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Experimental and Theoretical Expansion of the Phenomenology of Thermoelectricity

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EXPERIMENTALANDTHEORETICALEXPANSIONOFTHEPHENOMENOLOGYOFTHERMOELECTRICITY

Experimental and Theoretical Expansion of the Phenomenology of Thermoelectricity

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I. Introduction

The symmetry of the exposure on the p-n junction of the luminous flux and the heat flow is fundamentally different.

For the photoelectric effect, because of the small of the photon momentum, one can confine ourselves to a nonpolar and even isotropic consideration of the effect of light on the p-n junction and, as a consequence, a parametric description of the effect on its characteristics. That is why for the photo-effect within the framework of the linear E-N phenomenology we allow a parametric approach.

For the heat flow, its polar, object-changing effect determines the parameters of the EMF of the macroscopic response. In the longitudinal thermoelectric effect, the polarity of the heat flow determines the polarity of the EMF (for electrons and holes - different), in the transverse thermoelectric effect, the polarity of the heat flow determines the polarity of the EMF similarly, and the orientation of the heat flux relative to the symmetry axes of the crystal or the artificial object affects the magnitude of the EMF.

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Therefore, the Parametric Supplement to the theory of p-n junction allows, albeit roughly, but quantitatively, to describe the photo-effect - in the first approximation, the current-voltage bias. Whereas a similar parametric approach for thermoelectric effects gives only the correct sign of thermo-EMF, and their values tend to infinity at arbitrarily small displacement currents of the p-n junction. But since estimates of the magnitude of the current above the potential barrier on the basis of the Richardson formula gave negligible currents, then these "infinite" EMFs were neglected, assuming their output power to be immeasurable - below the Nainquist noise.

However, experiments on p-n junctions have shown that the values of thermoelectric currents, even at small temperature drop at the p-n junction, are much higher than the Richardson estimates. A rigorous ballistic model of the motion of electrons over a potential barrier showed that the Richardson formula itself, historically constructed to describe the arc current between carbon electrodes at 3000 degrees, is applicable only to the diffuse approximation for large temperature differences and lowers the current value by orders of magnitude for small temperature drop. Thus, both from experiments and from theory it follows that for small temperature differences at the p-n junction we have not infinite but giant EMF and completely measurable currents much higher than thermal noise.

Thus, for the heat flow in the sharply inhomogeneous media, there is no basis for neglecting the additional force: to the E-N phenomenology of the temperature force or to the thermoelectric E-T phenomenology of the concentration force. And to build a non-linear description of processes corresponding to effects on a micro- and nano-scale, one must begin with a strict definition of the zero and first approximations, i.e. with linear phenomenology, which includes both electric, thermal and concentration forces.

Earlier attempts to combine the phenomenology of macroscopic thermoelectricity with the description of contact phenomena were carried out formally, without a general phenomenological approach. The formal transfer of particular models of potential barriers, as will be shown below, did not give a complete self-consistent description and led to contradictions of its individual fragments.

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Located on the neighboring chapters of the monographs on the physics of semiconductors and semiconductor devices of the theory of thermoelectricity [1] and contact phenomena [2] could not not be crossed in any way (could not fail to intersect), since Ideally (and in theory!) the thermoelectric device has two contacts of materials with different types of conductivity (Figure 1a).

