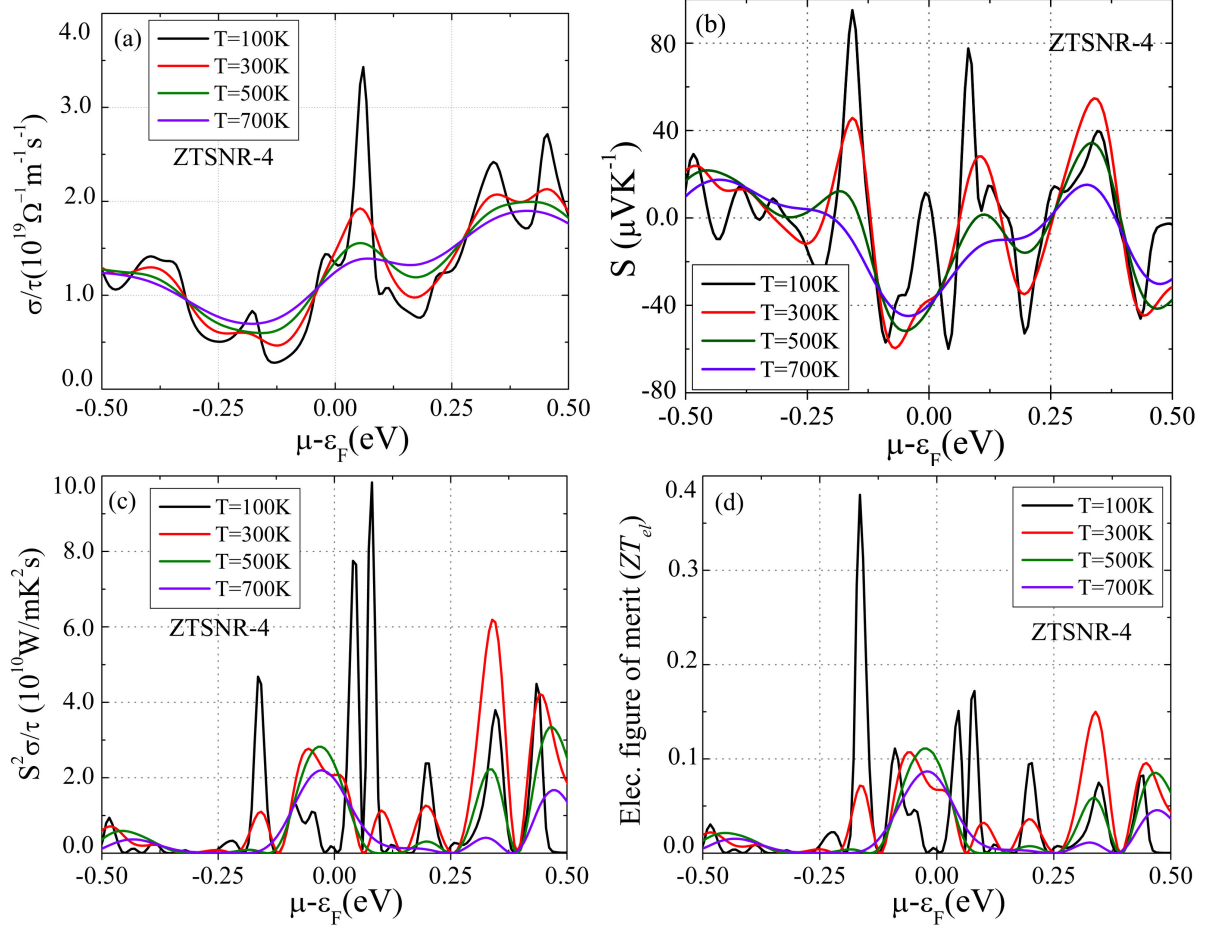
Figure 12: Variation of the (a) electrical conductivity, (b) Seebeck coefficient, (c) Power factor, (d) Electronic figure of merit of ZTSNR (N=4) with respect to the chemical potential.

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