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# Exchange-correlation effects in magnetic-field-induced superconductivity

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Motivated by recent experiments on the organic superconductor $\lambda$-$(BETS)_2FeCl_4$ we study magnetic-field-induced superconductivity, from the point of view of current-density-functional theory. It is found that both Meissner and Pauli pair breaking are suppressed by an exchange-correlation contribution to the vector potential, arising at the sites of the magnetic ions.

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The antagonistic nature of superconductivity and magnetism manifests itself via the pair breaking effect of external and internal magnetic fields. In the case of *external* magnetic fields this pair breaking can be due to the coupling of the field to orbital currents (the Meissner effect), or due to its coupling to the spins (Pauli, or paramagnetic, pair breaking). In the case of *internal* fields, pair breaking is due to magnetic ions in the lattice. These ions, too, in general make a spin and an orbital contribution to the system's susceptibility. On their own, both external and internal magnetic fields are normally adverse to superconductivity.

Intriguingly, under suitable circumstances these two detrimental agents, when present simultaneously, can compensate each other and give rise to superconductivity at extremely high external magnetic fields.[1] This phenomenon is known as the Jaccarino-Peter effect (JPE). At the heart of the JPE is the observation that in the presence of magnetic ions in the lattice an externally applied magnetic field acts in two ways on the conduction electron spins: directly, by its Zeeman coupling, and indirectly, by polarizing the magnetic ions whose exchange field in turn polarizes the conduction electrons. The JPE arises when the actions of the external and the exchange field on the conduction electrons cancel each other.[1,2] The present paper is devoted to a reanalysis of the JPE. It is pointed out that the conventional JPE scenario, just outlined, does not completely account for the experimental observations. A modification of this scenario is proposed and compared with available experimental data.

The JPE has very recently been observed in the organic conductor $\lambda$-$(BETS)_2FeCl_4$, for magnetic fields ranging from 18 to 41 T.[3,4] Before that it had already been seen in a variety of inorganic materials, most prominently in the pseudoternary chalcogenides $Sn_{1-x}Eu_xMo_6S_8$, where a JPE phase is observed between 4 and 23 T.[5–7] There is also strong experimental evidence that the JPE is at work in chalcogenides of the form $Pb_xEu_yMo_zS_8$,[8,9] in the transition-metal compounds $Mo_{1-x}Mn_xGa_4$,[10,11] in the heavy-fermion superconductors $CePb_3$ and $CeCu_2Si_2$,[12,13] and perhaps even in the high-$T_c$ cuprate $Gd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$.[14]

It is crucially important to recognize that in the standard JPE scenario[1,2] it is only the action of the external magnetic field on the electron *spins* that is compensated by the internal (exchange) field, not that on the *currents*. This imposes a severe restriction on the external field used for observation of the JPE: while it must be strong enough to cancel the exchange field arising from the magnetic ions, it must not simultaneously destroy superconductivity by its coupling to the orbital currents. One *ad hoc* way to reconcile these two constraints is to assume that in all systems in which the JPE has been observed the orbital upper critical field $H_{c2}$ is much higher than the external fields at which the JPE sets in.

In view of the recent experiments[3,4] on $\lambda$-$(BETS)_2FeCl_4$ it seems worthwhile to explore if this assumption is always necessary. In fact, in these experiments the JPE region in the phase diagram begins at 18 T, while $H_{c2}$ is about 3.5 T,[4] so that the relation between both fields is the opposite of what one would expect on the basis of the original theory of the JPE. Similarly, in the experiments[5–7] on $Sn_{1-x}Eu_xMo_6S_8$ the JPE manifests itself as a distinct phase in the temperature vs. magnetic field phase diagram, which appears for external fields higher than 4 T, whereas superconductivity first disappears, upon increasing the magnetic field from zero, at what appears to be a conventional orbital critical field of less than 1 T. The question thus poses itself: what protects the superconductor from *orbital* pair breaking in situations with *spin* compensation?

