

## Extension of Mott Formula in the Linearized Boltzmann Transport Equation to the Study of Thermoelectric Power of Electron in Metal

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### ABSTRACT

The focus of this paper is to compute and study thermoelectric power of electron in metals using Mott formula based on free electron theory to get an insight on the effect of strain on electron thermoelectric power in metals. Electron density parameter of strained metals and Poisson ratio of metals is obtained and used in this work unlike some researchers that assumed a value for Poisson ratio of all metals. This assumption make the electron density parameter of strained and unstrained metals to be equal and then reduce the computation to only one input parameter. The experimental and simulated value are in good agreement. This agreement is due to the usefulness of free electron theory in theoretical forecast of some properties of metals. The Poisson ratio is involved in simulation. Result obtained revealed that thermoelectric power of electron in metals goes up as the electron density parameter rises due to high-order contributions from electron scattering theory. Electron thermoelectric power in metals increases as the temperature increases due to electron thermal excitation from their mean position. The electron thermoelectric power rises as strain rises for metals.

### Keywords:

Thermoelectric power,  
Strain,  
Mott formula,  
Relaxation time,  
Boltzmann transport  
equation

### INTRODUCTION

Solid contains electrons and solids are grouped into metals, semiconductors and insulators Animalu, (1977). Metals are situated in unique position in the study of solids and exhibit striking characteristics that other solids lack Patterson and Bailey (2010). Such as good conductors of heat and electricity, ductility, malleability with shining on freshly surfaces Ashcroft and Mermin (1976). Insulators cannot transmit reasonable electric current while Semiconductor has intermediate characteristic between conductor and insulator Elliott (1997). Atoms in metal are closely packed so that their valence levels overlap and electrons find continuity from one atom to another. Electrons exhibit random motion in any direction in metals Uichiro (2003). Electrons in metals are depicted by wave packets of one-electron Bloch wave packets Ashcroft and Mermin (1976). Conductors conduct electric current and undergo joule heating during the flow of current Kittel (1976). Thermoelectric power of metal relied on scattering of carrier electrons in metal by deviations of crystal from perfect lattice Madelung (1995). Thermoelectric power

is the constant of proportionality between electric field and temperature gradient Uichiro (2003). A semi-empirical thermoelectric power in metal is examined by Mott since it links the absolute thermo-power to energy dependence of scattering cross section of lattice imperfection. He obtained scattering coefficients value in Taylor expansion with scattering cross section at Fermi level Rogalski and Palmer (2000). Thermoelectric effects result from the interference of electrical current and heat flow in various materials Kakani and Kakani (2004). Thermoelectric materials are used in thermoelectric systems for cooling or heating in niche applications and are being studied as a way to regenerate electricity from waste heat Rogalski and Palmer (2000). Thermoelectric phenomena shed much light on the interactions between electrons, phonons, impurities and other defects Ashcroft and Mermin (1976). The application of thermoelectric technology is hindered by low efficiencies and high costs Uichiro (2003). In the course of searching for thermoelectric power in different material, some brilliant successes have being recorded. Zhang et.al (2018) experimentally

measured the thermoelectric properties of super lattices at room temperature using a conventional d.c. four-probe method with a van der Pauw geometry. Result obtained agree with others which shows the validity of the method. Deet and Guenault (1977) experimentally investigated iron dissolved in gold and homovalent alloys using oxidation technique for low temperature thermoelectric power characteristic. Result obtained is in excellent agreement with other result. Kokalj and McKenzie (2021) use the finite temperature Lanczos method to compute the temperature dependence of thermopower based on Kelvin formula and the result obtained correlate with other results when compared. Aridi et. al (2021) presents the theoretical background of thermoelectric generator and its implementation in various fields. They also sheds light on the new technologies of the thermoelectric generator and their related challenges. They found that thermoelectric generator is efficient in hybrid heat recovery systems and that thermoelectric generator is a promising technology in power generation. Faraji et.al (2014) experimentally investigated continuous electricity production and found that thermoelectric technology has a good potential for reliable electricity generation. Uematsu et.al (2024) theoretically compute the anomalous enhancement of thermoelectric power based

