Thermoelectric power of inhomogeneous superconductors: Effects of field-induced intragrain granularity

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The temperature, magnetic-field, and oxygen-deficiency dependence of the thermoelectric power (TEP), S, of an inhomogeneous superconductor with field-induced intragrain granularity is considered via the superconductive-glass model. Below the phase-locking (grain-decoupling) temperature T_g, which defines the coherent properties of a Josephson-junction network, the TEP follows a linear T dependence, while above T_g (but below the single-superconducting-grain-depairing temperature T_c), S shows a semiconductorlike $1/T$ behavior. The model predicts the existence of a maximum of S near T_g that increases with the applied magnetic field and changes its sign above some threshold value of the oxygen-deficiency parameter. Attributing the intragrain granularity to clusters of oxygen defects (in the CuO plane), the calculated TEP behavior is found to be in qualitative agreement with the experimentally observed (below T_c) anomalous peak of the magneto-TEP in oxygen-deficient high-T_c superconductors.

I. INTRODUCTION

As it is now well-established, there are two main contributions to be expected in the observable behavior of the thermoelectric power (TEP), S, in high-T_c superconductors (HTS): vortex motion[1–6] and weak links.[7–13] Usually, the former contribution dominates in single crystals (and high magnetic fields), while the latter one is more pronounced in ceramics (and at low fields). However, the recent experimental observations[14–19] in oxygen-depleted HTS single crystals seem to break down this simple picture. Indeed, the magnetization curve of HTS single crystals was widely observed to exhibit an anomalous magnetic-field behavior, which has been attributed to the "field-induced intragrain granularity" in oxygen-deficient materials.[14,15,18] A "phase diagram" $H_m(\delta,T)$, that demarcates the multigrain onset as a function of temperature and oxygen deficiency, δ, reconstructed by Osofsky *et al.*,[15] allowed them to confirm that their single crystals exhibited behavior characteristic of homogeneous superconductors for $H<H_m$ and inhomogeneous superconductors for $H>H_m$. The granular behavior for $H>H_m$ has been related to the clusters of oxygen defects (within the CuO plane) that restrict supercurrent flow and allow excess flux to enter the crystal. At the same time, unusual behavior of the Seebeck and Nernst effects in the mixed state of slightly oxygen deficient $YBa_2Cu_3O_{7-\delta}$ was found[11] to indicate a strong departure of the vortices ("pancake vortices") from the standard Abrikosov type. Galffy, Freimuth, and Murek[12] argued that the large Seebeck effect they found in c-axis oriented epitaxial $YBa_2Cu_3O_{7-\delta}$ films can be attributed to dissipation due to "granularity", since the vortex contribution to the Seebeck voltage is by far too small to account for the observed value. Oxygen-deficiency dependence of the transport line energy carried by vortices has also been observed[5] in magneto-Seebeck effect measurements on single $YBa_2Cu_3O_{7-\delta}$ crystals. At last, a homogeneous versus fractal behavior deduced from the superconductivity fluctuations of the thermoelectric power of HTS has been discussed recently by Clippe *et al.*[13] in terms of the percolative superconductivity in granular materials.

In this paper a contribution to the longitudinal TEP, S, of inhomogeneous (due to field-induced intragrain granularity) superconductors is calculated within the framework of the superconductive glass (SG) model.[20] We find that below the phase-locking ("grain" decoupling) temperature $T_g(\delta,H)$, which defines the onset of coherent properties of a Josephson-junction (JJ) network, the TEP follows a linear T dependence, while above $T_g(\delta,H)$ (but below the single grain-depairing superconducting temperature T_c) S shows a semiconductorlike $1/T$ behavior. A maximum of S near $T_g(\delta,H)$ is found to increase with the applied magnetic field, changing its sign above some threshold value of the oxygen-deficiency parameter, $\delta_c(T,H)$. Attributing the intragrain granular behavior to clusters of oxygen defects (in the CuO plane), we compare the calculated TEP with the experimentally observed (below T_c) anomalous field-induced peak of the TEP in oxygen-deficient $YBa_2Cu_3O_{7-\delta}$.[1]

