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**SOME PECULIARITIES OF
THERMOPOWER ANISOTROPY IN
UNDEFORMED AND ELASTICALLY DEFORMED
n - Si AND *n* - Ge SINGLE CRYSTALS**

*In the framework of anisotropic scattering theory the mechanisms of origination of thermopower anisotropy in multi-valley undeformed and directionally deformed semiconductors (*n* -Si and *n* -Ge) in the presence of temperature gradient were analyzed in detail. Based on the use of the generalized Ohm's law the thermopower tensor was defined under $\nabla T \neq 0$.*

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Introduction

Electrophysical properties of semiconductors are essentially dependent not only on the anisotropy of dispersion law and the mechanisms of charge carrier scattering on phonons and impurity centres, phonon-phonon scattering, etc. (i.e. on microlevel anisotropy), but also on the reduced (for instance, by means of directional elastic deformation) anisotropy of the entire crystal [1 – 3]. A deeper insight into the mechanisms of origination of thermopower anisotropy, peculiarities of the energy spectrum of charge carriers is promoted by studies of thermoelectric properties of uniaxially elastically deformed semiconductors that are important from the standpoint of not only basic, but also applied science [4]. Practical importance of such studies is related to the possibility of creating (or increasing) by means of directional deformation of semiconductors the thermopower anisotropy which is used in the anisotropic thermoelements, eddy and galvanothermomagnetic energy converters [5]. It is exactly the efficiency of thermoelectric energy converters that are commonly referred to as modules that governs the possibilities of practical applications of thermoelectricity [6].

An essential drawback of thermoelectrically anisotropic materials used for industry is their low thermoelectric figure of merit. As long as one of the methods for improving the latter is to increase thermopower anisotropy, creation of high thermopower anisotropy by deformation will assure the possibility of designing artificial high-sensitivity anisotropic devices with higher efficiency.

The purpose of this work was to establish specific features of thermopower anisotropy in undeformed and elastically deformed *n* – Si and *n* – Ge single crystals, as well as to determine thermopower tensor in the presence of temperature gradient.

Anisotropic electron scattering in multi-valley n-type semiconductors Ge and Si

An appropriate idea of anisotropic electron scattering in a crystal appears when solving a kinetic Boltzmann-Lorentz equation for charge carriers in solids. This equation for steady-state conditions can be written as [7]:

$$\hat{D}n_{\vec{k}} + \sum_{\vec{k}'} W_{\vec{k}\vec{k}'} (n_{\vec{k}'} - n_{\vec{k}}) = 0, \quad (1)$$

where $n_{\vec{k}}$ is equilibrium electron distribution function; $\hat{D}n_{\vec{k}} = \frac{1}{kT} \frac{dn_{\vec{k}}^0}{dx_{\vec{k}}} (\nu_{\vec{k}}, \nabla\bar{\mu} + x_{\vec{k}} \nabla kT)$ is the so-called field term; $\nabla\bar{\mu} = \nabla\mu - e_0 E = \nabla(\mu + e_0\psi)$; ψ is electric field potential; e_0 is electron charge; $n_{\vec{k}}$ is the nonequilibrium addition to distribution function; $W_{\vec{k}\vec{k}'}$ is probability of electron scattering from the state with quasi-pulse \vec{k} to the state with quasi-pulse \vec{k}' .

Hereinafter we shall consider only elastic scattering whereby it can be considered that quasi-pulses \vec{k} and \vec{k}' are on the same constant-energy surface. In this case, scattering probability is a function of several values:

$$W_{\vec{k}\vec{k}'} = W(\varepsilon; \vartheta, \varphi, \vartheta', \varphi') \quad (2)$$

where ϑ, φ are spherical angles characterizing quasi-pulse direction prior to scattering, and ϑ', φ' – after scattering. In so doing, ε enters (2) as a parameter, since electron energy at scattering is not changed. Therefore, in the following the value of ε will not be entered into the notation of W function.

