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Effective Landau theory for crossover from thermal hopping to quantum tunnelling

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Abstract
In this paper the decay rate is obtained accurately near the crossover temperature. Because of the strong fluctuations near the crossover temperature a method called the effective Landau theory beyond the Gaussian semiclassical approximation is developed. We find that in the crossover region the transition rate of the first-order transition changes much more sharply than that of the second-order one. For both cases there is no discontinuity of transition rate for any of the order derivatives.

1. Introduction

The decay of metastable states in macroscopic systems is a fundamental problem in many areas of physics, such as macroscopic quantum tunnelling in a Josephson system [1, 2], violation of baryon-lepton in the Weinberg–Salam model [3, 4], nucleation theory in first-order phase transition theory [5, 6] and more recently, magnetic quantum resonant tunnelling [7–9]. The crossover from thermal hopping to quantum tunnelling has been studied intensively. Using the functional integral approach, Affleck first demonstrated that the transition can be found between the classical region and the quantum region [10]. Larkin and Ovchimikov also suggested this and gave a formula determining the boundary of first- and second-order transitions [11, 12]. The general conditions of transitions have been analysed by Chudnovsky [13].

Quite recently an effective free energy $F(E)$ for the quantum-classical transitions of the escape rate of a spin system or other system was advanced [14–25]. In the steepest descent approximation the transition rate is given by $\Gamma \sim e^{-S_{min}/T}$, where S_{min} is the minimum of the effective 'free energy'

$$F = S_{min} = E + TS(E) \tag{1}$$

where

$$S(E) = 2(2M)^{1/2} \int_{x_2(E)}^{x_1(E)} \sqrt{U(x) - E}\, dx$$

x_1 and x_2 are the turning points for the particle oscillating inside the inverted potential $-U(x)$.

The actual dependence of $S_{min}(T)$ goes along the minimum of these two actions (sphelaron and periodic instanton) and the transition occurs at $T = T_1$. The first derivative of $S_{min}(T)$ is discontinuous at T_1, providing that the crossover from thermal hopping to quantum tunnelling is the first-order transition, otherwise second-order transition.

The second-order transitions are common, whereas the first-order ones are exotic and have to be specially looked for. Qualitatively it is clear how $U(x)$ will look. The potential should change slowly near the top and the bottom, but will be rather steep in the middle. In this case, as for the rectangular barrier, tunnelling just below the top of the barrier is unfavourable, the thermal assisted tunnelling (TAT) is suppressed, and the thermal activation competes with the quantum tunnelling directly, leading to the first-order transition.

Quantum tunnelling of the magnetization (QTM) has become a focus of interest in physics and chemistry because it can provide a signature of quantum mechanical behaviour in a macroscopic system [7–9]. At a low enough temperature, it has been demonstrated that the vector of the magnetization formed by a large number of spins in a magnetic system can coherently tunnel between the degenerate minima of magnetic energy. Theoretical suggestions have led to a number of experiments which seem to support the idea of magnetic tunnelling. Since the $Mn_{12}Ac$ complex magnetic molecule provides a more suitable model for the magnetic quantum tunnelling, extensive works have been performed to demonstrate the QTM in large spin molecules [26]. On the other hand, the $Mn_{12}Ac$ molecule is one of the very few examples which could exhibit the first-order transition [14, 15]. It has been reported that $CrNi_6$ is the first example of a high-spin molecule where tunnelling is temperature independent at low-temperature, and the transition from the classical activated behaviour to the quantum one is sharp and consistent with first-order [27].

In the second part of this paper, we derive a compact formula for the decay rate: effective Landau theory for crossover which is valid for the entire range of parameters of the interesting problems in QTM. The quantum-classical transitions of the escape rates in the dissipation systems are investigated by the effective-mode method. In the next part, we discuss the tunnelling of second-order transitions which is well known. Applying the effective-mode method in the fourth part, we show how the first-order transitions occur. The results of the application of a previous method is developed for dealing with the quasi-zero modes up to sixth-order coupling and calculating the decay rate in the crossover region which is beyond the Gaussian semiclassical approximation [28–31]. Finally we show that the crossover theory may be useful to two familiar examples of QTM.