II. THE MODEL

The SG model is based on the well-known Hamiltonian of a granular (inhomogeneous) superconductor, which in the so-called pseudospin representation, has the form[20–26]

$$\mathcal{H}=-\frac{1}{2}\sum_{ij}J_{ij}S_i^+S_j^- + \text{H.c.} \ , \tag{1}$$

where

$$J_{ij}(\delta,T,H)=J(\delta,T)\exp[iA_{ij}(H)] \ , \quad S_i^+=\exp(+i\varphi_i) \ ,$$

$$A_{ij}(H)=\frac{\pi}{\phi_0}(\mathbf{H}\times\mathbf{R}_{ij})\mathbf{r}_{ij} \ , \quad \mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j \ , \tag{2}$$

$$\mathbf{R}_{ij}=(\mathbf{r}_i+\mathbf{r}_j)/2 \ .$$

This model describes the interaction between oxygen-rich superconducting grains [with phase $\varphi_i(t)$], arranged in a random two-dimensional (2D) lattice (modeling the CuO plane of oxygen-depleted $YBa_2Cu_3O_{7-\delta}$, where a glass-like picture is established[22–26]) with coordinates $\mathbf{r}_i=(x_i,y_i,0)$. The grains are separated by oxygen-poor insulating boundaries producing Josephson coupling with energy $J(\delta,T)$.[15] The system is under the influence of a frustrating applied magnetic field **H**, which is assumed to be normal to the CuO plane of HTS. According to the Ambegoakar-Baratoff expression for the temperature dependence of the Josephson energy,[20] near the single grain-depairing temperature, $T_c(\delta)$,

$$J(\delta,T)=J(\delta,0)[1-T/T_c(\delta)] .$$

The increase of the oxygen deficiency, δ, leads to the decrease of the Josephson energy (via the increase of the insulating layer between oxygen-rich grains). For small δ (such that $\delta \ll 1$) we can approximate the δ dependence of both the critical temperature and the Josephson energy by a linear law,[16,17] namely $T_c(\delta)=T_c(0)(1-\delta)$ and $J(\delta,0)=J(0,0)(1-\delta)$.

Following Mori's generalized theory of Brownian motion,[27] we calculate the static ($\omega=0$) contribution to the longitudinal TEP (Seebeck coefficient), $S(\delta,T,H)$, of an inhomogeneous superconductor with field-induced intragrain granularity within the above-mentioned model via the current-current, current-heat, and heat-current correlation functions[27–29]

$$S(\delta,T,H)=\frac{\overline{\langle \delta j(t)\delta Q(0)\rangle}_{\omega=0}+\overline{\langle \delta Q(t)\delta j(0)\rangle}_{\omega=0}}{2T\overline{\langle \delta j(t)\delta j(0)\rangle}_{\omega=0}} , \tag{3}$$

where

$$\delta Q(t)\equiv Q(t)-Q(\infty) , \quad \delta j(t)\equiv j(t)-j(\infty) , \tag{4}$$

$$\overline{\langle A(t)\rangle}_{\omega}\equiv \mathrm{Re}\int_0^{\infty} dt\, \exp(i\omega t)\overline{\langle A(t)\rangle} ,$$
$$\overline{A(\mathbf{r}_i)}\equiv \int d\mathbf{r}_i P(\mathbf{r}_i)A(\mathbf{r}_i) . \tag{5}$$

Here $j(t)$ is the x component of the Josephson current[21,26]

$$\mathbf{j}(t)=\frac{2ie}{\hbar}\sum_{ij} J_{ij}S_i^+S_j^-\mathbf{r}_{ij}+\mathrm{H.c.} \tag{6}$$

and $Q(t)=\mathbf{q}\mathbf{Q}(t)/q$ is the longitudinal part of the heat flux

$$\mathbf{Q}(t)=-\left[\frac{i\mathbf{q}}{q^2}\right]\frac{\partial \mathcal{H}(t)}{\partial t} , \tag{7}$$

obeying the conservation law $\partial_t\mathcal{H}(t)+\mathrm{div}\mathbf{Q}(t)=0$ [or, in q representation, $\partial_t\mathcal{H}(t)-i\mathbf{q}\mathbf{Q}(t)=0$]. The bar on the right-hand side of Eq. (3) denotes the configurational averaging [see Eq. (5)] over the randomly distributed grain coordinates, and $\langle \cdots \rangle$ means the thermodynamic averaging with the Hamiltonian (1).

