Normal-state Nernst effect of a high-critical-temperature superconductor

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We have analyzed the data of Clayhold *et al.* for the Nernst effect in the normal state of a high critical superconductor, i.e., $Tl_2Ba_2CaCu_2O_{8+\delta}$. This requested to derive a kinetic expression for the Nernst effect, an expression able to take into account inelastic scattering and magnetic-field dependence. This was done along a relaxation time formalism for the solution of the Boltzmann equation but leaving a *background term* which can be calculated by the most appropriate method. The final calculation leads to the evaluation of the *background term* resulting from the thermoelectric field-free effect. In order to do this we have considered a model of Livanov and Sergeev. The Nernst effect is explained by a simple two band model for electrons and holes with different mobilities. The resulting fit to the experimental data looks rather convincing. Several predictions are made thereafter. [S0163-1829(96)06921-4]

The Nernst effect (NE) is the off-diagonal term of the thermoelectric power tensor in presence of a magnetic field. NE is a very difficult property to measure, to calculate and to interpret. It is not even a quantity much reported upon for metallic systems. This is not totally surprising since the thermoelectric power [or Seebeck coefficient (S)] is already something quite complicated to calculate, the more so for a third rank tensor.

It is known, however, that the Nernst effect (represented by the Nernst coefficient Q) is a quite challenging problem[1] in high critical temperature superconductors (HTS's) which are furthermore anisotropic systems. It has been often examined below the critical temperature in the so-called mixed state though only recently was the anisotropy of the coefficient put into evidence.[2,3]

The understanding of the *background term* in such coefficients is, however, necessary before subtle physical effects are considered. The *normal, or high-temperature behavior* is thus of great interest. The most simple theory leads to

$$Q = \frac{\pi^2 k_B^2 T}{3m} \left(\frac{\partial \tau(\varepsilon)}{\partial \varepsilon} \right)_{\varepsilon = \varepsilon_F} \tag{1}$$

when the metallic limit ($k_B T \ll \varepsilon_F$) and a single type of carriers are assumed in standard notations, as recalled by Blatt.[4] Some more general treatment based on linear response theory can be found in the literature even including the role of fluctuations near, e.g., the superconductivity transition.[5]

Very recently Clayhold *et al.*[6] measured the Nernst effect in the normal state. They discussed the great difficulty in obtaining precisely signals of the order of nV for such systems. The measurements were made on a $Tl_2Ba_2CaCu_2O_{8+\delta}$ sample. Such features were seen by Pekala *et al.*[7] also, but were reported with less precision.

In the apparent absence of theory for the $Tl_2Ba_2CaCu_2O_{8+\delta}$ Nernst effect, we give here a theoretical work for such a multicarrier crossed heat and electrical current transport at high fields. The comparison with experimental data of Clayhold *et al.* looks very fine for the limited and reasonable set of assumptions used here.

The most simple method to determine a transport property is through the solution of the Boltzmann equation within a relaxation time formalism. However, the formal solution in general avoids inelastic-scattering contributions (relaxation time approximation) which are sometimes reintroduced later even though the theory had first missed the contribution. In principle, one should follow the formalism of Sorbello.[8] Another method of great interest is through a variational solution as discussed by Kohler[9] and Sondheimer,[10] popularized by Ziman,[11] and often used when a thermal gradient has to be taken into account. The method is much more complicated, however, when one has to include magnetic-field effects. A supposedly well investigated quantity like the magnetoresistance was written in a one page formula in the remarkable work of Garcia-Moliner.[12]

In fact, the relaxation time method is of interest for transport properties measured in a finite magnetic field if one notices that inelastic collisions are mainly relevant for the coefficients and terms pertaining to the thermal current, and leaving aside only the elastic collisions for the electric terms. In so doing (see some details in Appendix A), one can write the Seebeck and the Nernst coefficients as

$$QB = \frac{x}{1+x^2} \left(-S(0) + \frac{\pi^2 k_b^2 T}{2q\varepsilon_F} \right), \tag{2}$$

$$S(B) = \frac{1}{1+x^2} \left(S(0) + x^2 \frac{\pi^2 k_b^2 T}{2q\varepsilon_F} \right), \tag{3}$$

where B is the magnetic induction and $S(0)$ is obviously the Seebeck coefficient in absence of magnetic field. In the above formula, $x = q\tau B/m$, q is the carrier electric charge, m is the effective mass of the carriers, and τ is the field-free relaxation time (at the Fermi level δ_F). The above formulae are for isotropic systems.

