Persistent Currents in Charge-density Wave Systems

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(November 21, 2018)

The inductive exchange of carriers between closed Fermi surface sections subject to Landau quantization and open Fermi surface sections subject to charge-density wave (or spin-density wave) formation is shown to give rise to persistent currents. This mechanism may explain recent experimental data in certain organic conductors in high magnetic fields.

It is well known that a gradient in the orbital magnetization within a bulk metal corresponds to a net effective electrical current density \( j = \nabla \times \mathbf{M} \). Such currents can be true macroscopic flows of charged quasiparticles, created in response to changes of magnetic field (i.e. \( \partial \mathbf{H}/\partial t \)) and, in the case of superconductors, applied magnetic field \( \mathbf{H} \). They are considered to be “persistent” if they exist in equilibrium, and truly persistent currents are understood to exist only in superconductors [2]. In the type I superconducting phase, the Meissner effect screens all electromagnetic fields from within the bulk [2], while in the type II superconducting phase, net currents that permeate the bulk occur transverse to a vortex density gradient held in place by vortex pinning [2]. Long-lived currents [3] also occur in quantum Hall systems orthogonal to a chemical potential gradient \( \nabla \mu \). The equilibrium Hall electric field is sustained owing only to the absence of quasiparticle scattering processes orthogonal to the current [4].

In this paper, I identify a further mechanism for persistent currents involving the coexistence of orbital magnetism and a charge-density wave (CDW) or spin-density wave (SDW) modulation [5]. With the pinning of a CDW (or SDW) modulation [5], a situation can be realised whereby a differential chemical potential gradient \( 2 \nabla (\Delta \mu) \) exists between the CDW (or SDW) and additional ungapped sections of Fermi surface present. Currents result within the bulk if, and only if, the derivative \( \partial \mathbf{M}/\partial (\Delta \mu) \) is finite within the bulk. This quantity is vanishingly small in most metals at low magnetic fields [5], but can become quite significant when Landau levels are formed at high magnetic fields [5]. The development of the gradient \( 2 \nabla (\Delta \mu) \) relies on there being a continual relative exchange of carriers \( 2 \partial (\Delta N)/\partial |\mathbf{H}| \) between Fermi-surface sheets as the magnetic field is swept [5]. Again, this is a negligible effect at low magnetic fields, but becomes very significant when Landau levels are formed at high magnetic fields [5]. The mechanism for persistent currents in CDW (or SDW) systems, described here, will therefore be seen to be quite different than that predicted by Fröhlich [5].

A multiband metal consisting of separate one-dimensional (1D) and two-dimensional (2D) Fermi surface sections is an ideal system in which to model these effects. Fermi surfaces of this topology are found to occur in several organic metals [8][9], and effects consistent with the existence of persistent currents have been observed experimentally [10][11].

I begin by considering a metal that consists of only a single 2D pocket, characterized by a de Haas-van Alphen (dHvA) frequency \( F \) and cyclotron frequency \( \omega_c = eB/m^* \), where \( B = \mathbf{B} \cdot \hat{z} \). When the quasiparticles move in planes perpendicular to the unit vector \( \hat{z} \), Landau levels with eigenvalues \( \varepsilon = \hbar \omega_c (n + \frac{1}{2}) \) are formed, where \( n = 0, 1, 2, \ldots \). The chemical potential \( \mu \) is pinned to the highest occupied of these in order to conserve the total number of 2D carriers, \( N_{2D} \). It is instructive to consider contrasting situations at half-integral and integral Landau level filling factors \( \nu \). At half-integral filling factors (when \( \nu + \frac{1}{2} \) is an integer) the highest occupied Landau level is exactly half filled, while at integral filling factors (when \( \nu \) is an integer) \( \mu \) is in the transitional region between filled and empty levels. In the low temperature limit (\( T \rightarrow 0 \)), the corresponding susceptibility components \( \chi_{zz} = \partial M/\partial H \) are [12]

\[
\chi_{\text{half}} = \frac{\hbar \omega_c}{e \rho_{xy} \mu_0 H^2} F
\]

and

\[
\chi_{\text{int}} = -\frac{\pi \omega_c \tau}{4} \frac{\hbar \omega_c}{e \rho_{xy} \mu_0 H^2} F
\]  