2. Effective-Landau theory

In this paper we use an effective-mode method [17] to show first-order transition Landau theory. The Euclidean action is written as a functional integral over periodic paths where the path probability is weighted by

$$S = \int_0^{\hbar/k_BT} \mathrm{d}\tau \left[\frac{1}{2}M\dot{x}^2 + U(x)\right] + \frac{1}{2}\int_0^{\hbar/k_BT} \mathrm{d}\tau \int_0^{\hbar/k_BT} \mathrm{d}\tau' k(\tau - \tau')x(\tau)x(\tau') \qquad (2)$$

where

$$k(\tau) = \frac{k_BT}{M\hbar}\sum_{n=-\infty}^{\infty}\xi(\nu_n)\exp(\mathrm{i}\nu_n\tau)$$

$$\nu_n = 2\pi n k_B T/\hbar \qquad \text{and} \qquad \xi(\nu_n) = \gamma(\nu_n)|\nu_n|$$

is related to the frequency-dependent damping coefficient $\gamma(\nu_n)$. We use ω_R to denote the solution of the following equation $\omega_R^2 + \omega_R\gamma(\omega_R) = \omega_b^2$ where $\omega_b = \sqrt{-U''(x_b)/M}$

characterizes the width of the parabolic top of the well. In the classical limit, $1/\hbar \to \infty$, the steepest descent method is available:

$$\delta S[x(\tau)] = 0 \qquad x(0) = x(\hbar/k_BT). \tag{3}$$

The fluctuation modes about the saddle point are expanded using Ψ_n, $x = x_c(\tau) + \sum_n Y_n\Psi_n$, where Y_n are fluctuation amplitudes and Ψ_n are modes of the spectrum:

$$-\ddot{\Psi}_n + U[x_c(\tau)]\Psi_n = \omega_n^2\Psi_n \qquad \Psi(\hbar/k_BT) = \Psi(0). \tag{4}$$

According to the metastable-decay theory, the quantum tunnelling rate has the form $\Gamma = -\frac{2}{\hbar}\mathrm{Im}\,F$. Above the crossover temperature, T_c, the decay process comes from the thermal activation $\Gamma = (2/\hbar)(T_c/T)\mathrm{Im}\,F$ where $T_c = \omega_R/2\pi k_B$ [10]. In the ordinary case, the one-loop correction which results in a prefactor of the WKB leading order exponential, does not enhance the tunnelling significantly and the transition rate is dominated by the WKB leading order exponential. Near the transition point the imaginary part of the free energy has a common form:

$$\mathrm{Im}\,F = -\frac{k_BT}{2}\left(\frac{\omega_0}{\omega_R}\right)\frac{\omega_1^{(0)2}}{\Lambda}f_c \exp\left(\frac{-S_c}{\hbar}\right) \tag{5}$$

where S_c is just the WKB leading order exponent, $\frac{1}{2}$ is due to the negative mode, $\omega_n^{(0)2} = \omega_0^2 + \nu_n^2 + \nu_n\gamma(\nu_n)$, $\omega_n^{(b)2} = -\omega_b^2 + \nu_n^2 + \nu_n\gamma(\nu_n)$, $1/\Lambda$ comes from the two quasi-zero modes which need to be calculated carefully and

$$f_c[\omega_0, \omega_b] = \prod_{n=2}^{\infty}\left[\frac{\omega_n^{(0)}}{\omega_n^{(b)}}\right]^2 = \frac{\Gamma(2-(\lambda_b^+/\nu_1))\Gamma(2-(\lambda_b^-/\nu_1))}{\Gamma(2-(\lambda_0^+/\nu_1))\Gamma(2-(\lambda_0^-/\nu_1))} \tag{6}$$

where

$$\lambda_b^\pm = -\frac{\gamma(0)}{2} \pm \left[\frac{\gamma(0)^2}{4} + \omega_b^2\right] \qquad \lambda_0^\pm = -\frac{\gamma(0)}{2} \pm \left[\frac{\gamma(0)^2}{4} - \omega_0^2\right]$$

and

$$\omega_0 = \sqrt{-U''(0)/M}$$

$\Gamma(x)$ is the Gamma function.

The line of reasoning in a Gaussian semiclassical approximation is as follows. If the minima are separated by barriers the height of which is larger than $\mathrm{O}(k_BT)$, a Gaussian approximation around each minimum is applicable, and the semiclassical rate follows as a sum over these contributions. If the classical actions of these contributions differ by less than the order of $\hbar$, all must be summed, otherwise only the dominant ones must be kept. Some years ago Grabert and Weiss discussed the transition rate in the presence of dissipative effects of the environment in some detail. In addition to the zero mode near the transition point they found an unstable mode and calculated it carefully [17, 28–33]. Near the phase transition point the fluctuation modes about the saddle points include two dangerous modes which cannot be calculated by the Gaussian semiclassical approximation and it is necessary to consider higher-order couplings between the two dangerous modes [17, 28–33].