At low field ($x \ll 1$), the Seebeck coefficient $S(B)$ is very mildly different from the zero-field case $S(0)$—as always observed in the normal state of HTS's away from the critical temperature. At high field ($x \gg 1$), neglecting quantum effects (Landau levels), the linear correction term adds itself to the classical Mott dependence, i.e., is a sum of two terms. The

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Nernst effect QB at high field has a component proportional to T/x. At high temperature, the inverse of the relaxation time is linear in temperature and QB is thus expected to behave like T^2 for electron-phonon scattering.

The normal-state $Tl_2Ba_2CaCu_2O_{6+\delta}$. Nernst data[6] can now be analyzed. Let us recall that above the critical temperature (≈ 110 K) the Nernst coefficient is positive and decreasing. This is understood in terms of superconducting fluctuations,[5] not taken into account here, but known to exist rather far from T_c. From 150 to 200 K the Nernst coefficient becomes negative with a sharp variation. The coefficient smooths out, remains negative, and decreases above 200 and below 350 K.

We first take for granted that the conduction in HTS's is multiband. Several conduction models exist and we cannot review them nor quote them all here. We limit, in fact, our considerations to well-established models taking into account classical electrons and holes. One interesting case is to consider a mixed conduction with conducting holes in the CuO planes and electrons in the supposed semiconducting TlO planes.[13] We have attempted to use such a scheme for the Clayhold et al. data,[6] but the exponential behavior with temperature arising from the semiconducting contribution does not lead to a good fit.

Rather we will consider in a more phenomenological way that the hole and electron mobilities are different. Whatever the planes where they are, this is not in disagreement with the fact that the Hall coefficient remains positive between T_c and 250 K.[14] Indeed, in a mixed conduction model, the only way to explain such a behavior is to consider that the electron concentration is much smaller than the hole concentration. This can be taken as the main assumption, though a reasonable one, in the following. This in fact implies also that the thermoelectric power is essentially due to the holes according to the Kohler rule[4,11] and this is true in HTS's. The modeling of thermoelectric power in HTS's for specific hole transport $S_h(0)$ has been, e.g., realized by Livanov and Sergeev[15] considering the electron-phonon-impurity interference (EPII) process. The mixed conduction Nernst coefficient can be easily written in the same framework and then reads (see Appendix B)

$$Q = \frac{\mu_h^2(T_0)}{\mu_h(T)}\left[S_h(0) - \frac{\pi^2 k_B^2 T}{2e\varepsilon_F^e}\right] + \mu_h(T)\left[\frac{\pi^2 k_B^2 T}{2e\varepsilon_F^h} - S_h(0)\right],$$

(4)

where $S_h(0)$ is the hole thermoelectric power in absence of magnetic field, $\mu_h(T)$ is the hole mobility at temperature T, ε_F^h and ε_F^e are, respectively, the hole and electron Fermi energy and T_0 is a temperature defined in terms of μ_h and n_e/n_h which is the relative electron/hole concentration (B2). We repeat that $n_e \ll n_h$. The Fermi energy ε_F^h is fixed by band structure calculation to 2.4 eV.[13] ε_F^e is unknown and will be a fit parameter. $S_h(0)$ can be deduced from the formulae of Ref. 15. It depends on two free parameters: (i) S_0 which is the classical Mott diffusion thermopower measured at the Debye temperature taken here to be equal to $\theta = 400$ K so that $S_0 = 4$ μV K^{-1} and (ii) λ which is the electron-phonon coupling constant. The parameter λ has been chosen equal to 1.5 which is in the range of values (1–2.5) proposed by Livanov and Sergeev. In Ref. 15 a chemical parameter must
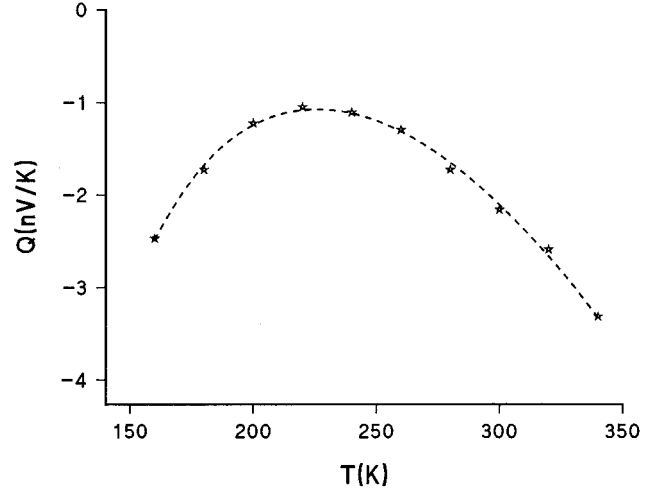

