Thermal conductivity of a granular superconductor

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The temperature and magnetic-field dependence of the thermal conductivity κ for a granular superconductor is considered via the superconductive glass model. Above the phase-locking temperature T_c (but below the single-grain superconducting temperature T_s) κ is found to be dominated by the Rayleigh-like grain-boundary-scattering mechanism, while below T_c the thermal conductivity follows a glasslike T^2 dependence. In the paracoherent state ($T_s > T > T_c$) the Wiedemann-Franz law appears to be nearly satisfied. The field dependence of κ is shown to be highly sensitive to the form of the grain-distribution function.

I. INTRODUCTION

The recently reported weak-link-induced analog of the fountain effect in high-T_c superconductors (HTS's) (Refs. 1–3), as well as other predictions concerning unusual thermoelectric effects due to the weak-link nature of these materials,[4–7] set up an interesting and corresponding problem on the thermal-conductivity (κ) behavior in granular HTS's. The unusual behavior of the Seebeck and Nernst effects in the mixed state of slightly oxygen-deficient $YBa_2Cu_3O_7$ (YBCO) has been found[8] to indicate a strong departure of the vortices ("pancake vortices") from the standard Abrikosov type. Galffy, Freimuth, and Murek[9] argued that the large Seebeck effect they found in *c*-axis-oriented epitaxial YBCO 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. It is also worthwhile to mention the recent experimental findings on the thermal conductivity of twinned and untwinned YBCO single crystals[10] and of twinned YBCO and tweeded Fe-doped YBCO polycrystals[11] (in high magnetic fields) which apparently show a strong departure from the usual linear behavior of the thermal magnetoresistivity (TMR), $W=\kappa^{-1}$, for conventional type-II superconductors.[12] Up to now, however, there have been practically no investigations concerning the TMR behavior in granular HTS's at low enough magnetic fields (below the first Abrikosov field) where the vortex-phonon-scattering mechanism is certainly inactive and where another scenario for the thermal conductivity is required.

In the present paper, a contribution to κ of granular superconductors based on the weak-link decoupling mechanism is calculated, both in zero and nonzero applied magnetic field. We find that above the phase-locking temperature T_c (which defines the coherent properties of the Josephson grain-boundary network) but below T_s, the critical temperature of the single grain, the temperature dependence of κ is dominated by the Rayleigh-like grain-boundary-scattering mechanism,[13] whereas below T_c the thermal conductivity follows the famous T^2 dependence[14] confirming the glasslike behavior of the Josephson phase network.[15–20] By accounting for the previously discussed temperature behavior of the excess magnetoconductivity of a granular superconductor,[21] the Wiedemann-Franz law is found to be nearly satisfied above T_c. The magnetic-field dependence of κ appears to be highly sensitive to the form of the grain-distribution function. In particular, a Gauss-like distribution law leads to a powerlike behavior of the TMR, while the Lorentzian form results in an exponential dependence for $\kappa(H)$.

II. MODEL, APPROXIMATIONS, AND THEORETICAL RESULTS

The model is based on the well-known Hamiltonian of a granular superconductor, which in the so-called pseudospin representation has the form[15–21]

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

where

$$J_{ij}(T,H) = J(T)\exp[iA_{ij}(H)] , \quad S_i^+ = \exp(+i\varphi_i) ,$$
$$A_{ij}(H) = \frac{\pi}{\phi_0}(\mathbf{H}\times\mathbf{R}_{ij})\cdot\mathbf{r}_{ij} , \quad (2)$$
$$\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j, \quad \mathbf{R}_{ij} = (\mathbf{r}_i + \mathbf{r}_j)/2 .$$

The model describes the interaction between superconducting grains [with phases $\varphi_i(t)$], arranged in a random three-dimensional (3D) lattice with coordinates $\mathbf{r}_i$ (modeling the distribution of CuO planes of oxygen-depleted $YBa_2Cu_3O_{7-\delta}$). The "grains" are separated by oxygen-poor insulating boundaries producing Josephson coupling characterized by an energy $J(T)$ which could depend on δ. (According to the Ambegaokar-Baratoff expression for the temperature dependence of the Josephson energy,[22] near the single "grain" depairing temperature $T_c(\delta)$, $J(\delta,T)$ is equal to $J(\delta,0)[1-T/T_c(\delta)]$. 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 in fact approximate the δ dependence of both the critical temperature and the Josephson energy by a linear law, namely, $T_c(\delta)=T_c(0)(1-\delta)$ and $J(\delta,0)=J(0,0)(1-\delta)$. In the following, though, the label δ will not be further mentioned. It will be understood hereafter that in making the appropriate configurational averages (see below) the distributions of T_c and J may depend on δ.)