on Boltzmann transport theory. They obtain enhancement rate defined as power factor. Robinson (1967) extend the theory of free electron to the study of thermoelectric power. Result obtained agree quite well with other result. Stepanoy et.al (2023) developed a theoretical and experimental models for thermoelectric power in high-temperature superconductors (HTSCs). Result obtained shows that the modified two-band model with an additional temperature term. This result is in excellent agreement with other result. Domenicali and Otter (1954) describe semi-empirical theory of thermoelectric power in metal alloys based on Mott formula and others. They observed that, Mott formula relates absolute thermo power to the energy dependence of the scattering cross section. Yongjie (2023) examine an ion–electron thermoelectric synergistic effect using an ion–electron conductor. And find that Electrons/holes can drift under the electric field generated by thermo-diffusion of ions. This result is in agreement with the result of other work. In this work, the dependence of electron density parameter and strain on thermoelectric power of electron in metals is investigated and studied. This work will be a guide to theorist and experimentalist on the behavior of electron thermoelectric power in metal. The metals were selected based on their technology application.

### Theoretical Consideration

Here, model for computing electron thermoelectric power is developed using Mott formula in sequential transport Boltzmann equation combined with relaxation time based on free electron theory. If the electrical current density  $J$  is expressed as linear functions of electric field  $E$  and temperature gradient  $\nabla T$  in the forms

$$J = L_{EE}E + L_{ET}\nabla T \quad (1)$$

and  $U$  the current density is expressed as

$$U = L_{TE}E + L_{TT}\nabla T \quad (2)$$

where  $L_{EE}$  is electrical conductivity,  $L_{ET}$  and  $L_{TE}$  are interconnected through the relation  $L_{ET} = -L_{TE}/T^4$ . The thermal conductivity  $\kappa$  is express as  $\kappa = -(L_{TT} - L_{TE} \cdot L_{ET}/L_{EE})$  using equations (1) and (2) Uichiro (2003).

Taking

$$\frac{\partial f_0}{\partial T} = -\left(\frac{\partial f_0}{\partial \epsilon}\right) \left[ \left(\frac{\epsilon - \zeta}{T}\right) + \frac{\partial \zeta}{\partial T} \right] \quad (3)$$

Where  $f_0(\epsilon_k, T) = 1/\{\exp[(\epsilon_k - \zeta)/k_B T] + 1\}$  and  $\zeta$  is the chemical potential. And the net change in electron distribution  $\partial f/\partial t$  is obtain as

$$\frac{df}{dt} = \left(\frac{\partial f}{\partial t}\right)_{diffusion} + \left(\frac{\partial f}{\partial t}\right)_{field} + \left(\frac{\partial f}{\partial t}\right)_{scatter} \quad (4)$$

The electron distribution change due to diffusion is Rogalski and Palmer (2000)

$$\begin{aligned} \left(\frac{\partial f(k)}{\partial t}\right)_{diffusion} &= \frac{f(r, k, t) - f(r, k, t - \Delta t)}{\Delta t} = \frac{f(r - v_k \cdot \Delta t, k, t - \Delta t) - f(r, k, t)}{\Delta t} \\ &= -v_k \cdot \frac{\partial f(r, k)}{\partial r} = -v_k \cdot \nabla f(r, k) \end{aligned} \quad (5)$$

Where  $v_k \cdot \nabla f(r, k) = v_{k_x} \cdot (\partial f/\partial x) + v_{k_y} \cdot (\partial f/\partial y) + v_{k_z} \cdot (\partial f/\partial z)$  and using  $f(r, k, t) = f\left(r, k - \left(\frac{\partial k}{\partial t}\right) \Delta t, t - \Delta t\right)$ , the changes in electron distribution in equation (5) become

$$\begin{aligned} \left(\frac{\partial f(k)}{\partial t}\right)_{field} &= \frac{f(r, k, t) - f(r, k, t - \Delta t)}{\Delta t} = \frac{f\left(r, k - \frac{\partial k}{\partial t} \Delta t, t - \Delta t\right) - f(r, k, t - \Delta t)}{\Delta t} \\ &= -\left(\frac{\partial k}{\partial t}\right)_{field} \cdot \frac{\partial f(k)}{\partial k} = -\frac{(-e)}{\hbar} (E + v_k \times B) \cdot \frac{\partial f_k}{\partial k} \end{aligned} \quad (6)$$

