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Pressure-induced coexistence of superconductivity and magnetism in organic conductors κ-(BEDT–TTF)$_2$X

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Abstract

We study within a mean field theory the interplay between superconductivity and antiferromagnetism in κ-(ET)$_2$X compounds. The hydrostatic or chemical pressure induces first-order phase transitions between superconducting, antiferromagnetic and mixed states in which superconductivity and antiferromagnetism coexist. In this work, we compare the pressure effects in experiments and nesting quality according to our calculations. We describe the κ-phase of the organic conductors by a two-band model exhibiting nesting properties governed by the ratio $\frac{t_1}{t_2}$. When we apply pressure in these compounds, we modify the Fermi surface shape and consequently the strength of the antiferromagnetic fluctuations depending on the nesting properties. Our theoretical description seems to explain various experimental data, for which, to our best knowledge, no clear theoretical interpretation has been given so far.
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1. Introduction

Depending on the species of the anion X and pressure, the layered organic superconductors κ-(BEDT–TTF)$_2$X (they are called κ-(ET)$_2$X hereafter) can exhibit ferromagnetic, antiferromagnetic, insulating, or superconducting ground state. Indeed, the ground state of κ-(ET)$_2$Cu [N(CN)$_2$] Cl material is an insulating AF phase [1,2], while the ground state of κ-(ET)$_2$Cu [N(CN)$_2$] Br and κ-(ET)$_2$Cu(NCS)$_2$ is a SC phase. In the former with X = Cu [N(CN)$_2$] Cl, the applied pressure suppresses antiferromagnetic order and stabilizes a novel state in which superconductivity and antiferromagnetism coexist for a pressure range 200 bar $\leq P \leq$ 400 bar [3]. For high pressures only unconventional superconductivity survives [3]. More recently Kimura et al. [4] have observed the pressure-induced phase transition of deuterated κ-(ET)$_2$Cu [N(CN)$_2$] Br (that changes its ground state from superconductivity to antiferromagnetism by deuteration) from the antiferromagnetic to superconducting state through an inhomogeneous phase coexistence by using optical reflection spectroscopy. The coexistence of the antiferromagnetism and superconductivity phases was observed in the pressure range below 5 MPa. However by deuterization of κ-(ET)$_2$Cu [N(CN)$_2$] Br, the ground state is gradually pushed from the superconducting state toward the antiferromagnetic state [4,5].

In the case of X = Cu [N(CN)$_2$] Br, the superconducting ground state is in the vicinity of the phase boundary because it changes to an antiferromagnetic state after deuteration of the two ethylene end groups [6]. The perfectly deuterated one is believed to be just on the boundary (the so-called Mott boundary) between the antiferromagnetic and superconducting states. When the sample is rapidly cooled, the introduction of disorder to the system results in an antiferromagnetic ground state. After applying pressure, the ground state changes from antiferromagnetic to superconducting through a re-entrant superconductivity phase that has been detected by measuring the electrical resistivity [7]. This re-entrant phase is believed to be an inhomogeneous phase consisting of antiferromagnetic and superconducting phases similar to X = Cu [N(CN)$_2$] Cl, although no direct evidence regarding this inhomogeneous

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phase has been reported. In their recent paper, the Kanoda group reports the direct observation of this phase coexistence using infrared reflection spectroscopy under external pressure [4].

The ground state of the quasi-two-dimensional material organic conductor κ-$(ET)_2X$ (with X $=$ Cu $[N(CN)_2]$ Cl, Cu $[N(CN)_2]$ Br, $Cu(NCS)_2$ and so on), is determined by the universal parameter $\frac{U}{W}$, where $U = g^0_{\mathrm{AF}}$ (the antiferromagnetic fluctuation energy which couples electrons occupying the same band according to our model) and W are the on-site Coulomb energy and bandwidth, respectively. These types of materials are classified as Mott insulator systems. In the case X $=$ Cu $[N(CN)_2]$ Br, the superconducting state directly changes to the antiferromagnetic insulating state below about 11 K with increasing $\frac{U}{W}$ at around $\frac{U}{W} = 1$ [6,8,9]. The $\frac{U}{W}$ parameter can be experimentally controlled by the deuteration and cooling rate [10].

