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Effects of Nonparabolicity on Electron Thermopower of Size-Quantized Semiconductor Films

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Effects of nonparabolicity of energy band on thermopower, in-plane effective mass and Fermi energy are investigated in size-quantized semiconductor films in a strong while non-quantized magnetic field. We obtain the expressions of these quantities as functions of thickness, concentration and nonparabolicity parameter. The influence of nonparabolicity is studied for degenerate and non-degenerate electron gases, and it is shown that nonparabolicity changes the character of thickness and the concentration dependence of thermopower, in-plane effective mass and Fermi energy. Moreover, the magnitudes of these quantities significantly increase with respect to the nonparabolicity parameter in the case of strong nonparabolicity in nano-films. The concentration dependence is also studied, and it is shown that thermopower increases when the concentration decreases. These results are in agreement with the experimental data.

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As is known, in the case of size quantization the energy of electrons becomes the function of thickness. On the other hand, the electron effective mass is the function of energy, in general Refs. [1,2] and therefore it is the function of thickness. The size dependence of electron effective mass which is predicted in Ref. [3] experimentally was observed on GaAs quantum wells in Refs. [4,5]. The materials such as GaAs, the semiconductors of the types $A^{III}B^{V}$, Cd_xHg_{1-x}, Te, $Zn_xHg_{1-x}Se$, have a nonparabolic energy band. Therefore, the electron effective mass is the function of energy. Consequently, in thin films of thickness, which satisfy the condition of quantization that the effective mass will be size dependent. Thus every quantity which can be expressed by electron effective mass is also size dependent. One of these quantities is the electron thermopower of thin films in a strong, while non-quantized magnetic field, directed perpendicularly to the film surface in the case of degenerate electron gas. In this situation, the effect of nonparabolicity was studied in Refs. [6,7] for the electron thermopower and in-plane effective mass without a detailed analysis. In the case of arbitrary temperature the thermopower cannot be expressed wholly by the electron effective mass. However, in this case also, thermopower depends on thickness and concentration.

In this Letter, a general expression of thermopower of nonparabolic semiconductor thin films at arbitrary temperature in a strong while non-quantized magnetic field is obtained. It is shown that, in the case of degenerate electron gas, the expression of thermopower reduces to the one obtained in Ref. [6]. The thickness and concentration dependences of thermopower, Fermi energy and electron effective mass are investigated in detail. The effects of nonparabolicity on effective mass, Fermi energy and thermopower are estimated. It is shown that the effect of nonparabolicity changes the character of thickness and concentration dependences of these parameters. Further, it is noted that in the limit of ultrathin films (quantum wells) with strong nonparabolic energy band, these parameters become more significant than in bulk materials. On the other hand, in the case of ultrathin films (under the Fermi level there is only one sub-band) the behavior of investigated parameters with respect to thickness is nonmonotonic. For example, the thermopower as a function of thickness has a minimum at $d_{\min} = (\frac{5\pi}{n_{el}})^{1/3}$, while as a function of concentration monotonically decreases with concentration. Throughout this study, the expression of thermopower increase (decrease) concerns its absolute value.

In the case of nondegenerate electron gas, we also obtain the expression of thermopower as functions of thickness and concentration. The effect of nonparabolicity is estimated in the limit of nanofilms with a strong nonparabolic energy band.

The nonmonotonic behavior of thermopower and Fermi energy experimentally was observed in Refs. [8–10,13,14].

Consider the semiconductor thin film having thickness d which satisfies the condition of size quantization. Assume that the film is in a strong while non-quantized magnetic field, directed along the normal direction to the surface. Consider the following en-

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ergy momentum relationship in a film,[6]

$$B(\varepsilon) = \frac{\hbar^2 k_{\text{II}}^2}{2m_n} + \frac{\hbar^2\pi^2}{2m_n d^2} n_z^2, \tag{1}$$

where $B(\varepsilon)$ is the function of energy; $k_{\text{II}} = \sqrt{k_x^2 + k_y^2}$ is the momentum in the plane, m_n is the electron effective mass at $k = 0$; the z axis is perpendicular to the film surface, and $k_z = \frac{\pi}{d} n_z, n_z = 1, 2, \ldots$.

