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Fluctuation thermopower above the superconducting transition temperature

Yong Lu† and Bruce R Patton

Department of Physics, The Ohio State University, Columbus, OH 43210, USA

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Abstract. The fluctuation contribution to the thermopower above the superconducting transition temperature T_c is calculated including all leading p–h asymmetric corrections. A fluctuation term is obtained which produces a peak in the thermopower at T_c in the dirty limit. The result is compared to some recent experiments on the high-T_c superconductors.

1. Introduction

Due to their very short intrinsic correlation length, the high-T_c copper oxide superconductors have demonstrated fluctuation effects in a much wider temperature range above T_c [1]. Among various fluctuation effects studied, the thermopower showed behaviour which was not well described by the previously published theory [2, 3, 4]. The correction to the normal-state diffusion thermopower due to superconducting fluctuations above T_c was studied theoretically by Maki using the time-dependent Ginzburg–Landau theory and shown to be dominated by the paraconductivity in all dimensions, resulting in a decrease in magnitude of the thermopower towards zero at T_c [2]. This prediction could not be experimentally verified for many years, due to the very narrow region of observable fluctuation effect in conventional superconductors and the difficulty in maintaining a tiny and precise temperature difference across a sample. Recently, by measuring the thermopower of $YBa_2Cu_3O_{7-\delta}$ with a high-temperature resolution near the superconducting T_c, Howson *et al* showed that the magnitude of the thermopower has a peak at the transition [3, 4]. This experimental result motivated us to re-examine Maki's theory. By carrying out a more complete microscopic calculation, we have found a new leading fluctuation correction to the normal-state thermopower near the T_c [5].

In this paper, our microscopic calculation for the fluctuation thermopower is first described, then the result is presented in comparison with Maki's result and with the experimental data, and some concluding remarks are made at the end.

2. The microscopic calculation

Although this study can be generalized to more realistic cases, we limit ourselves here to a simple situation: carriers of charge ($-e$) with free-electron-like dispersion interacting

† Current address: Department of Radiation Oncology, University of Chicago Medical Center, MC 0085, Chicago, IL 60637, USA.

through a weak BCS attraction ($-V_0$) in the presence of N_{imp} randomly distributed impurities with a potential U_{imp}.

The thermopower S can be generally expressed in terms of the electrical conductivity σ and the transport coefficient L^{21} [6]:

$$S = \frac{1}{T}\frac{L^{21}}{\sigma} \tag{1}$$

where T is the temperature and L^{21} is defined by $j^Q = L^{21}E$, where j^Q is the heat current and E is the electric field. In the case of an isotropic (s-wave scattering), which we assume for simplicity, the type of vertex correction shown in figure 1(a) vanishes and the normal-state conductivity is given by $\sigma_n = ne^2\tau/m$, where τ is the impurity scattering relaxation time. Within the linear response regime, L^{21} is obtained from the retarded correlation function $K^R_{j^Q j}$ of the heat current j^Q and the electrical current j:

$$L^{21} = -\lim_{\nu\to 0}\lim_{q\to 0}\frac{1}{\nu}\,\mathrm{Im}K^R_{j^Q j}(q,\ \nu) \tag{2}$$

where Im denotes the imaginary part. The retarded correlation function is the analytic continuation of the corresponding Matsubara correlation function

$$K_{j^Q j}(q,\ \mathrm{i}\nu_m) = -\int_0^\beta \mathrm{d}t\,\mathrm{e}^{\mathrm{i}\nu_m t}\langle T j^Q(q,\ t) j(-q,\ 0)\rangle \tag{3}$$

where $\beta \equiv 1/T$, $k_B \equiv 1$, $\nu_m = 2m\pi/\beta$, T is the imaginary-time (t) ordering operator, and $\langle\rangle$ denotes the thermal average. The current operators, including impurity contribution, can be derived from the conservation laws for charge and energy as [7]

