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Temperature and Electron concentration dependent Thermoelectric Power in Wide Band Gap semiconductor GaN Nanowire

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Abstract: ***Nanostructures have a significant promise as potential building blocks for the next generation thermoelectric devices. While the thermal transport properties of bulk materials have been intensely studied, the understanding of nanostructure thermoelectric properties and their interrelation is still incomplete. In the calculated temperature range, the thermoelectric power (TEP) was found to be linearly dependent on temperature, suggesting the degenerate nature of the GaN semiconductor nanowire similar to that of ZnO semiconductor nanowire. Observed negative values of TEP indicating that, majority charge carriers are electrons in GaN semiconductor. Linear dependence of TEP with temperature shows that TEP is only due to electron diffusion and not due to phonon-drag effect. Also observed the calculated TEP values are in good agreement with the experimental results in the overall temperature range 10 – 300 K.***
Keywords: Semiconductor nanowire, GaN, Mott relation, Thermoelectric power, wide band gap semiconductor.

I. INTRODUCTION

Solid-state thermoelectric devices which convert thermal energy to electrical energy. Operated in a reverse mode, thermoelectrics can use electricity to provide heating or cooling. Thermoelectrics provide localized control of energy transfer and distributed electricity generation. They have no moving parts and thus operate silently. To date, they have been reliable although new applications are challenging the reliability of current thermoelectric device technology. There are also considerable challenges associated with thermoelectric technology. The conversion efficiency is low, and performance is highly dependent on the device operating temperature, requiring specialized materials and devices for each application. System integration is particularly challenging as the devices require multiple materials with highly differentiated mechanical, thermal, and electrical properties. The abundance of recent reviews on thermoelectrics from materials physics to applications demonstrates the newfound popularity of this technology [1,2]. Thermoelectrics rely on the Seebeck effect for a material in which electrons are the dominant charge carriers. When a temperature gradient is applied across a material, electrons with more energy on the hot side shift to the cold side. An electric field develops which prevents further electron flow. The Seebeck coefficient, or thermopower, is a measure of the voltage developed in response to the temperature drop across the material.

The intensive research on thermoelectrics in the past two and a half decades was largely inspired by the seminal work by Dresselhaus et al. in 1993, which predicted that quantum confinement effects can lead to a drastically enhanced thermoelectric power factor in low-dimensional materials [3]. It was also quickly realized that the thermal conductivity of nanowires could be reduced remarkably, and that the combined effects of enhanced power factor and reduced thermal conductivity provide an extremely promising route to achieve high thermoelectric figures of merit (ZT) [4,5] These insights motivated extensive efforts on a full spectrum of research spanning nanowire synthesis, property characterization, and device integration.

Thermoelectrics describes direct conversion between thermal and electrical energy, which encompasses three separately identified physical phenomena: the Seebeck effect, the Peltier effect, and the Thomson effect [6,7]. Nearly two centuries ago, in 1821−1823, Thomas Seebeck first observed that a magnetic field was generated if two dissimilar conductors were attached together and one of the junctions was heated. What he really found is that a temperature gradient across certain materials will create a voltage drop between the hot and cold ends. The Seebeck coefficient S, named after him, describes the thermoelectric voltage change rate with respect to temperature at a given temperature. A more convenient parameter is the relative Seebeck coefficient, usually simply called Seebeck coefficient, defined as $S_{ab} = dV/dT$, where S_{ab} is the relative Seebeck coefficient between materials a and b, V is the voltage drop, and T is temperature.

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Recently the potential applications for thermoelectrics that employ semiconducting nanowires have recently emerged [8,9]. TEP, the ratio of the electric field to the thermal gradient, is one of the central physical quantities that determine the efficiency of thermoelectric applications. In general, TEP of an electronic system depends on the electron and hole asymmetries at the Fermi level, and thus is sensitive to small changes in scattering rate and the density of states at the Fermi level [10]. Utilizing this sensitivity, the electrical properties of 1D nanostructures, such as carbon nanotubes CNTs and semimetallic Bi nanowires, have been investigated by TEP measurements [11,12]. In addition, TEP measurements have provided an effective approach to measure carrier concentration in nanowires [13]. The ongoing efforts for producing efficient thermoelectric devices currently include the investigation of nanowires [14] and nanotubes [15] or even atomic chains [16] and single molecules [17] contacted to nano- or bulk electrodes. Besides generating thermoelectricity, nanostructures of this type constitute the main building blocks of high performance temperature sensors and cooling devices.

