On the Nernst effect in fluctuating superconductors: Serbyn, Skvortsov, and Varlamov reply

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This is an extended Reply to Comment by A. Sergeev, M.Y. Reizer, and V. Mitin [arXiv:0906.2389] on our Letter [Phys. Rev. Lett. **102**, 067001 (2009)]. We explicitly demonstrate that all claims by Sergeev et al. are completely unfounded, because their underlying theoretical work contains multiple errors and inconsistencies. For this reason, there is no need to revise the existing theories of thermoelectric response in superconductors.

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INTRODUCTION

In a series of recent papers (see, Ref. [1] and references therein), Sergeev, Reizer, and Mitin (SRM) have argued that all existing results on thermoelectric response in fluctuating superconductors are fundamentally flawed and must be revisited. According to SRM, these previous allegedly incorrect works include the pioneering works of Ullah and Dorsey [2] and those of Ussishkin, Sondhi, and Huse [3–5], the relevant chapter in the book of Varlamov and Larkin [6], our recent Letter [7], and the related work of Michaeli and Finkelstein [8]. Note that these researches [2–8] have employed at least four different approaches to calculate the Nernst effect in a superconductor: Time-dependent Ginzburg-Landau theory [3, 5, 6], microscopic techniques based on the Matsubara diagrammatic technique [4, 7] and on the quantum kinetic equation [8], and finally simple phenomenological arguments, which relate the Nernst coefficient to the temperature dependence of the chemical potential for the carriers [7]. All these approaches consistently provide the same order of magnitude for the Nernst coefficient, which has been shown to much exceed that in a Fermi liquid (hence, we called the effect "giant" in our Letter). Such a giant Nernst signal has been detected in a variety of well-known experiments in the high-temperature cuprates [9] and conventional superconducting films [10, 11]. Therefore the existence of giant Nernst effect constitutes a solid experimental fact, which has been the actual motivation for the aforementioned theoretical works. However, SRM "strongly object" to the existence of these results and experimental facts, arguing [12] that the "numerous recent theories grossly overestimate the thermomagnetic coefficients" and calling them in the abstract of their recent comment [1] "ridiculously large." These strong claims of Sergeev *et al.* are based solely on their own alternative calculation approach, sketched by them in Ref. [12].

In 2009 SRM had escalated the concern about the existing theories of the Nernst effect, by posting a comment [1] on our Letter. Despite SRM's evident errors, their Comment has recently been accepted for publication in Physical Review Letters. This manuscript is an extended version of our Reply.

Our Reply contains the following: (i) In the first part, we analyze the paper [12], which is needed because all SRM's criticism on the existing theories is based on this single paper. Hence, a careful analysis of Ref. [12] is the only means for us to refute SRM's criticism of the works by us and others. As a result of this exercise, we are able to show in the first part of the Reply that Ref. [12] contain serious errors. We identify the main problems of SRM's treatment as likely originating from the combination of inconsistent use of a specific gauge in a single calculation, missing pieces in the relevant diagrams, and technical mistakes. (ii) Next, we suggest specific technical steps to remedy these problems of SRM and bring their approach in accordance with the exiting theories. But most importantly, we conclude that since the paper [12] is manifestly incorrect, there is no need to revise all previous existing theories. (iii) In the last part, we elaborate on the phenomenological Eq. (1) that we suggested in our Letter [7] as a simple intuitive argument behind the experimentally observed giant Nernst effect.

ANALYSIS OF PHYS. REV. B 77, 064501 (2008) BY SERGEEV ET AL.

Ref. [12] concentrates entirely on the discussion of "a gauge-invariant microscopic approach," but we show below that it is the very gauge-invariance that is explicitly violated in the SRM's treatment. Below we choose to focus on two specific technical problems (among many) that suffice to prove the inadequacy of their proposed calculational method and therefore make the criticism mute.

On the calculations of the Nernst effect in a normal metal by Sergeev et al.

One of only a few existing and commonly-accepted results on the Nernst effect is that of Sondheimer [13], who first calculated the effect in a Fermi gas back in 1948. Any technique that claims to be of relevance to more complicated Nernst phenomena, such as those due to superconducting fluctuations, must recover Sondheimer's formula as a basic "sanity check". In Sec. II of their work, SRM do consider the case of a normal metal but fail to explain the origin of Sondheimer's [13] formula in a comprehensible way. In addition, their discussion contains an error, which propagates into further analysis.