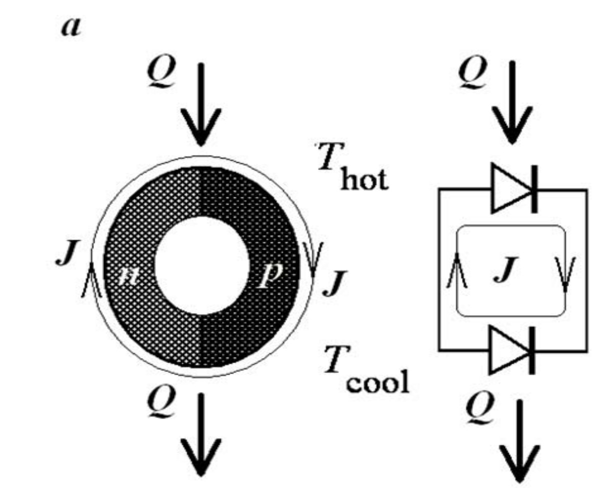

Figure 1: a - The thermoelement and its and equivalent circuit

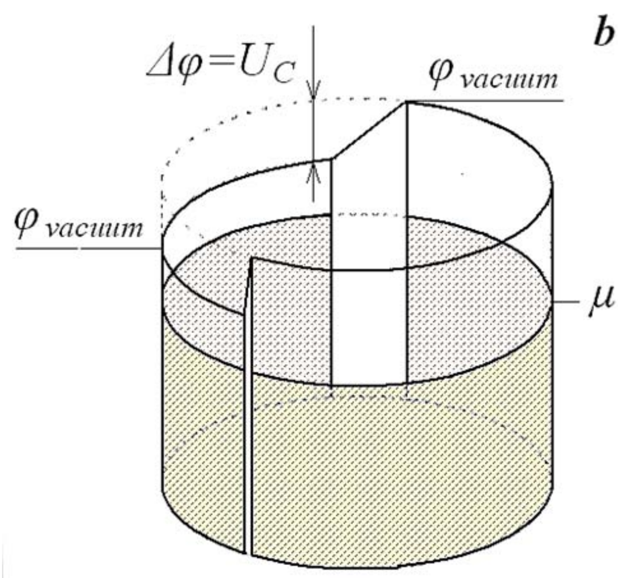

Figure 1: b –The location of the vacuum level (and the forbidden band edge in semiconductors) relative to the level of the chemical potential μ.

Therefore, it is quite natural that the macroscopic thermoelectric power of the material itself was tied to the dependence of the contact potential difference on temperature:

$$\Delta\varphi(T_{\text{hot}}) - \Delta\varphi(T_{\text{cool}}) = U_c(T_{\text{hot}}) - U_c(T_{\text{cool}})$$

(Figure 1b). This attempt led Peltier to the erroneous conclusion about the contact nature of the heat of Peltier he had discovered at the contact. And this erroneous representation, contrary to the principles of symmetry, [3] is included in many of the letters on thermoelectricity, although it has long been corrected [4, 5, 6].

In the framework of this erroneous conception, the attempt to take into account the concentration effects in thermoelectricity was reduced to the speculative application of correction: using a he "chemical" potential and the "observable" electric force F_E^* [3]. With the help of artificially introduced terms, in fact, the EMF of the p-n junction was not painted at a drop on it, but the difference between the EMF of hot and cold p-n junctions whose plates have the same temperature. In the framework of this approach, the equation for the flux of concentration simply "fitted" into the modified equation for an electric current with noncanonical coefficients a:

$$J_E = L_{EE}\cdot F_E + \frac{1}{e}\cdot L_{EE}(!)\cdot F_N + a_{ET}\cdot F_T = a_{EE}\cdot F_E^* + a_{ET}\cdot F_T \quad (1)$$

But we will not be distracted by the analysis of the nonstrict, non-canonical and boundedly applicable equation (1). Moreover, this equation was not used anywhere, except in a simplified and internally contradictory theory. And in macroscopic thermoelectric devices, the p-n junction was simply interfering, and its effect on both contacts (Figure 1) was leveled by a specially developed ohmic contact technology.

In the theory of the p-n junction, as was noted in the first part of this work [7], the small diffuse effects of Seebeck and Peltier and, corresponding to them, a small cross-sectional kinetic coefficient, were rightly dropped from consideration. And in practice, electronic devices based on p-n junctions also used ohmic contact, leveling the action of the opposing barrier layer.

As was shown in the first part, the analysis of non-canonical coefficients requires strict consideration of the conditions for their measurement and! their use is associated with a violation of a number of principles that are strictly applicable only to canonical coefficients. And the extension of the linear phenomenology of thermoelectricity proposed in this paper, the analysis of local effects essentially simplifies and makes more rigorous.

II. Materials and Methods

Samples used to study the appearing on the contacts, in particular in p-n junctions of local thermo-EMFs, the technology of their manufacture and various methods for their measurement, as well as the results of experiments and the first, trial attempts to explain them, described earlier in the cycle noted in the first part of the work, are reflected also in the reviews [8. 9].

III. Results

a) Result of previous experiments.