There has been experimentally observed that the Seebeck coefficient of GaN/AlGaN nanowires is more than twice as large as that for the GaN nanowires alone [18]. Also there has been a study of phonon drag in the AlGaN/GaN two-dimensional electron gas (2DEG) and observed that phonon drag does not contribute significantly to the thermoelectric behavior of devices with ∼100 nm GaN thickness, which suppresses the phonon mean free path. However, when the thickness is increased to ∼1.2 μm, up to 32% (88%) of the Seebeck coefficient at 300 K (50 K) can be attributed to the drag component. In turn, the phonon drag enables state-of-the-art thermoelectric power factor in the thicker GaN film, up to ∼40 mW m^{-1} K^{-2} at 50 K. By measuring the thermal conductivity of these AlGaN/GaN films. It is concluded that the magnitude of the phonon drag can increase even when the thermal conductivity decreases. Decoupling of thermal conductivity and Seebeck coefficient could enable important advancements in thermoelectric power conversion with devices based on 2DEGs [19].

Several groups have tried to tune the TE properties of bulk GaN materials by modification of electronic structure either by substitutions of additional elements such as In or Al at Ga site or by lowering the dimensions in the form of thin films or nanowires [20, 21]. Both p- and n-type materials were successfully achieved depending upon the amount or type of the additional impurity introduced in the pure GaN material [22]. Yamamoto et al. prepared bulk GaN material with micron size of particles, which showed a negative S value of −50 μV/K at room temperature that further increased with temperature [23]. The comparative study on both bulk and thin-film GaN, Kucukgok et al [24] reported a positive and large Seebeck coefficients which found to be decreased with increase in carrier concentrations. Also there has been an extensive study of phonon drag thermopower in silicene at equipartition regime at room temperature having high Fermi level, expecting contribution of thermopower by phonon-drag effect over the electron-diffusion thermopower [25].

II. THEORETICAL MODEL

We shall use the well-known Mott formula within parabolic band approximation to estimate the contribution of electrons towards TEP. The low temperature carrier diffusion TEP is [26]

$$\boldsymbol{S} = -\frac{\boldsymbol{\pi^2}}{\boldsymbol{3}}\frac{\boldsymbol{k_B^2 T}}{\boldsymbol{e}}\left|\frac{\partial \ln(\sigma(E))}{\partial E}\right|_{E=E_F} \qquad (1)$$

where $\sigma(\boldsymbol{E})$ is the energy dependence of conductivity in the relaxation time approximation. In what follows, the mean free path of the carriers is assumed to be independent of temperature, above equation (1) becomes

$$\boldsymbol{S} = -\frac{\boldsymbol{\pi^2 k_B^2 m^*}}{\boldsymbol{(3\pi^2)^{2/3}\hbar^2 e}}\frac{\boldsymbol{T}}{\boldsymbol{n^{2/3}}} \qquad (2)$$

With constant mean free path, and the method point to the scattering of carrier by impurities is dominant. We must point out that the linear response transport theory based on Fermi liquid description results in an exact expression for the most singular term in terms of damping rate and is valid for correlated electron systems as wide band gap semiconductors.

In the above equation (2), the right hand part was derived by assuming an energy-independent mobility, where n is the carrier concentration and $\boldsymbol{m^*}$ is the effective mass of the semiconductor material. It should be noticed that, above equation (2) is valid for the semiconductor sample studied in this article because the mean free path $\boldsymbol{l_m}$ is much larger than the interatomic distance and that is semi-classical approach is appropriate without a quantum correction since $\boldsymbol{k_F l_m} \approx 1$, where $\boldsymbol{k_F}$ is Fermi wave vector. This similar approximation has already been made in TEP analysis of semimetallic Bi nanowire [27] and small band gap semiconductors [28]. We used this approximation in wide band gap semiconductor GaN nanowire and compared with experimental results.