$$j(q) = -\frac{e}{2m}\sum_k (2k+q)c_k^+ c_{k+q} \tag{4}$$

$$j^Q(q) = \frac{1}{4m}\sum_k (2k+q)(\xi_k + \xi_{k+q})c_k^+ c_{k+q} + \frac{U_{imp}}{2m}\sum_j^{N_{imp}}\sum_p\sum_k \mathrm{e}^{\mathrm{i}p\cdot R_j}(2k + p + q)c_k^+ c_{k+p+q} \tag{5}$$

where $\xi_k = \mathcal{E}_k - \mu$ is the energy measured from the chemical potential μ, R_j is the position of the jth impurity, and $\hbar \equiv 1$.

The main difficulty in evaluating $K_{j^Q j}$ is that it vanishes if the system has particle–hole (p–h) symmetry. Its leading contribution therefore arises from numerous small p–h asymmetric terms [8]. In particular, to the first order in T/μ, there are p–h asymmetric corrections from: (i) the electronic density of states $N(\xi)$; (ii) the momentum k; (iii) limits of the energy integral from $(-\mu)$ to ∞ [9]; and (iv) the impurity scattering self-energy $\sum_{imp}$ and vertex correction Γ. We included in our calculation, as shown below in more detail, all corrections from (i) to (iv). For isotropic U_{imp}, the vertex correction shown in figure 1(a) vanishes in L^{21} as in the conductivity σ. The vertex corrections which need to be taken into account in L^{21} are the impurity scattering corrections to the j^Q operator as

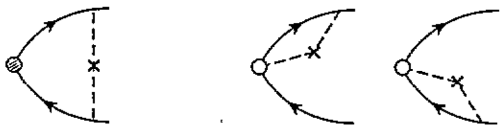

Figure 1. (a) The impurity vertex correction to σ and L^{21} which vanishes with isotropic impurity scattering and $q \to 0$; (b) impurity vertex corrections for the heat current in L^{21}. An open circle denotes a momentum vertex while a shaded one either a momentum or a momentum–energy vertex.

shown in figure 1(b) [10]. These corrections amount to replacing $(\xi_k + \xi_{k+q})$ in the heat current vertex by $[\xi_k + \sum_{imp}(\omega) + \xi_{k+q} + \sum_{imp}(\omega + \nu)]$.

With the above considerations, we have obtained L^{21} for the model system in the normal state (where the superconducting fluctuation is negligible):

$$L_n^{21} = -\frac{\pi^2}{3}\frac{ne\tau T^2}{m\mu} \tag{6}$$

which, together with the normal-state conductivity $\sigma_n = ne^2\tau/m$, gives the same free-electron thermopower $S_n = (1/T)L_n^{21}/\sigma_n$ as the Boltzmann equation result [11]:

$$S_n = -\frac{\pi^2 T}{3e\mu}. \tag{7}$$

As T approaches T_c, the fluctuation becomes more and more important. The thermopower can then be expressed as

$$S = \frac{1}{T}\frac{L_n^{21} + L_{fl}^{21}}{\sigma_n + \sigma_{fl}} \tag{8}$$

where the subscript fl indicates the fluctuation contributions. The behaviour of σ_{fl} has been extensively studied and is well understood both experimentally and theoretically [12]. For T sufficiently close to T_c and with certain amount of pair-breaking, σ_{fl} is dominated by the Aslamazov–Larkin (A–L) contributions, which diverge as $\epsilon^{(d-4)/2}$, where $\epsilon \equiv (T - T_c)/T_c$ [13]. In three dimensions ($d = 3$), it has the form

$$\sigma_{fl,AL} = \frac{1}{32}\frac{e^2}{\zeta(0)}\epsilon^{-1/2} \tag{9}$$

where $\zeta(0)$ is the superconducting correlation length at $T = 0$.