To regularize the divergent integral we have to add terms of higher-order in the amplitudes $Y_{\pm 1}$. After expanding the potential about the barrier top

$$U(x) = \Delta U - \frac{M\omega_b^2x^2}{2} + \sum_i c_ix^i \tag{7}$$

where $c_i = U^{[i]}(x = x_b)/i!$, we obtain the action

$$S[Y_n] = \frac{\hbar}{k_BT}\Delta U + \frac{1}{2}\frac{\hbar}{k_BT}M\left[\sum_{n=-\infty, n\neq\pm 1}^{\infty}\omega_n^{(b)2}Y_n^2\right] + \Delta S \tag{8}$$

where

$$\Delta S = \frac{\hbar}{k_B T}\left[\begin{array}{c}\frac{M\omega_1^{(b)2}}{2}(Y_1^2 + Y_{-1}^2) \\ +B_4(Y_1^2 + Y_{-1}^2)^2 + B_6(Y_1^2 + Y_{-1}^2)^3\end{array}\right] \tag{9}$$

with

$$B_6 = \frac{5}{2}c_6 - \frac{2c_4^2}{M\omega_3^{(b)2}} \qquad B_4 = \frac{3}{2}c_4 + \frac{9c_3^2}{2M\omega_b^2} - \frac{9c_3^2}{4M\omega_2^{(b)2}}$$

and

$$\omega_2^{(b)2} = 4\nu^2 - \omega_b^2 \simeq 3\omega_b^2.$$

Introducing the polar coordination: $\phi\cos\theta = Y_1$, $\phi\sin\theta = Y_{-1}$, we define the effective Landau function[5]

$$L = S[Y_{\pm 1}]k_B T/\hbar - \Delta U = \frac{M\omega_1^{(b)2}}{2}\phi^2 + B_4\phi^4 + B_6\phi^6 \qquad \frac{M\omega_1^{(b)2}}{2} \propto (T - T_c). \tag{10}$$

The analogy with the Landau model of phase transitions described by $F = a\phi^2 + b\phi^4 + c\phi^6$, now becomes apparent. Let us show the one-component order-parameter associated with the expansion:

$$L = \frac{\alpha}{2}\phi^2 + \frac{\beta}{4}\phi^4 + \frac{\gamma}{6}\phi^6 \qquad \alpha = M\omega_1^{(b)2} \qquad \beta = 4B_4 \qquad \gamma = 6B_6. \tag{11}$$

This is a typical O(2) global symmetry broken and ϕ is the order parameter. Because there is no spacial-dimension, the systems have no long-range order. The quasi-zero mode $\Psi_{-1} = \sqrt{2}\sin(\omega_R\tau)$ just takes place of 'soft mode' which restores symmetry and the zero mode of the Goldstone mode $\Psi_1 = \sqrt{2}\cos(\omega_R\tau)$ which reflects the freedom of phase. This is just the character of global O(2) broken symmetry.

This definition of order parameter gives the same crossover properties as before $\sqrt{(\Delta U - E)/\Delta U}$ [14]. Dimensionless temperature and energy variables $\theta = T/T_c$, $P = (\Delta U - E)/\Delta U$ have been introduced in terms of which the effective free energy near the top of the barrier ($P \gg 1$) becomes

$$F(P)/\Delta U \simeq 1 + AP + BP^2. \tag{12}$$

It is obvious that P takes the place of ϕ in this paper.

The boundary between the first-order transition and the second order one is as follows:

$$B_4 = \frac{3}{2}c_4 + \frac{9c_3^2}{2M\omega_b^2} - \frac{9c_3^2}{4M\omega_2^{(b)2}} = 0.$$

It is obvious that the dissipation may change the boundary between the first-order transition and the second-order one. B_4 is changed through the damping coefficient at frequency $\omega_2^{(b)2} = -\omega_b^2 + \nu_n^2 + \nu_n\gamma(\nu_n)$.

3. Crossover of second-order transition: $B_4 > 0$

First let us study the case of second-order transition ($B_4 > 0$).

Second-order transition occurs when the eigenvalue of the lowest mode is equal to zero $\lambda_1 = 0$ as temperature decreases, so it is defined as $T_c = \omega_R/2\pi k_B$. Above T_c, the decay

[5] The effective Landau function is just the fluctuation potential in [30].

