Strong Nonlinear Galvanomagnetic Effects in Thin Superconducting Films

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We show that in a certain range of parameters the interaction between vortices causes significant enhancement of galvanomagnetic effects in thin superconducting films, as opposed to thick films. In particular, the Nernst coefficient acquires a contribution exceeding the single-vortex term at relatively small external magnetic fields. The possibility of inducing the dc voltage due to either vortex-antivortex pair production by external ac currents or pair-breaking radiation is also discussed. Our results give a possible explanation for the experiments of A. Gerber and G. Deutcher.

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In normal metals, thermomagnetic effects appear to be hardly observable. However, these effects are enhanced by about 3 orders of magnitude in superconducting (SC) phases of metals due to magnetic vortices' motion [1,2]. Particularly, the Nernst effect accounts for an appearance of the transverse electric field E_y in the presence of both external magnetic field H_z and the temperature gradient $\nabla_x T$ as

$$E_y = N\nabla_x T\,, \tag{1}$$

where N is the Nernst coefficient. In the SC phase, this effect occurs because magnetic vortices of one sign of vorticity (e.g., "+") move in the external temperature gradient with some drift velocity V_x and produce the electric field in the y direction according to Josephson's formula [3]

$$E_y = c^{-1} V_x \Phi_0 N_+\,. \tag{2}$$

Here, $N_+ = H_z/\Phi_0$ is the vortex concentration; Φ_0 is the unit flux. The drift velocity turns out to be related to the entropy S_1 carried by one vortex [2] and the effective viscosity η [4], and leads to the expression for the Nernst coefficient as

$$N = (S_1/c\eta)H_z\,, \tag{3}$$

which turns out to be a linear function of the weak external magnetic field (in the absence of vortex pinning). Theoretical estimates of S_1 [2] give the correct order of magnitude for a voltage (1) as high as 10^{-6}–10^{-7} V/cm for $\nabla_x T \sim 1$ K/cm [5]. One should note that the estimate [2] was made for thick films, when the thickness d is much greater than the London penetration length λ_L.

The exact solution for S_1 for the magnetic field close to the upper critical field H_{c2} was given by Caroli and Maki [6]. In this limit, $S_1 \sim (H_{c2} - H)$ which yields

$$N \sim H_{c2} - H\,, \quad H \to H_{c2}\,. \tag{4}$$

It is easy to see that the solution [6] is applicable to both thick and thin films, when

$$d \le \lambda_L\,. \tag{5}$$

Indeed, near H_{c2} the order parameter tends to zero as $\sqrt{H_{c2} - H}$ which makes the result [6] exact, since the effects of vortex interaction are negligible.

It is known that the pair vortex-vortex interaction is essentially short ranged in thick SC films. However, as was shown by Pearl, this interaction becomes long-ranged in the case of thin films [7]. The force acting between the two vortices in the near-2D films [see condition (5)] is given as

$$F_{12} = \begin{cases} Q_{3D}^2/r^2, r \gg \lambda_\perp\,, & \lambda_\perp \equiv \lambda_L^2/d\,, \\ Q_{2D}^2/r, r \le \lambda_\perp \end{cases}$$

$$Q_{3D} \equiv \frac{\Phi_0}{2\pi}, \quad Q_{2D} \equiv \frac{\Phi_0}{2\pi\sqrt{2\lambda_\perp}}\,. \tag{6}$$

The long-distance behavior of the interaction turns out to be precisely the 3D Coulomb law with the effective charge Q_{3D}. It happens because in thin SC films the magnetic field appears not to be totally screened by the SC currents (see in [7], p. 60).

Usually, the properties of the 2D SC films are considered close to the SC transition temperature T_c, where the crossover parameter $\lambda_\perp$ in (6) can be very large and even greater than the film's largest dimension, R. In such a case, the 2D Coulomb gas model (CGM; see [8]), ignoring the 3D Coulomb "tails" and therefore considering the limit $\lambda_\perp \gg R$, appears to be a very useful description of the 2D vortex ensemble. As a consequence, the lower critical field H_{c1} for thin films happens to be several orders of magnitude smaller than for thick films [9]. In thin films, the vortex-antivortex pairs can be easily created either thermally, leading to the Berezinskii-Kosterlitz-Thouless transition (see [8]), or by applying electric current, causing some peculiarities of the resistive state [10–12].

The objective of the present paper is to show that the galvanomagnetic effects in thin films are significantly enhanced in comparison with similar effects in thick films in a certain range of parameters, where the 2D CGM [8,9] is still valid.

