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Seebeck effect in the mixed state of high-T_c superconductors

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The recent experimental observation of a pronounced Seebeck effect in the mixed state of high-T_c superconductors is explained by a model based on the counterflow of a quasiparticle current and a supercurrent in the presence of a temperature gradient. The relatively large magnitude of this effect is due to the negligible amount of flux pinning above the irreversibility line in high-T_c superconductors.

Following the pioneering paper by Ginzburg,[1] thermoelectric effects in superconductors have been the subject of a considerable number of experimental and theoretical studies. In a nonsuperconducting metal or semiconductor, thermoelectric voltages only arise as a result of the asymmetry in the diffusion of electrons and holes down the applied temperature gradient. However, in a superconductor a different mechanism must be active, since no voltage can be developed across the superconductor to prevent the thermoelectric current flow. In this case, the normal current density j_n is canceled locally by a counterflow of supercurrent with density j_s.[1] Because of this cancellation of the thermoelectric current in a superconductor, schemes for measuring the thermoelectric coefficient, based on inhomogeneous or anisotropic superconductor configurations, become necessary. A review of various experiments, such as a bimetallic superconducting ring or the presence of a Josephson weak link in the region of the temperature gradient, has been given by Van Harlingen.[2] In the case of the Josephson junction, it is only the supercurrent density j_s which changes the superconducting phase difference ϕ, thereby generating a thermoelectric voltage $V = \Phi_0 d\phi/dt$ along the direction of the temperature gradient (Seebeck effect).[3] Here Φ_0 is the flux quantum. Such thermoelectric dc and ac Josephson effects have been observed experimentally.[4]

The oxide-high-T_c superconductors represent interesting materials for investigating the thermoelectric effects, as pointed out already by Ginzburg.[5] One of the unique properties of the high-T_c superconductors is the broadening of the resistive transition in an applied magnetic field and the existence of the irreversibility line.[6–8] Recently, measurements of the Seebeck effect in this transition regime for an applied magnetic field B have been reported for Y-Ba-Cu-O (Refs. 9 and 10) and Bi-Sr-Ca-Cu-O (Refs. 11–13) based materials. But up to now, the study of the thermopower in a magnetic field has been performed mainly for polycrystalline samples,[9,11–13] and to our knowledge only limited data[10] are presently available for single crystals and none for epitaxial films.

The essential results of these measurements of the Seebeck effect can be summarized as follows. (a) Below the onset temperature of superconductivity: As measured until now only for polycrystalline samples, the thermoelectric power increases with increasing magnetic field, reaching values of typically 2–4 $\mu V/K$ at a magnetic field of 1–2 T. The nonzero values of the thermopower are restricted to the temperature regime of the resistive transition. The thermopower is about 1000 times larger than expected from the standard model of flux motion induced by the thermal force of a temperature gradient.[13] For Y-Ba-Cu-O single crystals[10] in an applied magnetic field, a nonzero thermopower was also noticed in the temperature range, where the resistive broadening occurred. (b) Above the onset temperature of superconductivity: The thermoelectric power smoothly joins the value of the normal state.

In view of the results summarized in (a), the authors of Ref. 13 claim that thermally induced flux motion is inadequate for explaining the observed behavior. Instead, they propose another mechanism such as fluctuations of the order parameter or an internal weak-link structure. However, we feel that such an explanation is unlikely, and is also unnecessary in view of our model discussed below.

In the following we show that all observations[9–13] can be explained straightforwardly from the established concepts dealing with the thermoelectric effects in superconductors.[1–5]

The counterflow of the quasiparticle current density j_n and of the supercurrent density j_s in the presence of a temperature gradient in the superconductor requires careful discussion of the origin of the voltage due to flux motion in a superconductor. Often two mechanisms are mixed up, namely a quantum-mechanical process due to phase slippage of the condensate wave function described by the Josephson relation[14–16] and an inductive process described by Faraday's law. As shown both experimentally and theoretically,[17] the inductive mechanism does not contribute to the time-averaged voltage since the total magnetic flux in the measuring circuit remains unchanged during flux motion. Only the temporal change of the quantum-mechanical phase produces the time-averaged voltage, and this phase change clearly can only be achieved by the supercurrent density. The fact that only the supercurrent density j_s causes temporal phase changes, even in the presence of a quasiparticle current density j_n considerably larger than j_s, has also been demonstrated experimentally.[18]

Having shown that in the counterflow of j_n and j_s due to a temperature gradient it is only the supercurrent density j_s which can interact with the vortices in a superconductor, the appearance of the Seebeck voltage in the mixed state can be understood immediately: it is of quantum-mechanical origin and associated with the spatiotemporal phase changes due to the vortex motion affected by j_s. A schematics of this mechanism is shown in Fig. 1.

Turning next to a more detailed discussion of the experimental results reported in Ref. 9–13 in terms of our model, we note specifically the following points. The existence of the Seebeck effect in the mixed state is explained. In addition, we see from Fig. 1 that reversal of the magnetic-field direction leaves the sign of the Seebeck voltage unchanged, as observed experimentally:[13] for the vortices of opposite sign their direction of motion is also reversed. Further, in the range $B \ll B_{c2}$ the Seebeck voltage is expected to increase monotonically with increasing magnetic field.