III. RESULTS AND DISCUSSION

We calculated theoretically the TEP for the wide band gap semiconductor GaN nanowire using the characteristic parameter, $\boldsymbol{m}^* = 0.20\,\boldsymbol{m_e}$, electron concentration $\boldsymbol{n} = 6.6\text{x}10^{25}$ m^{-3}. These calculated data is compared with the experimental results [29] as shown in Figure 1.

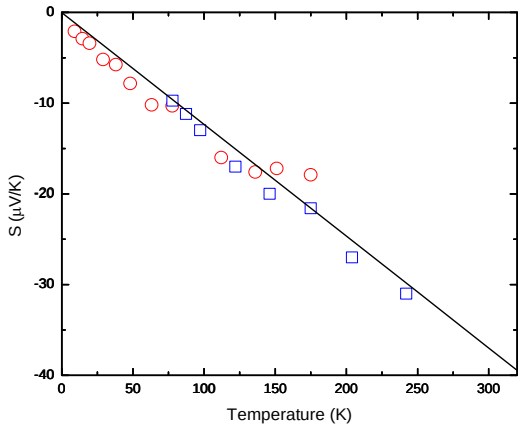


Figure 1: Comparison of TEP with experimental results of GaN nanowire [29]. The symbols show experimental data of different devices and solid line represents our theoretical calculations.

The experimental results of GaN nanowire which were grown by catalyst-free metallic-organic vapor-phase epitaxy. There are different devices used with different nanowire having diameter in the range 80 – 120 nm. We obtained a good agreement with the experimental results in the temperature range 10 – 300 K for different GaN devices. The measured TEP was about ~ -40 μV/K at room temperature, varying linearly with temperature. The negative values of TEP indicate that the carrier transport in GaN nanowire is contributed by electron diffusion, reflecting that the nanowire is n-type semiconductor. The overall temperature dependence, $\boldsymbol{S} \propto \boldsymbol{T}$, agrees well with the Mott relation, which was expected for metal or a heavy doped semiconductor [30]. The major relation in our calculations is that the values of TEP for the wide band semiconductor increase linearly with temperature. This linear dependence observation suggests that there is no appreciable phonon-drag induced TEP in this nanowire and thus TEP can only be described by thermal diffusion of the majority charge carriers. The calculated TEP at room temperature for GaN nanowire is -40μV/K which is nearly an order of magnitude less as compared to ZnO nanowire which was found to be -400μV/K at room temperature [31].

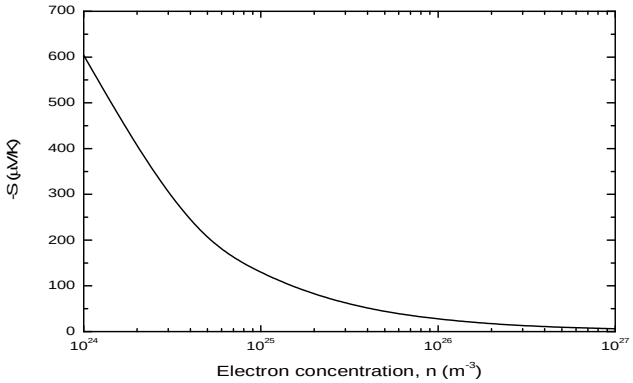


Figure 2: Variation of TEP with electron concentration at room temperature.

Figure 2 below shows the variation of thermoelectric power with carrier concentration at room temperature. It is observed that thermoelectric power increases with increase in concentration, same behavior observed in two-dimensional electron gas of ZnO. It is because in our formulation, TEP varies inversely proportional to carrier concentration.

