Pis'ma v ZhETF, vol.67, iss.9, pp.650 - 654

NONLINEAR SEEBECK EFFECT IN A MODEL GRANULAR SUPERCONDUCTOR

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Submitted 28 January 1998
Resubmitted 20 March 1998

The change of the Josephson supercurrent density j_s of a weakly-connected granular superconductor in response to externally applied arbitrary thermal gradient ∇T (nonlinear Seebeck effect) is considered within a model of 3D Josephson junction arrays. For $\nabla T > (\nabla T)_c$, where $(\nabla T)_c$ is estimated to be of the order of $\simeq 10^4$ K/m for YBCO ceramics with an average grain's size $d \simeq 10\,\mu$m, the weak-links-dominated thermopower S is predicted to become strongly ∇T-dependent.

PACS: 74.50.+r, 75.80.+q

A linear Seebeck effect, observed in conventional and high-T_c ceramic superconductors (HTS) and attributed to their weak-links structure (see, e.g., [1 – 7] and further references therein), is based on the well-known fact that in a Josephson junction (JJ) the superconducting phase difference $\Delta\phi$ depends only on the supercurrent density j_s (according to the Josephson relation $j_s = j_c \sin\Delta\phi$, where j_c is the critical current density). When a small enough temperature gradient ∇T is applied to such a JJ (with the normal resistivity ρ_n), the entropy-carrying normal current with density $j_n = S_0\nabla T/\rho_n$ is generated through such a junction, where S_0 is the thermopower (a *linear* Seebeck coefficient). This normal current density is locally cancelled by a counterflow of supercurrent with density j_s, so that the total current density through the junction $j = j_n + j_s = 0$. As a result [3], the supercurrent density $j_s = -j_n$ generates a nonzero phase difference $\Delta\phi$ via a transient Seebeck thermoelectric field $E_T = \rho_n j_n = S_0\nabla T$ induced by the temperature gradient ∇T. If in addition, an external current of density j_e also passes through the weak link, a non-zero voltage will appear when the total current density exceeds j_c, i.e., for $j_e = j_c \pm S_0\nabla T/\rho_n$.

In the present Letter, using a zero-temperature 3D model of Josephson junction arrays, a nonlinear analog of the thermoelectric effect (characterized by a non-trivial ∇T-dependence of the Seebeck coefficient S) in granular superconductors is considered. The experimental conditions under which the predicted behavior of thermopower can be observed in $YBCO$ ceramics are discussed.

The so-called 3D model of Josephson junction arrays (which is often used to simulate a thermodynamic behavior of a real granular superconductor) is based on the well-known tunneling Hamiltonian (see, e.g., [8 – 13])

$$\mathcal{H}(t) = \sum_{ij}^{N} J_{ij}[1 - \cos\phi_{ij}(t)], \tag{1}$$

and describes a short-range interaction between N superconducting grains (with the gauge invariant phase difference $\phi_{ij}(t)$, see below), arranged in a 3D lattice with coordinates

$\mathbf{r}_i = (x_i, y_i, z_i)$. The grains are separated by insulating boundaries producing Josephson coupling J_{ij} which is assumed [8] to vary exponentially with the distance $\mathbf{r}_{ij}$ between neighboring grains, i.e., $J_{ij}(\mathbf{r}_{ij}) = J(T)e^{-\boldsymbol{\kappa}\cdot\mathbf{r}_{ij}}$. For periodic and isotropic arrangement of identical grains (with spacing d between the centers of adjacent grains), we have $\boldsymbol{\kappa} = (1/d, 1/d, 1/d)$. Thus d is of the order of an average grain (or junction) size.

In general, the gauge invariant phase difference is defined as follows

$$\phi_{ij}(t) = \phi_{ij}(0) - A_{ij}(t), \tag{2}$$

where $\phi_{ij}(0) = \phi_i - \phi_j$ with ϕ_i being the phase of the superconducting order parameter, and $A_{ij}(t)$ is the so-called frustration parameter, defined as

$$A_{ij}(t) = \frac{2\pi}{\Phi_0}\int_i^j \mathbf{A}(\mathbf{r},t)\cdot d\mathbf{l}, \tag{3}$$

with $\mathbf{A}(\mathbf{r},t)$ the (space-time dependent) electromagnetic vector potential; $\Phi_0 = h/2e$ is the quantum of flux, with h Planck's constant, and e the electronic charge.

