Multicritical Phenomena of Superconductivity and Antiferromagnetism in Organic Conductor $\kappa$-(BEDT-TTF)$_2$X

Shuichi Murakami and Naoto Nagaosa

Department of Applied Physics, University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan

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We study theoretically the multicritical phenomena of the superconductivity (SC) and antiferromagnetism (AF) in organic conductors $\kappa$-BEDT salts. The phase diagram and the experimental data on the NMR relaxation rate $1/T_1$ is analysed in terms of the renormalization group method. The bicritical phenomenon observed experimentally indicates the rotational symmetry, i.e., SO(5) symmetry, of the SC and the AF. The critical exponent $x$ for the divergence of $1/T_1$ is well explained by $x = \nu(z - 1 - \eta)$ with the dynamical exponent $z = 3/2$ for the AF region while $z = \phi/\nu \sim 1.84$ at the bicritical point. These results strongly suggest that the origin of the SC is in common with that of the AF and that its symmetry is d-wave.

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In strongly correlated electronic systems, it often happens that superconductivity and magnetically ordered states touch or are near to each other. Heavy electron superconductors $\kappa$-[BEDT-TTF]$_2$Cu$_2$(NCS)$_2$ and high-$T_c$ superconductors $\kappa$-[BEDT-TTF]$_2$X$_2$ are examples of this phenomenon, where the Coulomb interactions are believed to be large compared with the bandwidth. In organic conductors, e.g., (TMTSF)$_2$X (X:anion) and $\kappa$-(BEDT-TTF)$_2$X, on the other hand, the magnitude of the Coulomb interaction is smaller because the molecular orbitals are extended. However, the bandwidth $W$ is also small ($\sim$ 1 eV) in these systems, and the electrons are half-filled when dimerization is considered. Therefore, it is expected that the Coulomb interactions play important roles in these compounds. In particular, in $\kappa$-(BEDT-TTF)$_2$X, recent experiments have revealed an interesting phase diagram including the bicritical phenomenon between the antiferromagnetism (AF) and the singlet superconductivity (SC) in the plane of temperature and a parameter $p$ controlling the ratio of the Coulomb repulsion $U$ and the bandwidth $W$. Above a characteristic temperature $T^*(U/W)$, the NMR relaxation rate, $T_1^{-1}$, increases as the temperature is lowered in a manner independent of $p$. Below $T^*(U/W)$, on the other hand, $T_1^{-1}$ diverges towards the AF transition temperature, while it exhibits a spin-gap-like behavior on the SC side.

It is a crucial theoretical issue to treat AF and SC in a unified fashion in the physics of strongly correlated systems. One of the most interesting proposals is based on the SO(5) symmetry, i.e., the rotational symmetry in the 5-dimensional order parameter space of AF and SC. The critical phenomena are useful for testing this idea because of their universality, and actually give the first evidence for it in these organic conductors.

It is well known that the Ginzburg criterion $|T - T_c| < (\frac{\xi}{\xi^c})^2$ specifies the critical region in 3D where the mean field theory breaks down. Here, $a$ is the lattice constant and $\xi$ is the correlation length, whose ratio is determined roughly as $\frac{\xi}{\xi^c} \sim \frac{\Delta}{W_{\text{eff}}}$, where $\Delta$ is the gap introduced in the electronic spectrum by the AF and/or the SC, and $W_{\text{eff}}$ is the reduced effective bandwidth by electron correlation. The mean field theory of the SDW for the AF and the BCS theory for the SC applies in the weak coupling regime, i.e., $\Delta \ll W_{\text{eff}}$, and the critical phenomenon is dominated by the mean-field-like behavior in this case. This occurs when the Coulomb interaction is weak ($U \ll W$) for the AF and/or the electron-phonon coupling is weak for the SC. In the strong coupling regime, on the other hand, we have a sufficiently large critical region and the critical behavior and the phase diagram are determined by the fluctuations beyond the mean-field theory.