The idea is that since the effective charges of "vortex Coulomb gas" and the crossover distance $\lambda_\perp$ in Pearl's formula (6) depend on the average SC density n_s [8], even a small gradient of n_s may strongly disturb the equilibrium of vortices, producing additional driving forces

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acting on the system. If this gradient is steady but nonequilibrium these driving forces can result in a steady current of vortices, giving rise to the voltage (2).

To describe this effect, we should derive an expression for the total force acting on the ensemble of vortices in the case when external sources produce a steady nonequilibrium distribution of normal components, resulting in the change $\Delta n_s = \tilde{n}_s - n_s$ of superconducting component density from the equilibrium value n_s to some nonequilibrium concentration $\tilde{n}_s$.

The infinitesimal work of external sources, creating some deviation δn_s can be represented as

$$\delta W = \int \xi \delta n_s \, dV \,, \tag{7}$$

where ξ has to be chosen so that the solution of the energy balance equation

$$\xi + \delta F(\tilde{n}_s)/\delta \tilde{n}_s = 0 \tag{8}$$

is satisfied for the steady-state solution $\tilde{n}_s$. In (8), F stands for the free energy of the superconductor which can be taken in Ginzburg-Landau form [7] as

$$F = \int d^2x \int_{-d/2}^{d/2} dz \left[\frac{\hbar^2}{4m} (\nabla \sqrt{n_s})^2 + \frac{\hbar^2}{4m} n_s \left(\nabla \Theta - \frac{2e}{\hbar c} \mathbf{A} \right)^2 + a n_s + \frac{b n_s^2}{2} \right] + \int d^2x \int_{-\infty}^{+\infty} dz \frac{(\mathbf{B} - \mathbf{H})^2}{8\pi} \,, \tag{9}$$

where $\mathbf{B}$ and $\mathbf{H}$ are total and external magnetic fields, respectively, $\mathbf{A}$ is the vector potential, and Θ is the phase of order parameter $\Psi = \sqrt{n_s} e^{i\theta}$. Parameters a, b, and m have the same meaning as in BCS theory [7]. In (9), the film is located in the (x,y) plane in the region $-d/2 \le z \le d/2$.

As follows from (7), the external force acting on the vortex ensemble is defined as $\mathbf{f} = \delta W / \delta \boldsymbol{\rho}$, or

$$\mathbf{f} = \int \xi \frac{\delta}{\delta \boldsymbol{\rho}} (\tilde{n}_s) dV \,, \tag{10}$$

where $\boldsymbol{\rho}$ is a center-of-mass coordinate for the vortex ensemble. If $\Delta n_s = \tilde{n}_s - n_s$ is small, then we can make an expansion in (8), which gives

$$\xi + (\delta^2 F / \delta n_s^2) \Delta n_s = 0 \,. \tag{11}$$

Substitution of Eq. (11) into (10) yields

$$\mathbf{f} = -\int \frac{\delta^2 F}{\delta n_s^2} \Delta n_s \frac{\delta}{\delta \boldsymbol{\rho}} (\tilde{n}_s) dV \,. \tag{12}$$

Since we are looking for linear response, we should substitute the nonperturbed solution $\tilde{n}_s \approx n_s$ into (12). A variation of (9) gives

$$b n_s + a + \frac{\hbar^2}{4m} \left(\nabla \Theta - \frac{2e}{\hbar c} \mathbf{A} \right)^2 - \frac{\hbar^2}{4m} \frac{\nabla^2 \sqrt{n_s}}{\sqrt{n_s}} = 0 \tag{13}$$

for such a solution. The vortices are well defined if the correlation length ξ, determining the size of vortex core, obeys the condition

$$\xi^2 N_+ \ll 1 \tag{14}$$

of low density of vortices. Therefore, the last term on the left-hand side of (13) can be omitted as being nonzero in the vicinity of cores only. It allows us to consider n_s as a smooth variable and to find $\delta^2 F / \delta n_s^2 = b$, which does not depend on center-of-mass coordinate $\boldsymbol{\rho}$ of the vortex ensemble. Taking into account that a also does not depend on $\boldsymbol{\rho}$, we obtain

$$\frac{\delta}{\delta \boldsymbol{\rho}} (n_s) = \left(-\frac{1}{b} \right) \frac{\delta}{\delta \boldsymbol{\rho}} \left[\frac{\hbar^2}{4m} \left(\nabla \Theta - \frac{2e}{\hbar c} \mathbf{A} \right)^2 \right] . \tag{15}$$