Our goal is to study the behaviour of L_{fl}^{21} with all p–h asymmetric corrections included to the first order in T/μ. We first examine the A–L contribution to L_{fl}^{21} shown in figure 2(a) since it is dominant in the presence of pair-breaking. The remaining contributions in (b) and (c) will be considered later. The contribution of figure 2(a) can be expressed as

$$\begin{aligned} K_{j^Q j}^{AL}(q, \mathrm{i}\nu_m) = \frac{-e}{8m^2}\frac{1}{\beta}\sum_Q\sum_{\Omega_l}\overline{\Lambda}(Q, \mathrm{i}\Omega_l;\, q, \mathrm{i}\nu_m)\Lambda(Q, \mathrm{i}\Omega_l;\, q, \mathrm{i}\nu_m) \\ \times\, t(Q, \mathrm{i}\Omega_l)\, t(Q + q, \mathrm{i}\Omega_l + \mathrm{i}\nu_m) \end{aligned} \tag{10}$$

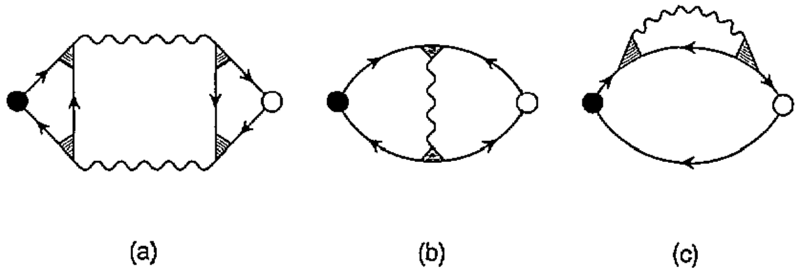

Figure 2. The fluctuation diagrams for L^{21}. The shaded triangles are the impurity vertex corrections Γ's. A filled circle denotes a momentum–energy vertex.

where $t(Q, \mathrm{i}\Omega_l)$ is the t-matrix for the Cooper pair scattering. The t-matrix has been calculated by Ebisawa and Fukuyama to the first order in T/μ [14]

$$t(Q, \mathrm{i}\Omega_l) = -\frac{1}{N(0)\{\epsilon + (\pi/8T)[1 + \mathrm{i}\lambda\Omega_l/|\Omega_l|]|\Omega_l| + \pi D Q^2/8T\}} \tag{11}$$

where the diffusion constant $D = \frac{1}{3}v_F^2\tau$ and the p–h asymmetric correction is given by

$$\lambda \equiv \frac{2\pi T N'(0)}{N(0)}\left[\frac{1}{N(0)V} + 1\right] \tag{12}$$

where $N'(0)$ is the derivative of the density of states at the Fermi level. The vertex functions in equation (10) are

$$\begin{aligned}\overline{\Lambda}(Q, \mathrm{i}\Omega_l;\ q, \mathrm{i}\nu_m) = \frac{2}{\beta}\sum_{\omega_n}\sum_k (2k+q)\big[\xi_k + \Sigma_{imp}(\mathrm{i}\omega_n) + \xi_{k+q} + \Sigma_{imp}(\mathrm{i}\omega_n + \mathrm{i}\nu_m)\big] \\ G_0(k, \mathrm{i}\omega_n)G_0(k+q, \mathrm{i}\omega_n + \mathrm{i}\nu_m)G_0(-k+Q, -\mathrm{i}\omega_n + \mathrm{i}\Omega_l) \\ \Gamma(\mathrm{i}\omega_n;\ -\mathrm{i}\omega_n + \mathrm{i}\Omega_l)\Gamma(\mathrm{i}\omega_n + \mathrm{i}\nu_m;\ -\mathrm{i}\omega_n + \mathrm{i}\Omega_l)\end{aligned} \tag{13}$$