For noninteracting fermions, the thermoelectric response is described by the Kubo-like diagram constructed of the heat and electric vertices and two Green functions. In the absence of a magnetic field, the heat vertex is $\xi_{\mathbf{p}}\mathbf{v} = \xi_{\mathbf{p}}\mathbf{p}/m$. For nonzero magnetic field, both the kinetic energy $\xi_{\mathbf{p}}$ and the momentum $\mathbf{p}$ itself, should be modified by the vector potential $\mathbf{A}$. SRM forget about the latter and thus come to a *wrong expression* for the heat current (Eq. (5) of Ref. [12]):

$$\mathbf{J}^h_{\mathrm{SRM}} = \sum_{\mathbf{p}} \mathbf{v}\xi_{\mathbf{p}} a^+_{\mathbf{p}} a_{\mathbf{p}} + \sum_{\mathbf{p}} \frac{e\mathbf{v}}{c}(\mathbf{v}\cdot\mathbf{A}) a^+_{\mathbf{p}} a_{\mathbf{p}}. \quad (1)$$

Further they forget to incorporate the vector potential $\mathbf{A}$ into the electric current to maintain gauge invariance. Then the correlator $\langle j^Q j^e \rangle$ is claimed to be calculated, but (i) a gauge in which calculations are performed is poorly defined and (ii) the external momentum is unspecified (below, we explain why it is important). We reiterate here that while SRM concentrate their work as well as their criticism of others on the gauge-invariance issue, which indeed is important, they fail to satisfy the gauge invariance in their own work, even at the non-interacting level. Their "gauge-invariant" expressions contain explicitly a vector potential, which is inserted into selected pieces of diagrams in an uncontrolled fashion and calculations are performed in an unspecified gauge.

On the calculations of the Nernst effect in a fluctuating superconductor by Sergeev et al.

In Sec. II of Ref. [12] Sergeev *et al.* analyze the heat current transferred by fluctuating Cooper pairs in the vicinity of T_c. They correctly obtain that the heat current is proportional to the gauge-invariant momentum (Eq. (11) of Ref. [12]):

$$\mathbf{B}^h_{\mathrm{SRM}} \propto \omega(\mathbf{q} + 2e\mathbf{A}/c), \quad (2)$$

and they claim that it is the term with $\mathbf{A}$ that is their key new finding and that this term was allegedly overlooked in all previous calculations. However, SRM again fail to include $\mathbf{A}$ in the electric vertex and draw the diagrams, extracting $\mathbf{A}$ from the propagators and from the heat vertex (2) only. Then they say: "Further calculations of the diagrams for Cooper pairs are similar to that for noninteracting electrons", thus inheriting all the inconsistencies from their normal-metal treatment. Even without that, *the expressions determining current* (1) and (2) *are different in their vector structure*: the first one contains $\mathbf{v}(\mathbf{v}\cdot\mathbf{A})$, while the second does not!

Correcting the problem with gauge invariance

Here we elaborate on the main technical problems of SRM's calculations and provide technical details on how to correctly enforce gauge-invariance for observables. Of course, thermal conductivity and any other linear response for a physical observable must be gauge-invariant independently of the means used to calculate it. In the framework of Kubo diagrammatic approach (both for a normal metal and a superconductor close to T_c), the relevant diagrams for thermoelectric response contain three sources of the vector potential/magnetic-field dependence: (A) Green functions/propagators, (B) heat current vertex, and (C) electric current vertex. The resulting expression is gauge invariant only if all these three sources are taken into account in a consistent fashion and within a specific gauge. In Ref. [12] it is claimed that the contribution (B) cancels the contribution (A) calculated by Ussishkin in Ref. [4]. However the consistency between calculations and gauge choices in the two parts of the same physical quantity is not discussed. Most importantly, contribution (C) is not even mentioned by Sergeev et al., which makes their conclusion erroneous. This contrasts sharply with our approach, where we follow the work of Galitski and Larkin [14], and from the outset use Landau representation that automatically takes into account all sorts of couplings to the magnetic field in a manifestly gauge-invariant fashion.

We have found that the use of Landau basis greatly simplifies the calculations and automatically circumvents the need to keep track of gauge invariance at intermediate steps. However, near T_c, the use of this method is not required and correct result can be obtained via perturbative expansions of all elements of the diagram in the vector potential. In this approach, care is required to keep the gauge invariance under control at all stages of the calculation. The correct starting point is to consider the correlator

$$\Pi_{\alpha\beta}(\mathbf{k}, \mathbf{Q}) = \langle j^Q_\alpha(\mathbf{k} + \mathbf{Q}) j^e_\beta(-\mathbf{k}) \rangle, \quad (1)$$

where $\mathbf{Q}$ is the momentum associated with the vector potential: $\mathbf{A}(\mathbf{r}) = \mathbf{a}\, e^{i\mathbf{Q}\cdot\mathbf{r}}$. Setting $\mathbf{k} = 0$ implies that the momentum $\mathbf{Q}$ comes into the heat vertex, while setting $\mathbf{k} = -\mathbf{Q}$ corresponds to $\mathbf{Q}$ coming out of the electric vertex. The limit $\mathbf{k} \to 0$ and $\mathbf{Q} \to 0$ needs to be taken in the end.