Taking into account the equation of motion for the Josephson pseudospins $S_i^{\pm}$,[21,25]

$$\partial_t S_i^+ \equiv -\frac{1}{\hbar}\frac{\delta\mathcal{H}}{\delta S_i^-}=\frac{1}{\hbar}\sum_j J_{ij}S_j^+ , \tag{8}$$

from Eqs. (1)–(8) we get for the current-current, current-heat, and heat-current correlators

$$\overline{\langle \delta j(t)\delta j(0)\rangle}=\left[\frac{2edJ(\delta,T,H)}{\hbar}\right]^2[D(t)-L]\times[D(0)-L] , \tag{9}$$

$$\overline{\langle \delta j(t)\delta Q(0)\rangle}=\left[\frac{2edJ^3(\delta,T,H)}{q\hbar^2}\right][D(t)-L]\times[D(0)-L]^2 , \tag{10}$$

$$\overline{\langle \delta Q(t)\delta j(0)\rangle}=\left[\frac{2edJ^3(\delta,T,H)}{q\hbar^2}\right][D(t)-L]^2\times[D(0)-L] , \tag{11}$$

respectively, where

$$J(\delta,T,H)\equiv\overline{J_{ij}(\delta,T,H)}=J(\delta,T)\int d\mathbf{r}_i P(\mathbf{r}_i)\exp[iA_{ij}(H)] . \tag{12}$$

Here $d=\sqrt{\overline{r_{ij}^2}}$, and $L(\delta,T,H)$ is the order parameter of the SG model which is defined via the phase-phase correlator[25] $D_{ij}(t)=\langle S_i^+(t)S_j^-(0)\rangle$,

$$L(\delta,T,H)\equiv \lim_{t\to\infty} D(t) , \quad D(t)\equiv\sum_i D_{ii}(t) . \tag{13}$$

To obtain Eqs. (9)–(11) we have used the so-called "mean-field approximation"[29,30] assuming that $\overline{A(\mathbf{r}_i)B(\mathbf{r}_j)}\simeq\overline{A(\mathbf{r}_i)}\,\overline{B(\mathbf{r}_j)}$. In the so-called "mode-coupling approximation,"[29,31] $D(t)$ obeys the self-consistent master equation[25–27]

$$\frac{d^2D(t)}{dt^2}+\Omega^2D(t)+\int_0^t d\tilde{t}\,K(t-\tilde{t})\frac{dD(\tilde{t})}{d\tilde{t}}=0 , \tag{14}$$

with $K(t)$ being a memory (feedback) kernel, and Ω is a characteristic frequency of the Josephson-junction network.

When there is no temporal correlations between grains (i.e., in the so-called paracoherent state[25]), the memory kernel has a "white noise" form $K(t)=2\gamma\delta(t)$, where $\gamma=k_BT/\hbar$ and $\delta(t)$ is the Dirac delta function. In this case the master Eq. (14) results in a Debye-like decay of uncorrelated paracoherent state, namely $D(t)=\exp(-t/\tau)$, where $1/\tau=\Omega=\gamma$. Such a situation is realized above some critical (phase-locking) temperature T_g when the coherent state within the JJ network is destroyed completely, so that the order parameter $L=0$. Below T_g, the situation changes drastically due to the superconducting correlations occurring between grains. In general,[25,29] the coherent part of the memory kernel is defined via the "velocity-velocity" correlator $K_{ij}(t)=\langle \dot{S}_i^+(t)\dot{S}_j^-(0)\rangle$. Thus, taking into account the equation of motion (8), the memory kernel below T_g can be presented in the form

$$K(t)\equiv\sum_i K_{ii}(t)=2\gamma\delta(t)+\Omega^2_{coh}(\delta,T,H)D(t)\ . \quad (15)$$