The current density  $J$  is expressed as

$$J = \frac{(-e)}{4\pi^3} \iiint v_k f(k) dk = \frac{(-e)}{4\pi^3} \iiint v_k [f(k) - f_0(k)] dk = \frac{(-e)}{4\pi^3} \iiint v_k \phi(k) dk \tag{7}$$

where  $\iiint v_k f_0(k) dk = 0$ , the linearized Boltzmann transport equation is Uichiro (2003)

$$\left(-\frac{\partial f_0}{\partial \varepsilon}\right) v_k \cdot \left[-\left(\frac{\varepsilon(k) - \zeta}{T}\right) \nabla T + (-e) \left(E - \frac{\nabla \zeta}{(-e)}\right)\right] = -\left(\frac{\partial f}{\partial t}\right)_{scatter} + v_k \cdot \frac{\partial \phi}{\partial r} + \frac{(-e)}{\hbar} (v_k \times B) \cdot \frac{\partial \phi}{\partial k} \tag{8}$$

During scattering, electron distribution change become Rogalski and Palmer (2000)

$$\left(\frac{\partial f(k)}{\partial t}\right)_{scatter} = \sum_{k'} \{Q(k, k') f(k') [1 - f(k)] - Q(k, k') f(k) [1 - f(k')]\} \tag{9}$$

where  $Q(k, k')$  is transition probability in scattering event. The term  $f(k') [1 - f(k)]$  in the bracket shows that electron of state  $k'$  is scattered into vacant state  $k$  and increases  $\left(\frac{\partial f(k)}{\partial t}\right)_{scatter}$ , whereas the second term  $f(k) [1 - f(k')]$  decreases it.

Equation (7) is rewritten by inserting the relaxation time approximation due to application of electric field to metal, then

$$J = \frac{e^2}{4\pi^3} \iiint \tau v_k (v_k \cdot E) \left(-\frac{\partial f_0}{\partial \varepsilon}\right) dk = \frac{e^2}{4\pi^3} \iiint \tau v_k (v_k \cdot E) \left(-\frac{\partial f_0}{\partial \varepsilon}\right) \frac{dS d\varepsilon}{|v_{k\varepsilon}|} \tag{10}$$

If

$$-\left(\frac{\partial f}{\partial t}\right)_{scatter} = \frac{f(r, k) - f_0(\varepsilon_k, T)}{\tau} = \frac{\phi(r, k)}{\tau} \tag{11}$$

where  $\tau$  is relaxation time and  $\phi(r, k)$  is deviation of the distribution function from equilibrium.

Substituting equation (11) into equation (8) gives

$$f(r, k) - f_0(\varepsilon_k, T) = \left(-\frac{\partial f_0}{\partial \varepsilon}\right) \tau v_k \cdot \left[-\left(\frac{\varepsilon(k) - \zeta}{T}\right) \nabla T + (-e) \left(E - \frac{\nabla \zeta}{(-e)}\right)\right] \tag{12}$$

where  $E$  is not zero, since the thermal conductivity is measured for a specimen on open electrical circuit, the term  $\varepsilon(k) - \zeta$  appears to represent the flow of heat, the term  $\frac{\nabla \zeta}{(-e)}$  implies that chemical potential gradient gives rise to additional field to induce a diffusional current.

Hence, we consider  $\left(E - \frac{\nabla \zeta}{(-e)}\right)$  to serve as an effective electric field. The linearized transport Boltzmann equation (13) is inserted into the expression for electrical current density given by equation (7), then

$$J = \frac{e^2 \tau}{4\pi^3 \hbar} \iint v_k v_k \left(-\frac{\partial f_0}{\partial \varepsilon}\right) \frac{dS}{v_{k\perp}} d\varepsilon \cdot E' + \frac{(-e) \tau}{4\pi^3 \hbar} \iint v_k v_k \left(\frac{\varepsilon - \zeta}{T}\right) \left(-\frac{\partial f_0}{\partial \varepsilon}\right) \frac{dS}{v_{k\perp}} d\varepsilon \cdot (-\nabla T) \tag{13}$$

where  $E' = E - [\nabla \zeta / (-e)]$ . From equation (14),  $\sigma = L_{EE}$  holds under the isothermal condition. Equation (1) leads to

$$E = Q \nabla T \tag{14}$$

where

$$Q = \begin{pmatrix} L_{ET} \\ L_{EE} \end{pmatrix} \tag{15}$$

where  $Q$  is the thermoelectric power and the numerator  $L_{ET}$  in equation (15) is obtain from equation (14) as

$$-L_{ET} = \frac{(-e) \tau}{4\pi^3 \hbar} \iint v_k v_k \left(\frac{\varepsilon - \zeta}{T}\right) \left(-\frac{\partial f_0}{\partial \varepsilon}\right) \frac{dS}{v_{k\perp}} d\varepsilon \tag{16}$$