2. Basic considerations and the model

Unconventional mechanisms of superconductivity are one of the most challenging issues in solid state physics. Not only that there is a possibility of accomplishing high-T_c as in the cuprates, but also the variety of pairing symmetries provides rich physics. Recently, it has become increasingly clear that organic materials can provide various stages for unconventional pairings. Moreover, in these layered organics, Shubnikov–de Haas oscillation experiments have established the existence of a well-defined Fermi surface, demonstrating the Fermi liquid character of the low energy excitation. The large enhancement of the effective mass revealed by the specific heat as well as magnetic susceptibility measurements suggests the strong electron correlation effect in the normal state. In particular, κ-(BEDT–TTF)$_2$X is of special interest in that superconductivity occurs in proximity to the antiferromagnetic ordered state in the phase diagram [3,8]. Since some of these unusual properties suggest analogies with high-T_c cuprates [8,11], it was pointed out by many authors that the antiferromagnetic spin fluctuation should play an important role in the occurrence of superconductivity [12–17]. Theoretically, previous studies on the Hubbard model with the fluctuation exchange approximation [12–17] or quantum Monte Carlo calculation [16] have suggested that superconductivity in κ-(BEDT–TTF)$_2$X is similar to the d-wave superconductivity in the cuprates.

The κ-$(ET)_2X$ organic superconductors are among the most complex systems studied in condensed matter physics. As a good starting point for modeling, the strong correlation effects in these salts should be the Hubbard or the $t - J$ models at low doping concentrations. In fact, the Hubbard model is, perhaps, the simplest model that can describe strongly correlated physics and is therefore an important starting point for a complete and general description of strong correlations. In the present work, we assume that well-defined quasi-particles exist at low temperature and can be treated in a Landau–Fermi liquid approach. Theoretically, some analytical calculations have supported spin-fluctuation mediated pairing in the Hubbard model on lattices representing κ-(BEDT–TTF)$_2$X [12–17]. However, numerical evidences supporting such a possibility are yet to come. We consider a two-dimensional Hubbard model and we propose to treat the electron–electron interaction in a mean field approximation, with two different symmetry-breaking order parameters : the first is an effective attractive electron–electron interaction term between electrons of the band $(-)$, which breaks the gauge symmetry. We do not specify the microscopic origin of this term leading to superconductivity. The second, the origin of which is the exchange interaction, is an electron–hole coupling term between electrons of the band $(+)$, leading to a time-reversal symmetry-breaking SDW state. The mean field Hamiltonian H includes the non-interacting electron term H_0, the superconducting term H_{SC} and the magnetic term H_{AF}

$$H_0 = \sum_{k,s,i} \epsilon^i_{k,s} c^+_{i,k,s} c_{i,k,s} \tag{1}$$

$$H_{\mathrm{SC}} = -\sum_{k,p,q,s,i} \frac{g_{\mathrm{SC}}(k,p,q)}{N} c^+_{i,k+q,s} c_{i,k,s} c^+_{i,p-q,-s} c_{i,p,-s} \tag{2}$$

$$\begin{aligned} H_{\mathrm{AF}} = & -\sum_{k,p,q,s,i,j\neq i} \frac{g_{\mathrm{AF}}(k,p,q)}{N} c^+_{i,k+q,s} c_{i,k,s} c^+_{j,p-q,-s} c_{j,p,-s} \\ & -\sum_{k,p,q,s,i,j\neq i} \frac{g_{\mathrm{AF}}(k,p,q)}{N} c^+_{j,k+q,s} c_{i,k,s} c^+_{i,p-q,-s} c_{j,p,-s} \end{aligned} \tag{3}$$

where $\epsilon^i_{k,s}$ is the non-interacting electron dispersion relation, $c_{i,k,s}$ indicates the electron annihilation operator and i corresponds to the band $(\pm)$. The coefficients g_{SC} and g_{AF} are respectively the superconductivity and the magnetic coupling constants.

We study within a mean field theory with renormalized parameters the coexistence of superconductivity and antiferromagnetism in κ-(BEDT–TTF)$_2$X organic superconductors. These quasi-two-dimensional compounds can be described by a two-band model which depends on two parameters t_1 and t_2 [15]. We have considered that the superconducting state is induced by spin fluctuations with a d-wave gap symmetry. We have studied the dependence on the ratio $\frac{t_1}{t_2}$ of the superconducting and the magnetic energy gap in the mixed phase.