(1)

respectively, where \( \rho_{xy} \) is the off-diagonal component of the resistivity tensor, \( M = \mathbf{M} \cdot \hat{z} \) and \( H = \mathbf{H} \cdot \hat{z} \). Note that the susceptibility at integral filling factors is extremely sensitive to Landau level broadening that results from a finite relaxation time \( \tau \). It follows that \( M \) and \( \mu \) are related via the differential

\[
\frac{\partial \mathbf{M}}{\partial \mu} = \frac{N_{2D}}{\mu_0 H} \hat{z} = \frac{1}{e \rho_{xy}} \hat{z},
\]

(2)

where both \( e \) and \( \rho_{xy} \) are negative for electrons and positive for holes [13].

When 1D carriers are added to the system, they primarily function as a charge reservoir [4]. At half-integral filling factors, carriers are transferred from the 1D sheets to the 2D pocket as \( H \) is increased, causing the magnetization to increase more quickly than in Equation (1).
At integral filling factors, the inverse situation becomes true. The presence of 1D states in the Landau gaps prevents $\mathbf{M}$ and $\mu$ from jumping abruptly between Landau levels. The susceptibilities therefore become

$$
\chi_{\text{half}} = \frac{g_{1D} \hbar \omega_c}{(g_{1D} + g_{2D}) \epsilon \rho_{xy} \mu_0 H^2} F
$$

and

$$
\chi_{\text{int}} = -\frac{g_{1D} \hbar \omega_c}{(g_{1D} + g_{2D}) \epsilon \rho_{xy} \mu_0 H^2} F,
$$

where $g_{1D}$ and $g_{2D}$ represent the density of states of the 1D sheets and 2D pocket respectively.

The process by which the 1D sheets function as a charge reservoir becomes perturbed upon opening of a gap. For the purpose of modeling these perturbations, it is convenient to consider a simplified system in which the Fermi surface nesting gaps the entire 1D density of states and the effects of Zeeman splitting of the 1D bands are negligible. The latter is true in SDW systems and also in CDW phases in which the spin up and spin down bands nest independently.

By far the most important consideration is that the CDW (or SDW) phase remains stable over the field range where the dHvA oscillations occur. Persistent currents are anticipated only in the case of an incommensurate CDW (or SDW) in which the nesting vector $\mathbf{Q}$ can adjust itself in order to minimise the total free energy of the system. They are not expected in the case of a commensurate CDW phase that is pinned to the crystal lattice. Following the solution of the gap equation obtained by Maki and Tsuneto, adapted to include the effects of a shift in the chemical potential (labelled here as $\Delta\mu$) in Reference, the change in free energy in forming such a CDW (or SDW) groundstate can be written as

$$
F = -g_{1D} \left[ \frac{\Delta \mu^2}{2} - (\Delta \mu)^2 \right] - F_{2D}^r(\Delta\mu).
$$

The first two terms of Equation are minimised when the zero-temperature gap $2\Delta_0$ that opens on the 1D sheets is centred about $\mu$, so that an offset $\Delta\mu$ of the gap relative to $\mu$ costs energy. The third term, $F_{2D}^r(\Delta\mu)$, accounts for an additional (either positive or negative) change in free energy of the 2D Landau level levels incurred by this offset. The offset translates directly into an increase in the number of states $\Delta N_{1D} = g_{1D} \Delta\mu$ accommodated by the 1D sheets, balanced by an equal and opposite reduction in the number of states contained in the 2D pocket. If $Q$ is the component of the optimum nesting vector $\mathbf{Q}$ parallel to the mean Fermi velocity $v_F$ of the 1D sheets, $\Delta\mu$ also results in a shift in this $Q$ given by $\Delta Q/\Delta\mu = Q_{1D}/N_{1D}$, where $N_{1D}$ is the equilibrium number of 1D states.