The fact that the JPE has been observed in physically very different systems, ranging from organic conductors and ternary chalcogenides to heavy fermion and high-temperature superconductors, suggests that the answer to this question is intrinsically tied to the physics of the JPE itself, and not a special feature of one particular type of material. To investigate these issues, we first restate the compensation between the external and the internal magnetic field in the framework of spin-density-functional theory (SDFT), which maps the many-body problem in the presence of the external field $\mathbf{H}_{ext}(\mathbf{r})$ on a single-body problem subject to the effective field[15]

$$\mathbf{H}_s(\mathbf{r}) = \mathbf{H}_{ext}(\mathbf{r}) + \mathbf{H}_d(\mathbf{r}) + \mathbf{H}_{xc}(\mathbf{r}). \quad (1)$$

Here the exchange-correlation (xc) magnetic field $\mathbf{H}_{xc}(\mathbf{r})$ is the SDFT counterpart to the internal exchange field of traditional theories of magnetism.[16] It is defined as the functional derivative

$$\mathbf{H}_{xc}(\mathbf{r}) = -\frac{\delta E_{xc}[n,\mathbf{m}]}{\delta \mathbf{m}(\mathbf{r})}, \quad (2)$$

where $E_{xc}[n,\mathbf{m}]$ is the xc functional of SDFT. $\mathbf{H}_d(\mathbf{r}) = \nabla \times \mathbf{A}_d(\mathbf{r})$ is a Hartree-like term arising from dipolar interactions. In the language of SDFT the JPE spin compensation would be described by saying that internal and external fields cancel each other on the average,[17] i.e.,

$$\mathbf{H}_{xc}(\mathbf{r})+\mathbf{H}_d(\mathbf{r})=-\mathbf{H}_{ext}(\mathbf{r})+\delta\mathbf{H}(\mathbf{r}), \quad (3)$$

for some range of densities and temperatures, so that the effective magnetic field $\mathbf{H}_s$ vanishes up to at most a remaining field $\delta\mathbf{H}$ that is much smaller than the critical field for Pauli pair breaking, $\mathbf{H}_P$, and thus not strong enough to destroy superconductivity paramagnetically. Once $\delta\mathbf{H}$ is of the order of $\mathbf{H}_P$ the cancellation (3) ceases to protect the superconductor from the external field, and the material becomes normal. This latter transition has already been observed experimentally.[4,5]

To study orbital currents, in particular diamagnetic pair breaking, one needs to go beyond SDFT and employ current-density-functional theory (CDFT).[18–20] CDFT is based on the many-body Hamiltonian

$$\hat{H}=\hat{T}+\hat{U}+\hat{V}-\int d^3r\,\mathbf{m}(\mathbf{r})\cdot\mathbf{H}_{ext}(\mathbf{r})$$
$$-\frac{q}{c}\int d^3r\,\mathbf{j}_p(\mathbf{r})\cdot\mathbf{A}_{ext}(\mathbf{r})+\frac{q^2}{2mc^2}\int d^3r\,n(\mathbf{r})\mathbf{A}_{ext}(\mathbf{r})^2, \quad (4)$$

where $\hat{T}$, $\hat{U}$, and $\hat{V}$ are the operators for the kinetic, interaction, and potential energy, respectively, $n(\mathbf{r})$ is the particle density, $\mathbf{m}(\mathbf{r})$ the spin magnetization, and $\mathbf{j}_p(\mathbf{r})$ is the paramagnetic current density. By construction, the CDFT single-particle equations reproduce the current, particle, and spin densities of the many-body Hamiltonian (4), and this property carries over to the extension of CDFT to the superconducting state.[21,22] In the CDFT single-particle equations[23] enter the effective magnetic field $\mathbf{H}_s^C(\mathbf{r})=\mathbf{H}_{ext}(\mathbf{r})+\mathbf{H}_d(\mathbf{r})+\mathbf{H}_{xc}^C(\mathbf{r})$, where

$$\mathbf{H}_{xc}^C(\mathbf{r})=-\frac{\delta E_{xc}^C[n,\mathbf{m},\mathbf{j}_p]}{\delta\mathbf{m}(\mathbf{r})} \quad (5)$$