FIG. 1. Normal-state Nernst effect of $Tl_2Ba_2CaCu_2O_{8+\delta}$; data from Clayhold et al. (Ref. 6), fit from text. ε_F^e has been taken equal to 0.3 eV.

be also given. ΔZ is the charge of scattering centers associated with ions O^{2-} in the CuO planes, $\Delta Z = -2$. With these parameter values, $S_h(0)$ behaves like $6.5 \times 10^{-6} - 2.16 \times 10^{-8}$ T V K^{-1}.

The main scattering process for the hole mobility is certainly the hole-acoustic phonon scattering. The correction due to the EPII is expected to be negligible for the hole mobility in the range of temperature that we consider.[16] The hole mobility can be then calculated from the Grüneisen formula for the hole resistivity $\rho_h = \rho_0(\theta/T)^5 F_5(T/\theta)$ where $F_5(T/\theta)$ is the Fermi-Dirac integral. The electrical resistivity, ρ_0 has been taken equal to 1.2×10^{-3} Ω cm from experimental data.[17] With a hole concentration equal to 5.10^{21} cm^{-3} (Ref. 18) the mobility is then obtained easily from $\mu_h = 1/(ne\rho_h)$.

One should notice that such parameter values only influence the magnitude of the relative terms. If the theory was based on unphysical assumptions, the predicted behavior would turn out to be way off from the data. We have used the above values to calculate the normal-state NE coefficient for the sample examined by Clayhold et al. (Fig. 1). The fit is very good. This confirms that both the parameters are not only good estimates and in agreement with the experimental data of other authors, but also that the theoretical expression makes sense. As a test of the main assumption, we obtain the only *free parameter*, i.e., $n_e/n_h = 3.2 \times 10^{-5}$ which is very small, as expected. A prediction resulting from the above data is that the Hall coefficient should be negative above 500 K. If the physical considerations that we have made stay valid at such temperatures.

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APPENDIX A

For independent electrons, the Boltzmann equation can be solved using an *a priori* relaxation time $\tau(\vec{k})$. In such a formalism the resolution of the Boltzmann equation is very simple compared to the formalism of Garcia-Moliner and Simons.[19] It can be shown that this latter method implies the existence of a relaxation time but different from the one deduced by the relaxation time approximation. A relaxation time formalism method is therefore justified.

After some algebra we can write for an isotropic three-dimensional system the following expressions for the Onsager coefficients:

$$\sigma_{xx} = \frac{q^2}{3\pi^2 m} \int_0^\infty k^3 \frac{\partial f_0(\vec{k})}{\partial k} \frac{\tau(\vec{k})}{1+x^2} \, dk, \tag{A1}$$

$$\sigma_{xy} = \frac{q^3 B}{3\pi^2 m^2} \int_0^\infty k^3 \frac{\partial f_0(\vec{k})}{\partial k} \frac{\tau^2(\vec{k})}{1+x^2} \, dk, \tag{A2}$$

$$L_{xx} = -\frac{q}{3\pi^2 m} \int_0^\infty k^3 \frac{(\varepsilon - \varepsilon_F)}{T} \frac{\partial f_0(\vec{k})}{\partial k} \frac{\tau(\vec{k})}{1+x^2} \, dk, \tag{A3}$$

$$L_{xy} = -\frac{q^2 B}{3\pi^2 m^2} \int_0^\infty k^3 \frac{(\varepsilon - \varepsilon_F)}{T} \frac{\partial f_0(\vec{k})}{\partial k} \frac{\tau^2(\vec{k})}{1+x^2} \, dk. \tag{A4}$$