$$\begin{aligned}\Lambda(Q, \mathrm{i}\Omega_l;\ q, \mathrm{i}\nu_m) = \frac{2}{\beta}\sum_{\omega_n}\sum_k (2k+q)G_0(k, \mathrm{i}\omega_n)G_0(k+q, \mathrm{i}\omega_n + \mathrm{i}\nu_m) \\ \times G_0(-k+Q, -\mathrm{i}\omega_n + \mathrm{i}\Omega_l)\Gamma(\mathrm{i}\omega_n;\ -\mathrm{i}\omega_n + \mathrm{i}\Omega_l) \\ \times \Gamma(\mathrm{i}\omega_n + \mathrm{i}\nu_m;\ -\mathrm{i}\omega_n + \mathrm{i}\Omega_l)\end{aligned} \tag{14}$$

where the single-electron Green's function $G_0(k, \mathrm{i}\omega_n) = [\mathrm{i}\omega_n - \xi_k - \Sigma_{imp}(\mathrm{i}\omega_n)]^{-1}$, and the Γ is the impurity vertex correction at each end of a t-matrix, whose Q-dependence is of higher order and is thus neglected in the following.

The vertex functions $\overline{\Lambda}$ and Λ are evaluated in the dirty limit ($1/T\tau \gg 1$). The p–h asymmetric corrections are included for $d = 3$ as follows. The sum over k becomes

$$\sum_k \to \int_{-\mu}^{\infty} \mathrm{d}\xi\ N(\xi) = N(0)\int_{-\mu}^{\infty} \mathrm{d}\xi\ \sqrt{1 + \frac{\xi}{\mu}}. \tag{15}$$

Carrying out the integral for the standard impurity self-energy exactly yields [15]

$$\Sigma_{imp}(\mathrm{i}\omega_n) = -\frac{\mathrm{i}}{2\tau}\left[1 + \frac{\mathrm{i}\omega_n}{2\mu} + \frac{\mathrm{i}}{4\tau\mu}\frac{|\omega_n|}{\omega_n}\right]\frac{|\omega_n|}{\omega_n} \tag{16}$$

and for the vertex Γ yields

$$\Gamma(\mathrm{i}\omega_n;\ -\mathrm{i}\omega_n + \mathrm{i}\Omega_l) = \frac{\Sigma_{imp}(\mathrm{i}\omega_n) + \Sigma_{imp}(-\mathrm{i}\omega_n + \mathrm{i}\Omega_l)}{\Sigma_{imp}(\mathrm{i}\omega_n) + \Sigma_{imp}(-\mathrm{i}\omega_n + \mathrm{i}\Omega_l) - 1/4\tau^2\mu}. \tag{17}$$

Equations (15), (16) and (17), together with

$$k = mv_F\sqrt{1 + \frac{\xi}{\mu}} \tag{18}$$

are used in evaluations of $\overline{\Lambda}$ and Λ.

The result of somewhat lengthy calculation of $\overline{\Lambda}$ and Λ, to the first order in T/μ, is [5]

$$\overline{\Lambda}(Q,\ \mathrm{i}\Omega_l;\ 0,\ \mathrm{i}\nu_m) = -\frac{\pi m N(0) v_F^2 \tau}{3T} Q(2\mathrm{i}\Omega_l + \mathrm{i}\nu_m) - \frac{\pi m N(0) v_F^2}{6T\tau\mu} Q \equiv \overline{\Lambda}_1 + \overline{\Lambda}_2 \tag{19}$$

$$\Lambda(Q,\ \mathrm{i}\Omega_l;\ 0,\ \mathrm{i}\nu_m) = -\frac{\pi m N(0) v_F^2 \tau}{3T} Q. \tag{20}$$

All p–h asymmetric corrections in Λ are of the form Ω_l/μ or ν_m/μ and vanish as $T \to T_c$ and $\nu \to 0$; thus only the leading term (p–h symmetric) is shown in equation (20), which would yield the A–L result $\sigma_{fl.AL}$. The heat-current vertex $\overline{\Lambda}$ in (19), on the other hand, has a non-negligible p–h asymmetric term $\overline{\Lambda}_2$ besides the p–h symmetric $\overline{\Lambda}_1$. In Maki's calculation [2], only $\overline{\Lambda}_1$ was used and p–h asymmetry came only from the t-matrices (as shown in equations (11) and (12)), which resulted in $L^{21}_{fl.AL} \propto \epsilon^{(d-2)/2}$ and implies a decrease in magnitude of S as $T \to T_c$. In the present calculation, since $\nu \to 0$ and $\Omega \to \epsilon$ after the analytic continuation, we find that $\overline{\Lambda}_2 \gg \overline{\Lambda}_1$ as $T \to T_c$. Therefore, the dominant contribution near T_c arises from $\overline{\Lambda}_2$ instead of $\overline{\Lambda}_1$.