In this paper, we study theoretically the critical phenomena of the AF and the SC in $\kappa$-(BEDT-TTF)$_2$X from the viewpoint that this system is in the strong coupling regime and nontrivial critical phenomena are observed. The fluctuation of both the AF and the SC order parameters are treated in terms of the renormalization group (RG) method, employing $\epsilon = 4 - d$ expansion. The critical phenomena occurs at finite temperature and hence is three dimensional. The anisotropy of the system is taken into account by proper rescaling and $\xi = \sqrt{\xi_{\|}^2 + \xi_\perp^2}$, and the critical region is discussed later. Classification of the scaling trajectories leads to three types of multicritical phenomena (Fig. 1): (a) tetracritical, (b) bicritical, and (c) tricritical phenomena. The bicritical phenomenon (b) is unstable towards tetra- and tricritical ones, and is realized only when there is SO(5) symmetry. Furthermore, the dynamic critical phenomena are studied, particularly for the NMR relaxation rate $T_1^{-1}$, and its critical exponent $x$ is in reasonable agreement with the $\epsilon$-expansion result in both the AF region and at the bicritical point. Thus, the analysis of multicritical phenomena gives quantitative evidence of SO(5) symmetry to a good accuracy without adjustable parameters, and it strongly suggests...
that the SC is realized by the Coulomb interaction as well as the AF and its pairing symmetry is d-wave.

Let us consider a generic Ginzburg-Landau model for a system with competing AF and SC order:

\[ H = \int d^3r \left[ \frac{1}{2} g_\parallel |\vec{\sigma}|^2 + \frac{1}{2} \nabla |\vec{s}|^2 + \frac{1}{2} r_\perp |\vec{s}|^2 + \frac{1}{2} |\nabla \vec{s}|^2 + u|\vec{s}|^4 + 2w|\vec{s}|^2|\vec{s}|^2 + v|\vec{s}|^4 \right], \tag{1} \]

where $\vec{\sigma}$ and $\vec{s}$ are the order parameters of the SC and the AF, respectively. We normalize the order parameters so that the coefficients of the gradient terms are $1/2$, and the other coefficients are roughly given in the unit where $a = 1, W = 1$ by $g_\parallel \approx (T-T_{SC})/T_{SC}, r_\perp \approx (T-T_{AF})/T_{AF}$, $u \approx \xi_{SC}^{-2}, v \approx \xi_{AF}^{-2}$, while $w$ represents the competition of the AF and the SC, and is expected to be positive. This model was proposed in refs. [10][11] in the context of the SO(5) theory for high-$T_c$ superconductors [9], and was also used in [12] for the heavy fermion compound CeCu$_2$Si$_2$ ($x \approx 1$) to discuss the effect of disorder. This model has two types of mean field phase diagrams [13]. When the competition of the AF and the SC is not strong, i.e., $w^2 < uw$, there exists the coexistence of the AF and SC, and the phase diagram looks like Fig.1(a). When $w^2 > uw$, the AF and the SC are separated by a first-order phase transition line and the phase diagram looks like Fig.1(b). One might be tempted to interpret the phase diagram and the bicritical phenomenon observed experimentally in terms of this mean field picture. However, there are several experimental facts which imply that the system is in the strong coupling regime at least for the AF: (i) the large saturation value of the staggered magnetization $M_s \approx 0.3 \mu_B$ [14], which is nearly the value for the 2D Heisenberg AF, (ii) fluctuation of the staggered moments are observed as the broadening of the NMR line shape well above $T_{c,AF}$ [9] and (iii) the spin-gap behavior is observed in $(TT)$ [1], well above $T_{c,SC}$. Hence, the critical region near the bicritical point should be large provided that $\xi_{AF} \sim 1$.