Should $Q$ be uniform throughout the sample as a result of cooling it in the presence of a constant magnetic field, a finite value of $\Delta\mu$ may already be required in order to minimise Equation owing to the $F_{2D}(\Delta\mu)$ term. Such an effect has already been considered to explain the suppression of oscillations in $\mu$ observed experimentally in certain organic metals. I will not consider this effect in detail for two reasons: first, it is not expected to operate at half-integral and integral filling factors where $\mu = F_{2D}(\Delta\mu) = \partial F_{2D}(\Delta\mu)/\partial \mu = 0$; second, I reveal below that it is unlikely that a uniform $\Delta\mu$ is maintained throughout the entire sample.

If the sample is field-cooled at $\mu = \Delta\mu = 0$, the starting condition of the sample corresponds to the point labelled ‘0’ in Fig. If the CDW (or SDW) is pinned, this pinning initially prevents $Q + \Delta Q$ from changing in response to a change in the applied magnetic field $\Delta H$. The optimum value of $Q$ does, however, change, leading to relative difference $\Delta Q$ between the actual and optimal values, and finally to a shift in $\Delta\mu$. Since the density of states at $\mu$ in the gap on the 1D sheets is zero, the sample begins by responding like a 2D metal in which there are no 1D states. From this point on, the magnetization becomes hysteretic. Consequently, the irreversible change in the magnetization is characterised by an irreversible susceptibility given by the difference between Equations and ; hence

$$
\chi_{\text{irr.half}} = -\frac{g_{1D} \hbar \omega_c}{(g_{1D} + g_{2D}) \epsilon \rho_{xy} \mu_0 H^2} F,
$$

and

$$
\chi_{\text{irr.int}} = -\left[ \frac{\pi \omega_c}{4} - \frac{g_{1D}}{(g_{1D} + g_{2D})} \right] \hbar \omega_c \frac{F}{\epsilon \rho_{xy} \mu_0 H^2}.
$$

As expected for magnetic hysteresis, $\chi_{\text{irr}}$ is always negative.

For a sample without boundary conditions, the magnetization would continue to change with the susceptibility given by Equation, giving rise to the dotted line in Figure. This would occur until $\Delta\mu$ reaches the thermodynamic limit, $\Delta\mu_{\text{lim}} = \pm \Delta\mu_0/\sqrt{2}$, when, upon Equation, is no longer negative. At this point, the magnitude of the irreversible magnetization would saturate at a maximum value, $M_{\text{irr,lim}} = \pm \Delta\mu_{\text{lim}} \times \partial M/\partial \mu = \pm \Delta\mu_0 2\rho_{xy}$, without any currents being induced within the bulk of the sample. Just as in superconductors and quantum Hall systems, however, a net magnetization cannot exist within the bulk without incurring a current $\mathbf{j}$ immediately inside its surface. Here, the current is carried by 2D carriers, and wherever these exist,

$$
\mathbf{j} = -\nabla(\Delta\mu) \times \frac{\partial \mathbf{M}}{\partial \mu},
$$

where $-\nabla(\Delta\mu)$ is the chemical potential gradient experienced by the 2D pocket. In order to maintain the equilibrium condition $\nabla \mu = 0$, an equal and opposite charge polarization field $\nabla(\Delta\mu)$ must also form on the nested 1D
Fermi surface sheets. Since CDWs (and SDWs) are electrostatically limited, rather than increasing indefinitely, $\nabla (\Delta \mu)$ must eventually be limited by the characteristic threshold electric field

$$\nabla (\Delta \mu)_{\text{lim}} = eE_t$$

required for its depinning. This equates to a critical current density

$$j_c = \frac{E_t \times \hat{z}}{\rho_{xy}}$$

for the carriers in the 2D pocket within the sample.