The potential relevance of the coexistence of superconducting and antiferromagnetic orders in organic systems has been justified by the proximity of the two orders in the P–T phase diagram [3,4] and has spurred theoretical suggestions of d-wave Cooper pairing mediated by antiferromagnetic fluctuations [14, 15]. Such an idea, which implies nodes in the superconducting order parameter, is strongly supported by NMR [18], tunnelling [19], thermal conductivity [20] and magnetic penetration depth experiments [21]. Theoretically, a simplest many-body Hamiltonian to incorporate the coexistence of superconductivity and antiferromagnetism is the Hubbard model. We consider a simplified dispersion relation of the conduction electron bands that reproduce band structure calculations and fit the Shubnikov–de Haas measurements [22]

$$\epsilon_{k(\pm)} = 2t_1(\cos k_z c - \cos k_{z_0} c) \pm 4t_2 \cos\left(\frac{k_x a}{2}\right) \cos\left(\frac{k_z c}{2}\right), \tag{4}$$

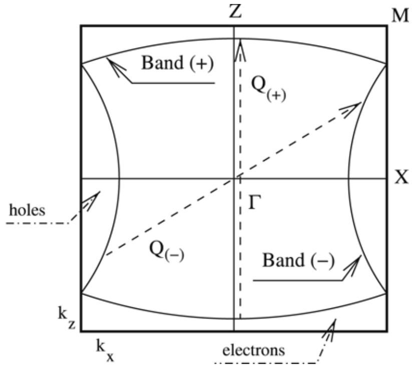

Fig. 1. Fermi surface of κ-$(ET)_2X$ with $Q_{(-)}$ which is the best nesting vector for the band (−) for an arbitrary value of the ratio $\frac{t_1}{t_2}$. For the band (+), the best nesting vector is $Q_{(+)}$.

where (k_x, k_z) is the conducting plane that can be different from one compound to another (it corresponds to the plane (k_x, k_y) for X = $Cu(NCS)_2$). t_1 and t_2 denote the inter-dimer transfer integrals in the **c** and **a** directions, respectively. k_{z0} changes with the anion X; for example $k_{z0} = 0.7\frac{\pi}{c}$ for X = $Cu[N(CN)_2]Br$ for which the best nesting quality corresponds to the nesting ratio $\frac{t_1}{t_2} = 0.4$ [15]. The (+) and (−) signs result in the quasi-one-dimensional and quasi-two-dimensional sections of the Fermi surface, respectively [14,15,22]. The Fermi surface displays nesting property particularly for the first band (+) [15]. Moreover, a change of the ratio $\frac{t_1}{t_2}$ induces a modification of the nesting property so that it affects the strength of spin fluctuations in the system. We show in Fig. 1 the Fermi surface topology of the κ-phase of organic compounds for an arbitrary nesting ratio $\frac{t_1}{t_2}$. The quasi-two-dimensional sections of the Fermi surface are labeled by the signs (±).

It is important to mention that our theoretical study of the P–T phase diagram is carried out strictly at two-dimension and it is evident that a small hopping within the third direction would not make any significant change in the pressure effects and would allow antiferromagnetic or superconducting phase at low temperature. The transfer integral in the third direction is much smaller than those within the planes and results in a slight warping of the Fermi surface perpendicular to this direction in the k-space. The validity of this picture of the Fermi surface as a three-dimensional object is discussed in detail by Singleton et al. [23]. The calculations that are presented in this work were undertaken in order to yield a phenomenological and accurate description of the pressure effect on the interplay between superconductivity and antiferromagnetism in the κ-phase of organic superconductors.

On the other hand, there is now a body of accumulating experimental evidence that organic superconductors have d-wave gap [18–21]. So, in our study of the pressure effects on the interplay between superconducting and antiferromagnetic orders in κ-$(ET)_2X$ compounds, we consider a superconducting order consistent with $d_{x^2-z^2}$-wave symmetry. As regards, it is required that the antiferromagnetic order parameter does not vanish on the nodes of the superconducting one. Therefore, the magnetic order considered here corresponds to the s-wave symmetry. The superconducting and antiferromagnetic coupling potentials which display the gap symmetries are approximated by

$$g_{SC}(k,k') = g^0_{SC} f_{SC}(k) f_{SC}(k'), \quad (5)$$

$$g_{AF}(k,k') = g^0_{AF} f_{AF}(k) f_{AF}(k'), \quad (6)$$

with

$$f_{SC}(k) = \cos(k_x a) - \cos(k_z c), \quad (7)$$

$$f_{AF}(k) = 1. \quad (8)$$

The coupled gap equations in the mixed phase are written as

$$\frac{1}{g^0_{SC(AF)}} = \sum_k f^2_{SC(AF)}(k) \frac{th\left(\frac{E(k)}{2k_B T}\right)}{2E(k)}, \quad (9)$$

where

$$E^2(k) = \left[\epsilon_{k(-)}\right]^2 + \left[\Delta^0_{SC} f_{SC}(k)\right]^2 + \left[\Delta^0_{AF} f_{AF}(k)\right]^2. \quad (10)$$