By expanding equation (16) same way as equation (12), then

$$-L_{ET} = \frac{(-e) \tau}{4\pi^3 \hbar} \cdot \frac{1}{T} \left\{ \left[ \int v_k v_k (\varepsilon - \zeta) \frac{dS}{v_{k\perp}} \right]_{\varepsilon=\zeta} + \frac{\pi^2}{6} (k_B T)^2 \left[ (\varepsilon - \zeta) \frac{\partial^2}{\partial \varepsilon^2} \left( v_k v_k \frac{dS}{v_{k\perp}} \right) + 2 \int \frac{\partial}{\partial \varepsilon} \left( v_k v_k \frac{dS}{v_{k\perp}} \right) \right]_{\varepsilon=\zeta} + \dots \dots \right\} \tag{17}$$

where the terms involving  $(\varepsilon - \zeta)$  vanish at Fermi energy  $\zeta$  and,  $-L_{ET}$  become

$$-L_{ET} = \frac{\pi^2}{3} (k_B T)^2 \cdot \frac{1}{T} \left\{ \frac{(-e) \tau}{4\pi^3 \hbar} \left[ \frac{\partial}{\partial \varepsilon} \left( v_k v_k \frac{dS}{v_{k\perp}} \right) \right]_{\varepsilon=\zeta} \right\} \tag{18}$$

The denominator  $L_{EE}$  in equation (15) is electrical conductivity and is express as

$$L_{EE} = \left[ \frac{(-e)^2 \tau}{4\pi^3 \hbar} \int v_k v_k \frac{dS}{v_{k\perp}} \right]_{\varepsilon=\zeta} = [\sigma(\varepsilon)]_{\varepsilon=\zeta} \tag{19}$$

Therefore, substituting equation (18) and (19) into equation (15), the thermoelectric power  $Q$  is obtained as Uichiro (2003)

$$Q = \frac{\pi^2}{3(-e)} k_B^2 T \left[ \frac{\partial \ln \sigma(\varepsilon)}{\partial \varepsilon} \right]_{\varepsilon=\zeta} \tag{20}$$

where  $\sigma(\epsilon)$  the electrical conductivity and  $T$  is temperature. In this work, thermoelectric power of electron in metals is computed and studied using equation (20). The metals were selected based on physical constant required for computation.

### RESULT AND DISCUSSION

In figure 1, electron density parameter dependence on thermoelectric power of electron is plotted for pure metals at 300K. Figure 1 indicates that thermoelectric power of electron in metals increases as the electron density parameter increases. Which revealed that thermoelectric power of electron in metals relies on electron mobility, electronic concentration and valence electron density. In figure 1 most of the metal whose electron thermoelectric power is computed and studied have more of their electron in the region of high density limit than the low density limit, this could be due to the high-order contributions in electron scattering theory which play a vital role in thermoelectricity, change in the Fermi surface and electronic structure of metals in this region of density limit. The pattern display in figure 1 also revealed that the thermoelectric power of electron in metals also relied on chemical potential gradient, thermoelectric field, free electron energy behavior and mean electronic velocity. Furthermore, the trend metals demonstrated in figure 1 indicate that there is a good agreement between computed and experimental value. In Figure 2, electron density parameter dependence on electron thermoelectric power is plotted for pure metals at different temperature. Figure 2 revealed that the electron thermoelectric power of metals increases as the temperature increases. This suggest that as the temperature increases, electrons in their valence band in metals becomes more thermally excited and move away from their equilibrium position which increase the nearest neighbor distance between interacting electron which their-by cause the rise in electron thermoelectric power of metals as the temperature increases. The rise