Let us first assume that scattering probability depends only on scattering angle θ :

$$W_{\vec{k}\vec{k}'} = W(\cos\theta), \quad (3)$$

in which case

$$\cos\theta = \cos\vartheta \cos\vartheta' + \sin\vartheta \sin\vartheta' \cos(\varphi - \varphi'), \quad (4)$$

It means that scattering probability does not depend on the way the angle formed by pulses \vec{k} and \vec{k}' is oriented inside a crystal (Fig. 1). Exactly such scattering is commonly referred to as isotropic. It is noteworthy that sometimes isotropic scattering shall mean the case when $W_{\vec{k}\vec{k}'}$ does not depend on θ at all. However, in reality this case is just a variety of isotropic scattering.

Anisotropic scattering shall mean the case when $W_{\vec{k}\vec{k}'}$ depends on the angles $\vartheta, \varphi, \vartheta', \varphi'$ in any way different from dependence (3). Hereinafter we shall also assume that constant-energy surface is square-shaped (sphere, ellipsoid). It also includes the Kane nonparabolicity. Then carrier velocity components $\nu_x \sim \sin\vartheta \cos\varphi$; $\nu_y \sim \sin\vartheta \sin\varphi$; $\nu_z \sim \cos\vartheta$ linearly depend on first-order spherical harmonics. Solution of the kinetic equation for isotropic scattering is of dissimilar nature than for the case of anisotropic scattering.

With the isotropic scattering $W(\cos\theta)$ can be expanded into a series in the Legendre polynomials:

$$W(\cos\theta) = \sum_l W_l P_l(\cos\theta). \quad (5)$$

And then, using known theory of summation of spherical functions

$$P_l(\cos\theta) = \sum_m Y_{lm}(\vartheta, \varphi) Y_{lm}^*(\vartheta', \varphi'), \quad (6)$$

Eq.(5) can be represented as

$$W(\cos\theta) = \sum_{lm} W_l Y_{lm}(\vartheta, \varphi) Y_{lm}^*(\vartheta', \varphi'). \quad (7)$$

Solution of the kinetic equation in this case should be sought for in the form of a series in spherical harmonics:

$$n_{\vec{k}}' = \sum_{lm} X_{lm}(\varepsilon) Y_{lm}(\vartheta, \varphi), \quad (8)$$

in so doing, $X_{lm}(\varepsilon)$ need to be defined. One can make sure that substitution of Eq. (8) into Eq. (1) yields the following relationship for X_{lm} :

$$L_l X_{lm}(\varepsilon) = D_m \delta_{l1}, \quad (9)$$

where L_l is in a definite way calculated through coefficients W_l . It follows from here that all $X_{lm}(\varepsilon)$, except for $X_{1m}(\varepsilon)$, are equal to zero, which yields known solution of the kinetic equation with the isotropic scattering.

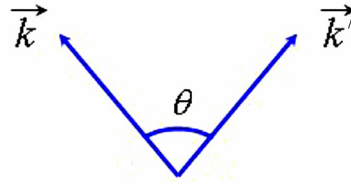


Fig. 1. Angle formed by quasi-pulses \vec{k} and \vec{k}' .

In the case of the anisotropic scattering the expansion of $W_{\vec{k}\vec{k}'}$ in spherical harmonics is of the form

$$W_{\vec{k}\vec{k}'} = \sum_{l'l'm'm'} W_{l'l'}^{m'm'} Y_{lm}(\vartheta, \varphi) Y_{l'm'}^*(\vartheta', \varphi'). \quad (10)$$

And, hence, the fundamental difference between the anisotropic and isotropic scattering is that in the case of the anisotropic scattering the expansion coefficients of $W_{l'l'}^{m'm'}$, first, depend on m and, second, are non-diagonal both in l and in m . As a result, with the anisotropic scattering for unknown functions $X_{lm}(\varepsilon)$ we obtain a system of coupled equations, rather than an autonomous equation

$$\sum_{l'm'} L_{l'l'}^{m'm'} X_{l'm'}(\varepsilon) = D_m \delta_{l1}. \quad (11)$$

And to calculate the kinetic coefficients, one need not know the entire distribution function assigned by series (8), but only the so-called abbreviated function

$$\bar{n}_{\vec{k}}' = \sum_m X_{1m}(\varepsilon) Y_{1m}(\vartheta, \varphi), \quad (12)$$

since, due to the foregoing, for charge carriers with a quadratic constant-energy surface charge carrier velocities are proportional to first-order spherical harmonics.