The conclusion from this cycle of studies, qualitatively confirmed by microscopic analysis, is shown in Figure 2

Experiments have shown that the local thermo-EMF corresponds to a maximum in the dimensional dependence of the thermoelectric power α shown in Figure 2. At the maximum, the volt-watt sensitivity is 3-4 orders of magnitude higher than for the Seebeck macroscopic thermoelectric effect. The experimentally observed thermo currents exceeding in the region of the maximum of the size dependence of the estimate based on Richardson's formula by two orders of magnitude were explained within the framework of the ballistic model [10].

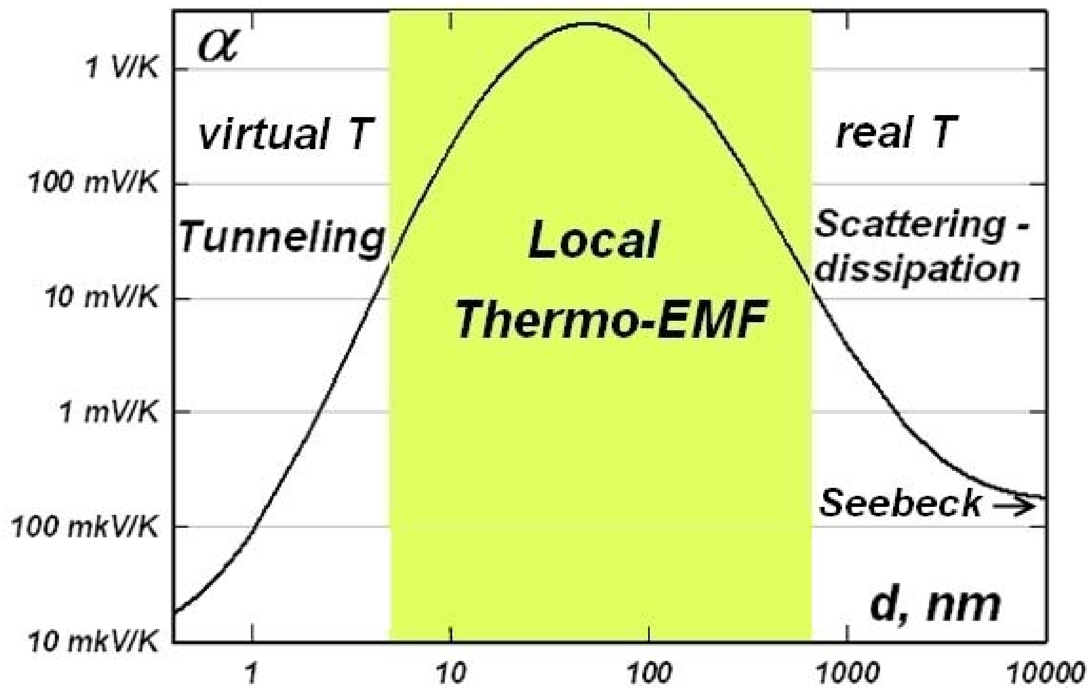

Figure 2: Model (Planck) thermoelectric power dependence on the thickness of the potential barrier

b) Resonance studies of local thermo-EMFs

In this paper, we present only those experimental results that relate to the resonance recorded on local thermo-EMF. Resonance is registered in the figures shown in Figure 3 detectors developed on the basis of longitudinal local thermo-EMFs. When continuous irradiation was applied to the detector, thermo-EMFs, antiphase photo-EMFs were registered.

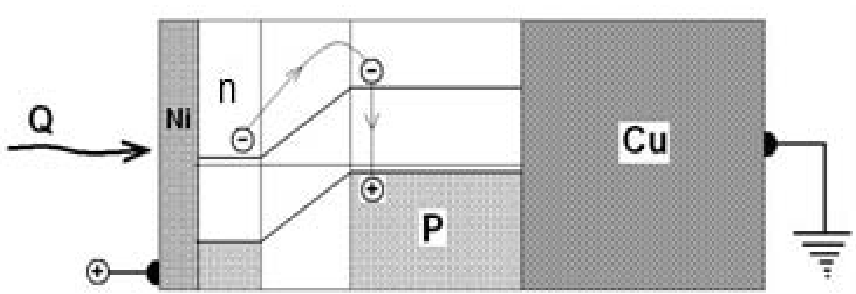

Figure 3: Scheme of generation of the longitudinal thermo-EMF at the p-n junction and its measurement (on the left) and the prototype of the detector under investigation (right)

The frequency dependence of the amplitude of the alternating signal generated by local thermo-EMF at the high-frequency edge of the frequency characteristic of the photoelectric effect for a given p-n junction (without an absorbing metal coating) is observed as a hump resonance (Figure 4).