[the superscript $C$ serves to distinguish $\mathbf{H}_{xc}^C$ from $\mathbf{H}_{xc}$ introduced in Eq. (2)—the two are in general not the same functional[24]], and the effective vector potential,

$$\mathbf{A}_s(\mathbf{r})=\mathbf{A}_{ext}(\mathbf{r})+\mathbf{A}_d(\mathbf{r})+\mathbf{A}_{xc}(\mathbf{r}), \quad (6)$$

whose exchange-correlation contribution is defined as

$$\mathbf{A}_{xc}(\mathbf{r})=-\frac{c}{q}\frac{\delta E_{xc}^C[n,\mathbf{m},\mathbf{j}_p]}{\delta\mathbf{j}_p(\mathbf{r})}. \quad (7)$$

$\mathbf{A}_d(\mathbf{r})$ describes the dipolar interactions. In general these interactions are much weaker than the xc effects, and one normally neglects $\mathbf{H}_d(\mathbf{r})$ and $\mathbf{A}_d(\mathbf{r})$ in Eqs. (1) and (6).[18–20] However, $\mathbf{A}_d(\mathbf{r})$ includes the Ampere term[19] which describes the current-current interactions that are responsible for the selfconsistent screening of the induced currents in the Meissner phase below $H_{c1}$.[21,22] We thus keep the dipolar terms in the equations.

Equation (3) expresses the fact that the basis for the absence of Pauli (paramagnetic) pair breaking is a mutual cancellation between internal and external magnetic fields. It is the main purpose of the present paper to point out that a similar cancellation between the vector potentials,

$$\mathbf{A}_{xc}(\mathbf{r})+\mathbf{A}_d(\mathbf{r})=-\mathbf{A}_{ext}(\mathbf{r})+\delta\mathbf{A}(\mathbf{r}), \quad (8)$$

can explain the absence of orbital (diamagnetic) pair breaking in the experiments listed above. Here $\delta\mathbf{A}(\mathbf{r})$ allows for imperfect cancellation, but is not strong enough to destroy superconductivity (i.e., $\delta\mathbf{H}=\nabla\times\delta\mathbf{A}$ is much smaller than the paramagnetic and diamagnetic critical fields $\mathbf{H}_P$ and $\mathbf{H}_{c2}$). Since the currents enter the Hamiltonian via their coupling to the vector potential, Eq. (8) implies ''current compensation,'' just as Eq. (3) implies spin compensation.

Initially, Eq. (8) can be interpreted as the CDFT formulation of the condition, already emphasized in earlier work in the field,[2,11,25] that to observe the JPE no significant orbital pair breaking may take place at the fields at which spin compensation happens. As such, Eq. (8) is a direct consequence of the experimental data. However, once the conditions for absence of paramagnetic and diamagnetic pair breaking are formulated in the language of CDFT, as is done in Eqs. (3) and (8), an additional piece of information becomes available: Within the formulation of CDFT proposed in Ref. 24 one can show quite generally [cf. Eq. (14) of that work, or Eq. (33) of Ref. 26] that the magnetic correlations described by $\mathbf{H}_{xc}^C$ and $\mathbf{A}_{xc}$ are not independent, but related in the same way as the external fields, i.e.,

$$\mathbf{H}_{xc}^C(\mathbf{r})=\nabla\times\mathbf{A}_{xc}(\mathbf{r}). \quad (9)$$

Hence Eq. (8) implies, upon taking the curl,

$$\mathbf{H}_{xc}^C(\mathbf{r})+\mathbf{H}_d(\mathbf{r})=-\mathbf{H}_{ext}(\mathbf{r})+\delta\mathbf{H}(\mathbf{r}), \quad (10)$$

which is the CDFT form of Eq. (3). The single Eq. (8) thus embodies both spin compensation and the absence of orbital pair breaking. This means that the magnetic ions, which dominate $\mathbf{A}_{xc}$, *simultaneously protect the superconductor from both the diamagnetic and the paramagnetic pair breaking action of the external fields*. Orbital pair breaking is then less critical for the JPE than it appears on the basis of the original theory.[1,2]