![FIG. 1. Schematic phase diagrams of the model (1) as a function of $t \sim (2r_\parallel + 3r_\perp)/5$ and $g \sim (r_\parallel - r_\perp)$. The thick and the thin lines represent first-order and second-order lines, respectively. “S.G.” denotes a region with a spin-gap behavior. (A)-(C) correspond to materials discussed later in the text. (a) $uw > w^2$: tetracritical, (b) $uw = w^2$: bicritical, (c) $uw < w^2$: tricritical.](image)

Let us proceed to the analysis of the fluctuation in terms of the RG method. The RG recursion relations for $u, v, w$ up to order $\epsilon$ are written as [13]

\[
\begin{align*}
\frac{d u}{d \ell} &= \epsilon u - \frac{(n_\parallel + 8)u^2 + n_\perp u w^2}{2\pi^2} \tag{2} \\
\frac{d v}{d \ell} &= \epsilon v - \frac{(n_\parallel + 8)v^2 + n_\parallel u w^2}{2\pi^2} \tag{3} \\
\frac{d w}{d \ell} &= \epsilon w - \frac{(n_\parallel + 2)u + (n_\perp + 2)v + 4w}{2\pi^2}.
\end{align*}
\]

It should be noted that the recursion relations for $u, v, w$ do not involve $r_\parallel, r_\perp$ up to this order. There are six fixed points [13], and a stable fixed point depends on the number of components $n_\parallel$ and $n_\perp$ of the vectors $\vec{\sigma}$ and $\vec{s}$. If $n = n_\parallel + n_\perp$ is smaller than $n_c = 4 - 2\epsilon + O(\epsilon^2)$, a Heisenberg fixed point with $u = v = w = 0$ is the only stable one. This fixed point becomes unstable when $n$ exceeds $n_c$, and the so-called bicritical fixed point with unequal values of $u, v, w$ becomes stable instead.

In the present case with $n_\parallel = 2, n_\perp = 3$, the only stable fixed point is the biconical one $(u^*_B, v^*_B, w^*_B) = 2\pi^2(0.9056, 0.0847, 0.0536)$. However, not all points will flow to this fixed point through the RG (Fig.2). There exists a curved surface, or a “separatrix”, $F(u, v, w) = 0$, which divides the entire parameter space into two regimes: one of convergence to the fixed point $(u^*, v^*, w^*)$, and the other in which the RG flows run away into an unstable region. Numerical analysis shows that this curved surface is well approximated by the surface $uw = w^2$ in the vicinity of the isotropic line $u = v = w$; thus, we can roughly say $F(u, v, w) \sim uw - w^2$.

![FIG. 2. Schematic diagram for the RG flow of the model (1) in the $u/w/v$ plane. Broken lines represent the flow. Points “B” and “II” are the bicritical and the Heisenberg fixed points, respectively. The shaded region is the unstable region of the model.](image)

If $F(u, v, w) \sim uw - w^2 > 0$, the RG flow converges to the biconical fixed point $(u^*_B, v^*_B, w^*_B)$ and the phase diagram shows a tetracritical behavior (Fig.1(a)), as is predicted by the mean-field approximation [13]. From the RG recursion relations of $r_\parallel$ and $r_\perp$ with this biconical fixed point, we obtain $r_B = 0.5 + 0.132\epsilon$ and $\alpha_B = -0.0278\epsilon$. The four second-order phase boundaries are written as $|g| \sim |t|^{\phi_B}$ with the crossover exponent $\phi_B = 1 + 0.135\epsilon$, where $t \sim 2r_\parallel + 2r_\perp$ and $g \sim (r_\parallel - r_\perp)$ are scaling fields corresponding to the temperature and the anisotropy between the AF and SC phases, respectively.
We shift the definitions of $t$ and $g$ properly so that the tetracritical point corresponds to $t = g = 0$. The values of the exponents are different from the ones in [13], which might be due to their approximation of smallness of $\frac{n_1 - n_0}{n_1 + n_0}$. A deviation of $(u, v, w)$ from the biconical fixed point is irrelevant and affects only the size of the tetracritical region.