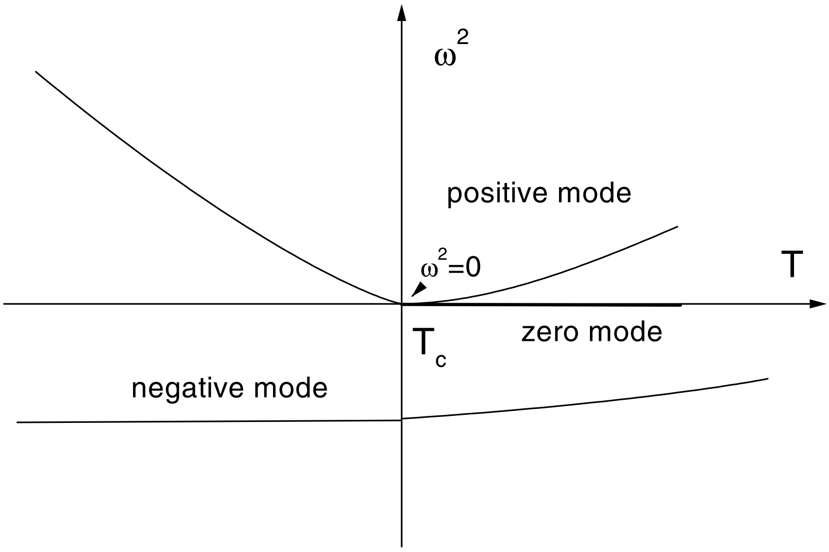

Figure 1. Modes near the crossover temperature of the second-order transition.

process is dominated by the saddle point, called a sphelaron $x = x_b$. Considering the fluctuation modes around it, we have the periodic paths near the saddle point [17, 28–33]

$$x = x_b + Y_0 + Y_{-1}\sqrt{2}\sin\frac{2\pi}{L}\tau + Y_1\sqrt{2}\cos\frac{2\pi}{L}\tau + \cdots$$
$$+Y_{-n}\sqrt{2}\sin\frac{2\pi n}{L}\tau + Y_n\sqrt{2}\cos\frac{2\pi n}{L}\tau\ldots. \qquad (13)$$

There is a mode with negative eigenvalue $\omega_0^{(b)2} = -\omega_b^2 = -U''/M$ which is the key mode giving a contribution to the imaginary part of the free energy. The eigenvalues of the two lowest positive modes are $\lambda_1 = \omega_1^{(b)2} = \omega_{-1}^{(b)2} = \nu_1^2 - \omega_R^2$. A second-order transition occurs when the eigenvalue of the lowest mode is equal to zero $\lambda_1 = 0$ as temperature decreases, so it is defined that: $T_c = \omega_R/2\pi k_B$. Near the transition point, the eigenvalue of the lowest positive mode is:

$$\lambda_1 = -2\omega_R^2\varepsilon \qquad (14)$$

where $\varepsilon = (1 - (T/T_c))$.

Below T_c, the saddle point is called a periodic instanton or thermon. Near T_c, this kind of classical periodic trajectory of thermion may be written as a Fourier series

$$x_c(\tau) = \sum_{n=0}^{\infty}[X_n\cos(\nu_n\tau) + X_{-n}\sin(\nu_n\tau)] \qquad (15)$$

The periodic paths near the saddle point are $x = x_c(\tau) + \sum_n Y_n\Psi_n = \sum_n(Y_n + X_n)\Psi_n$. We define the amplitudes into another form $Y_n' = Y_n + X_n$. There are also two dangerous modes about this saddle point near $T_0^{(2)}$: one is a quasi-zero mode which is associated with amplitude fluctuations of the periodic instantons, with the eigenvalue and eigenstates of $\omega_{-1}^{(b)2} = 4\omega_R^2\varepsilon$ and $\Psi_{-1} = \sqrt{2}\sin(\omega_R\tau)$; the other represents phase fluctuations and gives a large contribution to the partition function with $\omega_1^{(b)2} = 0$ and $\Psi_1 = \sqrt{2}\cos(\omega_R\tau)$. Figure 1 shows the modes near the crossover temperature of the second-order transition.

The action is reduced to

$$S[Y_{\pm 1}] - \frac{\hbar}{k_B T}\Delta U = \frac{\hbar M\omega_1^{(b)2}}{2k_B T}(Y_1^2 + Y_{-1}^2) + \frac{\hbar}{k_B T}B_4(Y_1^2 + Y_{-1}^2)^2 \tag{16}$$

which is equivalent to the effective Landau function

$$L = S[Y_{\pm 1}]k_B T/\hbar - \Delta U = \frac{M\omega_1^{(b)2}}{2}\phi^2 + B_4\phi^4 \qquad \frac{M\omega_1^{(b)2}}{2} \propto (T - T_c). \tag{17}$$

Above T_c there is only one minimum $\phi_c = 0$; below T_c, $\phi_c = 0$ is not stable. There is another one $\phi_p = \sqrt{M\omega_1^{(b)2}/4B_4}$ which represents the periodic instanton solution $x = \sqrt{2}\phi_p \sin(\omega_R \tau)$.