These conclusions have a number of immediate consequences for experimental and theoretical work on the JPE: (i) The details of the system do not enter the arguments leading to Eqs. (8)–(10), which are rather general and not tied to particular features of the system's electronic structure. This generality may explain why the JPE could be observed in the physically very different systems listed above. (ii) The mechanism for spin compensation is located at the magnetic ions, whose exchange-correlation magnetic field cancels the external one. Relation (9) implies that the origin for compensation of the induced currents must be located at the same place. The search for a physical explanation of current compensation can thus concentrate on the magnetic ions and, in particular, their correlations. (iii) Within CDFT $\mathbf{A}_{xc}$ is not a pure exchange field, but encompasses *all* current-related interaction effects not already contained in $\mathbf{A}_d$. The search for mechanisms by means of which current compensation can be accomplished is thus not limited to the conventional exchange interaction. This observation enables us to consider a wider range of mechanisms (examples are discussed below) and partially explains the negative result of an earlier similar theory (see below).

To further explore this CDFT interpretation of the JPE we provide, in the remainder of this paper, first a simple illustration of the cancellation embodied in Eq. (8), then discuss an earlier, closely related, theory, and finally compare the three consequences listed above with experiments and earlier theory on the JPE in λ-$(BETS)_2FeCl_4$ and $Sn_{1-x}Eu_xMo_6S_8$.

*Illustration of Eq. (8)*: *a London superconductor*. To illustrate in a simple case how Eq. (8) implies absence of orbital pair breaking recall the phenomenological London equation, according to which current and vector potential in a homogeneous superconductor are related by $\mathbf{j} = -qn_s\mathbf{A}_{ext}/(mc)$, where $n_s$ is interpreted as the number density of superconducting electrons. Since the full physical current is $\mathbf{j} = \mathbf{j}_p - qn\mathbf{A}_{ext}/(mc)$, where the first part is the paramagnetic current, entering the functionals of CDFT, and the second the diamagnetic current, one obtains for $\mathbf{j}_p$

$$\mathbf{j}_p = \frac{q(n-n_s)}{mc}\mathbf{A}_{ext}. \qquad (11)$$

From Eqs. (6) and (8) we obtain $\mathbf{A}_s = 0 + O(\delta)$, where the terms of order $\delta$ are by assumption not strong enough to destroy superconductivity and will be neglected below. The CDFT Kohn-Sham equations[18–20] for $\mathbf{A}_s = 0$ necessarily yield $\mathbf{j}_p = 0$. When substituted into Eq. (11) (with $\mathbf{A}_{ext} \neq 0$, since a nonzero external magnetic field is applied), $\mathbf{j}_p = 0$ implies $n = n_s$, which shows that the magnetic field has not broken any Cooper pair via its coupling to the orbital currents. We thus see explicitly that Eq. (8) implies absence of orbital pair breaking.

*Earlier theory*. A cancellation of orbital effects that is similar (but not identical) to Eq. (8) has been considered already in one of the earliest papers in the field,[25] and rejected as impossible. These investigations were performed within a simplified (pre-CDFT) single-particle treatment of the orbital currents. The authors of Ref. 25 assume that the exchange field $\mathbf{A}_x$, contained in $\mathbf{A}_{xc}$, must cancel both $\mathbf{A}_d$ and $\mathbf{A}_{ext}$, and assert that this is impossible since the exchange interaction is short ranged, while the dipolar interaction is long ranged. Hence, according to Ref. 25, no orbital cancellation can take place. From the present point of view this argument is not conclusive, because the correlation part $\mathbf{A}_c$ of $\mathbf{A}_{xc}$ may well be long ranged since it need not arise from the exchange interaction of Ref. 25. Moreover, all dipolar interactions are suppressed by the relativistic prefactor $(v/c)^2$, which makes them much smaller than typical exchange effects, so that they can hardly play a decisive role for effects dominated by $\mathbf{A}_{xc}$ and $\mathbf{H}_{xc}$. Indeed, in view of the experiments listed above the assumption that no cancellation of orbital effects takes place seems untenable.

