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Enhanced paramagnetic limit of the upper critical magnetic field for superconductors with charge-density waves

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Abstract

The paramagnetic limit H_p of the upper critical magnetic field H_{c2} in superconductors with charge-density waves has been derived in the framework of the self-consistent approach. The obtained quantity H_p always exceeds the Clogston–Chandrasekhar value H_p^{BCS} . Relevant experimental data for inorganic and organic superconductors with $H_{c2} > H_p^{BCS}$ are analysed and are shown to be in qualitative agreement with the proposed theory.

1. Introduction

The paramagnetic destruction of spin-singlet superconductivity was discovered long ago theoretically by Clogston [1] and Chandrasekhar [2]. In the framework of the original Bardeen–Cooper–Schrieffer (BCS) theory of superconductivity [3] they obtained a limit

$$H_{\rm p}^{\rm BCS} = \Delta_{\rm BCS}(T=0)/\mu_{\rm B}^* \sqrt{2} \tag{1}$$

from above for the upper critical magnetic field H_{c2} at zero temperature, T. Here $\Delta_{BCS}(T)$ is the superconducting energy gap and μ_B^* is the effective Bohr magneton. There are two reasons [4] why the renormalized quantity μ_B^* does not coincide with its bare value $\mu_B = \frac{e\hbar}{2mc}$, where e is the elementary charge, \hbar is Planck's constant, m is the electron mass and c is the velocity of light. First, the effective band mass m^* of the quasiparticle differs from m. Second, exchange–correlation Fermi-liquid effects enhance the paramagnetic (Pauli) susceptibility relative to its electron gas value. Paramagnetic effects may become significant for superconductors, in particular, in the case when an external magnetic field H is parallel to a superconducting film, whose thickness d is much smaller than the magnetic field penetration depth λ_L . Then the

magnetic field profile inside the film is almost uniform and the diamagnetic (Meissner) response is considerably suppressed [5, 6].

This conclusion may be violated for a large concentration of strong spin-orbit scattering sites, when the spins of the electrons constituting the Cooper pairs are flipped [6–9]. Then the actual $H_{c2}(T = 0)$ starts to exceed [10] the classical Clogston-Chandrasekhar bound (1). Such an enhancement of H_{c2} has been observed in Al films coated by monolayers of Pt [11]. The Pt atoms served there as strong spin-orbit scatterers due to their large nuclear charge Z. On the other hand, a similar contamination of another superconductor, the A15 compound V₃Ga, exhibiting the Pauli paramagnetic effect in the absence of impurities [12], altered neither $H_{c2}(0)$ nor the Zeeman splitting of the tunnel conductance [13]. Therefore, the spin-orbit mechanism of overcoming the Clogston-Chandrasekhar limit remains open to investigation.

At the same time, this limit may be exceeded due to a quite different effect, namely, the presence of a spin-singlet dielectric gap $|\Sigma|$ on the nested, d, sections of the Fermi surface (FS) [14–16]. The corresponding order parameter $\Sigma = |\Sigma|e^{i\varphi}$ describes chargedensity waves (CDWs), which reduce the energy of the reconstructed insulating or metallic phase below the critical temperature of the structural transition T_d . Here φ is the phase of the CDW, usually pinned by defects or the background crystal lattice in sub-threshold electrostatic fields [17, 18]. The expected increase of the calculated limiting paramagnetic field H_p for CDW superconductors, as compared to H_p^{BCS} , is intimately associated with paramagnetic properties of the normal CDW phase, which are very similar to those for BCS swave superconductors [19–22]. Nevertheless, microscopic backgrounds of these two formally similar many-body phenomena are appreciably different [23, 24].

It is important to underline that the assumed superconductivity is possible below a certain critical temperature, $T_c < T_d$, only if the CDW gapping of the electron spectrum is partial [25–27], i.e. some non-nested, nd, FS sections remain untouched by CDWs in the range $T_c < T < T_d$ and the distorted phase stays metallic. In incommensurate Peierls insulators, gapped by electron–phonon forces, the pinned phase φ may be arbitrary [17, 18], whereas for commensurate excitonic insulators, gapped by Coulomb interaction, it may be either 0 or π [23, 28].

