LOW-ENERGY SPIN FLUCTUATIONS OF THE HEAVY-FERMION COMPOUND CeNi₂Ge₂: ORIGIN OF NON-FERMI LIQUID BEHAVIOR*

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Neutron scattering shows that non-Fermi-liquid behavior of the heavy-Fermion compound CeNi₂Ge₂ is brought about by development of lowenergy spin fluctuations with an energy scale of 0.6 meV. They appear around antiferromagnetic wave vectors $(\frac{1}{2}\frac{1}{2}0)$ and $(00\frac{3}{4})$ at low temperatures, and coexist with high-energy spin fluctuations with an energy scale of 4 meV and a modulation vector $(0.23, 0.23, \frac{1}{2})$. The energy dependence of the spin fluctuations is a peculiar character of CeNi₂Ge₂ which differs from typical heavy-Fermion compounds, and suggests importance of low-energy structures of quasiparticle bands.

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Non-Fermi-liquid (NFL) behavior has been investigated in a number of f-electron systems in recent years [1]. In heavy-Fermion systems, large mass enhancements are brought about by fluctuations of the spin degrees of freedom participating in quasiparticles. When certain spin fluctuations are slowed down, the Fermi liquid (FL) description breaks down and NFL behaviors appear. For many NFL compounds, the slowing down is related to proximity to magnetic phases or quantum critical points (QCP), $T_{\rm N} = 0$, which requires tuning by chemical substitutions or static pressures.

A typical NFL compound $CeCu_{5.9}Au_{0.1}$ has been intensively studied experimentally [2]. The NFL behaviors of this system have also been discussed theoretically from viewpoints of QCP, where an important question is

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whether the critical singularity is explained by the standard spin fluctuation theory of an itinerant character [3] or by a fixed point with a localized spin character [4]. Unfortunately, inevitable disorder effects on QCP by chemical tuning give some difficulty in the interpretation of NFL. Among many NFL compounds a few examples, such as CeNi₂Ge₂ [5] and YbRh₂Si₂ [6], are stoichiometric and provide opportunities of studying NFL behaviors or QCP in clean limits. The compound CeNi₂Ge₂ belongs to paramagnetic heavy-Fermion compounds with enhanced $C/T \simeq 350 \text{ mJ/K}^2$ mole and $T_{\rm K} \simeq 30$ K [7], implying similarity to the typical FL CeRu₂Si₂. However precise measurements at low temperatures, for example $C/T \propto \ln(T_0/T)$, revealed that CeNi₂Ge₂ shows NFL behaviors with an energy scale much smaller than $T_{\rm K}$ [5]. In this study, we directly measured magnetic excitations of CeNi₂Ge₂ using neutron scattering and have found low-energy spin fluctuations which are the origin of the NFL behaviors.

A single-crystalline sample of 2.2 cm³ in volume was grown using the isotope ⁵⁸Ni to avoid the large incoherent elastic scattering of natural Ni nuclei, which is essentially important for observing spin excitations in a low energy range. Neutron scattering experiments were performed on the triple-axis spectrometer ISSP-HER installed at JRR-3M JAERI (Tokai). The typical energy resolution using a fixed $E_{\rm f} = 3.1$ meV condition was 0.1 meV (FWHM) at the elastic position.

It has been found that a pronounced spin fluctuation grows around a wave vector $\boldsymbol{Q} = (\frac{1}{2}\frac{1}{2}1)$. Constant-Q scans at this wave vector in the temperature range 1.5 < T < 20 K are shown in Fig. 1(a), together with those at $\boldsymbol{Q} = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$. At 20 K the both data are well described by the Lorentzian form $\operatorname{Im}\chi_{\mathrm{L}}(\boldsymbol{Q}, E) = \chi_{\mathrm{L}}(\boldsymbol{Q})E\Gamma_{\boldsymbol{Q}}/(E^2 + \Gamma_{\boldsymbol{Q}}^2)$ with $\Gamma_{\boldsymbol{Q}} \simeq 4 \text{ meV} = 44 \text{ K in}$ agreement with the previous neutron scattering experiments [7,8]. The spin fluctuation of this energy scale is antiferromagnetic with a characteristic wave vector $\mathbf{k}_1 = (0.23, 0.23, \frac{1}{2})$ [8]. The same spectral shape persists down to 1.5 K for $\boldsymbol{Q} = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$. On the other hand at $\boldsymbol{Q} = (\frac{1}{2}\frac{1}{2}1)$, the spectral weight in a low energy range increases below 10 K. Since the NFL behaviors of $CeNi_2Ge_2$ show salient features in this T range, we can conclude that this low-energy spin fluctuation is relevant to NFL. It should be noted that its characteristic wave vector $\mathbf{k}_2 = (\frac{1}{2}\frac{1}{2}0)$ is near the antiferromagnetic wave vector of $Ce(Ni_{1-x}Pd_x)_2Ge_2$, indicating the proximity to an antiferromagnetic phase. Since the observed spectral shapes in Fig. 1(a) show an additional peak structure below 1.5 meV, we tried to parameterize the data by adding either another Lorentzian or a Gaussian term to $\text{Im}\chi_{L}(\boldsymbol{Q}, E)$. Only the latter form