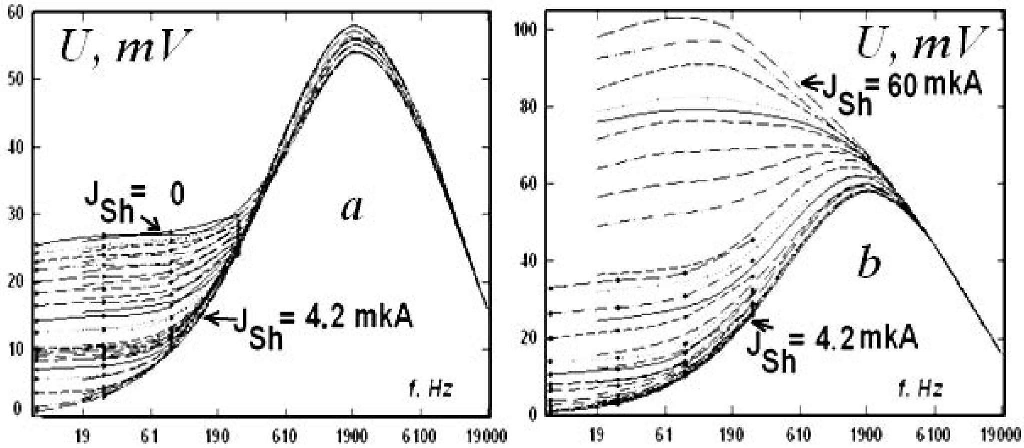

Figure 4: Dependence of the frequency response of local thermo-EMF on the bias current through the p-n junction

When the p-n junction is shifted by a direct current, it is possible to achieve complete compensation of the DC voltage due to the local thermo-EMF by the antiphase voltage of the external current source. In this case, the amplitude of local thermo-EMF at low frequencies passes through zero (Figure 4a), and with a further increase in current increases in antiphase (Figure 4b). With a displacement current determined for a particular p-n junction configuration, the resonance is manifested in the classical form (Figure 5).

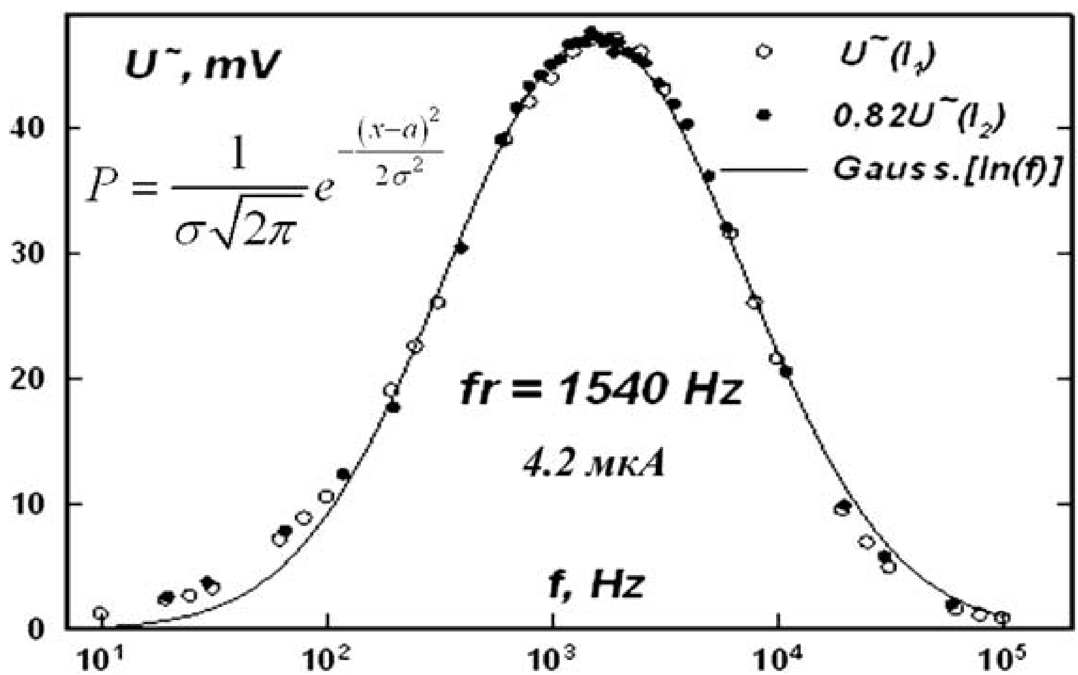

Figure 5: The resonance of the local thermo-EMF measured in the p-n junction, measured at two powers of the modulated heat flow and the corresponding Gaussian distribution