If $F(u, v, w) \sim uv - w^2 < 0$, on the other hand, although the mean-field approximation predicts a bicritical behavior [13], it is not the case. Through the RG flow, $u$ or $v$ decreases rapidly to become negative, and the model [14] becomes unstable. There are always higher-order terms which stabilize the system, though they are omitted in [14]. Accordingly, the phase transition between the disordered and the ordered phases becomes first order (fluctuation-induced first-order transition [15]), but if the quadratic anisotropy $|g|$ is sufficiently large, the transition becomes second order again [17]. Thus, a first-order transition line between two ordered phases branches at the triple point and extends until these two branches terminate at tricritical points (Figs. 1(c)). If $(u, v, w)$ is near the surface $w = w^2$, the length of the first-order lines on the normal-SC or the normal-AF phase boundaries will be small, since it takes a long time to flow into an unstable region.

Only in the case $w = w^2$ does the RG analysis predict a bicritical behavior (Fig. 1(b)). This behavior is governed by the Heisenberg fixed point $\nu_H = \nu_{H1}^t = \nu_{H1}^\tau = 2\pi^2/13$, which is stable only on the surface $w = w^2$, and the corresponding exponents are $\nu_H = 0.5 + 0.135\epsilon, \phi_H = 1 + 0.192\epsilon, \phi_{\parallel} = -0.0385\epsilon$. The experimentally observed bicritical behavior, combined with the above discussion, strongly suggests $w = w^2$ to a good accuracy, which corresponds to the rotational symmetry in the five-dimensional order parameter space of $(\sigma, \tilde{s})$. It is interesting to note that only if $w = w^2$ is the model [14] smoothly related to the SO(5) NLσ model in [13,15] in the limit $-r_1, -r_2, u, v, w \to \infty$ with their ratios fixed.

Now let us consider the dynamic critical phenomena [19]. The NMR relaxation rate [18] exhibits a divergence toward the transition in the AF side, or a spin-gap behavior in the SC side, while these two cases show a similar behavior above $T^*$. This is reminiscent of the bicritical crossover behavior of the model [14]. The NMR linewidth is proportional to $(T - T_{c,AF})^{\alpha}$ on the AF side of the normal phase with $\alpha = \nu(z - 1 - \eta)$, where $z$ is a dynamic critical exponent [14]. When the system is in the vicinity of $T_{c,AF}$, but not near the bicritical point, its dynamic critical behavior is governed by that of the AF isotropic Heisenberg model. Macroscopic variables describing slow dynamics are the staggered magnetization $\tilde{s}$ and the conserved uniform magnetization $\tilde{m}$, giving $z = d/2 = 3/2$. Thus, the exponent is $x_{AF} = 0.315$ up to $O(\epsilon^2)$ [20]. On the other hand, when the bicritical fixed point governs the dynamic critical phenomena, the value of $z$ changes, and so does the exponent $x$. In the bicritical region, the SC order parameters $\tilde{\sigma}$ enter the set of macroscopic variables. The free energy $F$ has a scaling form:

$$F(t, g, \tilde{\sigma}, \tilde{s}, \tilde{m}) = t^{2-\alpha} \Phi \left( \frac{g}{t^{\alpha}}, \frac{\tilde{\sigma}}{t^{\beta}}, \frac{\tilde{s}}{t^{\gamma}}, \frac{\tilde{m}}{t^{\zeta}} \right),$$

where $\tilde{\beta} = 2 - \alpha - \phi_H$. $t$ is proportional to $T - T_{c, BP}$, where $T_{c, BP}$ denotes the temperature at the bicritical point. Following the power-counting argument of the bicritical behavior in the spin-flop AF [21], we get $z = \phi_H/\nu_H \sim 1.84$, resulting in $x_{BP} = 0.584$, where we used $\phi_H \sim 1.313, \nu_H \sim 0.714$ up to $O(\epsilon^2)$ [20]. This value of $x_{BP}$ at the bicritical point increases as a function of $n$: $x_{BP} = 0.528, 0.558, 0.584, 0.607$ for $n = 3, 4, 5, 6$, respectively. On the other hand, in approaching the SC phase it does not diverge but exhibits a spin-gap behavior below a characteristic temperature $T_{5, G.}^\parallel \sim |g|^{1/\phi_H}$ because of the singlet formation. Throughout the entire critical region of the normal phase, $T_{c}^{-1}$ has a scaling form $T_{c}^{-1} = t^{x_{BP} f}(g/t^{\phi_H})$.