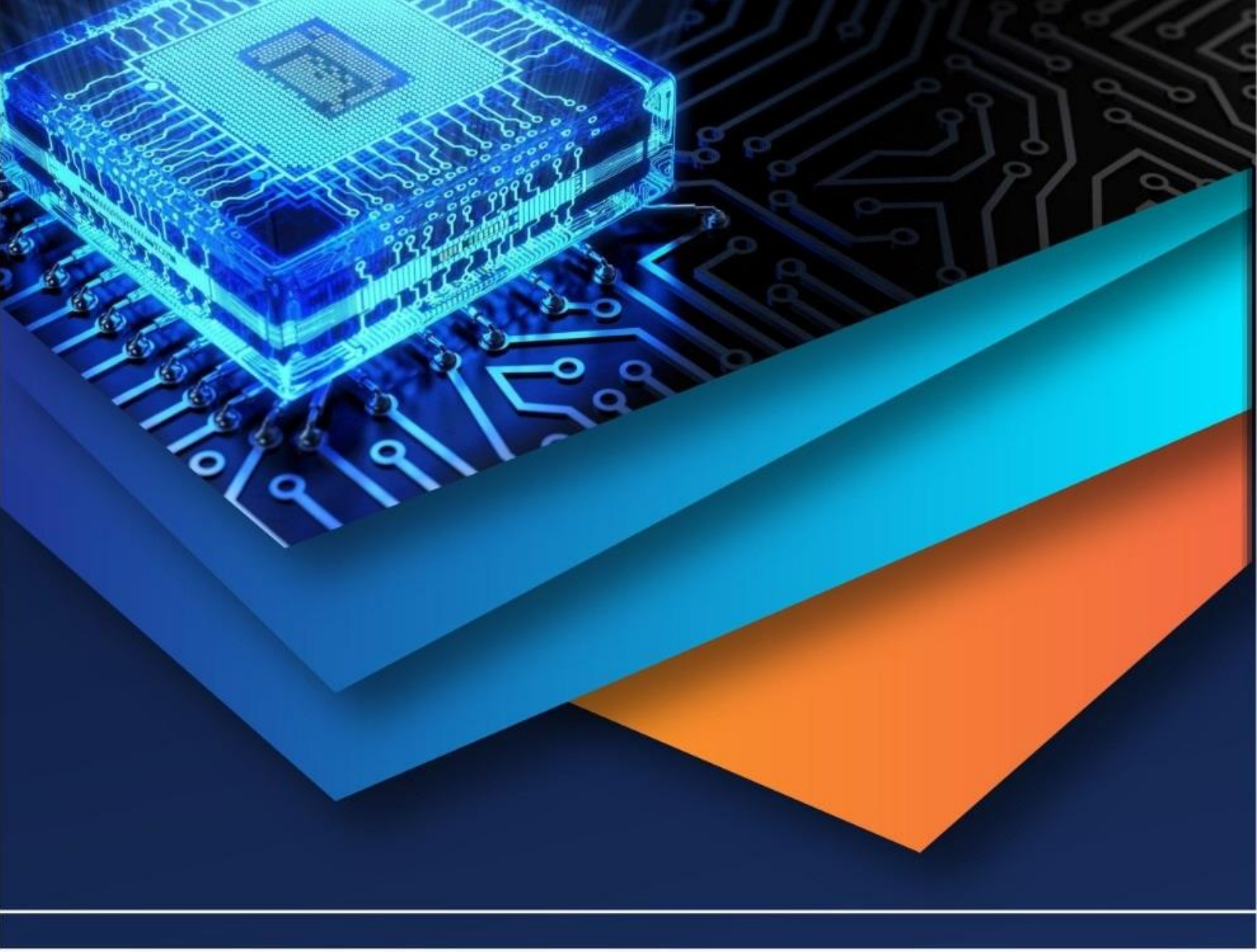
IV. CONCLUSIONS

We calculated TEP using basic semiclassical Mott formula to compare the experimental results of wide band gap semiconductor ZnO nanowire for various devices and obtained a good agreement in the temperature range 10 – 300K. The measured and calculated TEP increases linearly with temperature showing that TEP is due to electron-diffusion process. The negative sign of TEP values indicates that the majority carriers in ZnO are electrons. The calculated value of TEP at room temperature is about -400 μV/K.

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