The shape of the resonance is close to the normal (Gauss) (one-dimensional) distribution - probability density P, but! of the logarithm of frequency

$$U = U^{\max} e^{-0.5\left(\frac{x-x_0}{B}\right)^2}, \quad U^{\max} = 47.2056, \quad B = 1.5118 \Leftrightarrow \sigma \mapsto 1.0690 \tag{2}$$
$$x = \ln(f), \quad x_0 = 7.3456 = \ln(f_r) \Leftrightarrow a$$

Since any of the velocity components of electrons in thermal motion is normally distributed, the shape of the resonance directly indicates its statistical nature. In this case, the dependence on the logarithm of the frequency is more general, more corresponding to an unlimited frequency range than the dependence simply on the frequency, which describes well the narrow frequency resonances. Correction of Gaussian distribution is beyond the scope of this paper.

On the other hand, the very occurrence of any resonance is determined by the dependence on the frequency of the phase shift between the flow (displacement) and the driving force. In the simplest electrical resonance, the phase shift is given by reactive elements. In the resonance of local thermo-EMF in the p-n junction, in addition to its reactive electrical parameters, it is necessary to take into account the phase shifts of the electric and thermal forces, the integral effect of which forms the electron flux.

IV. Discussion

And so, for large concentration gradients in thermoelectric phenomenology, it is necessary to use a three-component linear thermoelectric E-N-T phenomenology rather than the traditional system of two coupled equations of generalized thermodynamic forces and flows. That is, it is necessary to use the system extended to three equations, which in the one-dimensional case has the following form:

$$\left.\begin{aligned} J_E &= L_{EE}\cdot F_E + L_{EN}\cdot F_N + L_{ET}\cdot F_T \\ J_N &= L_{EN}\cdot F_E + L_{NN}\cdot F_N + L_{NT}\cdot F_T \\ J_T &= L_{ET}\cdot F_E + L_{NT}\cdot F_N + L_{TT}\cdot F_T \end{aligned}\right\} \quad (3)$$

In the extended system of equations (3), in addition to the direct effects that are traditionally taken into account in thermoelectricity: the drift current - $L_{EE}\cdot F_E \to \sigma\cdot E$ and the heat flux - $L_{TT}\cdot F_T \to (\kappa + \mathrm{K}^*)\cdot \Delta T$, the diffusion flux- $L_{NN}\cdot F_N \to D\cdot\Delta T$, which has long been "allowed" in the theory of the p-n junction, but so far "forbidden" in the thermoelectric theory, is strictly taken into account. Additional effects-terms of the equations, crosswise with the diffusion flux are also taken into account.

Moreover, as an additional direct coefficient, each additional cross-ratio corresponds to effects already known from other sections of physics, but not fully taken into account or not considered in thermoelectricity (and p-n junction theory).

In addition to the cross thermoelectric current $L_{ET}\cdot F_T \to \alpha\cdot\sigma\cdot\Delta T$, two additional flows are taken into account in the system of equations (3). An electro-diffusion current $L_{EN}\cdot F_N \to R\cdot\Delta N$ is taken into account, which, in the theory of the p-n junction, must be taken into account, but, as shown above, strictly speaking, was not taken into account. And we took into account the Sorud-Duffur thermo diffusion current $L_{NT}\cdot F_N \to \Phi\cdot\Delta N$, which was only remembered in the physics of semiconductor devices when studying the degradation of p-n junctions, but was not taken into account in its work.

It is to these three equations (2) for the conducting medium (as shown in the first part, after subtracting the phonon and plasma parts - is discarded K^*), one can apply the Onsager symmetry principle, which, in the first approximation, reduces the number of independent coefficients for an ideal medium to 6: up to three direct and three cross.

In principle, it is possible to reduce the number of independent coefficients, since all 3 direct (diagonal) drift coefficients are interrelated - they are determined by the mutual friction of the respective streams about each other.

