Anomalous Magnetism of UPt$_3$: The Possibility of Oscillating Antiferromagnetic Moment

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UPt$_3$ exhibits a paradoxical behavior in its antiferromagnetism. Namely, the neutron scattering experiments surely detect the antiferromagnetic ordering below the transition temperature $T_N \sim 5K$, while no trace of the transition is seen in the NMR experiments and in the specific heat at $T_N$. For resolution of this paradox, we propose the “SDW state” which oscillates with a finite frequency larger than those of the NMR and smaller than those of the neutron scattering. For demonstrating such possibility we seek the divergent point of susceptibility, and offer a preliminary calculation of the frequency dependence of “magnetization”.

KEYWORDS: UPt$_3$, oscillating SDW state, mode-mode coupling theory of nested spin fluctuations

The heavy fermion compound UPt$_3$ has paradoxical magnetic properties. Neutron scattering experiments show the onset of antiferromagnetic order at $T_N=5K$, with the presence of ordered moment equal to 0.02$\mu_B$ at low temperatures. On the other hand, NMR experiments cannot detect any trace of the ordered moment and the specific heat has no signal of the transition at all. These experimental facts are considered to be well established. These apparently contradictory results may be understood naturally if the “antiferromagnetism” detected by neutron scattering is not a true order but fluctuates with some finite frequencies smaller than $\sim 10^{11}$/s, characteristic of the neutron scattering, but greater than $\sim 10^{7}$/s, which are those of the NMR measurements. However, the correlation length, $\xi$, is about 300Å which is so long that it is nearly equal to the resolution limit of the neutron scattering experiments. In addition, it can be observed over a wide temperature range from the “transition temperature” 5K down to zero temperature, this fact indicates that the spin fluctuation does not obey the ordinary scaling law, $\xi \propto |(T-T_c)/T_c|^{-\nu}$. We consider that this apparent paradox indicates a novel phenomenon and we propose here as its solution that the “SDW state” oscillates with a finite frequency.

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The localized spin part and the $\Pi$ is given as

$$\Pi = \Pi_0 + \Pi_1$$

where the dynamical susceptibility for the localized spin $\chi_s(Q + q, i\omega_m)$ without the coupling to the itinerant fermion is given as

$$\chi_s^{-1}(Q + q, i\omega_m) = \chi_s^{-1}(i\omega_m) - J(Q + q),$$

where $\chi_s(i\omega_m)$ is the local susceptibility and $J(Q + q)$ the RKKY-type interaction among the localized component of spins.

The fermion polarization $\Pi$ is given as $\Pi = \Pi_0 + \Pi_1$ where $\Pi_0$ is the bare electron-hole polarization part and the $\Pi_1$ is the mode-mode coupling term, which are given as

$$\Pi_0(Q + q, i\omega_m) = -g^2 T \sum_{n,p} G(p, i\epsilon_n)G(p + Q + q, i\epsilon_n + i\omega_m),$$

$$\Pi_1(Q + q, i\omega_m) = -g^4 T \sum_{n,p}^2 G_4\chi_s(Q + q', i\Omega_{m'}),$$

where $G(p, i\epsilon_n)$ is the Green function for the itinerant fermion, $\zeta$ is the renormalization amplitude, and $g$ is the coupling constant between localized spin fluctuations and fermions and is related to the fermion coherent temperature $zg \sim T_0$. The mode-mode coupling coefficient $(G_4)$ is given as

$$G_4 = G_4^s - G_4^w,$$

where

$$G_4^s = T \sum_n \sum_p G^2(p, i\epsilon_n)G(p + Q + q, i\epsilon_n + i\omega_m)$$

$$\times G(p + Q + q', i\epsilon_n + i\Omega_{m'}),$$

$$G_4^w = T \sum_n \sum_p G(p, i\epsilon_n)G(p + Q + q, i\epsilon_n + i\omega_m)$$

First we explore the possibility that the divergence of staggered susceptibility occurs at a finite frequency rather than at zero frequency. If it is possible, it strongly suggests the existence of the “order” which may oscillate with some finite frequency. The “order” we mean here is a kind of spin fluctuation which can be regarded as an antiferromagnetic state if observed for a very short time interval like the neutron scattering investigate, in other words, we may call it a “dynamical order”. We search the origin of such an exotic state for the nesting property of the Fermi surface which causes strong frequency dependence to the mode-mode coupling coefficient. The nesting property of the Fermi surface of UPt$_3$ is partly supported by the band structure calculations and the dHvA effect.$^3$ We calculate the frequency dependence of “transition temperature” using the mode-mode coupling scheme with a mode coupling coefficient which has a peculiar frequency dependence.

