Phenomenological theory in reentrant uranium-based superconductors

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We develop a phenomenological theory for the family of uranium-based heavy fermion superconductors ($URhGe$, $UCoGe$, and $UTE_2$). The theory unifies the understanding of both superconductivity (SC) with a weak magnetic field and reentrant superconductivity (RSC) that appears near the first-order transition line with a high magnetic field. It is shown that the magnetizations along the easy and hard axis have opposite effects on superconductivity. The RSC is induced by the fluctuation parallel to the direction of the magnetic field. The theory makes specific predictions about the variation of triplet superconductivity order parameters $\hat{d}$ with applied external magnetic fields and the existence of a metastable state for the appearance of the RSC.

Heavy-fermion superconductors $UCoGe$, and $URhGe$, as promising spin triplet superconductors, have drawn significant attentions. The spin triplet pairing is supported by their highly anisotropic upper critical fields which greatly exceed the Pauli limit along all three crystallographic directions (for $UCoGe$ only the a and b-axis) [1–6], and the coexistence of ferromagnetism (FM) and superconductivity (SC) [7–10] in the absence of magnetic fields.

Very recently, another uranium-based superconductor $UTE_2$ has been found. Considerable researches have been conducted on it [11–16]. In the light of these findings, the new superconductor shares many common features with the previous counterparts, such as highly anisotropic upper critical fields and reentrant superconductivity (RSC) under high magnetic fields. However, unlike the previous ones, there is no sign of ferromagnetic order in $UTE_2$ down to 25 mK [13, 14]. In all these superconductors, the superconductivity transition temperature, $T_{SC}$, is first suppressed by the magnetic field ($h_y$) perpendicular to both the hardest (x) and easy axis (z). But when the magnetic field is strong enough, the $T_{SC}$ arises again [15–21].

The difference between these superconductors brings new challenges and calls for a unified understanding of these materials. On the basis of Landau phenomenological theory and weak-coupling theory for $URhGe$ given by Mineev [22, 23], the jump of the magnetic moment $m_{z0}$ enhances the fluctuations along the easy axis to induce the RSC. This mechanism can not be applied to understand the RSC in $UTE_2$ [15, 16] because $UTE_2$ has no magnetic order along the easy-axis [6, 14–16]. The increase of the fluctuation along the easy axis cannot be the only cause of the appearance of the RSC. Experimentally, it has also been found that both the longitudinal (along the easy axis) and the transverse (along the magnetic field) fluctuations exist near the RSC region in $URh_{0.9}Co_{0.1}Ge$ by $^{59}Co$ nuclear magnetic resonance (NMR) measurements [24].

Herein, we generalize the phenomenological theory of the spin fluctuation feedback effect (SFEE) proposed by Amin et al. [25] to explain the physics in the family of uranium-based superconductors. We show that the decrease of $T_{SC}$ in a weak magnetic field and the appearance (disappearance) of the RSC near the first-order transition in $URhGe$, $UCoGe$, and $UTE_2$ can be understood in a unified manner. In the weak magnetic field region, $T_{SC}$ decreases with the decrease of static magnetic order along easy axis and the increase of magnetic moment along field directions. In the strong field region, the RSC is caused by the fluctuations along magnetic field directions. However, RSC can be killed by destroying the metastable state near a first order transition and a sudden increase of magnetic moment along the field directions. Our theory further predicts the d-vector of the RSC in these superconductors and the metastable RSC state during the magnetic-hysteresis-loop, providing a sound theoretical basis for further investigation of the RSC in a microscopic theory.

We first focus on the SC and RSC in ferromagnetic uranium-based superconductors, and take $URhGe$ as an example. The phase diagram of $URhGe$ is sketched in Fig.1. In the weak magnetic fields region, the SC coexists with FM, we can write the total free energy containing both magnetic ($\vec{m}$) and spin-triplet superconductivity ($\vec{d}$) order parameters as:

$$f_{sc-m} = A_{i|x} (m_i)^2 + B_{ij} (m_i)^2 (m_j)^2 - h_y m_y + K_{i|x} (m_i)^2 |\vec{d}|^2 + K_{2i} m_i (i\vec{d} \times \vec{d})^i, \tag{1}$$

where $i,j = x, y, z$ and $A_{i|x} < 0, A_{i|z} > 0, A_{1y} > 0 , B_{ij} > 0$ to ensure the FM ground state. Positive $K_{1i}, K_{2i}$ are the amplitudes of the couplings between the FM and SC order parameters [25]. The first line of Eq.1 is the magnetic part of the total free energy, the second line are terms coupling magnetic moment and superconductivity.