In terms of mathematics, construction of anisotropic scattering theory comes down exactly to development of methods for determination of $X_{1m}(\varepsilon)$ from system (11) [8, 9], and the difference

between the anisotropic and isotropic scattering in conceptual sense reduces to the fact that in the case of anisotropic scattering the relaxation properties of carrier system are determined not by one coefficient L_1 , but by certain combinations of the entire collection of expansion coefficients $L_{ll'}^{mm'}$.

Exactly for this reason instead of the relaxation time one should introduce the relaxation time tensor. Naturally, this changes the temperature and concentration dependences of the kinetic coefficients, their dependences on the magnetic field intensity and other external influence.

Scattering anisotropy in multi-valley semiconductors can be due to different reasons, namely scattering potential anisotropy (piezoelectric semiconductors, scattering on ions with dielectric constant anisotropy), phonon spectrum anisotropy, carrier energy spectrum anisotropy. In the latter case charge carrier scattering is described by a series of parameters, for instance, not by deformation potential constant (like in the case of isotropic scattering), but by the tensor of deformation potential constants, not by effective mass value, but by components of effective mass tensor, etc. Anisotropic scattering theory offers the possibility to find a dependence of experimentally measured values on the above parameters, which cannot be achieved through use of isotropic scattering theory.

Determination of the thermopower tensor

The Seebeck effect (or thermoelectric effect) discovered in 1823 consists in the origination in a semiconductor in the presence of temperature difference of electric field which in this case is called thermoelectric.

The generalized Ω law relating the tensors of current density vector \vec{j} and electric field intensity $\vec{\varepsilon}$ in the presence of temperature gradient $grad T \equiv \nabla T$, for the anisotropic media can be written as [9]:

$$\vec{j} = \hat{\sigma} \vec{\varepsilon} - \hat{\sigma} \hat{\alpha} \nabla T, \quad (13)$$

where $\hat{\sigma}$ and $\hat{\alpha}$ are second-rank tensors of electric conductivity and thermopower, respectively. If we introduce notation

$$\hat{b} = \hat{\sigma} \hat{\alpha}, \quad (14)$$

Eq.(13) can be written as follows:

$$\vec{j} = \hat{\sigma} \vec{\varepsilon} - \hat{b} \nabla T, \quad (15)$$

or in components

$$j_i = \sigma_{ik} \varepsilon_k - b_{il} \nabla_l T, \quad (16)$$

where

$$b_{il} = \sigma_{im} \alpha_{ml}. \quad (17)$$

Thermoelectric field intensity is found in the absence of electric current. On entering $\vec{j} = 0$ into (13), we obtain the following expression for thermoelectric field intensity:

$$\vec{\varepsilon} = \hat{\alpha} \nabla T, \quad (18)$$

or in components

$$\varepsilon_i = \alpha_{ik} \nabla_k T. \quad (19)$$

Formula (18) giving a linear dependence of thermoelectric field intensity on temperature gradient is the first term of the expansion of $\vec{\varepsilon}$ in ∇T and defines thermoelectric effect in the framework of linear in ∇T theory, valid at low temperature gradients, when one can ignore expansion terms with higher powers.

Based on (14), the Seebeck coefficient tensor is determined as follows:

$$\hat{\alpha} = \hat{\sigma}^{-1} \hat{b}, \quad (20)$$

On the other hand, based on the kinetic theory, current density can be represented as

$$\vec{j} = e \int_0^{\infty} \psi_1(E) \vec{v} g(E_k) dE_k, \quad (21)$$

where e is electron charge, \vec{v} is group carrier velocity, $g(E_k)$ is the density of states, E is full carrier energy, E_k is kinetic carrier density, ψ_1 is addition to equilibrium distribution function in the presence of disturbing fields, that is:

$$\psi = \psi_0 + \psi_1. \quad (22)$$

From the kinetic Boltzmann equation

$$\frac{\partial \psi}{\partial t} = \left(\frac{\partial \psi}{\partial t} \right)_{field} + \left(\frac{\partial \psi}{\partial t} \right)_{collision} \quad (23)$$

for the steady-state conditions and in the relaxation time approximation for ψ_1 (in conformity with [10]) we obtain