Now we expand all terms (two vertices and two propagators) to linear order in $\mathbf{A}$. In the most general form, this procedure generates four contributions $\Pi^{(m)}$ ($m = 1, \ldots, 4$) of the form (we retained the leading order in $\mathbf{k}$ and $\mathbf{Q}$):

$$\Pi^{(m)}_{\alpha\beta} = \mathcal{S}^{(m)}_{\alpha\beta\gamma\mu} k_\gamma a_\mu + \mathcal{T}^{(m)}_{\alpha\beta\gamma\mu} Q_\gamma a_\mu. \qquad (2)$$

Since $\mathcal{S}^{(m)}_{\alpha\beta\gamma\mu} \neq 0$, the relative contribution of these four terms retains the value of $\mathbf{k}$. This is a reason why it is important to specify $\mathbf{k}$ when calculating each of the four gauge-noninvariant pieces. Of course, $\sum_m \mathcal{S}^{(m)}_{\alpha\beta\gamma\mu} = 0$, indicating that there is a well-defined limit $\Pi_{\alpha\beta}(0, \mathbf{Q})$. The contributions $\mathcal{T}^{(m)}_{\alpha\beta\gamma\mu}$ are generally non-zero and explicitly gauge-dependent, but their sum is certainly gauge-invariant:

$$\Pi_{\alpha\beta} \propto (Q_\alpha a_\beta - Q_\beta a_\alpha) \int d^d q \, q^2 [L_+^2 L_- - L_+ L_-^2], \qquad (3)$$

where $L_+ = L(i\varepsilon_n + i\omega_\nu, q)$ and $L_- = L(i\varepsilon_n, q)$ are the fluctuation propagators. After summing over the Matsubara frequencies, ε_n, and performing an analytic continuation in ω_ν we come to the following result for the off-diagonal thermomagnetic response:

$$\tilde{\beta}^{xy}_{\text{C.p.}} \propto \int d^d q \, q^2 \int d\omega \, \omega \frac{\partial B(\omega)}{\partial \omega} |L(\omega, q)|^2 \, \mathrm{Im}\, L(\omega, q), \qquad (4)$$

where $B(\omega) = \coth(\omega/2T)$ is the bosonic equilibrium distribution function, and

$$L(\omega, q) = -\frac{8T_c/\pi\nu}{(8/\pi)(T - T_c) - i\omega + Dq^2} \qquad (5)$$

is the retarded propagator for Cooper pairs. Evaluating the integrals in Eq. (4), one immediately finds a finite value for Nernst coefficient $\propto (T - T_c)^{d/2-2}$ even in the absence of the particle-hole asymmetry, in accordance with all previous works in the field [2–8].

Thermomagnetic response: bosons vs. fermions

To clarify the relation between the thermomagnetic responses of fluctuating Cooper pairs in the vicinity of T_c [Eq. (4)] and that of normal electrons, β^{xy}_n, we find it instructive to present the latter in a similar form:

$$\beta^{xy}_n \propto \int d^d p \, p^2 \int d\varepsilon \, \varepsilon \, \frac{\partial F(\varepsilon)}{\partial \varepsilon} \, |G(\varepsilon, p)|^2 \, \mathrm{Im}\, G(\varepsilon, p), \qquad (6)$$

where $F(\varepsilon) = \tanh(\varepsilon/2T)$ is the fermionic equilibrium distribution function, and

$$G(\varepsilon, p) = \frac{1}{\varepsilon - \xi_{\mathbf{p}} + i/2\tau} \qquad (7)$$

is the retarded electron Green function. The similarly looking expressions (4) and (6) lead to essentially different results for the Nernst coefficient for electrons and Cooper pairs. For free fermions in the absence of the particle-hole asymmetry, momentum integration in Eq. (6) gives an ε-independent constant, and the subsequent energy integral vanishes by oddness. On the other hand, for fluctuating Cooper pairs the integrals in Eq. (4) lead to a finite Nernst coefficient even in the absence of the particle-hole asymmetry. The same conclusion can be readily achieved within the time-dependent Ginzburg-Landau approach [6].