The system is under the influence of a frustrating applied magnetic field $\mathbf{H}$, which is assumed to be normal to the *ab* plane of HTS's where the glasslike picture is established for the "carrier" (or phase) distribution, i.e., the pseudospins.[17-20]

According to linear-response theory,[23] we can calculate the weak-link-induced (static $\omega=0$) thermal conductivity normal to the c axis, within the above-mentioned model of a granular superconductor, via the Kubo formula

$$\kappa(T,H)=\frac{1}{2k_B T^2 V}\int_0^\infty dt\,\overline{\langle \delta Q(t)\delta Q(0)\rangle_\omega}\ , \tag{3}$$

where

$$\delta Q(t)\equiv Q(t)-Q(\infty)\ ,\qquad Q(t)=d\,\partial_t\mathcal{H}(t)\ . \tag{4}$$

Here $Q(t)$ is the longitudinal part of the energy-flux density $\mathbf{Q}(t)$,

$$\mathbf{Q}(t)=-\left[\frac{i\mathbf{q}}{q^2}\right]\frac{\partial\mathcal{H}(t)}{\partial t}\ , \tag{5}$$

obeying the conservation law $\partial_t\mathcal{H}(t)+\mathrm{div}\mathbf{Q}(t)=0$; V is the volume of the system, and d is the characteristic length of variation for the thermal flux flow [which is of the order of the weak-link thickness, $d=(\overline{r_{ij}^2})^{1/2}$]. The overbar [e.g., in the right-hand side of Eq. (3)] denotes configurational averaging over the randomly distributed grain coordinates and, specifically,

$$\overline{\langle A(t)\rangle_\omega}\equiv \mathrm{Re}\int_0^\infty dt\,\exp(i\omega t)\overline{\langle A(t)\rangle}\ ,\qquad \overline{A(\mathbf{r}_i)}\equiv\int d\mathbf{r}_i P(\mathbf{r}_i)A(\mathbf{r}_i)\ . \tag{6}$$

Taking into account the equation of motion for the Josephson pseudospins $S_i^{\pm}$,[15-21]

$$\partial_t S_i^+\equiv-\frac{1}{\hbar}\frac{\delta\mathcal{H}}{\delta S_i^-}=\frac{1}{\hbar}\sum_j J_{ij}S_j^+ \tag{7}$$

from Eqs. (1) and (5), we get an expression containing the overall average of a linear combination of four pseudospin correlators [times a J^4 factor as seen from Eq. (7)] for the energy flux-energy flux-density correlator in Eq. (3). At this stage it is customary to decouple the averaging of the "grain distribution" (represented by the "scattering potentials" J_{ij}) from the carrier ("spins" or phases) motion as in a common random-phase approximation.[24(a)] The argument is based on the different types of relaxation time for these physical quantities: The relaxation of the scattering potentials is (obviously) quasi-instantaneous (in some sense the carriers see a static array), while the relaxation time of the "carriers" is finite and has, in principle, to be taken self-consistently from the equation of motion; i.e., the "carrier" (inverse) relaxation time is defined through the scattering mechanism, i.e., through the characteristic frequency of the Josephson network, $\tau^{-1}=2eI_cR_N/\hbar$, where I_c is the Josephson critical current and R_N is the resistance between grains in their normal state. However, it is also customary to let this characteristic frequency go to zero afterwards and to consider the steady state of Eq. (3), i.e., let $\omega\to 0$ in Eq. (6), in order to obtain the "experimentally corresponding" thermal conductivity.

Another approximation is also used in order to avoid the (usually rarely known in fact) four-spin correlator: To obtain the final result, we thus also use the so-called "mean-field approximation"[24(b),25] assuming that $\overline{A(\mathbf{r}_i)B(\mathbf{r}_j)}\cong\overline{A(\mathbf{r}_i)}\,\overline{B(\mathbf{r}_j)}$. This might require greater care in the vicinity of a phase transition (see the corresponding work of Ref. 26 for magnetic systems), but is a standard procedure for random (quenched "disordered") alloys.[27]