On the basis of the itinerant-localized duality model,$^4$ the dynamical spin susceptibility $\chi(Q + q, i\omega_m)$ for the coherent spin fluctuations is given as

$$\chi(Q + q, i\omega_m) = \chi_s(Q + q, i\omega_m) + \chi_f(Q + q, i\omega_m),$$

where $\chi_s(Q + q, i\omega_m)$ is due to the localized components of spins, $\chi_f(Q + q, i\omega_m)$ to the itinerant fermions, and $Q$ is the commensurate antiferromagnetic wave vector.

The localized spin part $\chi_s(Q + q, i\omega_m)$ is obtained using the mode-mode coupling theory as

$$\chi_s^{-1}(Q + q, i\omega_m) = \chi_s^{-1}(Q + q, i\omega_m) - \Pi(Q + q, i\omega_m),$$

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$$G_4^w = T \sum_n \sum_p G(p, i\epsilon_n)G(p + Q + q, i\epsilon_n + i\omega_m)$$
\[ G_4(p + q - q', i\epsilon_n + i\Omega_m')G(p + q' - Q, i\epsilon_n - i\Omega_m'). \]

The polarization \( \Pi_0 \), given by eq.(2a), is expanded for small \( |q| \) and small \( \omega \) as,
\[ \Pi_0(Q + q, \omega) \approx N_f^{-1} \left[ 1 - A_f q^2 + iC\omega \right], \]
where we assume \( \Pi(Q, 0) \) to be \( N_f^{-1} \) with \( N_F \) being the effective density of states. The coefficients \( A_f \) and \( C \) are given as \( A_f = a_f v_F^2 / \left[ \max(T, h) \right]^2 \) and \( C = c_f / \max(T, h) \), respectively, where \( a_f \) and \( c_f \) are numerical constants, \( v_F \) is the renormalized Fermi velocity, and \( h \) is the parameter measuring the deviation from the perfect nesting condition. The nesting property of the systems is reflected in the strong temperature dependence of the coefficient \( A_f \) and \( C \) in contrast to the systems without nesting. So the retarded counterpart of \( \chi_s(Q + q, i\omega_m) \) is expressed as
\[ \chi_s(Q + q, \omega) = \frac{N_f}{A[\kappa(\omega)^2 + q^2] - iC\omega}, \]
where \( \kappa \) is the inverse of the correlation length, \( A = A_f + a_s J a^2 N_f \) where \( a_s \) are numerical constants due to the localized spins, \( J \) is the energy scale of the RKKY-type interaction and \( a \) is the lattice constant.

Due to the nesting property, \( G_4 \) in eq.(2b) not only has a temperature dependence but also has a strong frequency dependence. Here we calculate the mode-mode coupling coefficient assuming that there exists a ‘dominant frequency’ \( i\omega_m \) as well as a wave vector \( Q \). The mode-mode coupling coefficient, which is shown in Fig. 1, is obtained after performing frequency summations as,
\[ G_4 = z^4 g^4 \sum_p \left( \text{th} \frac{\xi_p}{2T} - \text{th} \frac{\xi_{p+Q}}{2T} \right) \frac{1}{(i\omega_m + \xi_p - \xi_{p+Q})^3} \]
\[ -z^4 g^4 \sum_p \left( \cosh^{-2} \frac{\xi_p}{2T} + \cosh^{-2} \frac{\xi_{p+Q}}{2T} \right) \frac{1}{(i\omega_m + \xi_p - \xi_{p+Q})^2} \]
When the perfect nesting condition \( \xi_{p+Q} = -\xi_p \) is fulfilled, we can see that \( G_4 \), given by eq.(6), has asymptotic dependence \( 1/\omega_m^2 \) as \( |\omega_m| \to \infty \). In Fig. 2 we also show the calculated result of
frequency dependence of (6) with the model dispersion $\xi_p = -2t(\cos k_x a + \cos k_y a) - \mu$ on the square lattice for various fillings. We can see the strong $\omega_m$ dependence which drastically decreases the mode-mode coupling coefficient with asymptotically proportional to $1/\omega_m^2$. This marked decrease in mode-mode coupling coefficient reflects the nesting property and is important for our discussion. Combining the asymptotic dependence of temperature, we can take the interpolating dependence

$$G_4 \simeq g_f^4 N_F \left( \frac{\tau^2}{\omega_m^4 + \tau^2} + \frac{\omega_m^2}{\omega_m^4 + \Gamma^2} \right)$$