In the weak magnetic field region, the magnetic part
of the total free energy can be simplified to be:

\[ f_m = A_1m_z^2 + B_zm_z^4 + A_2m_y^2 - Bh_ym_y, \quad (2) \]

in which the contribution from the x-axis and the mixed terms were ignored. The minimization of this free energy gives the magnetic moment: \( m_0^\alpha = (0, \frac{h_y}{2A_1}, -\frac{A_2}{2B_z}) \). To obtain the SFFE on superconductivity, we integrate out the magnetic fluctuation \( \delta \hat{m} \) (\( \hat{m} = m_0 + \delta \hat{m} \)) in \( f_{sc-m} \) and derive an effective superconductivity free energy,

\[ f_{sc} = \alpha|\vec{d}|^2 + K_{2z}m_zm_0 + \beta|\vec{d}|^4, \quad (3) \]

where \( \vec{d} = i\hat{d} \times \hat{d}, \alpha = \alpha_0(T-T_c0) + K_{1z}m_z^2 + \frac{K_{2z}}{2A_1m_0^2} + K_{1y}m_y^2 \) in which \( T_c0 \) is the critical temperature in the absence of the SFFE. By minimizing the free energy, we obtain the non-unitary superconductivity with order parameter \( \vec{d} = \frac{d_0}{\sqrt{2}}(1, -i, 0) \). This superconducting state has intrinsic z-polarized magnetic moment proportional to \( \vec{p} = -d_0^2\hat{z} \) [26] and the critical temperature [see supplement for detail]:

\[ T_{SC} = T_c0 - \left( \frac{K_{1z}m_0^2}{\alpha_0} + \frac{K_{1y}m_0^2}{8B_zm_0^2\alpha_0} \right) \frac{K_{1y}m_0^2}{\alpha_0} + \frac{K_{2z}m_0}{\alpha_0}. \quad (4) \]

For a weak ferromagnetic superconductor [23, 27], we can assume that \( m_z < \frac{1}{\sqrt{2B_z}} \). In this case, it can be seen from Eq.4 that either the decrease of \( m_z \) or the increase of \( m_y \) results in the decrease of \( T_{SC} \). Namely, \( T_{SC} \) decreases with increasing magnetic field \( h_y \), corresponding to SC phase of \( URhGe \).

Now we consider the strong magnetic field region to discuss the rotation of the d-vector and appearance of the RSC. Since the emergence of RSC is closely related to the first-order transition. The magnetic part of the free energy near the first-order transition line can be written as

\[ f_m = \alpha_zm_z^2 + \beta_zm_z^4 + \alpha_ym_y^2 + \beta_{yz}m_y^2m_z^2 - m_yh_y, \quad (5) \]

where \( \alpha_z < 0, \alpha_y > 0, \beta_z, \beta_{yz} > 0 \). Minimizing the free energy, we obtain the magnetic moments as \( m_z^0 = -\frac{\alpha_z+\beta_zm_z^0}{2\beta_z} \) and \( m_y^0 = \frac{h_y}{2(\alpha_y+\beta_ym_z^0)} \). Following the same procedure, by adding terms coupling magnetic moment and field with superconductivity on \( f_m \) and integrating out magnetic fluctuations in \( f_{sc-m} \), we obtain the effective free energy including the SFFE as

\[ f_{sc} = \alpha'|\vec{d}|^2 + K_{2y}m_y^4 + K_{2z}m_z^4 + \mu h_y, \quad (6) \]

where \( \alpha' = \alpha + K_{1y}m_0^2 + K_{1z}m_0^2 + K_{2z}m_0^2 + K_{1z}m_z^4 \) and the high order terms are not specified. Close to the first-order transition critical magnetic field \( h_{m} \), the variation of the last several terms dependent on \( m_z \) is small so that we can take it as a constant. The superconductivity with \( \vec{d} = \frac{d_0}{\sqrt{2}}(1, 0, \pm i) \) and \( \vec{p} = \pm d_0^2\hat{z} \) has the highest \( T_{SC} \). As a function of the magnetic field,