Taking the metallic limit approximation through Sommerfeld's lemma and keeping only the first relevant term we have

$$\sigma_{xx}(B) = \frac{1}{1+x^2} \sigma_{xx}(0), \tag{A5}$$

$$\sigma_{xy}(B) = \frac{x}{1+x^2} \sigma_{xx}(0), \tag{A6}$$

$$L_{xx}(B) = -\frac{q}{3\pi^2 m} \frac{\pi^2}{3} k_B^2 T \left(\frac{2m}{\hbar^2}\right)^{3/2} F_1, \tag{A7}$$

$$L_{xy}(B) = -\frac{q}{3\pi^2 m} \frac{\pi^2}{3} k_B^2 T \left(\frac{2m}{\hbar^2}\right)^{3/2} F_2, \tag{A8}$$

where

$$F_1 = \frac{3}{2} \varepsilon_F^{1/2} \frac{x}{1+x^2} + \varepsilon_F^{3/2} \frac{1-x^2}{(1+x^2)^2} \left(\frac{\partial \tau}{\partial \varepsilon}\right)_{\varepsilon = \varepsilon_F}, \tag{A9}$$

$$F_2 = \frac{3}{2} \varepsilon_F^{1/2} \frac{x}{1+x^2} + \varepsilon_F^{3/2} \frac{2x}{(1+x^2)^2} \left(\frac{\partial \tau}{\partial \varepsilon}\right)_{\varepsilon = \varepsilon_F}. \tag{A10}$$

The derivative of the relaxation time can be written in term of the zero magnetic field transport coefficients,

$$L_{xx}(B) = \frac{1-x^2}{(1+x^2)^2} L_{xx}(0) - \frac{q}{3m} k_B^2 T \varepsilon_F^{1/2} \tau \frac{x^2}{(1+x^2)^2} \left(\frac{2m}{\hbar^2}\right)^{3/2} \tag{A11}$$

and

$$L_{xy}(B) = \frac{2x}{(1+x^2)^2} L_{xx}(0) - \frac{q}{6m} k_B^2 T \varepsilon_F^{1/2} \tau \frac{(x^2-1)x}{(1+x^2)^2} \left(\frac{2m}{\hbar^2}\right)^{3/2}. \tag{A12}$$

The Nernst and Seebeck coefficient can then be written, to first order in $k_B T/\varepsilon_F$

$$QB = \frac{\sigma_{xx} L_{xy} - \sigma_{xy} L_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2} = \frac{x}{1+x^2} \left(-S(0) + \frac{\pi^2 k_b^2 T}{2q\varepsilon_F}\right), \tag{A13}$$

$$S(B) = -\frac{\sigma_{xx} L_{xx} + \sigma_{xy} L_{xy}}{\sigma_{xx}^2 + \sigma_{xy}^2} = \frac{1}{1+x^2} \left(S(0) + x^2 \frac{\pi^2 k_b^2 T}{2q\varepsilon_F}\right). \tag{A14}$$

The only relevant *background term* is $S(0)$ which can be calculated by any appropriate method for independent electrons and for any relevant model according to the physical interest.

APPENDIX B

According to the approximation made in the main text, the Nernst coefficient can be written in the following form:

$$QB = \frac{\sigma_{xx}^h (L_{xy}^h + L_{xy}^e) - L_{xx}^h (\sigma_{xy}^h + \sigma_{xy}^e)}{\sigma_{xx}^{h\,2}}. \tag{B1}$$

We will define a temperature T_0 at which $\sigma_{xy}^e = \sigma_{xy}^h$. We found that the only simple way to explain the decrease of the Nernst coefficient between 200 and 350 K is to consider that over these limits of temperature the electron mobility is sufficiently great so that $x_e \gg 1$. Taking into account the relation (A6) respectively for $x_e \gg 1$ and $x_h \ll 1$, one obtains the ratio of electron/hole concentration

$$n_e/n_h = \mu_h^2(T_0) B^2. \tag{B2}$$

Using formulae (B2), (A5), (A6), (A7), and (A8), one can write the Nernst coefficient Q as

$$Q = \frac{\mu_h^2(T_0)}{\mu_h(T)} \left[S_h(0) - \frac{\pi^2 k_B^2 T}{2e\varepsilon_F^e}\right] + \mu_h(T) \left[\frac{\pi^2 k_B^2 T}{2e\varepsilon_F^h} - S_h(0)\right]. \tag{B3}$$

To find the relative electron/hole concentration, the fit gives us the value of $\mu_h^2(T_0)/\mu_h(T)$ which allows us to determine the temperature T_0. We can then find the relative electron/hole concentration with relation (B2).

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