To calculate $L^{21}_{fl.AL}$ due to $\overline{\Lambda}_2$, we note that

$$\overline{\Lambda}_2 = \frac{1}{2\tau^2\mu}\Lambda. \tag{21}$$

Inserting equation (21) into equation (10), and comparing it to the expression for $\sigma_{fl.AL}$ [13], one can immediately write down the result for $L^{21}_{fl.AL}$:

$$L^{21}_{fl.AL} = -\frac{1}{4e\tau^2\mu}\sigma_{fl.AL} \tag{22}$$

with $\sigma_{fl.AL}$ given by equation (9). As $T \to T_c$, $L^{21}_{fl.AL}$ diverges as $\epsilon^{(d-4)/2}$, in the same way as $\sigma_{fl.AL}$ does.

Equation (22) was derived in the above for the A–L diagram in figure 2(a). We have also studied other fluctuation diagrams in figure 2 for their contribution to $L^{21}_{fl.AL}$. Figure 2(b) is referred to as *Maki diagram*, and is the leading term in σ_{fl} in the case of vanishing pair-breaking interaction [16]. In the dirty limit, we have derived an expression identical to equation (22) for the Maki diagram, and a similar expression but with a positive sign on the right-hand side for figure 2(c). Therefore, L_{fl}^{21} is related to σ_{fl} as in equation (22) regardless of whether the A–L diagram or the Maki diagram is more important.

The divergence of L_{fl}^{21} due to $\overline{\Lambda}_2$ given by equation (22) results in an increase in magnitude of S as T approaches T_c. From equations (8) and (22)

$$S = S_n \frac{\sigma_n + [3/(2\pi T\tau)^2]\sigma_{fl}}{\sigma_n + \sigma_{fl}} \cong S_n \left[1 - \frac{\sigma_{fl}}{\sigma_n} + \frac{3}{(2\pi T\tau)^2}\frac{\sigma_{fl}}{\sigma_n}\right]. \tag{23}$$

Equation (23) results from an expansion in terms of the small quantity σ_{fl}/σ_n, which is implied by the perturbative nature of the fluctuation theory. In equation (23), the second (negative) term arises from the fluctuation contribution to σ and the third (positive) one from the fluctuation contribution to L^{21}. In the dirty limit, when $1/T\tau > 2\pi/\sqrt{3}$ is satisfied, the third term in equation (23) dominates the second one and increases the magnitude of S. The increase becomes steeper as the system becomes dirtier.

3. Discussion

In the calculation by Maki [2], the contribution to L_{fl}^{21} came from the p–h asymmetry in the fluctuation propagators—t-matrices. In the present calculation, a large fluctuation contribution is obtained from the p–h asymmetry in the triangle momentum–energy vertex, which consists of single-electron Green's functions. One may view the former result as the thermopower of superconducting fluctuation pairs, and the latter result as the thermopower of the normal electrons with an enhancement caused by the propagation of superconducting fluctuations. The present calculation shows that the enhanced normal electron contribution to the thermopower is more important than the Cooper pair contribution at temperatures near T_c.