For arbitrary temperature, the thermopower (α_{f}) can be expressed as[6]

$$\alpha_{\text{f}} = (-\frac{k_0}{e}) \frac{\hbar^2}{2\pi k_0 T n_{\text{el}} d} \cdot \sum_{n_z} \int_0^{\infty} (-\frac{\partial f_0}{\partial \varepsilon}) \frac{(\varepsilon - \varsigma) k_{\text{II}}^3(\varepsilon) d k_{\text{II}}}{m_{\text{II}}(\varepsilon)}, \tag{2}$$

where k_0 is the Boltzman constant, e is the electron charge, f_0 is the Fermi function, ς is the chemical potential, T is the temperature, $m_{\text{II}}(\varepsilon) = m_n \frac{dB(\varepsilon)}{d\varepsilon}$ is the in-plane electron effective mass, and n_{el} is the electron density.

In the case of degenerate electron gas from Eq. (2) we can find the result of Ref. [6],

$$\alpha_{\text{f}}^{\text{np}} = \alpha_{\text{f}}^{\text{p}} \frac{m_{\text{II}}(\varepsilon_{\text{F}})}{m_n},$$

where ε_{F} is the Fermi energy, $\alpha_{\text{f}}^{\text{np}}$ and $\alpha_{\text{f}}^{\text{p}}$ are the thermopower of the films with nonparabolic and parabolic energy bands, respectively.

To study the effect of nonparabolicity on thermopower, in-plane effective mass and Fermi energy, it is necessary to determine $B(\varepsilon)$. We shall take it in a two-band approximation of the low Kane dispersion. Then, for the degenerate electron gas, from Eq. (2) and from the formula for concentration, one can find the expressions for weak and strong nonparabolicity. For weak nonparabolicity ($\gamma \ll 1$) we have

$$\alpha_{\text{f}}^{\text{np}} = \alpha_{\text{f}}^{\text{p}} (1 + \frac{\gamma}{2} f_{\bar{n}_z}(u)), \quad \varepsilon_{\text{F}} = \varepsilon_1(d_n) f_{\bar{n}_z}(u), \tag{3}$$

where $\gamma = 4\frac{\varepsilon_1(d_n)}{\varepsilon_{\text{g}}}$ is the parameter which determines the effect of nonparabolicity; $\gamma = 0$ is the parabolic case, $\gamma \neq 0$ is the nonparabolic case, $d_n = (\frac{\pi}{2n_{\text{el}}})^{1/3}$, $\varepsilon_1(d_n) = \frac{\pi^2\hbar^2}{2m_n d_n^2}$, ε_{g} is the energy gap, $u = \frac{d}{d_n}$, and $\bar{n}_z$ is the number of the subband which, at the bottom, coincides with the Fermi level and

$$f_{\bar{n}_z} = \frac{u}{\bar{n}_z} + \frac{\varphi(\bar{n}_z)}{\bar{n}_z u^2}, \quad \varphi(\bar{n}_z) = \frac{1}{6}\bar{n}_z(\bar{n}_z + 1)(2\bar{n}_z + 1).$$

Equation (3) determines the size and concentration dependences of the thermopower, in-plane effective mass and Fermi energy in the films of arbitrary thickness and weak nonparabolicity for degenerate electron gas. In this case the behavior of the Fermi energy, in-plane effective mass and thermopower versus thickness is nonmonotonic (the oscillating function of thickness) in general.

In the case of strong nonparabolicity ($\varepsilon_{\text{g}} \to 0$, $\gamma \gg 1$) one can find

$$\varepsilon_{\text{F}} = \varepsilon_1(d_n)\sqrt{f_{\bar{n}_z}(u)}, \quad \frac{\alpha_{\text{f}}^{\text{np}}}{\alpha_{\text{f}}^{\text{p}}} = 2\frac{\varepsilon_1(d_n)}{\varepsilon_{\text{g}}}\sqrt{f_{\bar{n}_z}(u)} = \frac{\gamma}{2}\sqrt{f_{\bar{n}_z}(u)}, \tag{4}$$

and in the case of ultrathin films (nanofilms) we also have

$$\varepsilon_{\text{F}} = \varepsilon_1(d_n)\sqrt{\frac{5}{2}}\frac{1}{u}, \quad \frac{\alpha_{\text{f}}^{\text{np}}}{\alpha_{\text{f}}^{\text{p}}} = \sqrt{\frac{5}{2}}\frac{\gamma}{2u}, \tag{5}$$

where $s = \sqrt{\frac{2}{3}}\frac{p}{\hbar}$, p is the matrix element which determines the interaction of conduction and valance bands, and

$$\varepsilon_1(d_n) = \begin{cases} \frac{\pi^2\hbar^2}{2m_n d_n^2}, & \gamma \ll 1, \\ \frac{\pi\hbar s}{d_n}, & \gamma \gg 1. \end{cases}$$

As seen from Eq. (4) the thermopower, in-plane effective mass and Fermi energy in the case of strong nonparabolicity are also the oscilation functions of thickness.