It is well known that there is a universal law in the crossover region of a second-order transition [17, 28–33]. The crossover region is defined as

$$|T - T_c| \leqslant T_c/\kappa \tag{18}$$

where $\kappa = (M\omega_R^2/2)\sqrt{1/k_B T B_4} \gg 1$. Above T_c, the transition rate is

$$\Gamma = \frac{\omega_0}{2\pi}\frac{\omega_1^{(0)2}}{\Lambda} f_c[\omega_0, \omega_b]\,\mathrm{e}^{-\Delta U/k_B T} \tag{19}$$

where $1/\Lambda = (\kappa\sqrt{\pi}/2\omega_R^2)\,\mathrm{erfc}(-\kappa\varepsilon)\exp(\kappa^2\varepsilon^2)$. Below T_c the tunnelling rate is

$$\Gamma = \frac{k_B T}{\hbar}\frac{\omega_0}{\omega_b}\frac{\omega_1^{(0)2}}{\Lambda} f_c[\omega_0, \omega_b]\,\mathrm{e}^{-\Delta U/k_B T}. \tag{20}$$

To show the universal law a quantity is defined as

$$y = \Gamma\exp(\Delta U/k_B T) \tag{21}$$

which is a function of ε but independent of the temperature T. With the quantity y, we have the universal law

$$y/y_0 = F(\xi/\xi_0) \tag{22}$$

where

$$F(\xi) = \mathrm{erfc}(\xi)\exp(\xi^2) \qquad \xi = T - T_c \qquad \xi_0 = T_c/\kappa$$

and

$$y_0 = (\omega_0/2\pi)((\omega_0^2 + \omega_b^2)/2)\sqrt{\pi/6k_B T c_4}\, f_c[\omega_0, \omega_b].$$

4. Crossover of first-order transition: $B_4 < 0$

Let us discuss the first-order transition for $B_4 < 0$ and show how it occurs.

Here phase I is the region dominated by quantum tunnelling and phase II by thermal activation. The Landau function:

$$L = \frac{\alpha}{2}\phi^2 + \frac{\beta}{4}\phi^4 + \frac{\gamma}{6}\phi^6 \qquad \alpha = M\omega_1^{(b)2} \qquad \beta = 4B_4 \qquad \gamma = 6B_6 \tag{23}$$

describes, when assuming $\beta < 0$ and $\gamma > 0$, a first-order transition between two phases denoted by I and II. The factor α changes sign at T_c. There indeed exists a phase boundary between the first- and second-order transitions, i.e. $\beta = 0$, at which the factor in front of ϕ^4 changes the sign.

From the Landau theory of first-order phase transitions, the temperature at which the two phases have the same free energy is

$$T_1 = T_c + \frac{3}{16}\frac{\beta^2}{\gamma a} \qquad a = M\omega_1^{(b)2}. \tag{24}$$

T_1 is always regarded as the crossover temperature for the first-order cases. At T_1, phase I and phase II are equally stable when the additional condition $L = 0$ is fulfilled. Below T_1, phase I becomes less stable than phase II, which exists until T_c. Above T_2 phase II does not exist until

$$T_2 = T_c + \frac{\beta^2}{4\gamma a}. \tag{25}$$

In the region $T_c < T < T_2 = T_c + (2/3M\omega_1^{(b)2})$ the two phases may coexist and the hopping process has two channels: one is above the barrier, the other below it.

4.1. Thermal hopping region: $T > T_2$

Above T_2, the decay process is dominated by the sphelaron ($\phi_c = 0$). Near $\phi_c = 0$, the quasi-zero modes have been discussed and the eigenvalue of the two lowest positive modes is: $\lambda_1 = -\omega_R^2\varepsilon$ where $\varepsilon = (1 - (T/T_c))$. We deal with the quasi-zero modes to consider the sixth term and integrate the order parameter

$$\begin{aligned}\frac{1}{\Lambda_{total}} = \frac{1}{\Lambda_{sphe}} &= \frac{1}{2\pi k_B T}\int_0^\infty \phi\,\mathrm{d}\phi \int_0^{2\pi} \mathrm{d}\theta \exp\left[-\left(\frac{\alpha}{2}\phi^2 + \frac{\beta}{4}\phi^4 + \frac{\gamma}{6}\phi^6\right)\Big/ k_B T\right] \\ &= \frac{1}{2k_B T}[B_6/k_B T]^{-1/3}\int_0^\infty \mathrm{d}t\ \exp[-(t^3 + \vartheta t^2 - 3\kappa'\varepsilon t)]\end{aligned} \tag{26}$$

where

$$\kappa' = \frac{M\omega_R^2}{3k_B T}[B_6/k_B T]^{-1/3} \qquad \vartheta = \frac{3\beta(k_B T)^{-1/6}}{2\gamma}.$$

Then above T_c, the transition rate is

$$\Gamma = \frac{\omega_0}{2\pi}\frac{\omega_1^{(0)2}}{\Lambda_{total}} f_c[\omega_0, \omega_b]\,\mathrm{e}^{-\Delta U/k_B T}. \tag{27}$$