As is known [10, 13], a constant electric field $\mathbf{E}$ applied to a single JJ causes a time evolution of the phase difference. In this particular case Eq.(2) reads $\phi_{ij}(t) = \phi_{ij}(0)+\omega_{ij}(\mathbf{E})t$ where $\omega_{ij}(\mathbf{E}) = 2e\mathbf{E}\cdot\mathbf{r}_{ij}/\hbar$ with $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ being the distance between grains. If, in addition to the external electric field $\mathbf{E}$, the network of superconducting grains is under the influence of an applied magnetic field $\mathbf{H}$, the frustration parameter $A_{ij}(t)$ in Eq.(3) takes the following form

$$A_{ij}(t) = \frac{\pi}{\Phi_0}(\mathbf{H}\wedge\mathbf{R}_{ij})\cdot\mathbf{r}_{ij} - \frac{2\pi}{\Phi_0}\mathbf{E}\cdot\mathbf{r}_{ij}t. \tag{4}$$

Here, $\mathbf{R}_{ij} = (\mathbf{r}_i + \mathbf{r}_j)/2$, and we have used the conventional relationship between the vector potential $\mathbf{A}$ and (i) constant magnetic field $\mathbf{H} = \mathrm{rot}\mathbf{A}$ (with $\partial\mathbf{H}/\partial t = 0$) and (ii) homogeneous electric field $\mathbf{E} = -\partial\mathbf{A}/\partial t$ (with rot $\mathbf{E} = 0$).

There are at least two ways to incorporate a thermal gradient ∇T dependence into the above model. Namely, we can either invoke an analogy with the above-discussed influence of an applied electric field on the system of weakly-coupled superconducting grains or assume a direct ∇T dependence of the phase difference (as it was recently suggested by Guttman et al [14]). For simplicity, in what follows we choose the first possibility and assume an analogy with the conventional Seebeck effect. Recall that application of a temperature gradient ∇T to a granular sample is known to produce a thermoelectric field [1, 2] $\mathbf{E}_T = S_0\nabla T$, where S_0 is the so-called *linear* (*∇T-independent*) Seebeck coefficient. Assuming that in Eq.(4) $\mathbf{E} \equiv \mathbf{E}_T$, we arrive at the following change of the junction phase difference under the influence of an applied thermal gradient ∇T

$$\phi_{ij}(t) = \phi_{ij}(0) - A_{ij}(t) \tag{5}$$

with the frustration parameter

$$A_{ij}(t) = \frac{\pi}{\Phi_0}(\mathbf{H}\wedge\mathbf{R}_{ij})\cdot\mathbf{r}_{ij} - \frac{2eS_0}{\hbar}\nabla T\cdot\mathbf{r}_{ij}t. \tag{6}$$

As we see, the above equation explicitly introduces a direct ∇T dependence into the phase difference, expressing thus the main feature of the so-called thermophase effect suggested by Guttman et al [14]. Physically, it means that the macroscopic normal thermoelectric voltage V couples to the phase difference on the junction through the quantum-mechanical Josephson relation $V \propto d\Delta\phi/dt$. Later on we shall obtain a rather simple connection between the thermophase coefficient $S_T \equiv d\Delta\phi/d\Delta T$ and the conventional linear Seebeck coefficient S_0.

To consider a nonlinear analog of the Seebeck effect (characterized by a ∇T-dependent thermopower S), we recall [10, 13] that within the model under consideration the supercurrent density operator $\mathbf{j}_s$ is related to the pair polarization operator $\mathbf{p}$ as follows (V is a sample's volume)

$$\mathbf{j}_s = \frac{1}{V}\frac{d\mathbf{p}}{dt} = \frac{1}{i\hbar V}[\mathbf{p}, \mathcal{H}], \tag{7}$$

where the polarization operator itself reads

$$\mathbf{p} = \sum_i^N q_i \mathbf{r}_i. \tag{8}$$

Here $q_i = -2en_i$ with n_i the pair number operator, r_i is the coordinate of the center of the grain.

Finally, in view of Eqs.(1)-(8), and taking into account a usual "phase-number" commutation relation, $[\phi_i, n_j] = i\delta_{ij}$, we find

$$< \mathbf{j}_s(\nabla T) > = \frac{2e}{\hbar d^3}\sum_{ij}^N \int_0^\tau \frac{dt}{\tau}\int_0^\infty \frac{d\mathbf{r}_{ij}}{V} J_{ij}(\mathbf{r}_{ij}) \sin\phi_{ij}(t)\mathbf{r}_{ij} \tag{9}$$

for the thermal gradient induced value of the averaged supercurrent density. Here a temporal averaging (with a characteristic time τ) accounts for a change of the phase coherence during tunneling of Cooper pairs through the barrier, while integration over the relative grain positions $\mathbf{r}_{ij}$ is performed bearing in mind a short-range character of the Josephson coupling energy, viz. $J_{ij}(\mathbf{r}_{ij}) = J(T)f(x_{ij})f(y_{ij})f(z_{ij})$ with $f(u) = e^{-u/d}$.