From Equation (6), one can see that the electrodynamics has parallels with the quantum Hall effect 1, with the exception that there exists zero net electric field in equilibrium: the quantity $E_t$ instead parametrizes the charge polarization field required to slide the CDW (or SDW), that ultimately leads to dissipation. The absence of a net electric field implies that the effect, described here, is not limited only to integral Landau level filling factors. The inductive behaviour of the sample is, in contrast, much more like that of a type II superconductor, but with an irreversible susceptibility $\chi_{irr}$ that can depart significantly from the ideal diamagnetic value $-1$. As $\Delta H$ increases, the surface region in which $E_t$ is polarized propagates further into the sample. The inductive response of the sample to a change in applied magnetic field $\Delta H$ is therefore indistinguishable from the critical state model developed by Bean 21, provided $E_t$ is finite for all directions within the planes. This is certainly the case in CDW (or SDW) systems for which $Q$ occurs at an oblique angle with respect to the lattice vectors, where Fermi surface nesting results in charge (or spin) modulation that is two-dimensional.

If one considers a cylindrical sample of radius $r$, with $\hat{z}$ aligned along its axis, following Bean 21, the irreversible magnetization becomes

$$M_{irr} = -\chi_{irr} \left[ \Delta H - \frac{(\Delta H)^2}{H^*} + \frac{(\Delta H)^3}{3H^{*2}} \right].$$

This region is identified by point ‘1’ in Fig. 1. In this model, $H^*$ is the characteristic coercive field while $\chi_{irr}H^*$ is the maximum magnetic field that can be screened within the sample. When $\Delta H = H^*$, corresponding to point ‘2’ in Fig. 1, the irreversible magnetization reaches its saturation limit for a cylindrical sample,

$$M_{irr} = \frac{\chi_{irr} H^*}{3} = -\frac{|j_c r|}{3} \hat{z}.$$  

When the direction in which the field is changed is reversed at point ‘3’ in Fig. 1, the initial susceptibility is, once again, given by Equation (6). A similar cubic law should apply to that in Equation (1) in the reversal state, but with double the interval in $\Delta H$ being required to reverse the polarity of $\nabla (\Delta \mu)$. The magnetization saturates again at point ‘4,’ and a full hysteresis loop results on further cycling of $\Delta H$.

I now compare the predictions of this model with recent measurements made on samples of the organic conductor $\alpha$-(BEDT-TTF)$_2$K$_3$Hg(SCN)$_4$ at high magnetic fields. This material has a Fermi surface which nearly matches that of the model 13. At low temperatures, $T \lesssim 8$ K, and low magnetic fields, $\mu_0H \lesssim 23$ T, $\alpha$-(BEDT-TTF)$_2$K$_3$Hg(SCN)$_4$ is believed to possess a commensurate CDW phase, transforming to one that is incommensurate at high magnetic fields $\mu_0H \gtrsim 23$ T 13 14 15 16 17 18 22 23. It is within the high magnetic field phase where effects consistent with persistent currents have been reported 13 14.

I begin my comparison by considering the irreversible susceptibility. On computing $\chi_{irr}$, using Equation (6) and the established parameters ($F \approx 670$ T, $m^* \approx 2\, m_e$, $N_{2D} \approx 1.6 \times 10^{26}$ m$^{-3}$, $g_{1D} \approx g_{2D}$ and $\tau \approx 2$ ps 22 for $\alpha$-(BEDT-TTF)$_2$K$_3$Hg(SCN)$_4$), the model estimates ($\chi_{mod}$) listed in Table I are found to agree rather well with the experimental values ($\chi_{exp}$) 4 both at half-integral and integral filling factors.

I now estimate the threshold electric field from the saturation magnetization $M_{irr,\, \text{max}}$ (listed in Table I as $M_{\text{max}}$). This is found to be marginally higher at half-integral filling factors than at integral filling factors in Reference 14, but becomes comparable at the lowest temperatures 11. Making the cylindrical sample approximation, with $r \sim 0.5$ mm, one can estimate the mean scalar critical current $j_c$ using Equation (10) and the corresponding threshold electric field $E_t$ using Equation (8). I believe the estimates in Table I to be physically realistic because the total potential difference across the sample $\psi_t = rE_t$ does not exceed the maximum thermodynamic potential $\sqrt{2e}\Delta_0 \sim 0.7$ meV (where $\Delta_0 \sim 0.5$ meV 12) that can be sustained without destroying the CDW groundstate.