\[ T_{SC} = T_{c0} - \frac{1}{\alpha_0}\left( \frac{K_{1y}m_0^2}{\alpha_0} - |K_{2y} - \mu h_y| \right), \quad (7) \]

where \( \alpha_y' = \alpha_y + \beta_{yz}m_z^2 \). The phase diagram in the strong magnetic field region can be explained if we assume \( h_m < h_y \equiv \frac{K_{2y}m_0^2}{\alpha_0} \). In this case, from Eq.7, the \( T_{SC} \) increases with increasing magnetic field \( (h_y > 0) \) at first and then decrease when the magnetic filed \( h_y > h_m \). However, it is noteworthy that Eq.7 is valid only for the region which is on the left side of the first-order transition line and close to it. When the magnetic field continues to increase and exceeds the critical value \( h_m \), as will be analyzed next, the RSC disappears with increasing magnetic field.

The first order transition and the disappearing of RSC close to it can be further understood within our theory. With a strong magnetic field and a small \( m_z \) in the FM phase, the free energy to describe the first order transition can be written as [22, 23]

\[ f_m = -\frac{h_y^2}{4\alpha_y} + \alpha_zm_z^2 + \beta_zm_z^4 + \gamma_m^2m_z^6, \quad (8) \]

where \( \alpha_z' = \alpha_z + \frac{\beta_{yz}h_y}{4\alpha_y}, \beta_z' = \beta_z - \frac{\beta_{yz}^2h_y^2}{4\alpha_y^2}, \) and \( \gamma_m' = \frac{\beta_{yz}^2h_y^2}{4\alpha_y} \). So one can learn from Eq.8 that the magnetic field \( h_y \) modifies the coefficients of the free energy \( f_m \). Thus the magnetic moment dependence of the free energy changes with increasing magnetic field, as shown in Fig.2. From Eq.8, we can derive [see supplement for detail] the condition for the first-order transition, \( m_z^2 = \frac{-\beta_z' + \sqrt{\beta_z'^2 - 4\gamma_m'^2m_z^6}}{2\gamma_m'^2} \). As well as the condition that the local minima are broken: \( m_z^2 = \frac{-\beta_z'}{2\gamma_m'} \). Here as \( m_z > m_z^c \), there is a metastable state as displayed in Fig.2(e), corresponding to the state between the green and orange lines in Fig.1. During the up-sweep of magnetic field, the system can cross the first-order transition line, the magnetic moment \( m_z \) does not
implies with the same method as before: 

$$T_{SC} = T_{c0} - \frac{K_1}{2(6 \delta' m_z^2 \kappa) \alpha_0} - \frac{K_1 m_z^2 \alpha_0}{\alpha_0} + \frac{\mu |h_y|}{\alpha_0}.$$  

\( (9) \)

The second term in Eq.9 shows that if the metastable state is broken, namely, \( m_{z0} = m_{ze} \) (when \( m_{z0} = \frac{\alpha_0}{6 \delta'}, \)
as shown in Fig.2 (d)), \( T_{SC} \) reaches \(-\infty\), indicating the disappearance of RSC before the metastable state broken line.

Secondly, we consider the paramagnetic uranium-based superconductor, \( UTe_2 \) [6, 14]. For this material, there are several known experimental facts. The SC as the weak field region is initially suppressed by the increasing magnetic field \( h_y \). However, when the magnetic field is sufficiently strong, the RSC appears. Finally, when the magnetic field arrives at 34.9T [28], a first order transition occurs with the increasing jump of the magnetic moment \( m_{y0} \), and the RSC disappears simultaneously. The phase diagram of the \( UTe_2 \) is summarized in Fig.3.

Due to the absence of FM order, the free energy in our theory hardly supports non-unitary triplet SC states in the weak magnetic field region. Thus the non-unitary triplet state do not exist in \( UTe_2 \) when the magnetic field is not strong enough, consistent with the measurements of heat capacity and thermal conductivity. 

which indicates the point-node gap structure [29]. Similar to the method in the ferromagnetic uranium superconductors, the superconducting critical temperature can be derived:

$$T_{SC} = T_{c0} - \left( \frac{K_{1z}}{2A_{1z}} + \frac{K_{1y}}{2A_{1y}} \right) - K_{1y} m_{y0}^2,$$  

\( (10) \)

where \( T_{c0} \) is the superconducting critical temperature without the SFFE. Since \( m_{y0} \) increases with increasing \( h_y \), the Eq.10 implies \( T_{SC} \) decrease as shown in Fig.3.