Substituting (15) into (12) and integrating by parts, we finally get

$$\mathbf{f} = \int dV \, \nabla (\Delta n_s) \frac{\hbar^2}{4m} \left(\nabla \Theta - \frac{2e}{\hbar c} \mathbf{A} \right)^2 , \tag{16}$$

where the fact that the phase and the vector potential depend on $(\mathbf{x} - \boldsymbol{\rho})$ as $\Theta(\mathbf{x} - \boldsymbol{\rho})$, $\mathbf{A}(\mathbf{x} - \boldsymbol{\rho})$ was taken into account. Using the definition of electric current $\mathbf{J} = -c \, \delta F / \delta \mathbf{A}$ and introducing the 2D current $\mathbf{I} \equiv \int_{-d/2}^{d/2} dz \, \mathbf{J}$ we can rewrite (16) as

$$\mathbf{f} = \int d^2x \frac{2\pi}{c^2} \lambda_\perp \mathbf{I}^2 \frac{\nabla (\Delta n_s)}{n_s} \,. \tag{17}$$

The distribution of currents $\mathbf{I}(x)$ can be taken as a sum of currents produced by each vortex. Making use of Pearl's solution, we obtain [7]

$$\mathbf{I}(q) = \sum_j \frac{c \Phi_0}{2\pi} \frac{i \mathbf{q} \times \mathbf{n}}{q(1 + \lambda_\perp q)} e^{i \mathbf{q} \cdot \mathbf{x}_j} \,, \tag{18}$$

where $\{\mathbf{x}_j\}$ are coordinates of vortex cores, $\mathbf{n}_j = (0,0,\pm 1)$ is a unit vector along the z axis, and $\pm$ stands according to the vorticity of the jth vortex. The vortex ensemble may be treated as the charged 2D Coulomb gas [8,9], where screening is possible. However, as shown by Doniach and Huberman in [9], the net effective charge (read the net paramagnetic moment) in the presence of the external magnetic field remains unscreened at large distances. In our case, it means that the harmonic with $q \sim 1/R$ in (18) is not affected by screening (compare to [13]). Note that this harmonic gives the main contribution to (18). It allows us to omit the rest of the harmonics in (18) and after substitution of (18) into (17), to obtain

$$\mathbf{f} = \frac{\nabla \Delta n_s}{n_s} \frac{\Phi_0^2 \lambda_\perp N_+^2}{2\pi} \frac{R^2}{(1 + \lambda_\perp / R)^2} \,. \tag{19}$$

Now, we can estimate the drift velocity of the equilibrium ensemble of vortices by assuming that the motion is described in terms of viscous flow [4] with the total viscous force $\mathbf{f}_\eta = -\mathbf{V}_d \eta N_+ R^2 d$, balancing the external force $\mathbf{f}$. Consequently, after substituting $\mathbf{V}_d$ into (2), we obtain

$$E_y = N_1 \frac{\nabla_x(\tilde{n}_s - n_s)}{n_s}, \quad N_1 \equiv \frac{\Phi_0}{2\pi c\eta} \frac{\lambda_\perp H_z |H_z|}{(1+\lambda_\perp/R)^2 d}. \tag{20}$$

Note that in the 3D case the corresponding term disappears as $\lambda_L^2/R^2 \to 0$ due to the absence of long-range forces.

Strictly speaking, this relation is only valid far from the critical line $H_{c2}(T)$ in the phase diagram of a superconductor. One can easily see that a certain line $H = H^*(T) < H_{c2}(T)$ may exist, such that for $H \geq H^*(T)$ the omission of the last term in (13) is no longer valid and the solution should smoothly transform into (4), according to [6]. We can estimate $H^*(T)$ by implementing the requirement that in the presence of external field H_z the solution for the average density n_s in (13) does not significantly change. Integrating (13) over the film and taking (18) into account, we arrive at

$$H^*(T) = \frac{H_c}{2}\left(1+\frac{\lambda_\perp}{R}\right)\left(\frac{d}{\lambda_\perp}\right)^{1/2}\left(1-\frac{T}{T_c}\right)^{1/2}, \tag{21}$$

where the relation [7] $a = [H_c^2/4\pi n_s(0)](1-T/T_c)$ for the GL coefficient in terms of the critical field H_c and superconducting component density $n_s(0)$ at $T=0$ was employed.