Note that in the case of strong nonparabolicity the parameter s reads $\varepsilon_{\text{g}} = 2m_n s^2$ and, as is seen, it has a velocity unit ($s \approx 10^8$ cm/s) in Ref. [1].

It is easy to see from Eqs. (3)and (4) that in the situation that under the Fermi level there is only one sub-band, the Fermi energy as a function of thickness has a minimum at $d_{\min} = (\frac{5\pi}{n_{\text{el}}})^{1/3}$. Since $\frac{\alpha_{\text{f}}^{\text{np}}}{\alpha_{\text{f}}^{\text{p}}}$ and $\frac{m_{\text{II}}}{m_n}$ are the functions of ε_{F}, they also have the minimum at $d = d_{\min}$.

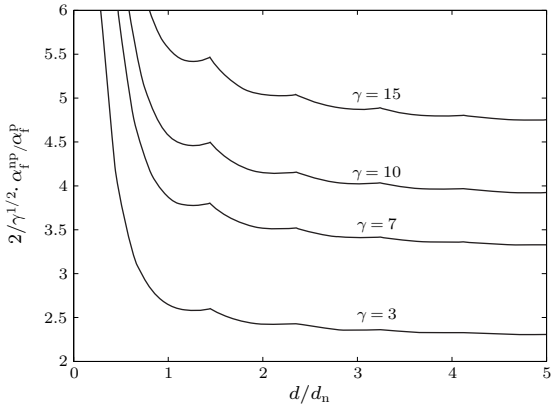


Fig. 1. The thickness dependence of $\frac{2}{\sqrt{\gamma}} \cdot \frac{\alpha_{\text{f}}^{\text{np}}}{\alpha_{\text{f}}^{\text{p}}} = \frac{2}{\sqrt{\gamma}} \cdot \frac{m_{\text{II}}}{m_n}$ for difference nonparabolicity (γ).

Graphic of thermopower (in-plane effective mass) for some values of parameter nonparabolicity ($\gamma = 3$, 7, 10 and 15) is given in Figs. 1 and 2.

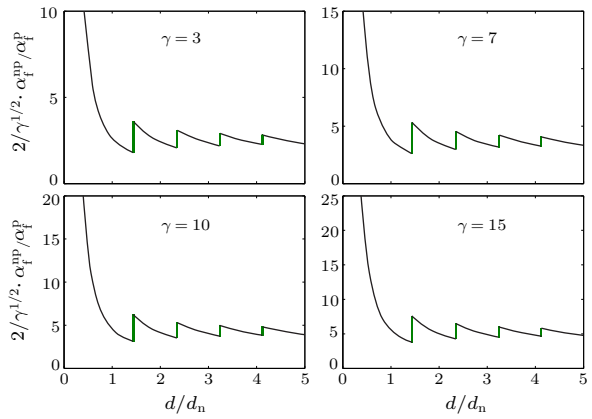


Fig. 2. The thickness dependence of $\frac{2}{\sqrt{\gamma}} \cdot \frac{\alpha_{\rm f}^{\rm np}}{\alpha_m^{\rm np}}$ for different γ ($\alpha_m^{\rm np}$ is the thermopower in bulk specimen).

The expression of $\alpha_{\rm f}^{\rm np}$ given in Eq. (2) can be rewritten in the following useful form for applications at arbitrary temperature. Thus in two-band approximation of the Kane model for arbitrary temperature we have

$$\alpha_{\rm f}^{\rm np} = (-\frac{k_0}{e})\frac{m_n k_0 T}{\pi\hbar n_{\rm el} d} \times \sum_{n_z}\{(1+2\beta\varepsilon^*_{n_z})[F_2(\eta_n) - \eta_n F_1(\eta_n)] + \beta[F_3(\eta_n) - \eta_n F_2(\eta_n)]\}, \quad (6)$$

where

$$F_r(\eta_n) = \int_0^\infty \Big(-\frac{\partial f_0}{\partial x}\Big) x^r dx$$

is the one parameter integral Fermi,

$$x =, \frac{\varepsilon - \varepsilon_{n_z}}{k_0 T}, \quad \eta_n = \frac{\varsigma - \varepsilon_{n_z}}{k_0 T}, \quad \varepsilon^*_{n_z} = \frac{\varepsilon_{n_z}}{k_0 T}, \quad \beta = \frac{k_0 T}{\varepsilon_{\rm g}},$$

where ε_{n_z} is the positive root of Eq. (1) at $k_{\rm II} = 0$.