$$\psi_1 = -\tau \frac{\partial \psi_0}{\partial E} \left[\frac{\mu - E}{T} \nabla T - \nabla(\mu + eV) \vec{v} \right], \quad (24)$$

where τ is relaxation time, μ is chemical potential, T is absolute temperature, V is electric potential. In formula (23) $\left(\frac{\partial \psi}{\partial t} \right)_{field}$ is a field term that defines the velocity of change in ψ distribution function as

a result of continuous electron motion in normal (geometric) space and in velocity space, $\left(\frac{\partial \psi}{\partial t} \right)_{collision}$ is a collision term that defines the velocity of change in ψ as a result of electron collision (scattering).

Substituting (24) into (21) and comparing the resulting expression to formula (15) written for the isotropic case, when b and σ are scalars, we shall find their (b and σ), and on the basis of (20) in the isotropic case for the Seebeck coefficient we shall find the relationship:

$$\alpha = \frac{k_0}{e} \left[\frac{\int_0^{\infty} \tau(E_k) \frac{\partial \psi_0}{\partial E} E \vec{v}^2 g(E_k) dE_k}{k_0 T \int_0^{\infty} \tau(E_k) \frac{\partial \psi_0}{\partial E} \vec{v}^2 g(E_k) dE_k} - \mu^* \right], \quad (25)$$

or

$$\alpha = \frac{k_0}{e} \left[\frac{\iiint_{-\infty}^{\infty} \tau \frac{\partial \psi_0}{\partial E} E \vec{v}^2 d^3 \vec{k}}{k_0 T \iiint_{-\infty}^{\infty} \tau \frac{\partial \psi_0}{\partial E} \vec{v}^2 d^3 \vec{k}} - \mu^* \right], \quad (26)$$

where k_0 is the Boltzmann constant, $\mu^* = \frac{\mu}{k_0 T}$ is reduced chemical potential. In (26) integrals are taken over quasi-pulse space.

In the general case of nonspherical surfaces of charge carrier energy constant, the relaxation time is a second-rank tensor. Having written (15) and (21) in the main axes of the inverse effective

mass tensor, for the components of thermopower tensor of electrons belonging to one minimum we obtain the following expression:

$$\alpha_{ii} = \frac{k_0}{e} \left[\frac{\iiint_{-\infty}^{\infty} \tau_{ii} \frac{\partial \Psi_0}{\partial E} E v_i^2 d^3 \vec{k}}{k_0 T \iiint_{-\infty}^{\infty} \tau_{ii} \frac{\partial \Psi_0}{\partial E} v_i^2 d^3 \vec{k}} - \mu^* \right]. \quad (27)$$

Mechanisms of origination of thermopower anisotropy

The anisotropy of electric conductivity, thermal conductivity and a number of other physical properties is typical (to a varying degree) of all noncubic crystals. However, thermopower anisotropy is a rather rare phenomenon, as long as for its manifestation a series of specific conditions must be met. For the electric conductivity anisotropy, for instance, it is sufficient to have either anisotropic energy spectrum or anisotropic scattering law, or both.

The situation is different with thermopower anisotropy. According to anisotropic scattering theory based on the method for solution of kinetic equation [8], the effect of scattering anisotropy on the kinetic coefficients for such most common scattering mechanisms as scattering on acoustic and optical phonons, on the ionized and neutral impurities (dipoles), is taken into account by the relaxation time tensor given below:

$$\tau_{ii} = \lambda_{ii} E_k^r. \quad (28)$$

In so doing, the tensor character of relaxation time is described by coefficients λ_{ii} that do not depend on energy, but are the functions of the effective mass anisotropy. Power exponent r depends on the type of charge carrier scattering in a crystal. At scattering on acoustic lattice vibrations and optical vibrations at higher than the Debye temperatures (θ) $r = -\frac{1}{2}$, at scattering on optical vibrations at $< \theta$ $r = 0$, at scattering on ionized impurities $r = \frac{3}{2}$.