The exceeding of the paramagnetic limit has been found to exist [16] for all possible values of the parameters inherent to the Bilbro–McMillan model [25]. That result, as is demonstrated below, remains correct in a more accurate approach. Nevertheless, our previous considerations [14–16] had a significant limitation. Specifically, the treatment of the superconducting phase with CDWs was not self-consistent, which made quite unexpectedly the whole problem *more* rather than less involved. In our current calculations we use the results of the self-consistent calculations of the thermodynamic properties [29] applied to a metal with two order parameters: a dielectric one $\Sigma(T)$, existing on the nested FS sections, and a superconducting one $\Delta(T)$, unique for both d and nd sections [25]. The ratio H_p/H_p^{BCS} , contrary to its counterpart in the non-self-consistent approach [16], turns out to be described by a simple analytical formula. We obtained a phase diagram in the parameter space for T = 0 and carried out its analysis in terms of the observed variables. Relevant experimental data were discussed.

2. Theory and results

In order to calculate the paramagnetic limit one should consider free energies F per unit volume for all possible ground state phases in an external magnetic field H. The parent non-reconstructed phase (actually existing only above T_d !), with both superconducting and electron–hole pairings switched off and in the absence of H, serves as a reference point.

At $T < T_d$, we deal with relatively small differences δF reckoned from this hypothetical 'doubly-normal' state [30].

Since we assume the Meissner diamagnetic response to be negligibly small as for the film geometry (see introduction), the external magnetic field *H* coincides with that inside the specimen and is almost uniform. Therefore, the additional energy of the paramagnetic phase in the magnetic field, when both Δ and Σ are equal to zero, takes the form [31]

$$\delta F_{\rm p} = -N(0)(\mu_{\rm B}^* H)^2.$$
⁽²⁾

Here N(0) is the total electron density of states per spin at the Fermi level.

The reconstructed superconducting state with the FS gapped both by superconductivity and CDWs constitutes another thermodynamic phase at $T < T_c$. Its free energy can be obtained from the following simple argument. In the adopted Bilbro–McMillan model [25] the order parameters $\Delta(T)$ and $\Sigma(T)$ satisfy the self-consistent equation system [29]. This system has a solution, which determines two different *T*-dependent gaps on nd and d sections of the FS. Specifically, there is the superconducting energy gap $\Delta(T) = \Delta_{BCS}(\Delta_0, T)$ below T_c on the nd sections, whereas the d sections are influenced by the effective gap $D(T) = \Delta_{BCS}(D_0, T)$. Here $\Delta_{BCS}(G, T)$ is the already mentioned Mühlschlegel gap function of the BCS theory with $G = \Delta_{BCS}(T = 0)$, so Δ_0 and D_0 are the values of the relevant gaps at T = 0. The effective gap D(T) is a combination of both order parameters

$$D(T) = \sqrt{\Delta^2(T) + \Sigma^2(T)}.$$
(3)

The value D_0 is equal to the parameter $\Sigma_* = \pi T_d/\gamma$, the bare CDW gap at T = 0 in the absence of superconductivity, and $\gamma = 1.7810...$ is the Euler constant.

The assumed equality of the superconducting gaps Δ_{nd} and Δ_d on the nd and dFS sections, respectively, is a consequence of the strong mixing of the electron spectrum branches by the matrix elements of the effective four-fermion interaction Hamiltonian, the interaction being a generalization of the virtual-boson-induced BCS-like contact attraction for an anisotropic system. In the framework of the Bilbro–McMillan model [25] adopted by us there is a possibility to consider two separate equations for Δ_{nd} and Δ_d [15]. Thus, one is forced to solve self-consistently a system of three equations for $\Sigma(T)$, $\Delta_{nd}(T)$ and $\Delta_d(T)$. Such a system is much more involved from the technical point of view. But specific physical consequences like those treated here and in [29] will not be qualitatively altered.

The justification of the two gap (Δ_{nd} and Δ_d) occurrence is very interesting in the context of the more general problem dealing with the coexistence in the same spatial region of two different superconducting gaps originating from two different pieces of the FS [32, 33]. This topic became a burning one because the hypothesis of two-gap superconductivity was claimed to happen in MgB₂ [34, 35], though there are sound objections to such a viewpoint [36–39].