(6)

where $\tau \equiv \sqrt{T^2 + h^2}$ and $g_f$ is a numerical constant from fermion contribution and is given as $g_f = 35\zeta(3)/8\pi^2$ for the 3D spherical band and $\Gamma$ is the parameter which is introduced so as to reproduce the correct asymptotic form. We choose this interpolating form so as to reproduce the correct asymptotic form concerning dependence on $\omega_m$ and $T$, and not to lead to unrealistic singularities when $G_4(\imath \omega_m)$ is analytically continued onto the real axis, i.e., $\imath \omega_m \to \omega + \imath \delta$. Considering that $\chi(Q, \imath \omega_m)$ takes real values and is as even function of $\omega_m$, we may be able to use eq.(6) as an appropriate form. With this frequency dependent mode-mode coupling coefficient, we follow the conventional mode-mode coupling theory. The self-consistent equation is derived from (2a) and (2b) as

$$y(\imath \omega_m) \equiv \frac{N_F}{Aq_B^2}(\chi_{s0}^{-1}(Q,0) - \Pi_0(Q,0)) + \frac{N_F}{Aq_B^2} G_4(\imath \omega_m) \sum_q T \sum_{m'} \chi_s(Q + q, \imath \omega_{m'})$$

(7)
Then we arrange it as follows,

\[
y(i\omega) = y_0 \frac{\tau^2}{1 + \alpha \tau^2} + F g(\omega) \frac{\tau^5}{(1 + \alpha \tau^2)^3} \tag{8}
\]

\[
\times \int_0^1 dx x^2 \int_0^\infty d\omega \frac{1}{\omega} \left[ \frac{1}{\omega} - 1 \right] \left[ y(\omega) + x^2 \right]^2 + \frac{\omega^2}{\left[ 2\pi \beta (1 + \alpha \tau^2) \right]^2},
\]

where

\[
g(\omega_m) = \frac{\tau^2}{\omega_m^4 + \tau^4} + \frac{\omega_m^2}{\omega_m^4 + \Gamma^2}, \tag{9}
\]

and \(t, \tau, h, \) and \(\omega_m\) have been normalized by dividing by \(N_F^{-1} \sim \epsilon_F\), and \(F \equiv g_f N_F^4 a^4 q_B^3 / 2\pi^4 a_f^4\) is the constant which is proportional to the strength of mode-mode coupling. \(y_0 \equiv (N_F/a_f) [\chi_s^{-1} (Q, 0) - \Pi_0 (Q, 0)]\) is the measure of the distance from the magnetic phase boundary at \(T = 0\). \(\alpha \equiv 4a_s J / \pi a_f \epsilon_F\) is the normalized RKKY-type interaction and \(\beta = a_f / 2 \pi c_f\) is the numerical factor. For the 3D spherical band, \(\alpha\) and \(\beta\) are given as \(\alpha = (96\pi / 7\zeta(3)) J / \epsilon_F\) and \(\beta = 7\zeta(3) / 8\pi^4\), respectively. \(q_B\) is the physical cut off wave number and is given as \(q_B^3 = 6\pi^2 / v_0\) where \(v_0\) is the volume per magnetic atom. \(y(\omega)\) in the integral (8) is analytically continued from \(y(i\omega_m)\) onto the real axis in complex \(\omega\) plane. The self-consistent equation for \(y(i\omega)\) is written as

\[
y(i\omega_m) = y_0 \frac{\tau^2}{1 + \alpha \tau^2} + F g(\omega_m) \frac{\tau^5}{(1 + \alpha \tau^2)^3} \Xi(T), \tag{10}
\]

where

\[
\Xi(T) \equiv \int_0^1 dx x^2 \int_0^\infty d\omega \frac{1}{\omega} \left[ \frac{1}{\omega} - 1 \right] \left[ y(\omega) + x^2 \right]^2 + \frac{\omega^2}{\left[ 2\pi \beta (1 + \alpha \tau^2) \right]^2}. \tag{11}
\]