Role of magnetization

Sergeev et al. [1] claim that the contribution of magnetization is not relevant for calculating the Nernst coefficient. This statement evidently contradicts the well established theory of thermomagnetic effects (see, e.g., [15]). According to the theory, the experimentally measured value of the thermoelectric tensor $\beta_{\alpha\beta}$ is given by a sum of the kinetic contribution, $\tilde{\beta}_{\alpha\beta}$, (which can be calculated using the Kubo-like approach, see above) and a thermodynamic contribution due to magnetization $\mathbf{M}$:

$$\beta^{\alpha\beta} = \tilde{\beta}^{\alpha\beta} + \epsilon_{\alpha\beta\gamma} c M_\gamma / T. \qquad (8)$$

The importance of the magnetization heat current for the Nernst effect was first demonstrated by Obraztsov in 1965 [16]. Later, the significance of this contribution to thermomagnetic response of electron systems has been emphasized by a number of authors in relation to the integer quantized Hall effect, interacting electron gas in a quantizing magnetic field [17], and the fluctuation Cooper pairs contribution to the Nernst effect within Ginzburg-Landau formalism [3, 6]. The crucial role of the magnetization contribution to the Nernst effect has recently been demonstrated in Refs. [7, 8] where it cancels the otherwise divergent kinetic contribution $\tilde{\beta}_{xy}$ at low temperatures above H_{c2}, thus being eventually responsible for the implementation of the third law of thermodynamics.

SRM argued that magnetization effects do not contribute to the heat transport in a magnetic field, and the second term in Eq. (8) should be omitted. This statement contradicts all known theories and is incorrect. As a side remark, we mention here that even if the magnetization contribution is omitted, the Nernst effect in fluctuating superconductors would still be giant provided that the error in calculating $\tilde{\beta}_{xy}$ made in Ref. [12] is corrected.

Finally, it also should be mentioned that the splitting of the Nernst coefficient (8) in a kinetic (Kubo-like) and thermodynamic (magnetization) contribution is completely analogous to that in the Hall conductivity: $\sigma_{xy} = \sigma^{\mathrm{I}}_{xy} + \sigma^{\mathrm{II}}_{xy}$ [18].

Summary

To summarize this part: pretending to have developed a gauge-invariant microscopic approach to heat transfer, the article [12] starts with a manifestly gauge non-invariant expression for the heat current [Eq. (5)] even for free electrons. It is precisely the incomplete account for the gauge invariance for why the authors of Ref. [12] came to the wrong conclusion that the nonzero Nernst effect due to fluctuation Cooper pairs necessarily requires presence of the particle-hole asymmetry in the underlying electronic band structure in a metallic phase.

ON THE QUALITATIVE FORMULA FOR THE NERNST COEFFICIENT

We take the opportunity here to discuss in more detail the qualitative expression for the Nernst coefficient,

$$N = \frac{\sigma}{nq^2c}\frac{d\mu}{dT}, \tag{9}$$

which was suggested in our Letter [7] and that was also criticized by SRM [1].

We note that the main purpose of our work was to present a technically challenging microscopic calculation of the Nernst coefficient for an extended range of magnetic fields and temperatures. These microscopic calculations do not rely on any phenomenology such as Eq. (9), but are strongly supported by it. The main purpose of including Eq. (9) in our paper was to provide a simple intuitive argument behind the giant Nernst effect in a fluctuating superconductor, which may be of value to the wide audience of PRL. And we believe that phenomenological Eq. (9) is a new interesting (qualitative) result that does convey the desired message.

Application to fluctuating Cooper pairs

Arguing for the validity of *phenomenological* Eq. (9) we used the notion of a drift velocity and ignored impurity scattering of the charge carriers, keeping in mind that the fluctuating Cooper pairs are not scattered by elastic impurities (all information about such scattering is included in the value of the effective coherence length or equivalently in the effective mass of the Cooper pairs). Equation (9) clarifies the physical origin of the observed anomaly and, moreover, sheds light on where to look for the giant Nernst effect: in systems with chemical potential strongly dependent on temperature.