*Experiments on the JPE in* λ-$(BETS)_2FeCl_4$. In connection with very recent experimental work[3,4] on the JPE in λ-$(BETS)_2FeCl_4$ a simple and explicit physical mechanism was proposed[4] by means of which the magnetic ions ($Fe^{3+}$) in between the BETS planes in λ-$(BETS)_2FeCl_4$ can simultaneously cancel the effects of the external in-plane magnetic field on the spin *and* suppress the induced orbital currents. Briefly, Hund's rule correlations imply that the polarized $Fe^{3+}$ ions have all spin up states of the Fe *d* shell occupied, so that these states are not available as intermediate states for transport of a singlet Cooper pair from one BETS layer to the next. Currents perpendicular to the layers are thus suppressed.[4] Interestingly, this proposal locates the mechanism which suppresses orbital pair breaking exactly where it is to be expected on the basis of the above arguments, namely at the magnetic ions [cf. conclusion (ii) above]. It also shows clearly how the magnetic ions can suppress orbital currents by interactions different from simple exchange [cf. conclusion (iii), above].

*Experiments on the JPE in* $Sn_{1-x}Eu_xMo_6S_8$. A different mechanism for protecting the JPE from orbital pair breaking has been proposed in Ref. 11 for the pseudoternary chalcogenides in which the JPE was first seen experimentally. Here the magnetic (Eu) ions act (apart from producing spin compensation) as scattering centers, reducing the mean free path and thereby increasing $H_{c2}$.[11] This allows the JPE to take place at fields above the value of $H_{c2}$ expected in the absence of magnetic ions.[5–7,11] Again, we see that the magnetic atoms suppress both, paramagnetic (Pauli) and diamagnetic (Meissner) pair breaking, and again the orbital cancellation is not simply due to their exchange interaction with the conduction electrons.

Two of the three physical mechanisms proposed in the literature to explain the absence of orbital pair breaking in experiments on the JPE are thus consistent with the general ideas developed here. To these two one can add the following alternative scenario for how breakdown of current compensation can imply breakdown of spin compensation: As soon as the external magnetic field is strong enough to produce spin-polarized currents in the sample, these currents will exert a torque[27] on the magnetic ions, which will affect their polarization. In extreme cases such torques can lead to a complete magnetization reversal,[28,29] but even in less extreme situations there is thus a negative feedback between the incipient currents and the magnetic ions needed to maintain spin compensation. Although it is not known at present whether this scenario is realized in nature, it provides a vivid illustration of the interplay between orbital effects and spin effects, and of how this interplay can affect the JPE.

*Outlook*. The main consequence of the above analysis is that the search for a mechanism for suppressing orbital currents in JPE-type[30] magnetic-field-induced superconductivity is simultaneously narrower and wider than is commonly thought. Narrower, because one only needs to consider the current-related effects of the magnetic ions or impurities; wider because these effects are not limited to simple exchange, but encompass a spectrum of other possibilities. On a more speculative note, it is worthwhile to point out that the JPE may well not be the only situation in nature in which similar cancellations take place, but might constitute a paradigm for other phenomena based on a complete or partial cancellation between $\mathbf{A}_{ext}$ and $\mathbf{A}_{xc}$. Only further research can show, for example, whether the transformation of electrons subject to huge magnetic fields into composite quasiparticles that do not feel any or only a much weaker effective field, observed in the fractional quantum Hall effect,[31] can be understood along similar lines within CDFT. Similarly, it may be interesting to explore the question whether the magnetic correlations which protect the superconducting JPE state from the influence of the otherwise pair-breaking external fields constitute another example for a *quantum protectorate* in the sense of Ref. 32.

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