To determine the chemical potential one can present the expression of electron concentration

$$n_{\rm el} = \frac{m_n k_0 T}{\pi\hbar^2 d}\sum_{n_z}\{(1+2\beta\varepsilon^*_{n_z})F_1(\eta_n) + \beta F_2(\eta_n)\}. \quad (7)$$

Solving Eqs. (6) and (7) together, one can find the thermopower at arbitrary temperature as functions of thickness and electron concentration. Due to the fact that the situation of degenerate electron gas is studied above, here we present the results only for nondegenerate electron gas ($-\eta_n \gg 1$) in the limit of strongly nonparabolic energy band $\beta \gg 1$. In this case, the Fermi integral expressed by Gamma function $\Gamma(\eta_n)$ are

$$F_r(\eta_n) = e^{\eta_n}\Gamma(r+1).$$

By using the relation $\Gamma(r+1) = r!$, the summation in Eqs. (6) and (7) with respect to n_z gives for $\alpha_{\rm f}^{\rm np}$ and $n_{\rm el}$,

$$\alpha_{\rm f}^{\rm np} = -\frac{k_0}{e}\Big\{3 - \eta + \frac{b^2 e^b(e^b+1)}{(e^b-1)(be^b+e^b-1)}\Big\}, \quad (8)$$

$$n_{\rm el} = \frac{\pi}{d^3}e^\eta \frac{be^b + e^b - 1}{b^2(e^b-1)^2}, \quad (9)$$

where $b = \frac{\pi\hbar s}{k_0 T d}$. Note that the parameter b is of energy nature, which determines the interval between size quantized energy levels. In fact, in the case of strong nonparabolicity the energy levels (at $k_{\rm II}$=0) become

$$\varepsilon_{n_z} = \frac{\pi\hbar s}{d} n_z,$$

and the distance between them is

$$\varepsilon_{n_z+1} - \varepsilon_{n_z} = \frac{\pi\hbar s}{d}. \quad (10)$$

Thus the parameter b is the ratio of energy gap between the levels ε_{n_z+1} and ε_{n_z} to $k_0 T$.

Equations (8) and (9) determine the thickness and concentration dependence of thermopower for the film with arbitrary thickness. We shall present here the expression of thermopower for two limits: in the case of bulk specimen ($d \to \infty$, $b \ll 1$) and ultrathin films (nanofilms $d \to 0$, $b \gg 1$). The result for the limit $d \to \infty$ is

$$\alpha_{\rm f}^{\rm np} = -\frac{k_0}{e}\Big\{4 + \ln(\frac{2k_0^3 T^3}{\pi^2\hbar^3 s^3 n_{\rm el}}) + \frac{\pi\hbar s}{2k_0 T d}\Big\}. \quad (11)$$

It follows from Eq. (11) that at $d \to \infty$, the last term becomes zero. Consequently, we find the thermopower in the bulk specimen as shown in Ref. [1]. In the second case when $b \gg 1$ (ultra thin films) for the thermopower we find the expression

$$\alpha_{\rm f}^{\rm np} = -\frac{k_0}{e}\Big\{3 + \ln(\frac{k_0 T}{\pi\hbar s n_{\rm el} d^2})\Big\}. \quad (12)$$

Note that for the non-degenerate electrons, $k_0 T \gg \hbar s n_{\rm el} d^2$. Therefore, as seen from Eq. (12), $\alpha_{\rm f}^{\rm np}$ increases when $n_{\rm el}$ and d decrease.