In order to study the pressure (α) effects on the superconducting and antiferromagnetic orders, we have carried out calculations of the free energy for all possible states, compared with that of the metallic phase F_0. The mean field expressions are given by

$$F_i - F_0 = \frac{(\Delta^0_i)^2}{g^0_i} - 2k_B T \sum_k Ln\left[\frac{ch\left(\frac{E_i(k)}{2k_B T}\right)}{ch\left(\frac{\epsilon_{k(-)}}{2k_B T}\right)}\right], \quad (11)$$

$$F_{AF/SC} - F_0 = \sum_{k;i}\left[\frac{(\Delta^0_i)^2}{g^0_i} - 2k_B T\, Ln \frac{ch\left(\frac{E(k)}{2k_B T}\right)}{ch\left(\frac{E_i(k)}{2k_B T}\right)}\right], \quad (12)$$

where

$$E_i(k) = \sqrt{\epsilon^2_{k(-)} + \left[\Delta^0_i f_i(k)\right]^2}, \quad (13)$$

and i = SC, AF.

3. Results and discussions

From our point of view, when we apply pressure in κ-$(ET)_2X$ systems, we modify the nesting ratio $\frac{t_1}{t_2}$, as was experimentally confirmed by Caulfield et al. [22], leading to the bandwidth $W = 4\alpha(t_1 + t_2)$. Indeed, the Fermi surface topology is modified and so the nesting properties. Consequently, comparing the effects of increasing the pressure in experiments, either by applying hydrostatic pressure or by anion substitution, and increasing α, $\frac{t_1}{t_2}$ or W according to our calculations, we find they are compatible if the pressure acts in the sense of increasing α and W.

Fig. 3 displays the dependence on pressure of the free energies of superconducting, magnetic and mixed states. We find that under pressure, there is a region of coexistence of the superconducting and the magnetic orders. This result is in agreement with the available experimental observations [2–4,24–26] showing an inhomogeneous region of coexistence of superconductivity and antiferromagnetism under pressure. In fact, an antiferromagnetic state was experimentally

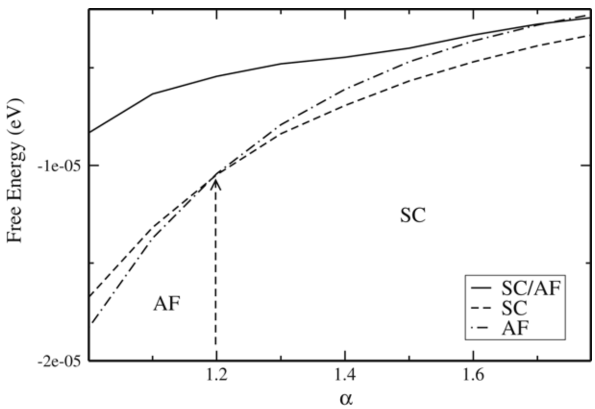

Fig. 2. Free energy of the superconducting, the antiferromagnetic and the mixed (SC/AF) states as a function of pressure for $\frac{t_1}{t_2} = 0.45$, $t_2 = 0.1$ eV, $T = 4$ K, $g^0_{SC} = 0.37$ eV, $g^0_{AF} = 0.29$ eV.