Here $\Omega_{coh}(\delta,T,H)=J(\delta,T,H)/\hbar$ and $J(\delta,T,H)$ is given by Eq. (12) above. In view of Eq. (13), a zero-frequency ($t\rightarrow\infty$) solution of the master Eq. (14) with the memory kernel (15) results in the nontrivial order parameter for the intragranular JJ network[25]

$$L(\delta,T,H)=1-\left[\frac{k_BT}{J(\delta,0,H)}\right]^2 . \quad (16)$$

The phase-locking temperature $T_g(\delta,H)$, below which the ensemble of grains undergoes the phase transition into the coherent state, is defined by the equation $L(\delta,T_g,H)=0$ which, due to Eq. (16), gives rise to $T_g(\delta,H)=J(\delta,0,H)/k_B$. As a result, the order parameter $L=1-[T/T_g(\delta,H)]^2$ gradually changes from 0 at $T\geq T_g(\delta,H)$ to 1 at $T=0$, thus describing the continuous phase transition. It is worthwhile to mention that the correlator $D(t)$ follows a simple Debye-like decay law only above $T_g(\delta,H)$, where $L=0$ (see above). Below $T_g(\delta,H)$ (where the order parameter $L\neq 0$) $D(t)$ can be presented in the form[25,26]

$$D(t)=L+1(-L)\Phi(t)\ . \quad (17)$$

Here the relaxation function, $\Phi(t)$, is supposed to be normalized, viz.

$$\frac{1}{\tau}\int_0^\infty dt\ \Phi(t)=1 \quad (18)$$

and obeys the following boundary conditions, $\Phi(0)=1$ and $\Phi(\infty)=0$, i.e., $D(0)=1$ [see Eq. (17)]. In principle,[25] the set of Eqs. (14)–(18) allows us to find a non-Debye relaxation behavior of the correlator $D(t)$ in the coherent state. We will not discuss this possibility in more detail, since in this paper we are only interested in a static ($\omega=0$) behavior of the TEP [see Eq. (3)].

Finally, taking into account Eqs. (3)–(18), the longitudinal TEP of inhomogeneous superconductor reads

$$S(\delta,T,H)=\left[\frac{k_B}{2e}\right]\left[\frac{\lambda}{2\pi d}\right]\left[\frac{J(\delta,T,H)}{k_BT}\right]\times[1-L(\delta,T,H)]\ . \quad (19)$$

Here we introduce the thermal flux wavelength $\lambda=2\pi/q$; the temperature, field, and oxygen-deficiency dependences of the Josephson energy $J(\delta,T,H)$ and the order parameter $L(\delta,T,H)$ are governed by Eqs. (12) and (16), respectively.

III. DISCUSSION

Above the phase-locking temperature $T_g(\delta,H)$, but below $T_c(\delta)$, the grains are in their decoupling state, and $L\equiv 0$. The ensemble of grains behaves as if it consists of independent oscillators obeying the Debye relaxation law $D(t)=\exp(-t/\tau)$. In this case, as it follows from Eq. (19), the TEP shows a semiconductorlike $1/T$ dependence[29,32] (slightly modulated by the factor $1-T/T_c(\delta)$ due to the temperature dependence of Josephson energy), namely

$$S(\delta,T,H)=\left[\frac{k_B}{2e}\right]\left[\frac{\lambda}{2\pi d}\right]\left[\frac{T_g(\delta,H)}{T}\right]\times\left[1-\frac{T}{T_c(\delta)}\right],\quad T_g(\delta,H)\leq T<T_c(\delta)\ . \quad (20)$$

Below T_g, where the coherent structure of the Josephson network is established and thus $L(\delta,T,H)\neq 0$ [viz., $L(\delta,T,H)=1-(T/T_g(\delta,H))^2$], the TEP of inhomogeneous superconductor follows a linear T dependence [see Eq. (19)]

$$S(\delta,T,H)=\left[\frac{k_B}{2e}\right]\left[\frac{\lambda}{2\pi d}\right]\left[\frac{T}{T_g(\delta,H)}\right]\times\left[1-\frac{T}{T_c(\delta)}\right],\quad T<T_g(\delta,H)\ . \quad (21)$$

Thus at $T=T_g(\delta,H)$ the TEP $S(\delta,T,H)$ has a field- and oxygen-deficiency-dependent maximum

$$S_m(\delta,H)\equiv S(\delta,T_g,H)=\left[\frac{k_B}{2e}\right]\left[\frac{\lambda}{2\pi d}\right]\left[1-\frac{T_g(\delta,H)}{T_c(\delta)}\right]. \quad (22)$$