in electron thermoelectric power in metals in figure 2 as temperature increases may also be due to the reduction in electron correlation between conducting electrons in metals at Fermi level. The form demonstrated by metals in figure 2 could be due to the scattering of electron from their periodic lattice site as the temperature increase that force the electron thermoelectric power to rise as the temperature go up. Figure 3 shows the plot of strain dependence of electron thermoelectric power in pure metals. In figure 3, the electron thermoelectric power in metals increases as the strain increases for all the metals whose electron thermoelectric power is computed and studied. This increase in electron thermoelectric power could be due to the effect of strain on the electron packing density and electron translational symmetry. The increase in the electron thermoelectric power as strain increases in figure 3 may be due to an increase in collision between the electron in metals which force the electron thermoelectric power to increase as strain increases because the hotter the region in which a collision occur, the faster and more energetic the electron emerge from the collision. The increase in the electron thermoelectric power as the strain increase in figure 3 could be due to the dependence of thermoelectric power on the details of how the current carrier in metals are scattered by various deviations of the crystal from perfect lattice. Among all the metals subjected to different strain in figure 3, Potassium has the highest electron thermoelectric power because Potassium has small electron ion core whose valence electron is much closer to the nucleus while Molybdenum and Tungsten has the lowest electron thermoelectric power because Molybdenum and Tungsten has small Fermi surface in most of completely filled zone. Another thing that may be responsible for the high effect of strain on the electron thermoelectric power of Potassium could be due to high effect of mechanical pressure on its valence electron density and characteristic metallic properties.

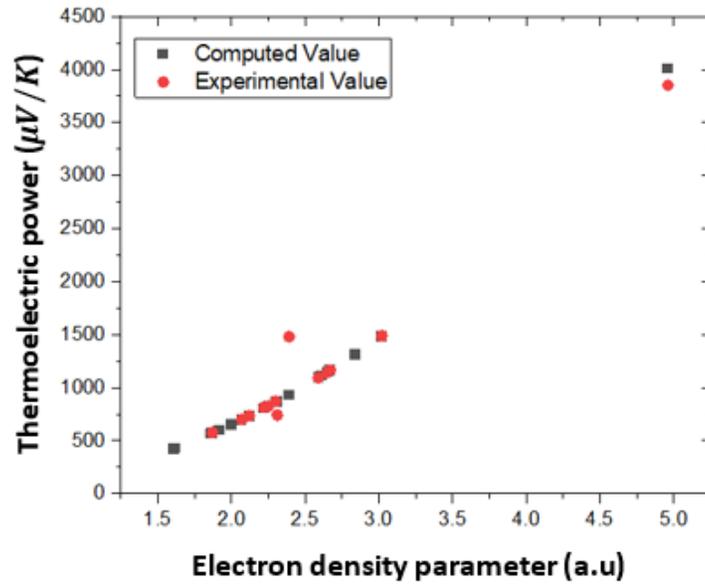


Figure 1: Plot of Electron Density Parameter Dependence of the Thermoelectric Power in pure Metals (T=300K)

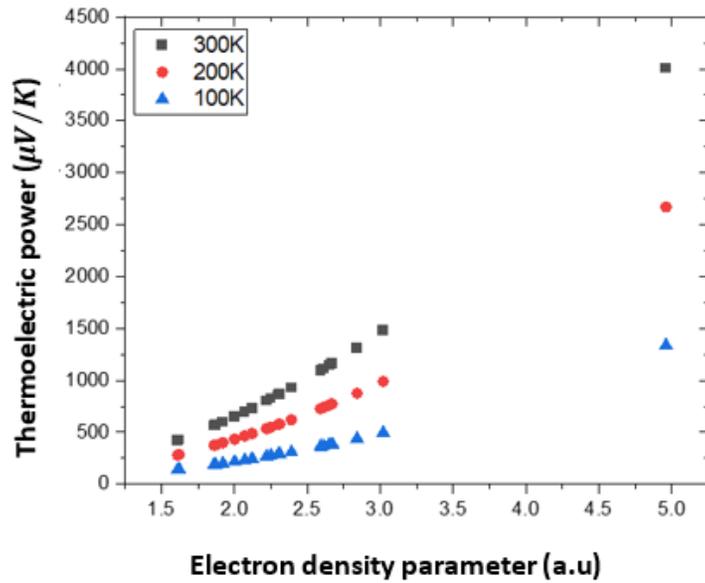


Figure 2: Plot of Electron Density Parameter Dependence of the Thermoelectric Power in pure Metals at different Temperature