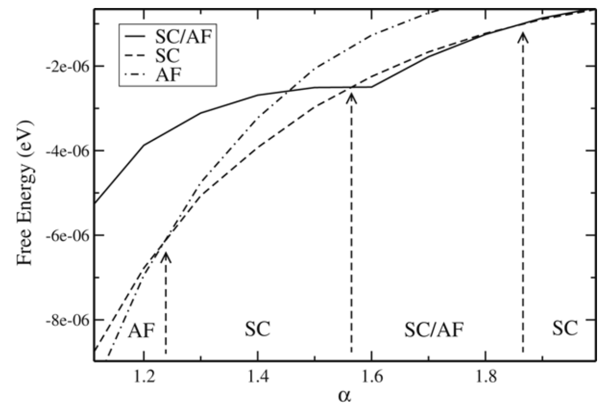

Fig. 3. Free energy of the superconducting, the antiferromagnetic and the mixed (SC/AF) states as a function of pressure for $\frac{t_1}{t_2} = 0.45$, $t_2 = 0.1$ eV, $T = 8$ K, $g^0_{SC} = 0.37$ eV, $g^0_{AF} = 0.29$ eV.

found below 27 K in κ-$(ET)_2$–Cu $[N(CN)_2]$ Cl [2] which is an insulator at ambient pressure. Under pressure, this salt undergoes a superconducting phase transition with T_c increasing with pressure until $T = 13$ K for around 300 bar.

According to our model, the antiferromagnetic spin fluctuations are well defined for lower values of the nesting ratio $\frac{t_1}{t_2}$. When the Fermi surface topology changes by increasing the nesting ratio $\frac{t_1}{t_2}$, we gradually enhance the strength of spin fluctuations (g^0_{AF}) related to the nesting quality and we move from an antiferromagnetically correlated system to a more usual metallic system.

Consequently, changing the nesting ratio $\frac{t_1}{t_2}$, corresponds to a change of the anion X and so from one compound to another. Indeed, the pressure axis in the P–T phase diagram is equivalent, according to our calculations, to the $\frac{t_1}{t_2}$ axis. This equivalence between pressure in experiments and $\frac{t_1}{t_2}$ according to our calculations is in good agreement with experimental measurements done on κ-$(ET)_2$Cu $[N(CN)_2]$ Cl, κ-$(ET)_2$Cu $[N(CN)_2]$ Br and κ-$(ET)_2$Cu$(NCS)_2$ [3,4,28,29].

We show in Fig. 2 (for the same value of the ratio $\frac{t_1}{t_2} = 0.45$ as in Fig. 3) that for the same Fermi surface topology corresponding to the same organic compound, when we change the temperature from $T = 8$ K to $T = 4$ K, the pressure effect is not the same. In fact, the mixed state cannot be stabilized under pressure for $T = 4$ K and the transition occurs from the antiferromagnetic state to the superconducting one.

The results shown in Figs. 2 and 3 provide a plausible explanation for the recent measurements of Lefebvre et al. [3] for X = Cu $[N(CN)_2]$ Cl and Kimura et al. [4] for X = Cu $[N(CN)_2]$ Br. In fact, $\alpha = 1$ in our calculations corresponds to the ambient pressure in experiment for the κ-$(ET)_2$–Cu $[N(CN)_2]$ Cl compound. It was shown experimentally that the SC and AF orders coexist together for $T = 8$ K [3]. More recently Kimura et al. have measured the decrease, with respect to the applied pressure, of the antiferromagnetic order parameter in the mixed state [4]. Our calculations of the dependence of the superconducting and the magnetic orders Δ^0_{SC} and Δ^0_{AF} as a function of α (see Figs. 7 and 8) are in good agreement with the experimental measurements of the two-order volume fractions in the sample [3,4].

By analyzing Figs. 2, 3 and 6, one can conclude that under pressure a phase transition occurs from the antiferromagnetic phase to the superconducting phase. When we apply pressure (α) on the antiferromagnetic ground state, the introduction of disorder to the system results in a superconducting state. By increasing pressure, the novel state changes to a mixing phase in which the SC and AF coexist. For high pressure, the re-entrant superconducting phase is concluded to originate from the phase coexistence of AF and SC. This transition from the coexisting phase to the SC one is consistent with the order parameter dependence on pressure that shows the reduction of antiferromagnetic order. These results are in agreement with Kimura et al. findings [4] showing the decrease of the volume fraction of the AF phase and the transition from the coexisting phase to the re-entrant superconducting one.