For quantitative comparison with the peak in S observed experimentally by Howson *et al*, we use $L_{fl.AL}^{21}$ for $d = 3$:

$$L_{fl.AL}^{21} = -\frac{7\zeta(3)}{3072\pi^2}\frac{1}{(T\tau)^2}\frac{e}{m\zeta^3(0)\epsilon^{-1/2}} \tag{24}$$

where $\varsigma(x)$ is the Riemann zeta function, to fit the experimental data. For a layered structure, $\zeta^3(0)$ in equation (24) should be replaced by $\zeta_x^2(0)\zeta_z(0)$. As shown in figure 3, the power law ($\epsilon^{-1/2}$) in equation (24) describes well the temperature dependence over a decade of temperatures. Using the experimental values $\zeta(0) = 30$ Å, $\zeta_z(0) = 10$ Å and $T_c = 92.6$ K, we get $T\tau = 0.1$ which is reasonable for $YBa_2Cu_3O_{7-\delta}$.

Even though our calculation is carried out for dirty metals for $d = 3$, the general result should qualitatively apply to other dimensions or to clean metals, as follows from the following symmetry argument. As $q \to 0$ and $\nu_m \to 0$, the general expression of $\overline{\Lambda}$ in equation (13) vanishes by symmetry for $Q = 0$; for $Q \neq 0$, $\overline{\Lambda}$ only vanishes by symmetry if $\Omega_l = 0$ *and* p–h symmetry is assumed. This indicates that $\overline{\Lambda}$ is proportional to Q but not to Ω_l as long as p–h asymmetry is present. Therefore in general, besides a term like $\overline{\Lambda}_1$, there should also be a term like $\overline{\Lambda}_2$ as introduced in equation (19). The term $\overline{\Lambda}_2$ is proportional to Q only and gives a more divergent contribution to L_{fl}^{21} near T_c than does $\overline{\Lambda}_1$.

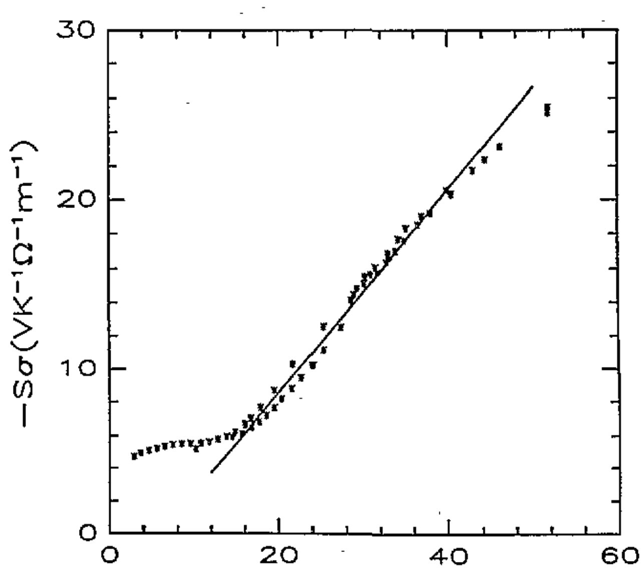

Figure 3. The linear dependence of $-S\sigma$ on $\epsilon^{-1/2}$ near T_c. The solid line is the theoretical fit to the experimental data [3] obtained using equation (24).

4. Summary

We have calculated the fluctuation thermopower near the superconducting transition from the lowest-order fluctuation diagrams and found a new leading contribution corresponding to the fluctuation-enhanced normal electron thermopower. In the dirty limit, this new contribution produces a peak in the thermopower which agrees quantitatively with the experimental data. We note that our theory is for T approaching T_c from above, and further study is necessary for the fluctuation effect on thermopower below T_c.

Acknowledgments

We gratefully acknowledge the helpful discussion with M A Howson and M Yu Reizer, and the generosity of M A Howson in allowing us to show his data in figure 3.

Note added in proof. While this work has been edited for publication, a paper by Varlamov and Livanov [17] was published which showed a *clean-limit* result similar to equation (23) of the present work. In contrast to [17], the present work is for the theoretically more challenging *dirty limit*—an extremely difficult problem as described by the authors of [17]—and is presumably more relevant to the high-T_c superconductors as T approaches T_c.

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