$$\operatorname{Im}\chi(\boldsymbol{Q}, E) = \operatorname{Im}\chi_{\mathrm{L}}(\boldsymbol{Q}, E) + \delta\chi(\boldsymbol{Q})\frac{\sqrt{\pi E}}{\Gamma_{\boldsymbol{Q}}} \mathrm{e}^{-(E/\Gamma_{\boldsymbol{Q}})^{2}}, \qquad (1)$$

provides reasonable fits, which are shown by solid lines in Fig. 1(a). The energy width of the Gaussian is 0.65 meV (HWHM) at T = 1.5 K.



Fig. 1. (a) Constant-Q scans at $\mathbf{Q} = (\frac{1}{2}\frac{1}{2}1)$. Solid lines are fits to the Lorentzian with $\Gamma_{\mathbf{Q}} = 4$ meV for data at T = 20 K and the function of Eq. (1) with an additional Gaussian for data below 10 K. The inset shows constant-Q scans at $\mathbf{Q} = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$ and a fit curve to the Lorentzian. (b) Constant-E scans along a line $\mathbf{Q} = (\frac{1}{2}\frac{1}{2}L)$ taken with E = 0.75 meV. Solid and dashed lines are a guide to the eye and an orientation factor of spin fluctuations in the *ab*-plane, respectively, for data at 1.5 K.

In Fig. 1(b) we show constant-E scans taken with E = 0.75 meV along a line $\mathbf{Q} = (\frac{1}{2}\frac{1}{2}L)$ below and above 10 K. From this figure one can see that a periodic modulation with peaks at integral L, implying an antiferromagnetic correlation with the wave vector \mathbf{k}_2 , is formed only at low temperatures. Second information extracted from these data is an anisotropy of the spin fluctuation. The slow Q variation of the intensity is brought about by an orientation factor $(1 + \hat{Q}_c^2)$ of the spin fluctuation in the *ab*-plane as shown by the dashed curve in Fig. 1(b).

A number of constant-E scans were carried out at 1.5 K in a wider Q range, to check the interesting possibility of two-dimensional (2D) spin fluctuations inferred from the resistivity exponent and to find out other spin fluctuations. A typical result is shown in Fig. 2 as a contour map of intensities with E = 0.75 meV in the (HHL) scattering plane. In this figure \mathbf{k}_2 corresponds the two X points $(\frac{1}{2}\frac{1}{2}0)$ and $(\frac{1}{2}\frac{1}{2}1)$. A simple rod-type scattering along $(\frac{1}{2}\frac{1}{2}L)$ of the 2D fluctuation has not been observed. There is, instead of this, another peak structure at a reduced wave vector $\mathbf{k}_3 = (00\frac{3}{4})$, which also grows only at low temperatures. Constant-Q scans at \mathbf{k}_3 show similar spectra to those of \mathbf{k}_2 with a slightly larger energy width of the Gaussian term of 0.9 meV (HWHM), which suggests secondary importance of \mathbf{k}_3 to NFL.



Fig. 2. A contour map of constant-*E* scans taken with E = 0.75 meV in the (*HHL*) scattering plane at T = 1.5 K. In hatched area, data were not observed owing to non-magnetic noise. Antiferromagnetic spin configurations, depicted in left and right sides, illustrate low-energy spin fluctuations with wave vectors $\mathbf{k}_2 = (\frac{1}{2}\frac{1}{2}0)$ (X point) and $\mathbf{k}_3 = (00\frac{3}{4})$, respectively, assuming spins along the *a*-axis. The wave vector $\mathbf{k}_1 = (0.23, 0.23, \frac{1}{2})$ is the position where the high-energy spin-fluctuation ($\Gamma_{\mathbf{Q}} \simeq 4$ meV) shows the maximum intensity [8].

It has been elucidated that a basic characteristic of CeNi₂Ge₂ is the existence of the low and high energy scales of the spin fluctuations with different Q dependences. In stark contrast with this, spin fluctuations of standard heavy-Fermion compounds, such as CeRu₂Si₂ and CeCu₆, possess continuous energy scale Γ_{Q} of the Lorentzian Im $\chi_{\rm L}$ modulated by RKKY interactions. For NFL CeCu_{5.9}Au_{0.1} chemical tuning reduces Γ_{Q} further to zero at the antiferromagnetic wave vector. On the other hand, the NFL behavior in CeNi₂Ge₂ is caused by the formation of the second low-energy spin-fluctuations, which is the Gaussian term of Eq. (1). Although the reason why this second spin fluctuation appear is not clear, at present, we may speculate that certain fine structures of quasiparticle bands, ex. nesting, formed well below $T_{\rm K}$ lead to the low-energy structure of spin excitations.

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