On substituting (28) into (27), coefficient λ_{ii} will reduce, and for α_{ii} we obtain a scalar expression. Thus, if there is one type of charge carriers and one scattering mechanism, the thermopower will be isotropic, whatever strong is the anisotropy of charge carrier effective mass. This result was used by the authors of [11] in the consideration of a multi-valley model. It is acceptable both for the isotropic dispersion law $E(\vec{k})$ and for the anisotropic parabolic and nonparabolic ellipsoidal law (the Kane model). Most likely, exactly this accounts for the fact that thermopower anisotropy is less common than the electric conductivity anisotropy.

If $E(k_i)$ is characterized by different dependence along different directions (nonparabolic and nonellipsoidal dispersion law), in this case (even with one type of carriers and one scattering mechanism) the thermopower anisotropy can be observed, as mentioned in [12].

It should be noted that at scattering on impurity ions the relaxation time in the form of (28) is some approximation. According to [8], coefficients λ_{ii}'' are energy dependent, this dependence being different for different directions in a crystal. On substituting (28) into (27), $\lambda_{ii}''(E)$ in this case will not be reduced, and owing to the fact that they are different for different directions, α_{ii} for

different i will differ from each other. That is, thermopower anisotropy will be observed in this case. This mechanism of thermopower anisotropy origination is typical of heavily doped semiconductors.

In the case of one type of carriers and two scattering mechanisms described by relaxation times $\tau_{ii}^{(1)} = \lambda_{ii}^{(1)} E^{r_1}$ and $\tau_{ii}^{(2)} = \lambda_{ii}^{(2)} E^{r_2}$, summation of inverse relaxation times will yield

$$\tau_{ii} = \frac{\lambda_{ii}^{(1)} \lambda_{ii}^{(2)} E^{r_1+r_2}}{\lambda_{ii}^{(1)} E^{r_1} + \lambda_{ii}^{(2)} E^{r_2}}. \quad (29)$$

Substitution of (29) into (27) will result in the expression

$$\alpha_{ii} = \frac{\int \frac{E^{r_1+r_2}}{\lambda_{ii}^{(1)} E^{r_1} + \lambda_{ii}^{(2)} E^{r_2}} \frac{\partial \Psi_0}{\partial E} v_i^2 (E - \mu) d^3 \vec{k}}{eT \int \frac{E^{r_1+r_2}}{\lambda_{ii}^{(1)} E^{r_1} + \lambda_{ii}^{(2)} E^{r_2}} \frac{\partial \Psi_0}{\partial E} v_i^2 d^3 \vec{k}}. \quad (30)$$

In this case the thermopower tensor does not reduce to a scalar and thermopower will be anisotropic. Thus, one of the reasons for origination of thermopower anisotropy can be the availability of two (or several) anisotropic scattering mechanisms. However, thermopower anisotropy caused by joint action of several scattering mechanisms is low and at scattering on phonons and ions, ions and dipoles, phonons, ions and dipoles is $0.1 \div 25.0 \mu\text{V/degree}$.

Another reason for thermopower anisotropy can be the presence of two (or greater number) charge carrier types with one scattering mechanism, if at least one group of carriers is characterized by anisotropic electric conductivity and their partial Seebeck coefficients are different. This is due to the fact that thermopower, unlike electric conductivity, is not an additive value. Really, let us denote through $j_i^{(1)}$ and $j_i^{(2)}$ current densities of "1" and "2" type carriers and write for them the generalized Ohm's law (16):

$$\left. \begin{aligned} j_i^{(1)} &= \sigma_{ii}^{(1)} \varepsilon_i - b_{ii}^{(1)} \frac{\partial T}{\partial x_i}, \\ j_i^{(2)} &= \sigma_{ii}^{(2)} \varepsilon_i - b_{ii}^{(2)} \frac{\partial T}{\partial x_i}. \end{aligned} \right\} \quad (31)$$

Total current density equals:

$$j_i = j_i^{(1)} + j_i^{(2)} = \left(\sigma_{ii}^{(1)} + \sigma_{ii}^{(2)} \right) \varepsilon_i - \left(b_{ii}^{(1)} + b_{ii}^{(2)} \right) \frac{\partial T}{\partial x_i} = \sigma_{ii} \varepsilon_i - b_{ii} \frac{\partial T}{\partial x_i}. \quad (32)$$