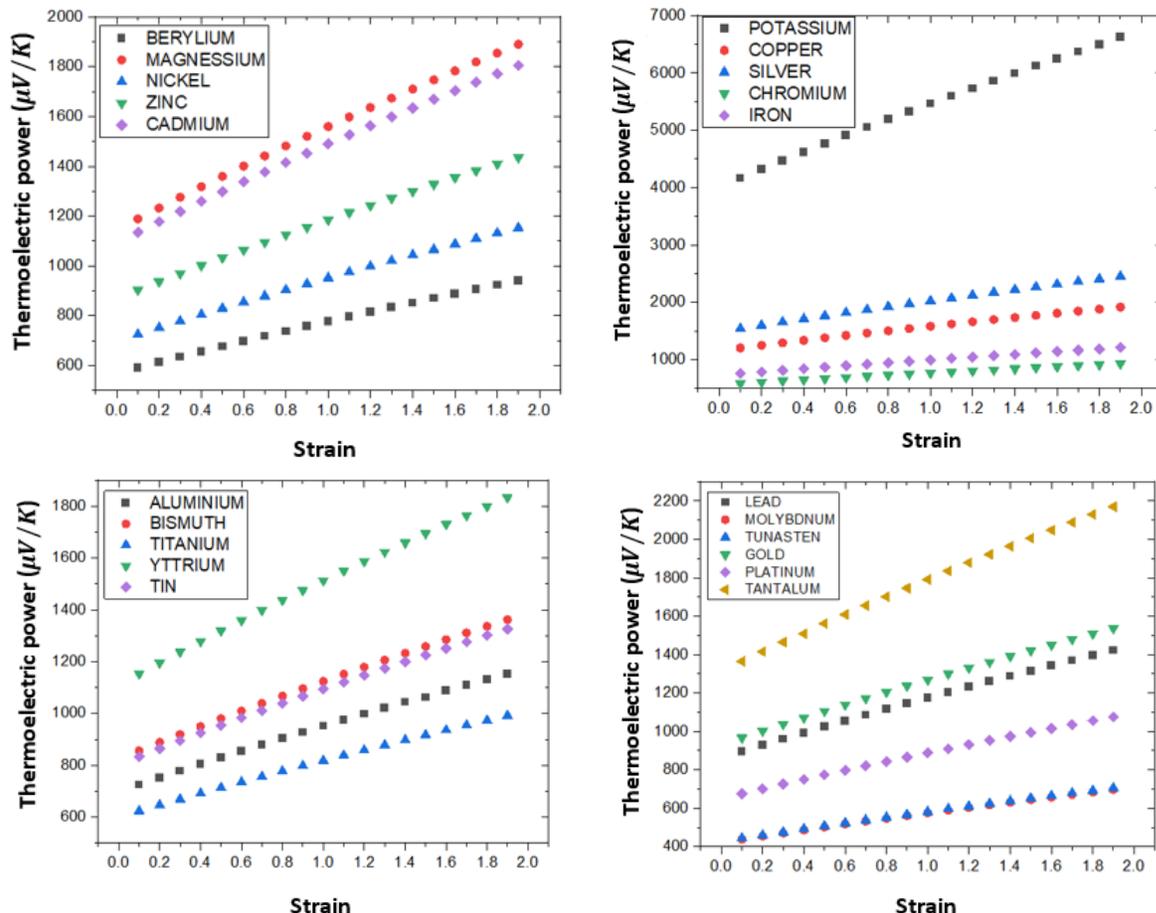


Figure 3: Plot of Strain Dependence of the Thermoelectric Power in pure Metals

Table 1: Thermoelectric power of Unstrained Metals

Metals	Electron Density Parameter $r_s$ (a.u)	Experimental Thermoelectric power ( $\mu V/K$ ) at 300K	Computed Thermoelectric power ( $\mu V/K$ )		
			Temperature at 300K	Temperature at 200K	Temperature at 100K
K	4.96	3851.09	4006.78	2671.18	1335.59
Cu	2.67	1166.41	1161.06	774.039	387.020
Ag	3.02	1489.57	1485.41	990.271	495.136
Be	1.87	577.367	569.528	379.685	189.843
Mg	2.65	1145.04	1143.73	762.486	381.243
Cr	1.86		563.453	375.635	187.818
Fe	2.12	735.474	731.987	487.991	243.996
Ni	2.07		697.866	465.244	232.622
Zn	2.31	742.023	869.072	579.381	289.691
Cd	2.59	1094.09	1092.52	728.349	364.175
Al	2.07	701.919	697.866	465.244	232.622
Bi	2.25	824.629	824.511	549.674	274.837
Ti	1.92		600.391	400.261	200.130
Y	2.61		1109.46	739.642	369.821
Sn	2.22	813.890	802.671	535.114	267.557
Pb	2.30	871.333	861.563	574.376	287.188
Mo	1.61		422.166	281.444	140.722

W	1.62		427.427	284.951	142.476
Au	2.39	1481.48	930.310	620.206	310.103
Pt	2.00		651.466	434.311	217.155
Ta	2.84		1313.62	875.744	437.872

**Table 2: Thermoelectric Power of Strained Metals**

Metals	$r_s(\text{A.U})$	Strain								
		0.2	0.4	0.6	0.8	1.0	1.2	1.4	1.6	1.8
K	4.96	4317.97	4617.99	4908.29	5190.00	5464.04	5731.18	5992.04	6247.18	6497.07
Cu	2.67	1251.23	1338.17	1422.30	1503.93	1583.33	1660.74	1736.34	1810.26	1882.68
Ag	3.02	1600.77	1712.00	1819.63	1924.06	2025.65	2124.68	2221.39	2315.98	2408.61
Be	1.87	613.761	656.407	697.671	737.716	776.657	814.640	851.718	887.985	923.502
Mg	2.65	1232.56	1318.20	1401.07	1481.48	1559.71	1635.96	1710.42	1783.25	1854.58
Cr	1.86	607.215	649.402	690.228	729.841	768.379	805.950	842.632	878.510	913.647
Fe	2.12	788.840	843.647	896.684	948.144	998.215	1046.97	1094.67	1141.28	1186.93
Ni	2.07	752.066	804.284	854.886	903.952	951.684	998.207	1043.65	1088.08	1131.60