It is worthwhile to mention that the temperature behavior of the TEP for inhomogeneous superconductor [given by Eqs. (20)–(22)] qualitatively correlates with analogous behavior of the TEP in a single tunnel junction, calculated recently by Amman, Ben-Jacob, and Cohn.[33] In fact, Laurent *et al.*[1] have made rather precise measurements of the magneto-TEP on the oxygen-deficient $YBa_2Cu_3O_{7-\delta}$. They found that below $T_c(\delta)=90$ K [the temperature which has been defined through the vanishing of the TEP, i.e., $S(T_c)=0$, cf. Eq. (20)], near $T=T_g(\delta,H)=84$ K in magnetic field $H=0.4$ T, the measured TEP exhibited a peak behavior with the maximum value of $S_m(\delta,H=0.4\ \text{T})=0.2\ \mu$V/K. Using these experimental values, Eq. (22) brings about the following estimate for the ratio between the thermal wavelength λ and the average grain size d, namely $\lambda/d\simeq 0.4$. To make the above comparison with the experimental data of Laurent *et al.*[1] more informative, we need to know the explicit field dependence of the phase-locking temperature $T_g(\delta,H)$ which, in turn, comes from the field dependence of the Josephson energy $J(\delta,0,H)$. The latter is defined via Eq. (12) and strongly depends on the form of the random distribution function $P(\mathbf{r}_i)$ as well as on the type of disorder.[34] Assuming, after Morgenstern, Müller, and Bednorz,[23] a site-type disorder allowing weak displacements of the grain sites at their positions of the original 2D lattice, i.e., within a radius $d=\sqrt{\overline{r_{ij}^2}}$, the new position is chosen randomly according to the normalized separable Gaussian distribution function $P(\mathbf{r}_i)=P(x_i)P(y_i)$, where

$$P(x)=\frac{1}{\sqrt{2\pi d^2}}\exp\left[-\frac{x^2}{2d^2}\right] \tag{23}$$

and using Eqs. (2) and (12), the explicit field dependence of the $T_g(\delta,H)$ reads

$$T_g(\delta,H)\equiv\frac{J(\delta,0,H)}{k_B}=\frac{T_g(\delta,0)}{\sqrt{1+H^2/H_0^2}} . \tag{24}$$

Here $T_g(\delta,0)=J(\delta,0,0)/k_B$, and $H_0=\phi_0/2s$ is a characteristic Josephson field with $s=\pi d^2$ an average JJ projection area. So, as it follows from Eqs. (21) and (24), at $T=T_g(\delta,H)$ the field dependence of the TEP peak, $S_m(\delta,H)$, shows the following asymptotic behavior

$$S_m(H)\propto\begin{cases}\dfrac{H^2}{2H_0^2}, & H\ll H_0\\[2mm] 1-\dfrac{H_0}{H}, & H\gg H_0 .\end{cases} \tag{25}$$

Such a behavior (increase at small fields and high-field saturation) qualitatively correlates with the experimentally found magnetic-field dependences of the HTS TEP in the mixed state.[2,6] To get a more quantitative description, however, the essential contribution to the observable magneto-TEP behavior due to the vortex motion[1-6] has to be taken into account. Assuming for simplicity that $T_g(\delta,0)\simeq T_c(\delta)$, we can estimate an average size of the oxygen-depleted region in the sample used in Ref. 1. Indeed, due to Eq. (24), using the above relation between H_0 and d, the experimental value of $T_g(\delta,H=0.4T)=84$ K results in $H_0\simeq 0.8$ T, which gives $d\simeq 20$ nm. By analogy with the critical temperature $T_g(\delta,H)$, we can introduce the critical field $H_g(\delta,T)$ as the solution of the equation $L(\delta,T,H_g)=0$, where the field dependence of the order parameter $L(\delta,T,H)$ is defined by Eq. (24). The result is

$$H_g(\delta,T)=H_0\sqrt{1-T/T_g(\delta,0)} . \tag{26}$$

Taking into account the δ dependence of the phase-locking temperature, $T_g(\delta,H)\simeq T_g(0,H)(1-\delta)$, Eq. (26) results in the following oxygen-deficiency behavior of the critical field