From condition of $j_i = 0$ for the thermoelectric field we have the expression

$$\varepsilon_i = \frac{b_{ii}}{\sigma_{ii}} \frac{\partial T}{\partial x_i} = \frac{b_{ii}^{(1)} + b_{ii}^{(2)}}{\sigma_{ii}^{(1)} + \sigma_{ii}^{(2)}} \frac{\partial T}{\partial x_i}. \quad (33)$$

Whence for the thermopower tensor we shall have the relationship:

$$\alpha_{ii} = \frac{b_{ii}^{(1)} + b_{ii}^{(2)}}{\sigma_{ii}^{(1)} + \sigma_{ii}^{(2)}}. \quad (34)$$

On substituting b_{ii} based on (17) and taking into account that thermopower of one group of carriers with one scattering mechanism is isotropic, we finally obtain:

$$\alpha_{ii} = \frac{\alpha^{(1)} \sigma_{ii}^{(1)} + \alpha^{(2)} \sigma_{ii}^{(2)}}{\sigma_{ii}^{(1)} + \sigma_{ii}^{(2)}}, \quad (35)$$

and the difference in two components of the thermopower tensor $\hat{\alpha}$ in this case is expressed by the formula

$$\Delta\alpha = \alpha_{11} - \alpha_{22} = \frac{(\sigma_{11}^{(1)} \sigma_{22}^{(1)} - \sigma_{11}^{(2)} \sigma_{22}^{(2)}) (\alpha^{(1)} - \alpha^{(2)})}{(\sigma_{11}^{(1)} + \sigma_{11}^{(2)}) (\sigma_{22}^{(1)} + \sigma_{22}^{(2)})}. \quad (36)$$

Eq.(36) suggests that even with the isotropic partial Seebeck coefficients the thermopower of the entire semiconductor can be anisotropic, if the electric conductivity of each carrier type, or at least one of them, is of anisotropic nature. In so doing, naturally, partial thermopowers must be different. This mechanism of thermopower anisotropy is apparently realized in semiconductors of orthorhombic syngony *CdSb* and *ZnSb* and in pure *Bi* in the area of mixed conductivity. The value of $\Delta\alpha$ in this case can reach $\sim 300 \mu\text{V}/\text{degree}$.

Thermopower anisotropy caused by the presence of several groups of charge carriers with different thermoelectric properties and anisotropic electric conductivities can be artificially created by directional deformation of multi-valley semiconductors of *n-Ge* and *n-Si* type. Really, directional elastic deformation of such crystals (except for directions $\vec{X} // \vec{J} // [100]$ in *n-Ge* and $\vec{X} // \vec{J} // [111]$ in *n-Si*) will lead to inequality of energy minima [13] and redistribution of electrons between the minima. As long as the kinetic properties of electrons belonging to different valleys (energy minima) are now different because of their different population, and the electric conductivity of each of these groups is strongly anisotropic, according to (35) the thermopower of the entire crystal will become anisotropic. The emergence of particularly high anisotropy can be expected in the area of mixed conductivity, as long as uniaxial deformation of *Ge* and *Si* leads to removed degeneration of the zones of light and heavy holes at point $\vec{k} = 0$ of quasi-pulse space and to the emergence of considerable electric conductivity anisotropy of holes. Thus, uniaxial elastic deformation can cause the emergence of thermopower anisotropy even in materials which in the undeformed state are fully isotropic thermoelectrically.

There is another mechanism of thermopower anisotropy origination related to the manifestation of the electron-phonon drag effect. With the anisotropic nature of charge carrier energy spectrum, the phonon thermopower is anisotropic. In so doing, thermopower anisotropy can occur even with one type (variation) of carriers and one scattering mechanism, unlike the diffusion thermopower that has been analyzed above.

Conclusions

1. The ways of using anisotropic scattering theory for the analysis of the kinetic effects in the presence and absence of temperature gradient on *n-Ge* and *n-Si* crystals under study were considered.
2. Based on the use of the generalized Ohm's law the thermopower tensor was defined under crystal conditions $\nabla T \neq 0$.

3. The most important mechanisms of thermopower anisotropy origination were analyzed in detail, and emphasis was placed on the peculiarities of this anisotropy in multi-valley semiconductors (*n-Ge* and *n-Si*) with their directional (uniaxial) elastic deformation.

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