Zn	2.31	936.571	1001.65	1064.61	1125.72	1185.15	1243.10	1299.67	1355.02	1409.22
Cd	2.59	1177.38	1259.19	1338.34	1415.16	1489.88	1562.71	1633.84	1703.41	1771.55
Al	2.07	752.066	804.321	854.886	903.952	951.684	998.207	1043.65	1088.08	1131.60
Bi	2.25	888.547	950.291	1010.02	1068.00	1124.39	1179.36	1233.04	1285.54	1336.96
Ti	1.92	647.017	691.979	735.478	777.689	818.754	858.779	897.869	936.103	973.548
Y	2.61	1195.63	1278.71	1359.09	1437.09	1512.98	1586.94	1659.17	1729.82	1799.01
Sn	2.22	865.013	925.116	983.272	1039.71	1094.60	1148.12	1200.37	1251.49	1301.54
Pb	2.30	928.481	992.988	1055.41	1115.99	1174.92	1232.36	1288.45	1343.31	1397.04
Mo	1.61	454.953	486.564	517.151	546.837	575.709	603.854	631.337	658.219	684.553
W	1.62	460.621	492.630	523.597	553.651	582.884	611.379	639.205	666.424	693.081
Au	2.39	1002.56	1072.23	1139.62	1205.04	1268.67	1330.69	1391.25	1450.50	1508.52
Pt	2.00	702.060	750.842	798.042	843.847	888.403	931.836	974.249	1015.73	1056.37
Ta	2.84	1415.64	1509.04	1609.18	1701.54	1791.37	1878.96	1964.48	2048.13	2130.05

**CONCLUSION**

In summary, this work clearly demonstrates the behavior of electron density parameter and linearly applied strain dependence on the electron thermoelectric power in pure metals. This study is based on Mott formula in the sequential transport Boltzmann equation combined with relaxation time based on free electron theory. Result obtained agree quite well with experimental value. The electron thermoelectric power of metals depends on electronic concentration and structure factor. Electron thermoelectric power of metals increases as temperature and strain increases. The increase in the electron thermoelectric power of metals as the temperature increases is due to the increase in the electron nearest neighbor distance between the conducting electrons in metals at Fermi level.

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