When $|-\kappa'\varepsilon| \gg 1$, the Gaussian semiclassical approximation becomes reasonable. Hence away from the crossover region $|-\kappa'\varepsilon| \gg 1$ we can drop the $t^3 + \vartheta t^2$ term from the fluctuation potential and have

$$\frac{1}{\Lambda_{total}} = \frac{1}{2k_B T}[B_6/k_B T]^{-1/3}\int_0^\infty \mathrm{d}t\ \exp(3\kappa'\varepsilon t) = \frac{1}{2M\omega_R^2\varepsilon}. \tag{28}$$

The decay rate then goes back to the classical one

$$\Gamma = \frac{\omega_0}{2\pi}\frac{\omega_1^{(0)2}}{2M\omega_R^2\varepsilon} f_c[\omega_0, \omega_b]\,\mathrm{e}^{-\Delta U/k_B T} \to \omega_0\,\mathrm{e}^{-\Delta U/k_B T}. \tag{29}$$

4.2. Quantum tunnelling region: $T < T_c$

Below T_c, the decay process is dominated by the periodic instanton $\sqrt{2}\phi_p \sin(\omega_R\tau)$ with the order parameter $\phi_p = (-\beta + \sqrt{\beta^2 - 4\alpha\gamma})/2\alpha$. Near this solution, the quasi-zero modes change and the eigenvalue of the lowest positive mode is:

$$\lambda_1 = -4\alpha\sqrt{\alpha\gamma T_2}/\left(\beta + \sqrt{\beta^2 - 4\alpha\gamma}\right)\sqrt{\varepsilon}. \tag{30}$$

From a similar calculation, we obtain the transition rate:

$$\Gamma = \frac{\omega_0}{2\pi}\frac{\omega_1^{(0)2}}{\Lambda_{total}} f_c[\omega_0, \omega_b]\,\mathrm{e}^{-\Delta U/k_B T} \tag{31}$$

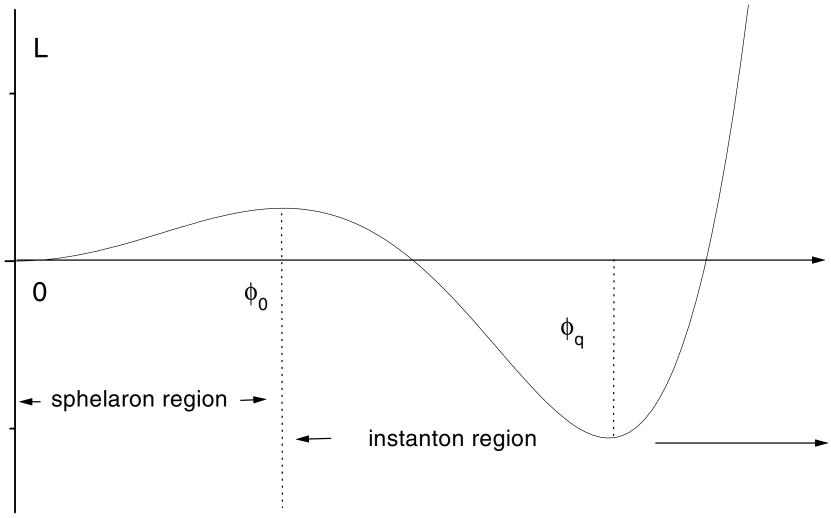

Figure 2. Instanton region $\infty > \phi > \phi_0$ and sphelaron region $0 < \phi < \phi_0$ during the first-order transition for $T_c < T < T_2$.

with

$$\frac{1}{\Lambda_{total}} = \frac{1}{\Lambda_{ins}} = \frac{1}{2k_B T}[B_6/k_B T]^{-1/3} \int_0^\infty \mathrm{d}t\ \exp[-(t^3 + \vartheta t^2 - 3\kappa'\varepsilon t)]. \tag{32}$$

Away from the crossover region $|-3\kappa'\varepsilon\phi_p| \gg \phi_p^3 + \vartheta\phi_p^2$, a Gaussian approximation around the instanton is applicable and the tunnelling rate is reduced to the standard form

$$\Gamma = \frac{1}{\Delta} \frac{\sqrt{\sum_{n\neq 0} \omega_n^{(0)2}}}{\sqrt{\sum_{n\neq 0,1} \omega_n^{(b)2}}} \mathrm{e}^{-S_c/\hbar} \tag{33}$$

where $1/\Delta = \sqrt{S_c/2\pi\hbar}(\hbar/k_B T)$. This is known as the Faddeev–Popov technique.