![Fig. 3](image-url)

**Fig. 3.** Log-log plot of $T_{c}^{-1}$ vs $(T - T_{c})/T$ for (A) $\kappa$-(BEDT-TTF)$_2$Cu[N(N$_2$)$_2$Cl] (solid squares), and (B) deuterated $\kappa$-(BEDT-TTF)$_2$Cu[N(N$_2$)$_2$]Br (open squares). (Data from [13].)

Fig. 3 shows the log-log plot of $T_{c}^{-1}$ vs $T - T_{c}$ for (A) $\kappa$-(BEDT-TTF)$_2$Cu[N(N$_2$)$_2$Cl] (solid squares), and (B) deuterated $\kappa$-(BEDT-TTF)$_2$Cu[N(N$_2$)$_2$]Br (open squares). The former is located in the AF region slightly distant from the bicritical point while the latter is nearly at the bicritical point, as shown in Fig 1(b). We selected $T - T_{c}$ instead of $T_{c}^{-1}$ as the abscissa, because the critical temperatures of these compounds are much lower than their classical values ($\sim J \sim 500$ K), indicating that they are close to a quantum critical point. This scaling by $T - T_{c}$ near the quantum critical point is obtained from an RG study of a quantum SO(5) NLσ model with a quadratic anisotropy, and a detailed discussion will be given in a subsequent paper [22]. The proximity to the quantum bicritical point is also consistent with the strong correlation, because $T_{c}$ is suppressed by the quantum fluctuation driven by the Coulomb interaction.
The estimated slope, i.e., the critical exponent $x$, in the region $1 \mathrm{~K} \lesssim T - T_c \lesssim 10 \mathrm{~K}$ gives $x_{AF} \sim 0.30 \pm 0.04$ for (A) and $x_{BP} \sim 0.56 \pm 0.04$ for (B). The values of $x$ are in reasonably good agreement with the above theoretical values ($x_{AF} = 0.314$, $x_{BP} = 0.584$), which supports that the critical region is wide ($\sim 10 \mathrm{~K}$) and $\xi_{AF} \sim 1$. Several points should be noted. (i) The fitting with $\frac{T - T_c}{T_c}$ underestimates the values of $x$ as $x \sim 0.24$ for (A) and $x \sim 0.40$ for (B); thus, it guarantees the validity of our fitting with $\frac{T - T_c}{T_c}$. (ii) Note that the data in $T - T_c \lesssim 1 \mathrm{~K}$ seems to deviate from the above fitting. We discard these data in the fitting for the following reasons. First, $T_c$ cannot be determined more accurately than $\Delta T_c \sim 0.5 \mathrm{~K}$ due to experimental limitations. This uncertainty largely affects the fitting within $T - T_c \lesssim 1 \mathrm{~K}$. Second, in real systems, SO(5) symmetry is approximate, and there is always a small deviation from $uv - w^2 = 0$, which alters the critical behavior very close to $T_c$. Here, it is important to note that the increase of the deviation through the RG flow is slow; in the linearization of $\frac{T - T_c}{T_c}$, the exponent $\epsilon/13$ of the growth is much smaller than the magnitude of the other two exponents $-8\epsilon/13, -\epsilon$. Therefore, the bicritical behavior governed by the SO(5) fixed point can be observed for a relatively wide range of $\{uv - w^2\}$. For example, $\{uv - w^2\} \lesssim 0.1 \cdot \max(uw, w^2)$ is sufficient to explain the observed bicritical phenomenon.