However, when the magnetic field is strong enough, from out theory, the non-unitary superconductivity can appear because of the \( K_2 \) coupling term in Eq.1. The free energy can be expressed as:

$$f_{sc-m} = A_{1z} m_z^2 + A_{1y} m_y^2 + K_{1z} m_z^2 |\vec{d}|^2 + K_{1y} m_y^2 |\vec{d}|^2 + K_{2y} m_y p_y - \mu h_y p_y.$$  

\( (11) \)

The superconducting critical temperature can be derived as:

$$T_{SC} = T_{c0} - \left( \frac{K_{1y} m_{y0}^2}{\alpha_0} + \frac{|K_{2y} - 2\mu A_{1y}|}{\alpha_0} \right) m_{y0},$$  

\( (12) \)

where \( \alpha_0 > 0 \). This parabolic function on \( m_{y0} \) explains the RSC close to the first order transition line in \( UTe_2 \).

Similar to the FM case, we can also describe the first-order transition and metastable state in \( UTe_2 \), which have been detected in experiment [28]. In this case, the magnetic part of the free energy in a strong magnetic field can be written as:

$$f_m = a_y m_y^2 - c_y m_y^3 + \frac{1}{2} b_y m_y^4 - \mu_1 h_y m_y + a_z m_z^2.$$  

\( (13) \)

We can derive [see supplement for detail] the condition for the first-order transition, \( h_{yc1} = \frac{c_y}{\mu_1} \left( \frac{a_y}{2c_y^2} - \frac{c_y^2}{b_y} \right) \) as well.
as the condition that the local minimum appears, $h_{yc2} = \frac{\Delta_y(8a_y b_y - c_y \Delta_y)}{36b_y^2 \mu_1}$, where $\Delta_y = 3c_y + \sqrt{9c_y^2 - 12a_y b_y}$. Here $h_{yc2} < h_{yc1}$, so the metastable state is located in the paramagnetic region as shown in Fig. 3. Then considering the fluctuation $\vec{m} = (0, m_y, \delta m_y, \delta m_z)$, we obtain the total free energy as follow:

$$f_{sc-m} = a_y \delta m_y^2 + (a_y + \mu_1 \frac{h_y}{m_y} + \mu_2 m_y^2) \delta m_y^2 + K_1 m_y^2 |\vec{d}|^2 + K_2 m_y |m_y|^2 + K_3 \mu_y m_y - \mu_1' h_y p_y,$$

(14)

with the superconducting critical temperature

$$T_{SC} = T_a - \frac{K_1 y}{2(\mu_1 \frac{h_y}{m_y} - a_y)} - K_1 m_y^2 + |K_2 m_y - \mu_1' h_y|,$$

(15)

When the first-order transition happens, the magnetic moment $m_{y0}$ increases abruptly. The second term in Eq.15 increases suddenly. As for the last two terms of Eq.15, the jump of the magnetic moment $m_{y0}$ can lead to $m_{y0} \gg \frac{K_2 m_y - \mu_1' h_y}{2K_2}$ which belongs to the right side of the first-order transition line. In this case, the $T_{SC}$ decrease abruptly as shown in Fig. 3. This explains the experimental observation of the sudden truncation of the RSC in $UTe_2$ upon the first order transition [15, 16].

In summary, we develop a phenomenological theory to describe the SC and RSC in uranium-based superconductors unified. The theory explains the global phase diagram of this family of superconductors. In our theory, the SC at weak magnetic region are suppressed with the increasing transverse magnetic field $h_y$, due to the energy cost from the mismatch of the induced transverse magnetic moment $m_{y0}$ with the $z$-polarized nonunitary SC order $p_z$ and the unitary SC order, for the ferromagnetic and paramagnetic superconductors, respectively. However, the RSC in both ferromagnetic and paramagnetic superconductors are induced by the fluctuation parallel to the magnetic fields, rather than the sudden jump of the magnetic moment upon the first order transition. Instead, the sudden jump of the magnetic moment indeed truncates the superconductivity and there should be a shift of the superconducting dome upon a magnetic-hysteresis-loop type of measurement.