4.3. Coexistence region: $T_c < T < T_2$

In the coexistence region $T_c < T < T_2$, the order parameter has two solutions: one is $\phi_c = 0$, the other is

$$\phi_p = \left(-\beta + \sqrt{\beta^2 - 4\alpha\gamma}\right)/2\alpha. \tag{34}$$

The condition $\beta^2 - 4\alpha\gamma < 0$ gives the high-temperature limit $T_2 = T_c + (\beta^2/4\gamma a)$ of the instanton solution $\sqrt{2}\phi_p \sin(\omega_R\tau)$.

In this temperature region $T_c < T < T_2$, fluctuations are so strong that contributions of all points are required to be considered. Instead of two points (ϕ_c and ϕ_p) which represent the sphelaron and periodic instanton, we have two regions: the instanton region and the sphelaron region (shown in figure 2). Another point $\phi_0 = (-\beta - \sqrt{\beta^2 - 4\alpha\gamma})/2\alpha$ divides the order parameter into two regions: $\infty > \phi > \phi_0$ is the instanton region, another $0 < \phi < \phi_0$ for small one is the sphelaron region. Remember that $\phi_c = 0$ has been called the 'sphelaron'; ϕ_p takes the place of the 'periodic instanton'.

First we integrate the order parameter's fluctuations in the sphelaron region $0 < \phi < \phi_0$

$$\frac{1}{\Lambda_{sphe}} = \frac{1}{2\pi k_B T}\int_0^{\phi_0}\phi\,\mathrm{d}\phi\int_0^{2\pi}\mathrm{d}\theta\,\exp\left[-\left(\frac{\alpha}{2}\phi^2+\frac{\beta}{4}\phi^4+\frac{\gamma}{6}\phi^6\right)\Big/k_B T\right]$$
$$= \frac{1}{2k_B T}[B_6/k_B T]^{-1/3}\int_0^{t_0}\mathrm{d}t\,\exp[-(t^3+\vartheta t^2-3\kappa'\varepsilon t)]. \tag{35}$$

where $t_0 = \sqrt{\gamma/6k_B T}\phi_0^2$. Near T_c, $1/\Lambda_{sphe}$ turns into zero as $\phi_0 \to (2a\gamma/\beta)(T-T_c) \to 0$ and the decay rate from thermal hopping disappears as $(T-T_c)^2$.

For the instanton region $\infty > \phi > \phi_0$, we have:

$$\frac{1}{\Lambda_{ins}} = \frac{1}{2k_B T}[B_6/k_B T]^{-1/3}\int_{\phi_0}^{\infty}\mathrm{d}t\,\exp[-(t^3+\vartheta t^2-3\kappa'\varepsilon t)]. \tag{36}$$

At T_2, although there are two zero modes around one instanton, the transition rate from it is not zero but

$$\frac{1}{2k_B T_2}[B_6/k_B T_2]^{-1/3}\int_{\phi_0}^{\infty}\mathrm{d}t\,\exp[-(t^3+\vartheta t^2)]. \tag{37}$$

In the range $T_c < T < T_2$ it is reasonable to integrate the parameter ϕ from zero to infinity which means that the fluctuation amplitudes $Y_{\pm 1}$ are not constrained. The total hopping rate is determined by two parts as

$$\Gamma_{total} = \Gamma_{sphe} + \Gamma_{ins} \simeq \frac{1}{k_B T\hbar}\left(\frac{\omega_0}{\omega_R}\right)\frac{\omega_1^{(0)2}}{\Lambda_{total}} f_c[\omega_0, \omega_b]\,\mathrm{e}^{-\Delta U/k_B T} \tag{38}$$

where

$$\frac{1}{\Lambda_{total}} = \frac{1}{2k_B T}[B_6/k_B T]^{-1/3}\int_0^{\infty}\mathrm{d}t\,\exp[-(t^3+\vartheta t^2-3\kappa'\varepsilon t)].$$

Considering the order parameter's strong fluctuations, we have the remarkable result that there is no discontinuity in the transition rate's first derivative with temperature. From figure 3, one can see that in the crossover region the transition rate of the first-order transition changes much more sharply than that of the second-order one.

If the minima, however, are not separated by sufficiently high barriers (and the corresponding classical action differs by less than $\hbar$), to speak of a 'sphelaron' and an 'instanton' region is simply not possible, for we cannot distinguish them. While in this case the formula of the total hopping rate has no change.