Now we clarify the issue related to the chemical potential of fluctuating Cooper pairs, $\mu_{\text{C.p.}}(T)$, which we introduced as an auxiliary concept within the phenomenological derivation of the Nernst effect. Indeed, it is known that in the thermodynamic equilibrium, the chemical potential of a system with a variable number of particles is zero, with photon and phonon gases being the textbook examples. A naïve application of this "theorem" to fluctuating Cooper pairs "gas" has lead SRM to a wrong conclusion that $\mu_{\text{C.p.}} = 0$ [1]. However, a delicate issue concerning Cooper pairs is that they do not form an isolated system but are composed of fermion quasiparticles which constitute another subsystem under consideration. According to the same textbook discussion [20], in a multicomponent system, the chemical potential of the i'th component, μ_i, can be defined as the derivative of the thermodynamic potential with respect to the number of particles of i-th sort:

$$\mu_i = (\partial\Omega/\partial N_i)_{P,V,N_j}\,, \tag{10}$$

provided the numbers of particles of all other species are fixed, $N_{j\neq i} = \text{const}$. In deriving the condition for thermodynamic equilibrium, one should now take into account that creation of a Cooper pair must be accompanied by removing two quasiparticles from the fermionic subsystem. This leads to $\mu_{\text{C.p.}} - 2\mu_n = 0$, where μ_n is the chemical potential of quasiparticles. Therefore, the equilibrium condition does not restrict $\mu_{\text{C.p.}}$ to zero, even though the number of Cooper pairs is not conserved.

The value of $\mu_{\text{C.p.}}(T) = T - T_c$ [7] can be found from Eq. (10) using the explicit temperature dependencies of fluctuation part of Gibbs potential Ω_{fl} and concentration of fluctuating Cooper pairs. After this Eq. (9) reproduces [7] the known results for fluctuating Nernst response near the classical superconducting transition point. This style of reasoning is in close analogy with the "pedestrian" approach to fluctuating paraconductivity [6], which employs the standard Drude formula with the assumption that the Ginzburg-Landau relaxation time plays the role of the scattering time. In this sense, Eq. (9) is an analogue of the classical Drude formula for electrical conductivity. This analogy also suggests that the domain of validity of Eq. (9) is constrained to temperatures close to T_c. In the opposite low-temperature limit, only microscopic calculations (such as presented by us in Ref. [7]) are reliable.

What concerns the applicability of Eq. (9) to a normal metal, one can easily check that it readily reproduces the Sondheimer's result.

CONCLUSIONS

To summarize, we list below certain points of the criticism of Ref. [1] together with our responses:

1. SRM claim that "the linear response calculation of β_{xy} does not require any magnetization correction" and present their own interpretation of various contribution to the heat current. — We cer-

tainly disagree with this statement. In such a delicate and controversial issue as heat transport, we consider counterproductive to discuss interpretations. Instead, one should either microscopically derive the expression for $\beta_{\alpha\beta}$ or check existing ones for possible inconsistencies. The crucial role of the magnetization contribution to β_{xy} has recently been demonstrated in Refs. [7, 8]: In particular, at $T \to 0$ it cancels the otherwise divergent $\tilde{\beta}_{xy}$, thus ensuring the implementation of the third law of thermodynamics. Therefore, omitting the magnetization contribution to β_{xy} as suggested by SRM will inevitably violate the fundamental law of thermodynamics.

2. SRM claim that the particle-hole asymmetry of the single-electron spectrum is necessary for a nonzero Nernst effect due to fluctuating Cooper pairs. — This wrong statement is solely based on the results of Ref. [12] which contains multiple errors, as discussed in details above.

3. SRM claim that our results [7] generalized to repulsive interaction in the Cooper channel would yield β_{xy} significantly exceeding that for non-interacting electrons. Further they claim that, "certainly, this effect is not known". — We believe that the first of these statements is correct and the Cooper-channel correction to β_{xy} is indeed nonzero in the absence of the particle-hole asymmetry. Similar effects have been theoretically predicted for fluctuation diamagnetism [21–23] and fluctuation conductivity [24]. However we disagree with the second statement of SRM: The relative values of various contributions to β_{xy} should be considered individually for each experiment.

4. SRM claim that, "according to textbooks [19], $\nabla\mu$ should always be included in the effective electric field". — That is true, the textbook condition of vanishing current, $E - \nabla\mu/e = 0$, is precisely our initial assumption of electroneutrality employed in the phenomenological treatment.

5. SRM claim that "a thermodynamic value of $\mu_{\mathrm{C.p.}}$ is always zero, because a number of pairs is not conserved". — This statement is wrong: The chemical potential of fluctuating Cooper pairs is nonzero in equilibrium since they do not form an isolated system.

Thus, it has been explicitly demonstrated that all SRM's ground-breaking claims, seemingly of fundamental importance, are completely unfounded, because the underlying theoretical work of SRM contains [12] multiple errors and inconsistencies. For this reason, there is no need to revise the existing theories of thermoelectric response in superconductors and in particular results of a microscopic analysis presented in our Letter [7]; they remain intact.

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