$$H_g(\delta,T)=H_0\sqrt{[\delta_c(T,0)-\delta]/(1-\delta)} . \tag{27}$$

Here we have introduced the critical oxygen deficiency, $\delta_c(T,H)$, which is defined as the solution of the equation $L(\delta_c,T,H)=0$ and has the form $\delta_c(T,H)=1-T/T_g(0,H)$. The physical meaning of this critical parameter is as follows. For $\delta>\delta_c(T,H)$ oxygen-rich superconducting grains are separated by oxygen-poor insulating boundaries so that there is no superconducting path through the sample. We notice that within the SG model, the critical field [Eq. (27)] plays in fact the same role as the "phase-boundary" field $H_m(\delta,T)$, which demarcates homogeneous and inhomogeneous behavior in single crystals[15] (see the Introduction). Using the fact[19] that for fully oxygenated $YBa_2Cu_3O_{7-\delta}$ $T_g(0,0)\simeq T_c(0)=92$ K, for the oxygen deficiency of the sample used in the experiments of Laurent *et al.*[1] [with $T_c(\delta)\simeq 90$ K], we get the value of $\delta\simeq 0.02$. In turn, for the critical oxygen deficiency near the TEP peak temperature, $T_p=T_g(\delta,H=0.4T)=84$ K, we obtain $\delta_c(T_p,H=0.4\ \text{T})\simeq 0.08$. Finally, using these estimates (including the above found value of the characteristic field $H_0=0.8$ T), Eq. (27) leads to the following critical-field estimate, $H_g(\delta,T_p)\simeq 0.2$ T. Thus, in agreement with the field-induced granularity interpretation, the peak of the TEP observed by Laurent *et al.*[1] indeed can be attributed to the manifestation of the inhomogeneous regime of the oxygen-deficient superconductor, since this peak occurs only when the applied magnetic field exceeds the phase-boundary field $H_g(\delta,T)$. It is interesting also to discuss a δ dependence of the TEP peak, $S_m(\delta,H)$, provided by the above-mentioned percolation picture. According to Eq. (20) and taking into account the definitions of δ and $\delta_c(T,H)$, the peak value of the TEP reads

$$S_m(\delta)=\left[\frac{k_B}{2e}\right]\left[\frac{\lambda}{2\pi d}\right]\left[\frac{\delta_c(T_p)-\delta}{1-\delta_c(T_p)}\right] . \tag{28}$$

Interestingly, the above expression qualitatively agrees with the experimentally observed[19] δ dependence of the TEP peak in oxygen-deficient single crystals. Namely, TEP shows a maximum at $\delta=0$ (fully oxygenated case), vanishes at $\delta=\delta_c$, and changes its sign at $\delta>\delta_c$. As it was found,[19] in zero applied magnetic field $\delta_c\simeq 0.1$, which reasonably correlates with the estimate $\delta_c\simeq 0.08$ that we found here above for field $H=0.4$ T.

In summary, the temperature-, magnetic-field, and oxygen-deficiency-dependent contribution to the thermoelectric power (TEP), $S(\delta,T,H)$, of inhomogeneous (due to field-induced intragrain granularity) superconductors has been calculated within the framework of the superconductive-glass (SG) model. Below the phase-locking (grain-decoupling) temperature $T_g(\delta,H)$, which defines the onset of coherent properties of Josephson-junction (JJ) network, the TEP follows a linear T dependence, while above T_g (but below the single grain-depairing superconducting temperature T_c) S shows a semiconductorlike $1/T$ behavior. The model predicts the existence of a maximum of $S(\delta,T,H)$ near $T_g(\delta,H)$, which increases with the applied magnetic field and changes its sign above some critical value of the oxygen deficiency (according to the percolationlike law). Attributing the intragrain granular behavior to clusters of oxygen defects[15] (arranged within the CuO plane), the calculated TEP was found to be in a qualitative agreement with the experimentally observed[1] (below T_c) field-induced anomalous peak of the magneto-TEP in oxygen-deficient $YBa_2Cu_3O_{7-\delta}$.

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