The 2D CGM assumes the limit $\lambda_\perp \gg R$, in which there is no limitation on H_z, since H^* in (21) is formally infinite. In the realistic case, when T is not too close to T_c, this field can be considerably smaller than H_c. It implies that the superposition principle for currents (18) is no longer valid for $H > H^*(T)$, as the superconducting density n_s changes significantly. This means that the 2D CGM is not applicable to this case. However, in this paper we will only attempt to estimate the maximum value of the coefficient N_1, supposedly achieved at $H \approx H^*(T)$. A substitution of (21) into (20) gives us

$$N_{\max} = \frac{\Phi_0}{c\eta}\frac{H_c^2}{4\pi}\left(1-\frac{T}{T_c}\right). \tag{22}$$

In (20), we can express $\nabla(\tilde{n}_s - n_s)$ in terms of some temperature gradient, if there is any in the system. For $T \to 0$, one finds $\nabla(\tilde{n}_s - n_s)/n_s \sim -e^{-\Delta/T}\nabla T$, where Δ stands for the superconducting gap. After comparing with (1), it results in $N \sim e^{-\Delta/T} \to 0$. For T near (but not too close to) T_c we have $\nabla(\tilde{n}_s - n_s)/n_s \sim -\nabla T/T_c$ and consequently

$$N = N_1/T_c \tag{23}$$

for Eq. (1). Taking into account typical values $H_c \sim 10^3\text{-}10^4$ G, $\eta = 10^{-6}\text{-}10^{-7}$ g/s, one arrives at the estimate

$$E_Y \leq (10^{-1}\text{-}10^{-4})\frac{\nabla T}{T_c}\ (\text{V/cm}). \tag{24}$$

Therefore, for temperature gradients as small as $0.1T_c$/cm the produced voltage may be of the order of mV/cm, exceeding the characteristic values of the Nernst voltage in thick films [1,2,5] by several orders of magnitude.

It is interesting to compare our result (23) and (20) for the Nernst coefficient to that obtained for thick films [1,2]. Using the results [1,2] and (23) and (20), one obtains the estimate

$$\frac{E^{(2D)}}{E^{(3D)}} \cong \frac{H}{H_c}\frac{\lambda_L^2(T)\lambda_L(0)}{\xi(0)(d+\lambda_L^2(T)/R)^2}, \tag{25}$$

where $E^{(2D)}$ and $E^{(3D)}$ stand for the values of the transverse electric field in thin and thick films, respectively. Let us take a case where $\lambda_L^2/d \ll R$ and $\lambda_L \cong 1000$ Å, $\xi \approx d \cong 100$ Å. From (25) we get $E^{(2D)}/E^{(3D)} \sim (H/H_c) \times 10^3$. In this case, (21) implies that $H^* \cong 0.1 H_c$. Thus, we have $E^{(2D)}/E^{(3D)} \sim 10^2$.

Analyzing (25), one arrives at the conclusion that for the fields as high as $H \geq H_{\mathrm{cr}}$ where

$$H_{\mathrm{cr}} = \frac{H_c^2}{H_{c2}}\left(1+\frac{\lambda_\perp}{R}\right)^2\frac{d}{\lambda_\perp}, \tag{26}$$

the collective contribution to the Nernst voltage (23) and (20) exceeds the one-vortex term (25). Note that the crossover field H_{cr} turns out to be smaller than H^* (21). This means that in this region our calculations and the 2D Coulomb gas model are still valid. However, when $\lambda_\perp$ become considerably large in comparison to the characteristic size of the system R, H_{cr} is of the same order as H_c implying that the interaction effects are insignificant in the very vicinity of T_c.

In case the nonequilibrium gradient in (20) exists due to a gradient ∇n_f in vortex-antivortex pair density created by an external current, the estimate for E_y turns out to be

$$E_y \leq -(10^{-1}\text{-}10^{-4})\xi^2\nabla n_f\ (\text{V/cm}), \tag{27}$$

where it was assumed that for the high density of pairs the lower estimate for $\tilde{n}_s$ should be $\tilde{n}_s \approx n_s(1-\xi^2 n_f)$ (compare with [8,13]).

It is worth noting that the external inhomogeneous pair-breaking microwave radiation may produce the steady nonequilibrium gradient of the normal component in a thin SC film. It should also produce the gradient of the SC density and, consequently, the driving forces acting on vortices. Then, (20) is still valid and the dc voltage is proportional to the square of the external magnetic field. In this case, the dc voltage does not contain the linear terms $\sim H_z$, as opposed to the Nernst effect.