First we have to determine the parameter \(\Xi(T)\) self-consistently from eqs.(10) and (11). Then we determine the “transition temperature” \(T_N(i\omega)\) from the equation, \(\chi_s(Q, i\omega) \sim 0\), i.e. \(y(i\omega) = 0\), for finite frequency \(i\omega\) for which \(T_N(i\omega)\) is expected to be higher than \(T_N(0)\). Namely, \(T_N(i\omega)\) is determined by the condition

\[
y_0 \tau + \frac{1}{2\pi \beta} \omega + F g(\omega) \frac{\tau^4}{(1 + \alpha \tau^2)^2} \Xi(T) = 0, \tag{12}
\]

where the frequency \(i\omega\) is analytically continued on the imaginary axis from the Matsubara frequency points \(i\omega_m\)’s.

The numerical solution of eq.(12) is shown in Fig. 3. We can see that the highest “transition temperature” \(T_N\) is obtained at the finite frequency \(i\omega_0\). The abrupt disappearance of the transition temperature in the high frequency region is due to the \(\omega\)-dependent structure of eq.(12), which may not be valid in this region. A realistic physical situation may be that the “transition temperature” once reaches its maximum at some frequency, \(i\omega_0\), and then gradually approaches zero with increasing frequency \(i\omega\). Hereafter we write the highest “transition temperature” as \(T_N(i\omega_0)\).
The reason for such frequency dependence of $T_N$ is easily understood by inspecting the structure of eq.(12). The mode-mode coupling term of eq.(12), which plays the role of reducing the transition temperature $T_N$, is suppressed by the factor $g(\omega)$, given by eq.(9), with increasing frequency $\omega$ so that the effect of increasing $T_N$ overwhelms the damping term arising from the electron-hole pair excitations, the second term of eq.(12), in some region of frequency.

In Fig. 3 we also show the result with large damping (small value of $\beta$ in eq.(12)), in which case the transition temperature takes the highest value at zero frequency like the ordinary SDW case. Thus we obtain a possible structure of the magnetic susceptibility that has divergence at some finite frequency under certain conditions. We recognize this as an instability due to the emergence of the dynamical component of the SDW moment. So the state we consider below is different from the usual paramagnetic phase and in this context it can be said that it is a phase transition but the order in this state cannot be defined for a long time interval.

By a simple argument we can expect that the “SDW moment” $M_Q$ has its peak at some finite value of real frequency $\omega$. If the mode-mode coupling coefficient given by eq.(8), which corresponds to the fourth-order term in GL expansion, has its peak at $\omega = 0$, the existence of zero-frequency component of “order parameter” involved more energy than the other components of frequency. In order to confirm the above prediction we calculate the SDW “moment” on the real $\omega$-axis.

For simplicity, we retain only one frequency component of the “order parameter”, $M_Q(\omega)$, and do not take into account the coupling between different frequency components of $M_Q(\omega)$ as the first approximation. In the presence of $M_Q(\omega)$, the self-consistent mode-mode coupling equation is given as

$$y_\perp = y_0 \frac{\tau^2}{1 + \alpha \tau^2} + \frac{F}{5} g(\omega) \frac{\tau^5}{(1 + \alpha \tau^2)^3} (3 \Xi_\perp + 2 \Xi_\parallel)$$
\[ \tau^2 \frac{g(\omega)}{1 + \alpha \tau^2} M_Q(\omega)^2 \]  
\[ y_{\parallel} = y_{\perp} + M_Q(\omega) \frac{\partial y_{\perp}}{\partial M_Q(\omega)} \]  

where the subscripts \( \perp \) and \( \parallel \) denote the transverse and the longitudinal components of susceptibilities, respectively, and \( y_{\perp} = N_F/A q_B^2 \chi_{\perp}(Q, \omega) \). We should note that the damping term, linear in \( i \omega \) vanishes when the wave vector of \( \chi_{\perp}(q, \omega) \) is just the antiferromagnetic wave vector \( Q \) in the presence of \( M_Q(\omega) \). The \( \Xi_\nu(T) \), \( \nu = \perp \) or \( \parallel \) is given as,

\[ \Xi_\nu(T) = \int_0^1 dx x^2 \int_0^\infty d\omega \frac{1}{\omega_\nu/\nu + 1} \left( y_{\nu}(\omega) + x^2 \right) + \left[ \frac{\omega_\nu}{2 \pi \beta (1 + \alpha \tau^2)} \right]^2. \]