Next, I consider the coercive fields required to reverse the polarity of the current. On applying the simple expression for $H^*$ in Equation (1), the values estimated in Table I compare favourably with experimental observations 14. The behaviour of the irreversible magnetization is also found to be very well described by a cubic law of the form given by Equation (4). Note that twice the interval in field $2H^*$ is required to reverse the polarity of the currents throughout the entire sample. Meanwhile, the maximum magnetic field that can be screened by the sample, $\chi_{irr} H^*$, is a small fraction of a millitesla.

Finally, I consider an odd situation that occurs at the minima and maxima of the dHvA oscillations where the orbital susceptibility $\chi = \partial M/\partial H$ vanishes. Consequently, there will be discrete fields at which $\chi_{irr} \rightarrow 0$ causing the irreversible magnetization never to reach saturation within the quarter period of the dHvA oscillations $\Delta H = \mu_0 H^2/4F$. Because there exists a chemical
potential gradient $\nabla (\Delta \mu)$ across the sample, however, the situation where $M_{\text{irr}} = 0$ can only exist in a narrow region of annular topology within the interior of the sample. The effect should still be observable, however, as drop in the volume-averaged saturation magnetization. This could provide an explanation for the drop in the saturation magnetization observed at intermediate filling factors at the lowest temperatures in Reference [1].

In conclusion, I have identified a mechanism for persistent currents in metals in which a CDW (or SDW) phase coexists with a strongly field-dependent orbital magnetization. The magnetic field drives an exchange of carriers between the 1D and 2D Fermi surface sheets, which has the effect of stretching or compressing the CDW (or SDW) like a concertina. Persistent currents result when pinning the CDW (or SDW) resists the concertina-like motions. The charge polarization field of the pinned CDW negates the Hall electric field that would normally give rise to the Hall effect [1]. However, because the CDW (or SDW) exists in a critical state on the brink of sliding, the magnetic properties resemble those of a type II superconductor [21]. Like a superconductor, currents exist only because of impurity pinning. The mechanism for persistent currents, described here, however, is neither the quantum Hall effect nor superconductivity, but one that is entirely different. I propose this effect as an explanation for persistent currents observed in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ [11,12].

The work is supported by the Department of Energy, the National Science Foundation (NSF) and the State of Florida. I would like to thank John Singleton, Albert Migliori and Arzhang Ardavan for stimulating discussions.

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FIG. 1. Magnetic hysteretic behaviour anticipated by the persistent current model for a cylindrical sample, as described in the text.

| filling | $\chi_{\text{mod}}$ | $\chi_{\text{exp}}$ | $M_{\text{max}}$ | $j_c$ | $E_c$ | $\mu_0 H^*$ | $\mu_0 \chi_{\text{exp}} H^*$ |
|---------|-----------------|-----------------|-----------------|--------|------|-------------|-----------------|
| half    | -0.7            | -0.8            | 80              | 50     | 0.61 | 0.3         | 0.4              |
| int     | -5              | -4              | 50              | 30     | 0.36 | 0.2         | 0.5              |

TABLE I. A comparison of the irreversible susceptibility of $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ expected for an ideal sample according to the model, $\chi_{\text{mod}}$, with that obtained experimentally, $\chi_{\text{exp}}$ [12]. Also listed are the maximum irreversible magnetization, $M_{\text{max}}$, obtained experimentally together with the critical current density, $j_c$, CDW depinning threshold electric field, $E_c$, total potential difference, $\nu_0$, coercion field, $\mu_0 H^*$, and screening field, $\mu_0 \chi_{\text{exp}} H^*$, calculated according to the model.
Figure 1 of Harrison et al