Now, let us consider the recent experiment [14] on the ac-to-dc conversion in thin superconducting granular films subjected to both static magnetic field and ac external currents. The dc voltage measured across the sample was dependent upon the external magnetic field **H** and amplitude of the applied ac current. As mentioned by the authors in [14], the voltage of the order of tens of microvolts was observed at ac frequencies as low as ~ 10 Hz ruling out an explanation in terms of the inverse Josephson effect. The results of [14] have several peculiar features: (i) For small external magnetic fields, the dc voltage has an odd component as a function of H_z. (ii) When the direction of the sample is reversed, the induced voltage changes sign. The direction of the sample was defined as a direction from the grounded side to the side subjected to external ac current [14]. (iii) The effect has a threshold with respect to the ac current amplitude and the typical values of the voltage vary from ~ 10 mkV/cm to ~ 1 mV/cm. (iv) The dc voltage has its maximum values for $H \approx H_c \approx 10^4$ G and $T \cong T_c$.

These features can be understood within the framework of the model presented above. Indeed, general symmetry and time-reversal considerations imply the presence of some steady current changing its sign after time reversal. This can be due to either the Nernst effect (enhanced, as it was shown above) in some uncontrollably small temperature gradient, or to inhomogeneity of vortex pair creation by the external ac current. Feature (ii) gives a hint of this possibility. The pair creation mechanism seems to be more realistic, because the simple analysis of data [14] shows that the dc voltage increases almost linearly with the current amplitude above the threshold. This behavior resembles the regime considered before in [10,11], when the density of vortex pairs increases linearly with current. As was emphasized in [10,11], the conditions for pair creation in thin films are very sensitive to different parameters of the film, including the temperature. Therefore, a small temperature gradient can produce a pair concentration gradient which in its turn causes the unipolar vortex motion and creates dc voltage.

Note that the typial size of a granule in [14] was of the order of 10^{-3}–10^{-4} cm with thickness of film being about 10^{-6} cm. In a dirty superconductor, one should expect λ_L to increase up to $\sim 10^4$ Å (see [7]). Going back to (21), we see that the range of applicability of the CGM expands to $H^* \cong H_c$. In this case, the crossover field $H_{\rm cr}$, given by (26), is of the order $H_{\rm cr} \cong H_c \times (\xi/\lambda_L) \ll H_c$. There is another interesting feature of the results [14], which can probably be prescribed to some vortex pinning inside a grain. The odd component of dc voltage as a function of the external magnetic field has well-pronounced peaks. As was mentioned by the authors [14], those peaks approximately corresponded to the penetration of one additional elementary flux into the grain. The reason for that might be a finite vortex pinning, which we did not take into account. One can expect that the penetration of one additional flux into the grain may change the balance between the collective forces considered above and pinning and cause a finite flow of vortices.

However, the more detailed interpretation of the results [14] requires some additional analysis, which should probably take into account some percolation properties of the samples. For example, here we only concentrated on the voltage component which was antisymmetric with respect to the external field. In [14], both symmetric and antisymmetric components were observed. For that symmetric component, our simple considerations of a "steady flux" flow may be reconsidered. In fact, for the odd component we had for the local electric field $\mathbf{E} \sim \mathbf{j}(|\mathbf{H}|) \times \mathbf{H}$ where $\mathbf{j}(|\mathbf{H}|)$ stands for the gradient of some thermodynamical variable. This seems to be the only possibility to satisfy both the features (i) and (ii) due to the general considerations of T invariance and feature (ii). However, for the even component we may have $\mathbf{J} \sim \mathbf{H} \times \mathbf{D}$, where $\mathbf{D}$ is some vector characterizing asymmetry of the sample. This gives $\mathbf{E} \sim \mathbf{D}(\mathbf{H})^2$. The more detailed analysis will be a subject of our further efforts.

In summary, the contribution to the Nernst coefficient in thin superconducting films due to vortex-vortex interaction was obtained. We also consider another situation, typical for thin films, where the external electric current applied to the system creates nonequilibrium gradient of vortex-antivortex pairs density. Our calculations are valid in the region where the 2D Coulomb gas model [8,9] is applicable. The contribution we obtain turns out to be several orders of magnitude larger than a single-vortex term. We compare our results with unusually large values obtained in the experiment [14] and find that our calculations give the same order of magnitude values. For this reason, we think that the more detailed experimental studies of galvanomagnetic effects in thin superconducting films would be of major interest.

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