To estimate the effect of non-parabolicity on thermopower, it is useful to take size-dependent parts of thermopower $\Delta\alpha = \alpha_{\rm f} - \alpha_m$. Then the ratio of these parts for nonparabolic and parabolic cases becomes

$$\frac{\Delta\alpha_{\rm f}^{\rm np}}{\Delta\alpha_{\rm f}^{\rm p}} = \frac{\ln(\frac{b^2}{2\pi e})}{\ln\sqrt{\frac{4\varepsilon_1^*}{e}}}, \quad (13)$$

where ε_1^* and b are given in Eqs. (6) and (8), respectively, and e is the base of the natural logarithm. The ratio Eq. (13) is defined for the nondegenerate electron gas in ultrathin films. One can show that for the reasonable values of thickness and temperature this ratio becomes more than one. For example, for d in a few of nm,

$$T \approx 300\,{\rm K}, \quad \frac{\Delta\alpha_{\rm f}^{\rm np}}{\Delta\alpha_{\rm f}^{\rm p}} \approx 3.$$

Thus in the case of nondegenerate electron gas, the effect of non-parabolicity reduces the enlargement of thermopower.

First of all, it is noted that the effect of non-parabolicity on thermopower for the degenerate electron gas is determined wholly by the in-plane electron effective mass, $\frac{\alpha_{\rm f}^{\rm np}}{\alpha_{\rm f}^{\rm p}} = \frac{m_{\shortparallel}}{m_n}$. The behavior of this ratio is given in Fig. 1. As is seen, there are the oscillations of this ratio with respect to thickness ($u = \frac{d}{d_n}$). The period of oscillations as a function of electron concentration is

$$\Delta d = \left(\frac{\pi}{2n_{\rm el}}\right)^{\frac{1}{3}}(u_{\bar{n}_z+1} - u_{\bar{n}_z}), \tag{14}$$

where $u_{\bar{n}_z} = [\bar{n}_z^3 - \varphi(\bar{n}_z)]^{\frac{1}{3}}$. Note that, the points $u_{\bar{n}_z}$ are the ones of maxima thermopower (of in-plane effective mass) and between these points ($u_{\bar{n}_z+1} \prec u \prec u_{\bar{n}_z}$) there is the point of minimum; $u_{\min} = (2\varphi(\bar{n}_z))^{\frac{1}{3}}$.

Figure 2 shows the oscillation behavior of the ratio $\frac{\alpha_{\rm f}^{\rm np}}{\alpha_m^{\rm np}}$ as a function of thickness. As is seen, the points $u_{\bar{n}_z}$ for this ratio are not the points of maxima, they are the jumping ones of it. In addition, between these points of jumping this ratio has no point of minimum, it is a monotonically decreasing function of thickness. Thus the effect of non-parabolicity changes the character of thickness dependence of the thermopower and in-plane effective mass. Figures 1 and 2 show the influence of non-parabolicity on the magnitude of thermopower and effective mass. As it is seen that thermopower (in-plane effective mass) increases with the non-parabolicity parameter γ. For example, the ratio $\frac{\alpha_{\rm f}^{\rm np}}{\alpha_{\rm f}^{\rm p}}$ at any point, that is, $u = \frac{d}{d_n} = 1.5$, is 3.7 at $\gamma = 7$, while this ratio is 7.9 at $\gamma = 15$. The rise of thermopower becomes significant in the case of ultrathin films and strong non-parabolicity ($u \ll 1; \gamma \gg 1$). In this case from Eq. (4) $\frac{\alpha_{\rm f}^{\rm np}}{\alpha_{\rm f}^{\rm p}} \approx \sqrt{\frac{5}{2}\frac{\gamma}{2u}}$. Thus if one takes $u \approx 0.2$ and $\gamma \approx 4$, the ratio $\frac{\alpha_{\rm f}^{\rm np}}{\alpha_{\rm f}^{\rm p}} \approx 15.8$; while for the same u and $\gamma = 16$ this ratio becomes four times more, i.e. $\frac{\alpha_{\rm f}^{\rm np}}{\alpha_{\rm f}^{\rm p}} \approx 63.2$.

Due to the Fermi energy also oscillated with respect to thickness like that in Fig. 1 we do not present the graphic of Fermi energy as a function of thickness.