In Fig. 1 we have restricted our discussion to the case where the magnetic field is oriented perpendicular to the direction of the temperature gradient. Turning now to the case where the directions of the magnetic field and of the temperature gradient are parallel to each other, the observation of a Seebeck voltage not much smaller than for the perpendicular case[13] can be understood in the following way. Due to the granular structure and the intragranular anisotropy of the polycrystalline sample, considerable misalignment of the vortices is to be expected. Because of this misalignment, there appear components of the magnetic flux within the superconductor perpendicular to the temperature gradient (and thereby to j_s). These perpendicular components are likely to exist with equal probability in the two opposite directions. However, it is important to note that these two components in opposite direction do not cancel each other in their contribution to the Seebeck effect, because they move in opposite directions due to their interaction with j_s (see Fig. 1), thereby generating a Seebeck voltage of the same sign. Of course, this situation is similar to the force-free configuration in resistance measurements, where such misalignment effects can also become important.

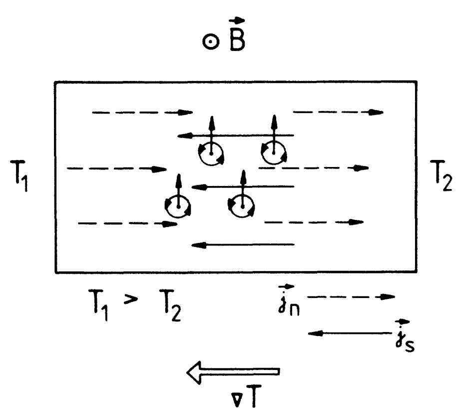

FIG. 1. Schematics of the component of the vortex motion yielding the Seebeck effect in the presence of a temperature gradient in a superconductor. The direction of the vortex motion is shown by the arrows. (The component of the vortex motion due to the thermal force and yielding the Nernst effect is not shown.)

The model we have strongly discussed differs from the standard phenomenological model for explaining the generation of thermoelectric voltages in the mixed state of a type-II superconductor based on the thermal force $f_{th} = -S_\phi \nabla T$ acting on each flux line.[19] Here S_ϕ is the transport entropy per unit length of flux line. The force f_{th} causes the motion of magnetic flux from the hot to the cold end of the sample, and a flux-flow voltage appears transverse to the temperature gradient and the direction of the applied magnetic field. This voltage corresponds to the Nernst effect in a superconductor. In the mixed state it is usually several orders of magnitude larger than in the normal state. A longitudinal thermoelectric voltage (along the direction of the temperature gradient) corresponding to the Seebeck effect can be generated from the thermal force f_{th} only by means of the Hall angle associated with the flux-flow process.[19] However, the Hall angle is usually very small, and as a consequence the Seebeck effect based on this mechanism is expected to be about 3 orders of magnitude smaller than the Nernst effect.

Recently, the Nernst effect has been observed in superconducting polycrystalline[20] and epitaxial[21] Y-Ba-Cu-O films and in polycrystalline Pb-doped Bi-Sr-Ca-Cu-O.[13] The Nernst effect can be explained in terms of the flux motion caused by the thermal force f_{th}, and its order of magnitude agrees with the value predicted by the time-dependent Ginzburg-Landau theory.[21] In contrast to the Nernst effect, the appearance of a Seebeck voltage in the mixed state of a high-T_c superconductor of the same order of magnitude as the Nernst voltage[13] cannot be explained in terms of the thermal force f_{th}, and the two-fluid model described above is more adequate.

An important difference between this mechanism of the Nernst effect and our model for the Seebeck effect is the following. For the Nernst effect only Abricosov vortices are assumed to move down the temperature gradient, thereby changing the phase difference and creating the Nernst voltage. However, for generating the Seebeck effect, in addition to the scheme shown in Fig. 1, at least two other phase-slip mechanisms, namely weak links and phase-slip centers, need to be discussed.[22]

In the vicinity of T_c a sharp peak of the thermopower strongly sensitive to an external magnetic field has also been observed for Y-Ba-Cu-O single crystals.[10] The authors of Ref. 10 discuss this peak in terms of a fluctuation effect. However, in view of an unreasonable value obtained for the electron diffusion constant, there remains some controversy. Our model suggests that the peak in the thermopower is likely due to Kosterlitz-Thouless (KT) behavior. The thermally excited unbound vortex-antivortex pairs existing in the temperature range between T_{KT} and T_c clearly contribute to the Seebeck effect due to their interaction with the supercurrent density j_s. Since the KT behavior is essentially a zero-field effect, in the presence of interaction between vortices the disappearance of the peak with increasing magnetic field is expected.

It is interesting that the KT behavior is not expected to contribute to the Nernst effect since the thermal force f_{th} is pointing in the same direction for the vortex and the antivortex. Hence, combined measurements of the Seebeck and the Nernst effect would be interesting for clarifying the KT contribution to the Seebeck effect.

It is interesting to note also that the supercurrent density j_s generated in a temperature gradient according to the scheme in Fig. 1 is not experiencing spatial redistribution effects due to inhomogeneous flux pinning or flux flow. This absence of redistribution effects is in contrast to the situation for an applied electric current flowing through a superconductor at constant temperature.

The Seebeck effect in the mixed state of high-T_c superconductors represents a beautiful demonstration of the fountain effect displayed by superfluids. Because of the negligible amount of flux pinning above the irreversibility line in high-T_c superconductors,[6–8] such a display of the fountain effect in the mixed state in terms of a Seebeck voltage is much more pronounced in these materials than in the classical superconductors.

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