Bearing these "random-phase–mean-field approximations," the correlator appearing in the thermal-conductivity definition reads

$$\overline{\langle\delta Q(t)\delta Q(0)\rangle}=\left[\frac{dJ^2(T)}{\hbar}\right]^2 f(H/H_0)[D(t)-L][D(0)-L]\ , \tag{8}$$

where the magnetic field enters only through the (static) average of the Josephson junction J_{ij} network [as can be seen by linearizing $J_{ij}(T,H)$ in the definition], i.e.,

$$f(H)=\int d\mathbf{r}_i P(\mathbf{r}_i)\exp[iA_{ij}(H)]\ . \tag{9}$$

Here $P(\mathbf{r}_i)$ denotes the (static) grain-distribution function and $L(T)$ is the order parameter of the above model, which is defined via the (in principle still time-dependent) phase-phase correlator $D_{ij}(t)=\overline{\langle S_i^+(t)S_j^-(0)\rangle}$ as

$$L(T)\equiv\lim_{t\to\infty}D(t),\quad D(t)\equiv\sum_i D_{ii}(t)\ , \tag{10}$$

but for which as explained above the $\omega\approx 0$ limit is taken (or equivalently by letting $t\to\infty$). However, note that the equation of motion enters for defining the time-dependent correlator value. In fact, in general, the time-dependent correlator $D(t)$ can be presented in the form[19,28]

$$D(t)=L+(1-L)\Phi(t)\ . \tag{11}$$

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

$$\frac{1}{\tau}\int_0^\infty dt\,\Phi(t)=1\ , \tag{12}$$

and obeys the boundary conditions $\Phi(0)=1$ and $\Phi(\infty)=0$, i.e., $D(0)=1$ [see Eq. (11)]. This relaxation time as explained above is taken to be finite and the thermal conductivity is supposed to be measured after a time τ_1 long enough ($\tau_1\gg\tau$) that the change in temperature (or the application of the thermal gradient in an ac method) has been washed out by the fluctuations.

Finally, the thermal conductivity of a granular superconductor reads

$$\kappa(T,H)=\kappa(T,0)f(H/H_0) , \tag{13}$$

where

$$\kappa(T,0)=\frac{d^2J^4(T)\tau}{2k_BT^2\hbar^2V}[1-L(T)]^2 . \tag{14}$$

The phase-locking temperature T_c, below which the ensemble of grains undergoes the phase transition into the coherent state,[15-21] is defined by the equation $k_BT_c=J(T_c)$ with $J(T)=J(0)(1-T/T_s)$, where $J(0)$ is the Josephson energy at $T=0$ and T_s is the single-grain superconducting temperature. Since usually[15-21] $T_c \ll T_s$, we can put $J(T)=J(0)$ with a quite good accuracy. The mode-coupling approximation[29] for the correlator $D(t)$ results in the following temperature dependence for the order parameter $L(T)=1-(T/T_c)^2$.

(i) *Above the phase-locking temperature,* but below T_s, 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 follows from Eq. (14), the thermal conductivity is dominated by the Rayleigh-like grain-boundary-scattering mechanism, namely,

$$\kappa(T,0)=\frac{d^2J^4(0)\tau}{2k_BT^2\hbar^2V} . \tag{15}$$

(ii) *Below* T_c, where the coherent structure of the Josephson network is established and thus $L(T)\neq 0$ [viz., $L(T)=1-(T/T_c)^2$], the thermal conductivity of a granular superconductor follows the law [see Eq. (14)]

$$\kappa(T,0)=\frac{d^2J^4(0)\tau}{2k_BT_c^4\hbar^2V}T^2 . \tag{16}$$

As is well known,[14,30,31] such a T^2 law for the thermal conductivity reflects a glasslike *response* of any disordered system. Thus such a behavior of κ in the system under consideration gives further evidence in favor of the so-called glassy behavior (for the phases, equivalent to the "carriers") induced by the Josephson-junction array,[15-21] the origin of which can be traced to the (somewhat static) oxygen inhomogeneous distribution.

III. WIEDEMANN-FRANZ LAW

Furthermore, it is interesting to compare the above-calculated thermal conductivity of a granular superconductor [Eq. (14)] with the longitudinal (normal to the c axis) excess conductivity $\sigma(T,H)=\sigma^{xx}(T,H)$ due to the electrical current-current correlations within the same model,[21]

$$\sigma^{\alpha\beta}(T,H)=\frac{1}{2k_BTV}\int_0^\infty dt\,\overline{\langle\delta j^\alpha(t)\delta j^\beta(0)\rangle} \quad (\alpha,\beta=x,y,z) , \tag{17}$$

where

$$\delta j^\alpha(t)\equiv j^\alpha(t)-j^\alpha(\infty) ,$$
$$\mathbf{j}(t)=\frac{2ie}{\hbar}\sum_{ij}J_{ij}S_i^+S_j^-\mathbf{r}_{ij}-\text{H.c.} \tag{18}$$

Repeating the same procedure we have used before to derive Eq. (14), we find that above T_c (where $L=0$) $\sigma(T,0)$ has the form [cf. Eq. (15)]:

$$\sigma(T,0)=\frac{2d^2J^2(0)\tau}{k_BT\phi_0^2V} . \tag{19}$$

Thus, in view of Eqs. (15) and (19), we find that, near T_c [in the paracoherent state, where $J(0)\approx k_BT$],

$$\frac{\kappa(T,0)}{\sigma(T,0)}=\left[\frac{\pi}{2}\right]^2\left[\frac{k_B}{e}\right]^2T . \tag{20}$$

This can be compared with the Wiedemann-Franz law for normal metals, $\kappa/\sigma=L_0T$, where $L_0=(\pi/3)^2(k_B/e)^2$ is the Lorentz number. It is worth noting that $\sigma\approx J^2$, and $\kappa\approx J^4$, but in the paracoherent state the ratio turns out to be, in a first approximation, J independent. We recall that the energy J is in principle δ dependent (see Sec. II).