Using an analysis of fluctuations of independent forces in a freely suspended sample similar to that described in the previous article, we obtain from the system of equations (3) the following system of equalities:

$$\left.\begin{aligned} 0 &= L_{EE}\cdot F_E + L_{EN}\cdot F_N + L_{ET}\cdot F_T \\ 0 &= L_{EN}\cdot F_E + L_{NN}\cdot F_N + L_{NT}\cdot F_T \\ 0 &= L_{ET}\cdot F_E + L_{NT}\cdot F_N + L_{TT}\cdot F_T \end{aligned}\right\} \quad (4)$$

The system of three equations (4), in addition to the trivial solution: $F_E = 0,\ F_N = 0,\ F_T = 0$, as well as

$$\left.\begin{aligned} -\left(L_{ET}^2 L_{NN} - 2L_{EN}L_{ET}L_{NT} + L_{EN}^2 L_{TT}\right) &= L_{EE}\left(L_{NT}^2 - L_{NN}L_{TT}\right) \\ +\left(L_{NT}^2 L_{EE} - 2L_{EN}L_{ET}L_{NT} + L_{EN}^2 L_{TT}\right) &= L_{NN}\left(L_{ET}^2 - L_{EE}L_{TT}\right) \\ +\left(L_{NT}^2 L_{EE} - 2L_{EN}L_{ET}L_{NT} + L_{ET}^2 L_{NN}\right) &= L_{TT}\left(L_{EN}^2 - L_{EE}L_{NN}\right) \end{aligned}\right\} \quad (5)$$

In this case, we find that the cross-correlation coefficients for three thermodynamic forces, similar to those considered in the first part of special cases with two forces, are associated only with a pair of corresponding direct coefficients, but they have an additional factor corresponding to the forces considered:

$$L^i_{EN} = C^i_{EN} \cdot \sqrt{L_{EE} \cdot L_{NN}}, \quad L^i_{ET} = C^i_{ET} \cdot \sqrt{L_{EE} \cdot L_{TT}}, \quad L^i_{NT} = C^i_{NT} \sqrt{L_{NN} \cdot L_{TT}} \qquad (6)$$

In expressions (6), the factors $\mathbf{C}^{\mathbf{i}}$, as in two-component phenology, are determined by the "hidden" parameters, which give a degeneracy for direct coefficients. It should be noted that the more general consideration of the three-component phenomenology revealed the interdependence of the "allowed" cofactors $\mathbf{C}^{\mathbf{i}}$, which was missed when considering the partial two-component phenomenology. The first 4 solutions ($\mathbf{i}$ - the solution number) for the factors $\mathbf{C}^{\mathbf{i}}$.

i	C^i_{EN}	C^i_{ET}	C^i_{NT}
1	-1	-1	+1
2	+1	-1	-1
3	-1	+1	-1
4	+1	+1	+1

(7)

$\mathbf{C}^{\mathbf{i}}$ and in the three-component case one simply determines the sign in the expressions for the cross-correlation coefficients, but the signs are "resolved" only in a combination shown in the matrix (7), which is in strict accordance with the Onsager symmetry principle and for two-component phenomenology (analogous solutions can be obtained for the Casimir principle of symmetry, but they are not thermo-electric).

The first four solutions providing a conditionally-single-phase balance of three forces correspond to real pair-cross coefficients with factors $\mathbf{C}^{\mathbf{i}} = \pm 1$. In this case, the counter-phase nature of the sum of the cross-terms in the equations of system (4) to the direct term is ensured so that the cross members themselves can only be either in phase with each other or in antiphase.

But, unlike the two-component cases, when the "hidden" parameters give only a sign depending on the type of charge carriers, for the three-component phenomenology for pair-cross-correlation coefficients, we have 8 additional solutions for "allowed" complex factors whose absolute magnitudes are equal $1/\sqrt{2}$.

i	C^i_{EN}	C^i_{ET}	C^i_{NT}
5	$-\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$-\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$-\frac{1}{4}+i\frac{\sqrt{7}}{4}$
6	$\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$-\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$\frac{1}{4}-i\frac{\sqrt{7}}{4}$
7	$-\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$\frac{1}{4}-i\frac{\sqrt{7}}{4}$
8	$\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$-\frac{1}{4}+i\frac{\sqrt{7}}{4}$
9	$-\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$-\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$\frac{5}{8}-i\frac{\sqrt{7}}{8}$
10	$\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$-\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$-\frac{5}{8}+i\frac{\sqrt{7}}{8}$
11	$-\frac{1}{4}-i\frac{\sqrt{7}}{4}$	$\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$\frac{1}{4}+i\frac{\sqrt{7}}{4}$
12	$\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$\frac{1}{4}+i\frac{\sqrt{7}}{4}$	$-\frac{1}{4}-i\frac{\sqrt{7}}{4}$