The deepest fundamental controversy in the magnesium diboride superconductivity is an observed two-gap survival in heavily Al-doped [40, 41], carbon-doped [42] and neutron-irradiated [43] samples, which contradicts the Anderson theorem [44]. The same reasoning can be also applied to partially dielectrized CDW superconductors. Indeed, even if Δ_{nd} and Δ_d differ in pure specimens, they should merge in dirty ones. As far as we know, all relevant substances under discussion in the section 3 are in the dirty limit. This strengthens arguments for the strong-mixing approximation and unique superconducting order parameter Δ .

Hence, on both parts of the FS, the BCS-like (but different!) gap functions are developed. The change of the free energy δF_s at T = 0 is determined by their zero-T values in the conventional manner [30]:

$$\delta F_{\rm s} = -N_{\rm nd}(0) \frac{\Delta_0^2}{2} - N_{\rm d}(0) \frac{\Sigma_*^2}{2}.$$
(4)

The quantities $N_{nd}(0) \equiv (1 - \mu)N(0)$ and $N_d(0) \equiv \mu N(0)$ are the partial densities of states on the nd and d FS sections, respectively. The control parameter μ of the dielectrized electron spectrum varies in the range $0 \le \mu \le 1$. For $\mu = 1$ the FS is completely gapped by CDWs and the ground state is insulating.

Finally, a paramagnetic superconducting CDW phase should be considered. In this phase, characterized by two order parameters $\Delta(T)$ and $\Sigma(T)$, the applied magnetic field induces Zeeman splitting of the quasiparticle spectrum. Both order parameters depend on magnetic field in a strange way, growing with H. Such a phase is a generalization of the metastable one, found theoretically by Sarma for BCS superconductors [45] (see also [30, 46]). A free energy of the paramagnetic superconducting CDW phase is higher than that given by equation (4) for all values of H up to the limiting value, when superconductivity ceases to exist, i.e. H_p [16], so that it cannot be realized in the system. Of course, the same is true for the Sarma phase in BCS superconductors.

It is worth noting that any orbital magnetic field effects favourable for the CDW state are not accounted for, because the values of H relevant to the problem concerned are considerably smaller than those, which reduce the dimensionality of the electron spectrum [47–52]. We also do not take into account the possibility of the Larkin–Ovchinnikov–Fulde–Ferrel (LOFF) non-homogeneous superconducting state [30, 53–57], although there are some hints that it might have been observed in low-dimensional organic compounds [58–61] and heavy-fermion superconductor CeCoIn₅ [62]. It is well to bear in mind that the existence of a CDW with the vector **Q** may promote the appearance of the LOFF structure [63, 64]. It may be the case in ferromagnetic superconductor ZrZn₂ [65]. On the other hand, in the non-superconducting CDW state the vector **Q** may be changed in large magnetic fields [19, 50, 66] in much the same manner as a new spatial modulation emerges in the LOFF phase for superconductors.

Thus, with the assumption of the order parameter homogeneity the procedure of the paramagnetic limit determination is formally the same as used by Clogston [1] and Chandrasekhar [2]. Namely, one should equate δF_p and δF_s . It leads to a basic relationship for the actual paramagnetic limit H_p :

$$\left(\mu_{\rm B}^* H_{\rm p}\right)^2 = \frac{1}{2} \left[(1-\mu) \,\Delta_0^2 + \mu \,\Sigma_*^2 \right] = \frac{1}{2} \left[\Delta_0^2 + \mu \left(\Sigma_*^2 - \Delta_0^2 \right) \right]. \tag{5}$$

Since $\Sigma_* = D_0 > \Delta_0$, which is a consequence of equation (3), the limiting magnetic field H_p in a CDW superconductor *always exceeds* the Clogston–Chandrasekhar value H_p^{BCS} .