IV. WEAK-FIELD DEPENDENCE

Finally, turning to the magnetic-field behavior of the weak-link-induced $\kappa(H)$, we stress that it strongly depends on the form of the grain-distribution function $P(\mathbf{r}_i)$. Since the calculation of $P(\mathbf{r}_i)$ is beyond the scope of our paper, as is the use of the real form $P(\mathbf{r}_i)$ for performing the averaging in Eq. (9), we restrict ourselves to two commonly used distributions for a theoretical illustration. Namely, let us consider the Gaussian and Lorentzian laws. If the randomness of the Josephson lattice is governed by Gauss-like fluctuations of the form

$$P(\mathbf{r}_i)=\frac{1}{\sqrt{2S}}\exp\left[-\frac{\mathbf{r}_i^2}{2S}\right] , \tag{21}$$

the configurational averaging in Eq. (9) leads to a quadratic field dependence of the TMR ($W=\kappa^{-1}$):

$$W(H)=W(0)+\frac{H^2}{H_0^2} . \tag{22}$$

Here $H_0=\phi_0/2S$ is the characteristic Josephson field, where $S=\pi r_g^2$ is the effective junction surface (projected area) and r_g is the grain size. On the other hand, a Lorentz-like distribution law

$$P(\mathbf{r}_i)=\frac{1}{\pi}\frac{r_g}{r_i^2+r_g^2} \tag{23}$$

results in an exponential behavior of the field-dependent thermal conductivity,

$$\kappa(H)=\kappa(0)\exp(-H/H_0) . \tag{24}$$

In more realistic situations when the anisotropy in grain orientations (with respect to an applied magnetic field) plays an important role, the above simple dependences for $\kappa(H)$ can be drastically modified. Moreover, Eqs. (8), (9), and (13) could allow one, at least in principle, to reconstruct the form of the distribution function $P(\mathbf{r}_i)$ of any particular ceramics using the experimental dependence of $\kappa(H)$ for this granular material.

V. CONCLUSION

To make more definite conclusions on the observability of the thermal conductivity given by Eqs. (15) and (16), let us estimate the magnitude of κ in such a model. Assuming that $V \approx r_g^3$ and $d \approx r_g$ and using the typical parameters for HTS granular superconductors,[32] $r_g \approx 10$ μm, $\tau \approx 10^{-9}$ s, and $T_c \approx 50$ K, we get $\kappa \approx 1$ W/mK. This value is quite comparable with other contributions to the observed thermal conductivity of HTS ceramics.[13]

In summary, the weak-link-induced thermal conductivity of a granular superconductor has been calculated within the superconductive glass model with the view of estimating the order of magnitude of such an effect for fields below the first Abrikosov field. The weak-link (oxygen-depleted) array is taken as a quenched disordered system, but the relaxation mechanism of the phases ("carriers") leads to a finite characteristic relaxation time, which in turn leads to the need to calculate the energy-flux–energy-flux correlator in terms of a pseudospin-pseudospin correlation function along lines of the mode-coupling formalism. In order to do so, a so-called random-phase approximation is made to decouple the weak-link array of Josephson exchange energies (J_{ij}) from the pseudospins (S_i). A molecular-field approximation is also made on the four-spin correlator, but the two-spin correlator is calculated in the mode-coupling approximation.

In the paracoherent state ($T_s > T > T_c$, where T_s is a single-grain superconducting temperature and T_c is the phase-locking temperature for the Josephson array), the Rayleigh-like grain-boundary-scattering mechanism has been found to dominate $\kappa(T)$, whereas below T_c (in the coherent state) $\kappa(T)$ follows a glasslike T^2-dependence law. The Wiedemann-Franz law appears to be nearly satisfied in the paracoherent state of a granular superconductor. Furthermore, the magnetic-field dependence of the thermal conductivity has been shown to be strongly sensitive to the form of the grain-distribution function. In particular, a Gauss-like distribution results in a quadratic power-law dependence of the thermal magnetoresistivity W, while the Lorentzian distribution leads to an exponential law for $\kappa(H)$.

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