(8)

$$
\begin{array}{cccc}
i & \mathrm{Arg}[C_{EN}^{i}] & \mathrm{Arg}[C_{ET}^{i}] & \mathrm{Arg}[C_{NT}^{i}] \\
5 & \pi - \mathrm{ArcTan}[\sqrt{7}] & \pi - \mathrm{ArcTan}[\sqrt{7}] & \pi - \mathrm{ArcTan}[\sqrt{7}] \\
6 & -\mathrm{ArcTan}[\sqrt{7}] & \pi - \mathrm{ArcTan}[\sqrt{7}] & -\mathrm{ArcTan}[\sqrt{7}] \\
7 & \pi - \mathrm{ArcTan}[\sqrt{7}] & -\mathrm{ArcTan}[\sqrt{7}] & -\mathrm{ArcTan}[\sqrt{7}] \\
8 & -\mathrm{ArcTan}[\sqrt{7}] & -\mathrm{ArcTan}[\sqrt{7}] & \pi - \mathrm{ArcTan}[\sqrt{7}] \\
9 & -\pi + \mathrm{ArcTan}[\sqrt{7}] & -\pi + \mathrm{ArcTan}[\sqrt{7}] & -\mathrm{ArcTan}\left[\frac{\sqrt{7}}{5}\right] \\
10 & \mathrm{ArcTan}[\sqrt{7}] & -\pi + \mathrm{ArcTan}[\sqrt{7}] & \pi - \mathrm{ArcTan}\left[\frac{\sqrt{7}}{5}\right] \\
11 & -\pi + \mathrm{ArcTan}[\sqrt{7}] & \mathrm{ArcTan}[\sqrt{7}] & \mathrm{ArcTan}[\sqrt{7}] \\
12 & \mathrm{ArcTan}[\sqrt{7}] & \mathrm{ArcTan}[\sqrt{7}] & -\pi + \mathrm{ArcTan}[\sqrt{7}]
\end{array} \quad (9)
$$

The "allowed" combinations of complex factors obtained from the vanishing of the fluctuation currents in the one-dimensional case (or in the isotropic case, for collinear thermodynamic forces) can be useful only for estimating the noise level in measurements on a constant heat flux.

But they directly point out that under certain "hidden" parameters conditions can be realized when the contributions from cross effects are shifted in time on the phase concerning the contribution of the direct effect in the equations for the fluxes (4). In this case, when the alternating heat flux is applied to the p-n junction, the resulting relationships give, without loss-damping, the dependence of the magnitude of the local thermo-EMF on the phases of these contributions.

It was checked that the shift of the resonance frequency to the low-frequency or high-frequency region requires connection of a capacitance or inductance by several orders of magnitude larger than the corresponding values typical for the p-n junction and measuring circuits.

Thus, it was shown that it is the phase shift of the contributions of different thermodynamic forces to the total fluxes that determines how with zeroing of the signal at low frequencies, so the resonance itself, and the resulting expressions (6) allow to make not only qualitative conclusions, but also to carry out quantitative calculations.

V. Conclusion

The local thermo-EMFs detected at the contacts were investigated in p-n junctions based on silicon, which is a contact with highly reproducible properties. In the frequency dependences of local thermo-EMF, Gauss resonance was recorded, which directly indicated the balance / imbalance of the phase of contributions from various thermodynamic forces to the signal.

The expansion of the phenomenology of thermoelectricity necessary to take into account the concentration force in the first, linear approximation gave a description of the dependence of the total fluxes on the phase balances / imbalances of contributions on direct and cross effects.

The obtained results already qualitatively allow to optimize micro- and nano-devices in terms of their efficiency, which is essential both for thermoelectric conversion and for any other elements of microelectronics.

The solutions of the extended system of equations (2) for non-zero flows needed for quantitative calculations of the efficiency of micro- and nano-devices will be presented in the next article.

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