The magnetization \( M_Q(\omega) \) is determined from the equation, \( \chi_{\perp}(Q, \omega, M_Q(\omega))^{-1} = 0 \), where the subscript \( \perp \) denotes the transverse component of susceptibility, and this condition is rearranged as,

\[ y_{\perp}(Q, \omega) = y_0 \tau + \frac{D}{1 + \alpha \tau^2} \omega^2 \]

\[ + \frac{F}{5} g(\omega) \tau^4 \left( 3 \Xi_{\perp} + 2 \Xi_{\parallel} \right) + \tau g(\omega) M_Q(\omega)^2 = 0. \]

Here we have inserted the \( \omega^2 \)-term in eq.(15) which is the leading term in \( \omega \) of the susceptibility for \( q = Q \) and the coefficient \( D \) should be determined by the band structure. We can determine \( M_Q(\omega) \) by solving the coupled equations (13) \( \sim \) (15). For simplicity we approximate \( \Xi_{\parallel} \) by \( \Xi_{\perp} \). This treatment is valid at low values of \( M_Q(\omega) \). Since we are detecting the frequency dependence of the “order parameter” \( M_Q(\omega) \) near the “transition temperature”, this approximation may be sufficient for our purpose. The plot of \( M_Q(\omega) \) is shown in Fig. 4. We can see that \( M_Q(\omega) \) has large values at finite frequency than at \( \omega = 0 \). Furthermore, \( M_Q(\omega) \) vanishes for low values of \( \omega \) (the case \( t = 0.1 \) in Fig. 4). Such a situation is expected. There exists no \( M_Q(0) \), the static component, so that it is not a true SDW state. We should note that a finite value of \( M_Q(\omega) \) is only meaningful in the time interval \( \sim 2\pi/\omega \), so that the existence of \( M_Q(\omega) \) indicates that there exists a spin fluctuation mode whose “correlation length” is extremely long and it as if there was a true SDW order when observed in the time interval \( \sim 2\pi/\omega \). The specific heat will not be changed by the appearance of \( M_Q(\omega) \) unless \( M_Q(0) \) appears because the specific heat jump in the usual phase transition mainly comes from the static component of magnetization. So far, we have assumed many parameters with arbitrary values. But we believe that the real behavior of UPt3 is just in the parameter region where this novel phenomenon can occur. The Plot of \( M_Q(\omega) \) has been obtained appears by assuming that there exists no coupling between the “order parameter” with different frequencies. The SDW “order” first develops at some finite frequency. With decreasing temperature, the other frequency component, including the static (zero-frequency) component of
the SDW state, should gradually develop in general. Indeed, the ordered component $M_Q(\omega)$ can induce other components through the coupling term such as $M_Q(\omega)^3 \cdot M_Q(-3\omega)$, where $M_Q(-3\omega)$ is the admixture frequency component. However, as long as the coupling term maintains its form similar to eq.(6), the induced order $M_Q(-3\omega)$ whose frequency is located in the frequency region where $T_N(3i\omega) = 0$, results in a considerable energy cost and hardly grows. When the temperature is decreased and the ‘ordered’ state develops sufficiently, the mode-mode coupling term no longer has the frequency dependence like eq.(9). Then the dominant frequency of the SDW moment may change from the value near the “transition temperature”. Therefore, there is a possibility that the specific heat jump at $T=20\text{mK}$ may be interpreted as that at this temperature the dominant frequency of the SDW moment has changed to the static component (i.e., $\omega=0$) and it reveals its own state as the ordinary SDW state.

In conclusion, existence of an “oscillating SDW” state can possibly be verified using a probe with time scale intermediate between neutron scattering and NMR: say, $\mu$SR measurements. A possibility of the “oscillating SDW” state can be seen in a recent numerical simulations for an extended Hubbard model with pair hopping terms. In the ordered state we can determined only the qualitative level near the ‘transition temperature’ and further investigation is required to say details of this new state of magnetism.

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\[ \log(G_n) = \log(\omega_n) \]

- \( n = 0.9 \)
- \( n = 0.8 \)
- \( n = 0.7 \)
$h=0.01, \alpha=10, F=10, y_0=-0.3$

$T^n$

$\beta=1.5$

$\beta=1.0$
$h=0.01, \alpha=10.0, D=0.3$ 

$F=10.0$