It is convenient now to introduce a primordial superconducting gap Δ_* at $\tilde{T} = 0$ in the absence of CDWs. The observable superconducting order parameter Δ_0 can be expressed in terms of the bare input parameters in the following way [29]

$$\Delta_0 = \Sigma_* \left(\frac{\Delta_*}{\Sigma_*}\right)^{\frac{1}{1-\mu}}.$$
(6)

Then the increase of the paramagnetic limit over the Clogston–Chandrasekhar value is given by the formula

$$\left(\frac{H_{\rm p}}{H_{\rm p}^{\rm BCS}}\right)^2 = 1 + \mu \left[\left(\frac{\Sigma_*}{\Delta_*}\right)^{\frac{2}{1-\mu}} - 1 \right]. \tag{7}$$

The level lines of H_p/H_p^{BCS} on the phase plane $(\Delta_*/\Sigma_*, \mu)$ are shown in figure 1. It is clear from the plots that the smaller the ratio between the superconducting and CDW coupling constants the larger the excess of the paramagnetic limit.

The dimensionless parameters Δ_* / Σ_* and μ are independent of one another. The latter can be determined, in principle, by resistive, specific heat, or optical experiments [27]. On the other hand, the bare gaps Δ_* and Σ_* are hardly measurable, because to get rid of either



Figure 1. Contour plot of the ratio H_p/H_p^{BCS} on the plane $(\Delta_*/\Sigma_*, \mu)$. Here H_p is the paramagnetic limit for superconductors with charge-density-waves (CDWs) and H_p^{BCS} is that for BCS spin-singlet superconductors, Δ_* and Σ_* are bare values of the order parameters in parent phases with Cooper or CDW pairings, respectively, and μ is the portion of the nested Fermi surface sections, where the CDW gap develops.

superconductivity or CDWs it is necessary to apply pressure, external magnetic field, or alloying. Therefore, various background electronic and crystal lattice properties would be inevitably altered, including gaps (some insight can be obtained from [24, 67, 68]). Moreover, for experimentalists to analyse the situation, it would be of benefit instead to deal with the observable properties. The corresponding formula can be directly obtained from equations (1) and (5). The quantity $D_0 = \Sigma_*$, as has been indicated above, is linked to the structural (excitonic) transition temperature T_d by the BCS relationship. The same is true for the pair Δ_0 and T_c [29]. Hence, it comes about that

$$\left(\frac{H_{\rm p}}{H_{\rm p}^{\rm BCS}}\right)^2 = 1 + \mu \left[\left(\frac{D_0}{\Delta_0}\right)^2 - 1\right] = 1 + \mu \left[\left(\frac{T_{\rm d}}{T_{\rm c}}\right)^2 - 1\right].$$
(8)

All quantities in equation (8) can be easily measured or inferred from the experimental data. The corresponding contour curves are displayed in figure 2. One can readily see that for typical $T_c/T_d \approx 0.05-0.2$ (some A15 compounds are rare exceptions [27]) and moderate values of $\mu \approx 0.3-0.5$, the augmentation of the paramagnetic limit becomes very large. Of course, this outcome may be essentially reduced by the spin–orbit scattering [11].

At first glance, there is a difficulty in estimating the input parameter μ in equation (8). This quantity is correlated to the independently measured critical temperatures T_d and T_c by a relationship

$$T_{\rm d}^{\mu}T_{\rm c}^{1-\mu} = T_*,\tag{9}$$

which stems from equation (6) and involves also a 'bare' superconducting critical temperature in the absence of the CDW distortion, $T_* = \frac{\gamma}{\pi} \Delta_*$. An actual experimental determination of the latter may be impossible if CDWs are tolerant of varying external conditions. Nevertheless, a *series* of measurements may easily overcome this uncertainty. Such a procedure was carried out and the values of the parameter μ were found, e.g., in [69], where resistance and thermoelectric power of Chevrel-phase samples of Eu_xMo₆S₈ were measured at different external pressures up to 20 kbar.



Figure 2. The same as in figure 1 on the plane $(T_c/T_d, \mu)$. Here T_c and T_d are the observed critical temperatures of the superconducting and CDW transitions, respectively.

Physically, the rise of H_p in CDW superconductors is quite natural. Both Cooper and electron-hole pairings are simultaneously depressed by the paramagnetic effect, whereas we consider the detrimental influence of the external field H on the superconducting gap only. Therefore, larger fields H are required to do the same job as in the absence of CDW-induced gap.