The remarkable feature of these findings is that by increasing the pressure (α) we change not only the bandwidth W but probably also the nesting properties by changing the nesting ratio $\frac{t_1}{t_2}$. It seems that increasing pressure is equivalent to increasing $\frac{t_1}{t_2}$ as can be concluded from Figs. 6, 3 and 2. In fact, we find that the less correlated system (less spin fluctuation strength g^0_{AF} corresponding to the lower value of the ratio $\frac{t_1}{t_2}$) corresponds to increasing the bandwidth (or equivalently the pressure) for a given $\frac{t_1}{t_2}$. Figs. 7 and 8 depict the dependence on the applied pressure (α) of the superconducting and the antiferromagnetic order parameters in the mixed state calculated for $T = 8$ K by resolving Eq. (9) written in the mixed phase. We confirm the enhancement of the antiferromagnetic order under pressure and the first-order nature of the transition from one state to another, consistent with the broad resistivity hump that manifests itself under pressure and leading to the superconductivity suppression in κ-$(ET)_2$–Cu$(NCS)_2$ and $\kappa(ET)_2$–Cu $[N(CN)_2]$ Br [27]. Moreover, the calculated pressure effect on superconducting and magnetic order parameters is in sound agreement with the evolution of superconducting and magnetic ones experimentally evaluated by Lefebvre et al. [3] and Kimura

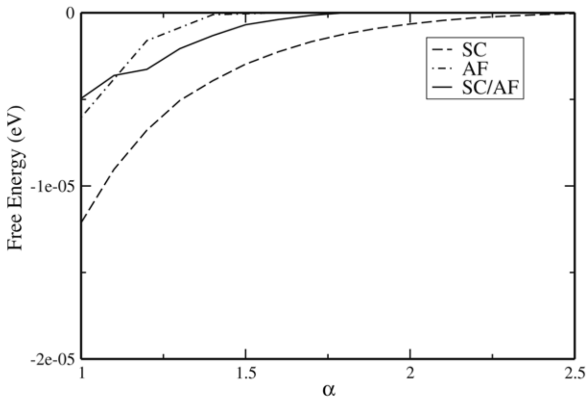

Fig. 4. Free energy of the superconducting, the antiferromagnetic and the mixed (SC/AF) states as a function of pressure for $\frac{t_1}{t_2} = 0.45$, $t_2 = 0.1$ eV, $T = 12$ K, $g^0_{SC} = 0.37$ eV, $g^0_{AF} = 0.29$ eV.

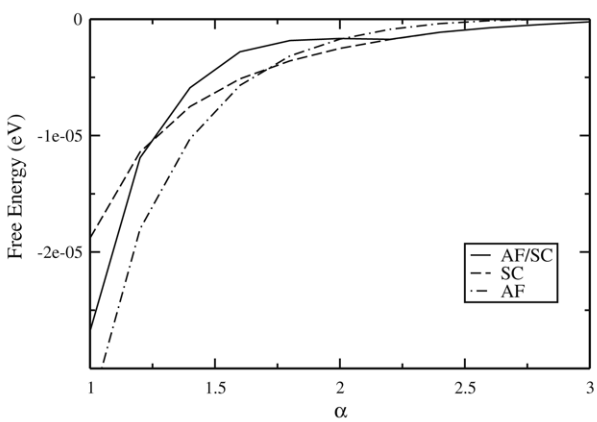

Fig. 5. Free energy of the superconducting, the antiferromagnetic and the mixed (SC/AF) states as a function of pressure for $\frac{t_1}{t_2} = 0.1$, $t_2 = 0.1$ eV, $T = 4$ K, $g^0_{SC} = 0.26$ eV, $g^0_{AF} = 0.22$ eV.

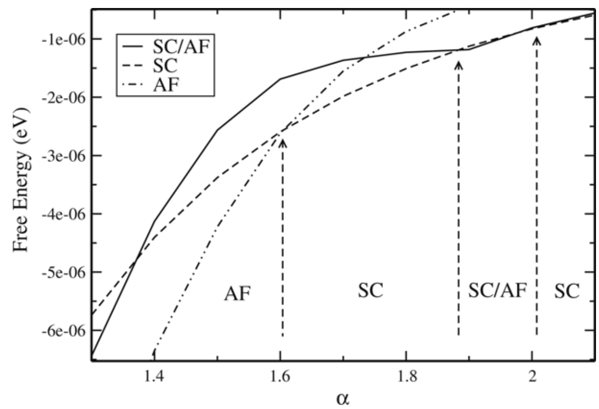

Fig. 6. Free energy of the superconducting, the antiferromagnetic and the mixed (SC/AF) states as a function of pressure for $\frac{t_1}{t_2} = 0.1$, $t_2 = 0.1$ eV, $T = 8$ K, $g^0_{SC} = 0.26$ eV, $g^0_{AF} = 0.22$ eV.