To discuss a true ∇T induced thermophase effect only, in what follows we completely ignore the effects due to a nonzero applied magnetic field (by putting $\mathbf{H} = 0$ in Eq.(6)) as well as rather important in granular superconductors "self-field" effects (see [12, 13] for discussion of this problem) and assume that in equilibrium (initial) state (with $\nabla T = 0$) $< \mathbf{j}_s > \equiv 0$, implying thus $\phi_{ij}(0) \equiv 0$. The latter condition in fact coincides with a current density conservation requirement at zero temperature [9]. As a result, we find that an *arbitrary* temperature gradient $\nabla_x T \equiv \Delta T/\Delta x$, applied along the x-axis to the Josephson junction network, induces the appearance of the corresponding (nonlinear) longitudinal supercurrent with density

$$j_s(\Delta T) \equiv < j_s^x(\nabla_x T) > = j_0 G(\Delta T/\Delta T_0), \tag{10}$$

where

$$G(z) = \frac{z}{(1+z^2)}. \tag{11}$$

Here, $j_0 = 2eJNd/\hbar V$, $(\nabla_x T)_0 \equiv \Delta T_0/\Delta x = \hbar/2ed\tau S_0$, and $z = \Delta T/\Delta T_0$.

Notice that for a small enough temperature gradient (when $\nabla_x T \ll \nabla_x T_0$), we recover a conventional *linear* Seebeck dependence $j_s(\nabla_x T) = \alpha(T)S_0\nabla_x T$ with $\alpha(T) =$

$(2ed/\hbar)^2 J(T) N\tau/V$. On the other hand, for this result to be consistent with the above-discussed conventional expression $j_s = S_0 \nabla_x T/\rho_n$, zero temperature coefficient $\alpha(0)$ should be simply related to the specific resistance ρ_n. Let us show that this is indeed the case. Using $J(0) = \hbar\Delta_0/4e^2 R_n$ for zero-temperature Josephson energy (where Δ_0 is a zero-temperature gap parameter), $V \simeq Nld^2$ for sample's volume (with l being a relevant sample's size), and taking into account that the normal state resistance between grains R_n is related to ρ_n as follows, $\rho_n \simeq (d^2/l)R_n$, the self-consistency condition $\alpha(0) = 1/\rho_n$ yields $\tau \simeq (l/d)^2(\hbar/\Delta_0)$ for the characteristic Josephson time.

As it follows from Eq.(10), above some threshold value $(\nabla_x T)_c \simeq 0.25(\nabla_x T)_0$ the supercurrent density starts to substantially deviate from a linear law suggesting thus the appearance of nonlinear Seebeck effect with ∇T-dependent coefficient $S(\nabla_x T) = S_0/(1+z^2)$ where $z = \nabla_x T/(\nabla_x T)_0$ and $S_0 \equiv S(0)$. Let us estimate an order of magnitude of this threshold value of the thermal gradient needed to observe the predicted nonlinear behavior of the thermopower in weak-links-bearing HTS. Using $S_0 \simeq 0.5\,\mu\mathrm{V/K}$ and $\Delta_0/k_B \simeq 90\,\mathrm{K}$ for thermopower and zero-temperature gap parameter in YBCO, and $l \simeq 0.5\,\mathrm{mm}$ for a typical sample's size [2, 4], we get $\tau \simeq 10^{-9}\,\mathrm{s}$ for the characteristic Josephson tunneling time (cf. [13]), and $(\nabla_x T)_c \simeq 10^4\,\mathrm{K/m}$ for the threshold thermal gradient in a granular sample with an average grain (or junction) size $d \simeq 10\,\mu\mathrm{m}$. Besides, taking $J(0) \simeq \Delta_0$ for a zero-temperature Josephson energy (in samples with $R_n \simeq \hbar/4e^2$), we arrive at the following reasonable estimate of the weak-links-dominated critical current density $j_0 = 2eJ/\hbar ld \simeq 10^3\,\mathrm{A/m^2}$ in YBCO ceramics. We believe that the above estimates suggest quite an optimistic possibility to observe the discussed nonlinear behavior of the thermoelectric power in (real or artificially prepared) granular HTS materials and hope that the effects predicted in the present paper will be challenged by experimentalists.

In conclusion, let us obtain the connection between the conventional (linear) Seebeck effect and the above-mentioned thermophase effect (which is linear by definition). According to Guttman et al [14], the latter effect is characterized by a nonzero transport coefficient $S_T = d\Delta\phi/d\Delta T$. In our particular case (with $\phi_{ij}(0) = 0$ and $\mathbf{H} = 0$), it follows from Eqs.(5) and (6) that

$$\Delta\phi \equiv \frac{1}{\tau}\int_0^\tau dt \sum_{ij} \frac{\phi_{ij}(t)}{N} \simeq \frac{e\tau S_0}{\hbar}\Delta T. \tag{12}$$

Hence, within our approach the above two ∇T induced *linear* effects (characterized by the transport coefficients S_T and S_0, respectively) are related to each other as follows

$$S_T \simeq \left(\frac{e\tau}{\hbar}\right) S_0 \simeq \left(\frac{e}{\Delta_0}\right)\left(\frac{l}{d}\right)^2 S_0. \tag{13}$$

To summarize, the change of the Josephson supercurrent density of a granular superconductor under the influence of an arbitrary thermal gradient (a nonlinear Seebeck effect) was considered within a model of 3D Josephson junction arrays. A possibility of experimental observation of the predicted effect in HTS ceramics was discussed.

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