Now, let us treat the case of non-degenerate electron gas. The expressions of thermopower in this case are given in Eqs. (8) and (9) for the films of arbitrary thickness and strong non-parabolicity. In the case of ultrathin films with the non-parabolic energy band, Eq. (12) is obtained from Eqs. (8) and (9), which determines the thickness and concentration dependences of thermopower. One can rewrite the argument of the natural logarithm in Eq. (12) using the parameter b as

$$\frac{k_0 T}{\pi\hbar s n_{\rm el} d^2} = \frac{1}{b n_{\rm el} d^3}.$$

Note that for the electron gas to be in a non-degenerate state it must satisfy the condition $bn_{\rm el}d^3 \ll 1$. On the other hand, for electrons with a strong non-parabolic energy band, the condition of size quantization is $b \gg 1$. At first glance, it might be considered that one of these conditions ($b \gg 1$) contradicts the other. However, it is easy to see that for the semiconductor thin films there is a sufficiently large interval for temperature, where these conditions together satisfy $n_{\rm el}d^3 \ll \frac{T}{T_d} \ll 1$, with $T_d = \frac{\pi\hbar s}{k_0 d} \approx 2,3(10^3 d)^{-1}$ (d and $n_{\rm el}$ taken in cm and cm^{-3}, respectively). For example, at the thickness $d \approx 10^{-6}$ cm, $n_{\rm el} \approx 10^{16}$ cm^{-3} we have $23 \ll T \ll 2300$ K. As is seen, the width of temperature interval, where the electron gas is in size quantized and non-degenerate states, is determined by thickness and concentration.

Now let us treat the experimental results reported in the literature. First, consider the concentration dependence of thermopower, which was studied experimentally in Ref. [11] on the nanocrystals of PbTe and in Ref. [12] on the films of InN. The results of these studies show that the thermopower increases when the concentration decreases. Such a behavior of thermopower is in agreement with Eqs. (4) and (5) obtained from Eq. (3). In fact, analysis of Eqs. (4) and (5) shows that for the films with a parabolic energy band and for ultrathin films with a strong non-parabolic energy band, thermopower changes as $n_{\rm el}^{-1}$. However, in thin films in arbitrary thickness and non-parabolicity, the concentration dependence of thermopower becomes much weaker than $n_{\rm el}^{-1}$. For example, at finite d and $\varepsilon_{\rm g}$ (d and $\varepsilon_{\rm g}$ are not around zero) the concentration dependence of thermopower leads to $n_{\rm el}^{-\frac{1}{2}}$. The parameters which determine the thickness dependence of thermopower also depend on concentration. In fact, the minimum point for thermopower of nano-films, as is noted above, is a function of concentration: $d_{\min} = (\frac{5\pi}{n_{\rm el}})^{1/3}$. The concentration dependence of the minimum point in nano-films of lead chalcogenides was measured in Ref. [8]. The results of that work are

$$\begin{aligned} &d_{1\,\min}({\rm PbS}) = 18.1\,{\rm nm}, \quad (n_{\rm el} = 2.5 \times 10^{18}\,{\rm cm}^{-3}) \\ &d_{2\,\min}({\rm PbSe}) = 18\,{\rm nm}, \quad (n_{\rm el} = 4 \times 10^{18}\,{\rm cm}^{-3}) \\ &d_{3\,\min}({\rm PbTe}) = 26.3\,{\rm nm}, \quad (n_{\rm el} = 0.72 \times 10^{18}\,{\rm cm}^{-3}). \end{aligned}$$

The data obtained with respect to formula $d_{\min}$ given above are

$$\begin{aligned} &d_{1\,\min}({\rm PbS}) = 19\,{\rm nm}, \\ &d_{2\,\min}({\rm PbSe}) = 15.7\,{\rm nm}, \\ &d_{3\,\min}({\rm PbTe}) = 22.05\,{\rm nm}. \end{aligned}$$

It can be seen that $d_{\min}$ increases with the electron concentration ($n_{\rm el}$) and there is a reasonable agreement with the experimental data. The thickness dependence of thermopower was measured in Ref. [9] on

thin films of PbTe/B_i, where the oscillating behavior was observed. The analysis of these oscillations shows that the period of oscillations decreases as a function of thickness, which is in agreement with Eq. (14). In fact, in view of Eq. (14), as the thickness $\bar{n}_z$ increases, $u_{\bar{n}_z+1} - u_{\bar{n}_z}$ decreases and consequently Δd decreases. The non-monotonic dependence of thermopower with thickness was also observed on PbTe nano-films in Refs. [13,14] and on PbS, PbSe, PbTe nanostructures in Ref. [8]. Non-monotonic thickness dependence of Fermi energy was also observed on StTe quantum wells in Ref. [10]. The experimental data are also in qualitative agreement with our results.

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