Our theory makes a few explicit predictions. First, we predict that the rotation of the spin-triplet pairing d-vector in different magnetic field regions. In ferromagnetic superconductors $UCoGe$ and $URhGe$, with increasing magnetic field, the d-vector rotates from $\frac{\vec{d}}{\sqrt{2}}(1, -i, 0)$ to $\frac{\vec{d}}{\sqrt{2}}(1, 0, \pm i)$. In $UTe_2$, the superconductivity is unitary at first. However when the magnetic field is high enough, it becomes a non-unitary superconductivity with a d-vector, $\frac{\vec{d}}{\sqrt{2}}(1, 0, \pm i)$. The rotation of the d-vector by magnetic field was studied in Sr$_2$RuO$_4$[30] whose spin-triplet pairing symmetry has been seriously questioned recently[31]. In principle, this prediction can be examined experimentally in superconducting junctions made by these materials. The d-vector can also be visualized from quasi-particle interference technique in STM experiments[32].

Second, we predict that it is the metastable state that ensures the extension of the RSC over the right side of the first-order transition line in $URhGe$ and $UCoGe$. This prediction can be checked by performing a magnetic-hysteresis-loop type measurement around the first order transition line. We can apply a strong magnetic field to destroy the metastable state at first and then reduce it to induce the RSC. The maximum of the upper critical magnetic field is predicted to have a magnetic-hysteresis-loop type of behavior. Namely, the maximum of the upper critical magnetic field is predicted to be much smaller than the one with a normal procedure that the field crosses the first-order transition line from its left side. The RSC dome upon down-sweep magnetic field would shift to the left of the first order transition line as depicted by the broken cyan line in Fig. 1.

Finally, we predict that the metastable state also exists in $UTe_2$ and affects the behavior of the RSC in $UTe_2$ because of the magnetic hysteresis [28]. The metastable state indicates the remaining large magnetic moment $m_{y0}$ during the down-sweep process. Since the RSC is truncated by the sudden increase of $m_{y0}$, during the up-sweep, the RSC would exist until the first order transition line. However, during the down-sweep, the magnetic moment does not decrease abruptly when the system crosses the first-order transition line so that the RSC will not appear until the system cross the metastable state broken line (the cyan down-sweep line in Fig. 3). (note: After we completed this paper, we notice that the magnetic-hysteresis-loop type of behavior near the first-order transition line in $UTe_2$ were detected in a recent experiment [33]. Although the experimental was performed under pressure, it is a strong support for our theory.)

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Supplemental Information

Superconducting critical temperature of the free energy with non-unitary part

Starting from Eq.(1) in the main text, we integrate out the magnetic fluctuations \( e^{-\int d^2xf_{sc}} = \int D(\delta \vec{m}) e^{-\int d^2xf_{sc-m}} \) to get the effective superconductivity free energy in Eq.(3):

\[
f_{sc} = \alpha |\vec{d}|^2 + K_{2z} \eta_2 m_{z0} + \beta |\vec{d}|^4,
\]

(A.1)

By minimizing the free energy and using this equation:

\[
\sum_i d_i \frac{\partial f_{sc}}{\partial d_i} = 0,
\]

we can get:

\[
\alpha |\vec{d}|^2 + 2\beta |\vec{d}|^4 + K_{2z} \eta_2 (id \times d^2)_z = 0.
\]

(A.2)

Substituting \( \vec{d} = (d_1, d_2 e^{i\phi}, d_3 e^{i\theta}) \) into Eq.A.2 gives this equation:

\[
\alpha (d_1^2 + d_2^2 + d_3^2) + 2\beta (d_1^2 + d_2^2 + d_3^2)^2 + 2K_{2z} \eta_2 d_1 d_2 \sin \phi = 0.
\]

(A.3)

We take:

\[
(d_1, d_2, d_3) = d_0 (\sin x \cos y, \sin x \sin y, \cos x),
\]

(A.4)

and obtain from Eq.A.3:

\[
d_0^2 = -\frac{2\beta}{\alpha + K_{2z} \eta_2 \sin x \sin 2y \sin \phi}.
\]

(A.5)

Where \( \beta > 0 \). To get the highest superconducting critical temperature, in Eq.A.5, for \( K_{2z} > 0 \), we choose: \( \sin^2 x = 1, \sin 2y = 1, \sin \phi = -1 \). So the d-vector is \( \frac{d_0}{\sqrt{2}} (1, -i, 0) \).