It is of interest that recently the enhancement of the paramagnetic limit has been also found theoretically for the model related to the CDW one and taking into account the Van Hove singularity of the two-dimensional electron density of states [70].

3. Discussion

To verify our theory it would be desirable to observe the coexistence between CDWs and superconductivity in the same samples where the Clogston–Chandrasekhar paramagnetic limit is exceeded. Unfortunately, such a direct verification is still lacking.

In principle, photoemission experiments might confirm simultaneous superconducting and CDW gapping of FSs and give FS momentum-space maps in the high- (ungapped) and low-T (gapped) states [71-73]. In particular, such measurements might verify or disprove the strong-mixing concept discussed in the previous section. There are, however, methodological difficulties, which can hamper the unambiguous identification of the magnitudes as well as directional and temperature dependences of Δ and Σ (see, e.g., the analysis in [74] as applied to $Bi_2Sr_2CaCu_2O_{8+\nu}$). Another important point is a three-dimensionality of the FS in cuprates [75]. If such warnings are ignored, the situation with gapping in photoemission spectra for $Bi_2Sr_2CaCu_2O_{8+v}$ (a high- T_c oxide, the most suspicious from the CDW point of view) looks as follows [72, 73]. The superconducting gap Δ has a d-wave momentum dependence in the $k_x - k_y$ plane with definite nodes. The same features are appropriate to pseudogaps, earlier identified by us as CDW ones [26, 27]. Therefore, a clear-cut division of the cuprates' FS into two parts, one non-nested and gapped by Δ and the other nested and gapped both by Σ and Δ (see equation (3)), is not confirmed so far. On the other hand, tunnel measurements demonstrate that for different high- T_c oxides superconducting gaps and pseudogaps have different T- and *H*-dependences [76–81], so that the unique symmetry properties for quite distinct objects look quite strange. It seems that one should still wait for more conclusive photoemission

experiments and also for the definite solution to the d-wave versus s-wave controversy (see recent discussions in [82–87]).

We would like to point out that the convincing evidence for the decisive destructive role of CDWs in superconducting La-based cuprates was obtained in neutron diffraction experiments [88, 89]. The CDWs manifest themselves there in addition to the essential antiferromagnetic correlations. See the relevant discussion in our review [27].

FSs and their gapping in layered dichalcogenides have been studied extensively by photoemission methods as well as by tunnelling. In particular, the tunnel measurements [90] for 2H-polytype compounds showed a conspicuous anticorrelation between Σ (or T_d) and $T_{\rm c}$. For 2H-NbSe₂, the CDW gap $\Sigma \approx 34$ meV is the smallest non-zero one, whereas $T_{\rm c}$ is 7.2 K. Nevertheless, Σ escaped detection by photoemission, although a much smaller superconducting gap was disclosed [91, 92]! The authors of [92] believe that this result is due to the fact that the nested FS portion (μ in our terms) is tiny. This explanation does not seem satisfactory, since all FS sheets and all directions in the \mathbf{k} -space were investigated. At the same time, a superconducting gapping was found for the Γ -centred [91] and K-centred [92] FS cylinders. Notwithstanding substantially different electron-phonon coupling strengths at various points of FS cylinders surrounding the K-point, $\Delta \approx 1$ meV is uniform there. This behaviour counts in favour of the strong-mixing paradigm. In the related compound 2H-TaSe₂, for which superconductivity is very weak ($T_c \approx 0.15$ K [93]) and Δ has still not been observed, a CDW gap was found around the K-point [94]. All the aforesaid means that the microscopic relationships between two types of gapping in layered dichalcogenides are far from being resolved.

Let us turn back to the paramagnetic properties of CDW superconductors. It seems quite plausible that the phenomenon predicted in this paper has already been observed in the C15 compound $Hf_{1-x}Zr_xV_2$, where $H_{c2}(T) = 230$ or 208 kG for x = 0.5 and 0.6, respectively, and $H_p^{BCS} \leq 190$ kG if the simplest possible estimation is made [95]. On the other hand, in these solid solutions the CDW gapping was directly found by resistive measurements [96].