Let us consider whether it is possible to have such a large ($\sim 10 \mathrm{~K}$) critical region in the quasi-2D system. Following $\frac{\partial}{\partial T}$, we can evaluate the critical region of the quasi-2D Heisenberg system as

$$\frac{|T - T_c|}{T_c} \lesssim \frac{kT_c}{J_{\parallel}} \sim \frac{1}{\ln(J_{\parallel}/J_{\perp})},$$

where the in-plane exchange $J_{\parallel}$ is larger than the inter-plane exchange $J_{\perp}$. Thus there is only logarithmic dependence on $J_{\perp}/J_{\parallel}$, and it does not significantly reduce the width of the critical region. In $\kappa$-(BEDT-TTF)$_2\mathrm{X}$ in a metallic region, the anisotropy of the conductivity is $\sigma_{\parallel}/\sigma_{\perp} \sim 100$, implying $t_{\parallel}/t_{\perp} \sim \sqrt{100}$ and $J_{\parallel}/J_{\perp} \sim 100$. Thus, the width (7) is not extremely small ($\sim 0.2 - 0.3$), and in the AF region ($T_c \sim 30 \mathrm{~K}$) the observed size of the critical region $5 \mathrm{~K} - 10 \mathrm{~K}$ is reasonable. In contrast, in quasi-1D systems, the width in (8) is $J_{\perp}/J_{\parallel}$; the quasi-one-dimensionality is effective in reducing the width of the critical region.

On the SC side, the experimental estimation of the coherence length $\xi_{SC}$ is still controversial, e.g., for the SC compound $\kappa$-(BEDT-TTF)$_2\mathrm{Cu(NCS)}_2$, the calculated in-plane coherence length $\xi_{SC}$ ranges from 31 Å in $\frac{\partial}{\partial T}$ to $\sim 180$ Å in $\frac{\partial}{\partial c}$. However, the rotational symmetry between the AF and SC further suggests that the correlation length $\xi_{SC}$ is also short ($\sim 1$). This is because when $\xi_{SC} \gg \xi_{AF}$, the scaling trajectory in Fig. 8 starts from the initial point ($u \ll v$) far away from the Heisenberg fixed point, and the deviation from the separatrix is easily magnified as the length scale becomes larger. It is also inferred that the observed spin-gap behavior is due to the large fluctuation of the SC order parameter in the SC compound $\kappa$-(BEDT-TTF)$_2\mathrm{Cu(NCS)}_2$ (C) in Fig. 1(b)).

One might wonder that the easy-axis anisotropy reduces the number of the components of the AF order parameters and stabilizes the bicritical phenomenon. However, the spin-orbit interaction is negligible in organic systems, and the spin anisotropy energy due to dipole-dipole interactions is estimated by the AF resonance to be $\sim 10^{-4} \mathrm{~K}$ for (TMTSF)$_2\mathrm{X}$. Assuming a similar value for $\kappa$-(BEDT-TTF)$_2\mathrm{X}$, and that $\xi_{AF} \sim 1$, this anisotropy is negligible in the temperature region of interest.

Finally, the implications of these results are discussed below. The rotational symmetry between AF and SC order parameters suggest that the mechanism of the AF and SC is common and that the underlying microscopic quantum model has enhanced dynamical symmetry, i.e., SO(5). Therefore, it is likely that the symmetry is d-wave. We believe that the (a) half-filling, (b) intermediate Coulomb interaction, and (c) nearly two-dimensional Fermi surface in $\kappa$-(BEDT-TTF)$_2\mathrm{X}$ makes the SO(5) symmetric model a promising candidate. When these conditions are largely violated, e.g., by doping carriers and/or the application of an external magnetic field, the SO(5) symmetry is broken and the bicritical phenomenon turns into tetra- or tricritical behavior [18].

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* e-mail: murakami@appi.t.u-tokyo.ac.jp

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