In the same way, for \( K_{2z} < 0 \), the d-vector is \( \frac{d_0}{\sqrt{2}} (1, i, 0) \).

First-order transition and metastable state broken in \( URhGe \) (\( UCoGe \))

We start from the free energy Eq.(8) about the magnetic moment in the main text:

\[
f_m = -\frac{h_y^2}{4\alpha_y} + \alpha'_z m_z^2 + \beta'_z m_4^2 + \delta'_z m_6^2.
\]

(A.6)

There are two non-zero local minima in this free energy which satisfy this equation:

\[
2\alpha'_z m_z^2 + 4\beta'_z m_4^2 + 6\delta'_z m_6^2 = 0.
\]

(A.7)

The solution of the Eq.A.7 is: \( m_{z0}^2 = \frac{-\beta'_z + \sqrt{\beta'_z^2 - 3\alpha'_z \delta'_z}}{3\delta'_z} \).

The local minimum will be broken at this condition: \( 3\delta'_z \alpha'_z = \beta'_z \) namely, \( m_{z0}^2 = \frac{-\beta'_z}{3\delta'_z} (\alpha'_z, \delta'_z > 0; \beta'_z < 0) \). The metastable broken line is shown in the Fig.1, and corresponds to the Fig.2(d). Moreover, the first order transition line is determined by these equations:

\[
\frac{\partial f_m}{\partial m_y} = 2a_y m_y + 2b_y m_y^3 - 3c_y m_y^2 - \mu y h_y = 0
\]

(A.9)

\[
f(m_{y1}) = f(m_{y3}).
\]

(A.10)

![Fig. A.1](image)

**FIG. A.1.** First-order transition in \( UTe_{2} \), from (a) to (d), the magnetic field increases gradually. (a) free energy\( f_m \) - magnetic momentum\( m_y \) in zero magnetic field. (b)\( f_m - m_y \) on the left side of first-order transition line. (c)\( f_m - m_y \) on the first-order transition line. (d)\( f_m - m_y \) on the right side of the first-order transition.
The relationship of the coefficients and the solutions is given by:

\[ m_1 + m_2 + m_3 = \frac{3c_y}{2b_y} \]
\[ m_1m_2 + m_2m_3 + m_3m_1 = \frac{a_y}{b_y} \]
\[ m_1m_2m_3 = \frac{\mu_1h_y}{c_y} \] \hspace{1cm} (A.11)

From Eq. A.10, this condition \( m_1 + m_3 = 2m_2 \) can be derived. By substituting it into the Eq. A.11, we can get the equations about \( m_1 \) and \( m_3 \):

\[ m_1 + m_3 = \frac{c_y}{b_y} \]
\[ m_1m_3 = \frac{\mu_1h_y}{c_y} \] \hspace{1cm} (A.12)

and the critical magnetic field: \( h_{yc1} = \frac{c_y}{\mu_1} \left( \frac{a_y}{b_y} - \frac{c_y^2}{2b_y} \right) \), which is corresponding to the magnetic field in the Fig. A.1 (c). The jump of the magnetic momentum is \( \Delta m_{y0} = \sqrt{\left(\frac{c_y}{b_y}\right)^2 - \frac{4\mu_1h_a}{c_y}} \).

However, when the magnetic field is small, the local minima and local maximum don’t appear, as shown in Fig. A.1 (a). So we can derive a critical magnetic field for the appearance of the local minima and local maximum. The critical magnetic field \( h_{yc2} \) satisfies these equations:

\[ 2a_ym_y + 2b_ym_y^3 - 3c_ym_y^2 - \mu_1h_y = 0 \]
\[ 2a_y + 6b_ym_y^2 - 6c_ym_y = 0 \] \hspace{1cm} (A.13)

The critical magnetic field is \( h_{yc2} = \frac{\Delta_y(8a_ymb_y - c_y\Delta_y)}{36b_y^2\mu_1} \), where \( \Delta_y = 3c_y + \sqrt{9c_y^2 - 12a_yb_y} \).