More recently necessary correlations between the increase of the paramagnetic limit and the CDW appearance have been revealed for organic superconductors. For example $H_{c2}(0)$ in the layered κ -(BEDT-TTF)₂Cu(NCS)₂ with $T_c \approx 10.4$ K and the FS prone to nesting [97, 98], overcomes the corresponding H_p^{BCS} [99]. At the same time, the *T*-dependence of the resistance for this compound demonstrates a high and wide peak in the range 85–100 K interrupting the metallic trend appropriate both to low and room temperatures. Most probably, this behaviour reflects the partial CDW gapping [100]. The competition between the CDW insulating state and superconductivity triggered by an external pressure *P* in the related compound (BEDT-TTF)₃Cl₂·2H₂O, can be considered as additional indirect evidence for the possible CDW presence in the superconducting state of κ -(BEDT-TTF)₂Cu(NCS)₂ [97].

 κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl is another charge-transfer salt with the κ -packing arrangement, where $H_{c2}(0)$ conspicuously exceeds H_p^{BCS} [101]. It is remarkable that this substance is an insulator at ambient pressure, but becomes metallic and superconducting for P > 0.3 kbar. In view of such a proximity between dielectric and superconducting phases, it seems quite possible that κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl retains nesting properties of its FS for higher *P*. The observed positive curvature of the $H_{c2}(T)$ in the neighbourhood of T_c [101], a feature appropriate to superconductors with density waves [102], agrees with the assumption made. At the same time, at larger P = 6 kbar the critical temperature T_c reaches a rather high value of 12.8 K [97]. In the framework of our model [25, 27] it corresponds to the FS distortion with $\mu \rightarrow 0$. The authors of [101] point out that spin–orbit scattering cannot lead to the exceeding of $H_{c2}(0)$ over H_p^{BCS} in the case discussed, since the Shubnikov–de Haas quantum oscillations in this compound are distinctly seen under pressure [97, 98]. In the layered superconductor α -(BEDT-TTF)₂NH₄Hg(SCN)₄ the value $H_{c2}(0)$ is comparable to H_p^{BCS} [103]. This salt with $T_c \approx 1$ K is the only superconductor from the family α -(BEDT-TTF)₂MHg(SCN)₄, while other sister compounds demonstrate the ground state of the density-wave type and $T_d \approx 8$ K for M = K, Tl or 10 K for M = Rb [97]. A comparison of critical temperatures shows that density-wave correlations are stronger than superconducting ones, which imply large Σ_*/Δ_* and hence favours the increase of the ratio H_p/H_p^{BCS} . It should be noted that the CDW nature of the low-T insulating state in nonsuperconducting salts stems from the observed paramagnetic effects [22, 51, 52, 104–106] not appropriate to the SDW phase [19].

Application of the external pressure *P* to the initially insulating compound α -(BEDT-TTF)₂KHg(SCN)₄ leads to a complete suppression of CDWs for $P > P_0 \approx 2.5$ kbar and an appearance of superconductivity with $T_c \approx 0.1$ K [107]. This agrees well with our concept and one should expect the exceeding of $H_{c2}(0)$ over H_p^{BCS} under pressure, similar to what has been revealed in κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl [101]. On the other hand, at pressures below P_0 the superconductivity [107], which is not covered by our theory [27]. However, this behaviour may also stem from experimental artefacts, such as non-attained thermal equilibrium or internal strains. In any case, magnetic studies of α -(BEDT-TTF)₂KHg(SCN)₄ would be very important to elucidate the nature of CDW, superconducting and superconducting + CDW phases.

A new oxide KOs₂O₆ with a defect pyrochlore structure and $T_c = 9.6$ K is the most recently synthesized superconductor with $H_{c2} > H_p^{BCS}$ [108]. Since many oxides exhibit structural metal–insulator transitions with low-*T* phases of the CDW nature [27, 109–111], it would be of interest to check whether in this compound CDWs really coexist with superconductivity.

To summarize, we obtained a formula describing the increase of the paramagnetic limit for $H_{c2}(0)$ in CDW superconductors over the Clogston–Chandrasekhar value of the BCS theory. The similarity of the paramagnetic properties for s-wave superconductors and CDW partially gapped metals and the interplay of two coexisting order parameters are responsible for the effect. There are strong experimental grounds to associate the observed experimental data with the proposed approach.

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