Another interesting problem is the universal law in the crossover region $|-\kappa'\varepsilon| < 1$ of first-order transition. Similar to that of the second-order transition, the universal law of first-order transition can be also defined with the quantity $y = \Gamma\exp(\Delta U/k_B T)$. With y, we have the universal law

$$y/y_0 = F(\xi/\xi_0) \tag{39}$$

where

$$F(\xi) = \int_0^{\infty}\mathrm{d}t\,\exp[-(t^3+\vartheta t^2-3\xi t)] \qquad \xi = T - T_c \qquad \xi_0 = T_c/\kappa'$$

and

$$y_0 = \frac{\omega_0}{12\omega_b}(\omega_0^2+\omega_b^2)[B_6/k_B T]^{-1/3}F(0)f_c[\omega_0, \omega_b] \tag{40}$$

with

$$F(0) = \int_0^{\infty}\mathrm{d}t\,\exp[-(t^3+\vartheta t^2)]. \tag{41}$$

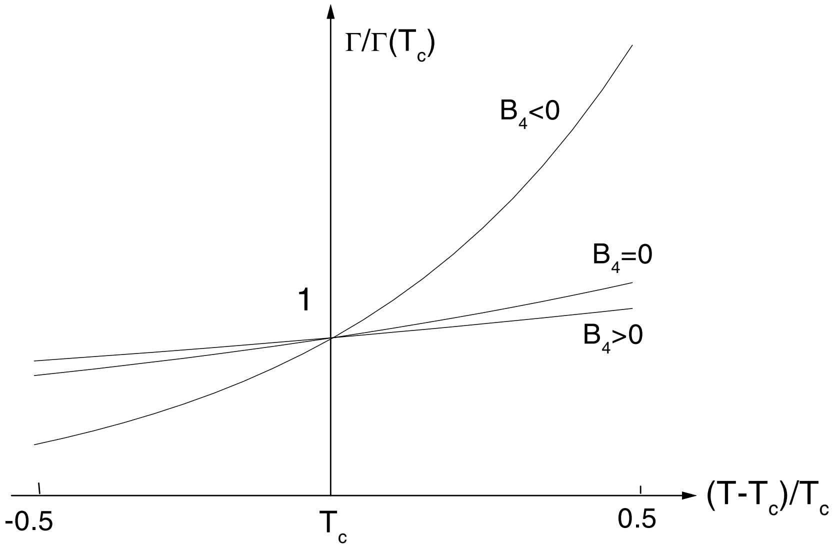

Figure 3. Transition rate near crossover temperature of different case: first-order transition $B_4 < 0$, second-order transition $B_4 > 0$ and the boundary of them $B_4 = 0$.

5. Application

The results can be easily used in $Mn_{12}Ac$ whose Hamiltonian $\mathcal{H} = -DS_z^2 - H_x S_x$ can be mapped onto a particle problem [14, 15], and the equivalent particle Hamiltonian is

$$\mathcal{H} = \frac{1}{4D}x^2 + S(S+1)D(h_x \cosh x - 1)^2 \tag{42}$$

where $h_x = H_x/2SD$. The equation $B_4 = S(S+1)Dh_x(h_x - 1/4)/2 = 0$ gives the phase boundary point $h_x = \frac{1}{4}$.

Another example is the biaxial anisotropic ferromagnetic model $\mathcal{H} = K_1 S_z^2 + K_2 S_y^2$ which describes XOY easy plane anisotropy and an easy axis along the x direction with the anisotropy constants $K_1 > K_2 > 0$. Mapped onto a particle problem, the equivalent particle Hamiltonian is

$$\mathcal{H} = \frac{1}{4K_1}\dot{x}^2 - K_2 S(S+1)\,\mathrm{sn}^2(x, \lambda) \tag{43}$$

where $\mathrm{sn}\,(\tau, \lambda)$ is the Jacobi elliptic function with modulus $\lambda = K_2/K_1$. From the equation $B_4 = K_2 S(S+1)(1-\lambda)(1-2\lambda)/2 = 0$, the phase boundary point is obtained as $\lambda = \frac{1}{2}$.

6. Conclusion

In this paper we have shown that the decay rate Γ can be accurately determined near the crossover temperature. Because the fluctuations of order parameter, ϕ, near the crossover temperature are very strong, one must use a method beyond the Gaussian semiclassical approximation. Based on this idea we develop the effective Landau theory of crossover phenomenon. The crossover temperature can be obtained easily

$$\begin{aligned} T_c &= \frac{\omega_R}{2\pi k_B} && \text{second-order transition} \\ T_1 &= T_c + \frac{B_4^2}{2B_6 M\omega_1^{(b)2}} && \text{first-order transition} \end{aligned} \tag{44}$$

where

$$B_4 = \frac{3}{2}c_4 + \frac{9c_3^2}{2M\omega_b^2} - \frac{9c_3^2}{4M\omega_2^{(b)2}} = 0$$

is the boundary between the first-order transition and the second-order one. With this theory [17, 28–33], not only is a new kind of global broken symmetry pointed out, but also the first-order transition is easily studied, especially the phase-coexistence of crossover is first proposed. After a detailed discussion, we find that there is no discontinuity of the transition rate's first derivative during the first-order transition.

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