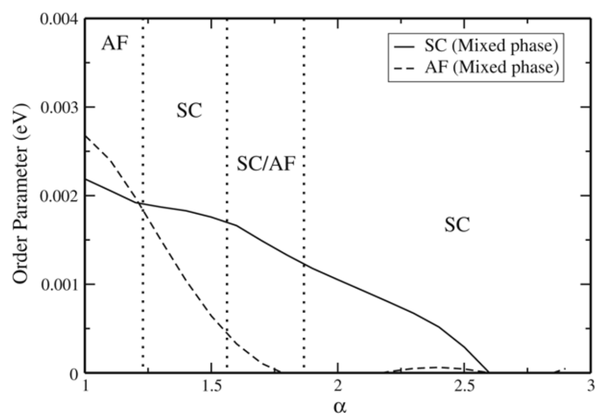

Fig. 7. Superconducting and antiferromagnetic order parameters calculated in the mixed state (SC/AF) as functions of pressure for $\frac{t_1}{t_2} = 0.45$, $t_2 = 0.1$ eV, $T = 8$ K, $g^0_{SC} = 0.37$ eV, $g^0_{AF} = 0.29$ eV.

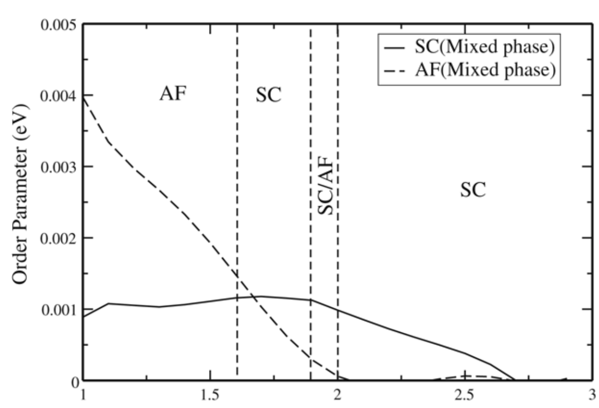

Fig. 8. Superconducting and antiferromagnetic order parameters calculated in the mixed state (SC/AF) as functions of pressure for $\frac{t_1}{t_2} = 0.1$, $t_2 = 0.1$ eV, $T = 8$ K, $g^0_{SC} = 0.26$ eV, $g^0_{AF} = 0.22$ eV.

et al. [4]. It is clear from the free energy curves, that for the same temperature, when we increase the nesting ratio $\frac{t_1}{t_2}$, the nesting quality is enhanced and so the antiferromagnetic spin fluctuations decrease and consequently the pressure range of the magnetic phase in the P–T phase diagram decreases. In fact, for $T = 4$ K and $\frac{t_1}{t_2} = 0.1$, the antiferromagnetic phase is stable until $\alpha \simeq 1.7$ (see Fig. 5). On the other hand this phase is stable for $\alpha \leq 1.2$ for $\frac{t_1}{t_2} = 0.45$ (see Fig. 2).

In Fig. 4 we show the evolution as a function of pressure of the three possible phases for $\frac{t_1}{t_2} = 0.45$ and $T = 12$ K. It appears that pressure does not change the superconducting state. This result is in accordance with Lefebvre et al. works, showing that for $X = Cu[N(CN)_2]Cl$, the sample is a superconductor for $T = 12$ K and remains in the same state under pressure.

4. Conclusion

The most important conclusion one can make from the present work is that when we apply chemical or hydrostatic pressure on organic compounds we modify the nesting quality by changing the nesting ratio $\frac{t_1}{t_2}$ and consequently the bandwidth W changes. Moreover, changing the nesting ratio $\frac{t_1}{t_2}$ in our model corresponds to the change of the anion X and so the ground state to be antiferromagnetic or superconducting. We confirm the analogy between applying pressure in experiments and increasing the bandwidth W or changing nesting properties of the Fermi surface directly linked with the strength of the antiferromagnetic fluctuations.

These results provide a plausible explanation for the coexistence of unconventional superconductivity and antiferromagnetism in the P–T phase diagram of κ-$(ET)_2X$ salts. Indeed, comparing the effects of increasing the pressure in experiments and increasing α, $\frac{t_1}{t_2}$ or W according to our calculations, we find they are compatible if the pressure acts in the sense of increasing W.

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