

Spin fluctuations and non-Fermi-liquid behavior of CeNi₂Ge₂

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Neutron scattering shows that non-Fermi-liquid behavior of the heavy-fermion compound CeNi₂Ge₂ is brought about by the development of low-energy spin fluctuations with an energy scale of 0.6 meV. They appear around the antiferromagnetic wave vectors ($\frac{1}{2}, 0$) 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, $\frac{1}{2}$). This unusual energy dependent structure of $\text{Im}\chi(\mathbf{Q}, E)$ in \mathbf{Q} space suggests that quasiparticle bands are important.

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Non-Fermi-liquid (NFL) behavior has been investigated in an increasing number of *d*- and *f*-electron systems in recent years.^{1,2} In usual heavy-fermion systems, although strong correlation effects of *f* electrons bring about a mass renormalization m^*/m by a factor of up to a few thousands, the systems remain in Fermi-liquid (FL) states, which are typically observed as $C/T = \text{const}$ and $\rho - \rho_0 \propto T^2$ at low temperatures. The large mass enhancement originates from fluctuations of the spin degrees of freedom of the *f* electrons participating in the quasiparticles. When spin fluctuations are slowed down by certain mechanisms, the FL description breaks down, and NFL behavior appears as, for example, $C/T \propto \ln(T_0/T)$ and $\rho - \rho_0 \propto T^x$ with $x < 2$.

A mechanism of NFL behavior is critical spin fluctuations near a quantum critical point (QCP), i.e., a zero-temperature magnetic phase transition, T_N (or T_C) = 0.²⁻⁴ Observation of a QCP requires tuning of the competition between quenching of spin by the Kondo effect and interspin coupling by Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions using chemical substitutions, static pressures, or magnetic fields.⁵ Recent experimental studies on critical behavior of CeCu_{5.9}Au_{0.1} (Refs. 5,6) posed an intriguing theoretical question: Is the singularity described by the standard spin-fluctuation theories^{3,4} or a locally critical quantum phase transition?^{2,7} For chemically substituted systems, disorders inevitably affect singularities, ranging from perturbative effects to disorder-driven NFL behaviors.⁸ Experiments using stoichiometric compounds showing NFL behavior without tuning, such as CeNi₂Ge₂ (Ref. 9) and YbRh₂Si₂,¹⁰ are thus expected to clarify the QCP or other mechanisms of NFL in the clean limit.

CeNi₂Ge₂, which crystallizes in a body-centered tetragonal structure (see Fig. 1), is a paramagnetic heavy-fermion compound with enhanced $C/T \approx 350$ mJ/K² mol.¹¹ It shows Kondo behavior with a temperature scale of $T_K \approx 30$ K (Ref. 11) and has a metamagnetic behavior at $H_M \approx 42$ T.¹² For $T < 5$ K, i.e., well below T_K , CeNi₂Ge₂ exhibits NFL behavior with $C/T \propto \ln(T_0/T)$ and $\rho - \rho_0 \propto T^x$, where $1 < x < 1.5$.⁹ CeNi₂Ge₂ also displays superconductivity near the QCP (Ref. 13) which may be spin-fluctuation mediated.¹⁴

The NFL behavior has been thought to be caused by the

spin fluctuations being slowed down by a QCP of an antiferromagnetic phase, which would be one of those observed in Pd, Rh, or Cu substituted compounds.¹⁵⁻¹⁷ However, previous neutron-scattering experiments¹⁸ on single crystalline CeNi₂Ge₂ disagree with this simple interpretation. The dynamical susceptibility is well described by the standard form

$$\text{Im}\chi_L(\mathbf{Q}, E) = \chi(\mathbf{Q}) \Gamma_Q \frac{E}{E^2 + \Gamma_Q^2}, \quad (1)$$

used in the spin-fluctuation theory.³ However, the energy scale $\Gamma_Q \sim 4$ meV $\sim k_B T_K$ shows only a weak \mathbf{Q} dependence. This is in contradiction with the QCP scenario, in which Γ_Q is expected to depend strongly on \mathbf{Q} and vanish at the antiferromagnetic wave vector $\mathbf{k}_1 = (0.23, 0.23, \frac{1}{2})$ at $T = 0$.

In this communication, we present neutron-scattering measurements that reveal a second type of spin fluctuations, which are shown to be characterized by a lower-energy scale and highly relevant to the NFL behavior. The main part of the measurements was performed on the triple-axis spectrometer HER at JAERI, equipped with a PG(002) monochromator and a horizontally focusing PG(002) analyzer. The typical energy resolution using a final energy of $E_f = 3.1$ meV was 0.1 meV [full width at half maximum (FWHM)] at the elastic position. Complementary measurements at lower energies were done on the IRIS time-of-flight spectrometer at RAL, with an energy resolution of 15 μ eV (FWHM). Single crystals were grown by the Czochralski method using isotopic ⁵⁸Ni, which is important to avoid the large incoherent elastic scattering of natural Ni. Four crystals with a total volume of 2.2 cm³ were aligned together and mounted in He flow or dilution cryostats. All the data shown are converted to the dynamical susceptibility and corrected for the magnetic form factor. It is scaled to absolute units by comparison with the intensity of the incoherent scattering from a vanadium sample.

A number of constant- E scans covering an irreducible Brillouin zone were performed to search for low-energy spin fluctuations at $T = 1.6$ K. Constant- E scans at $E = 0.75$ meV in the (*HHL*) scattering plane show (see Fig. 1)

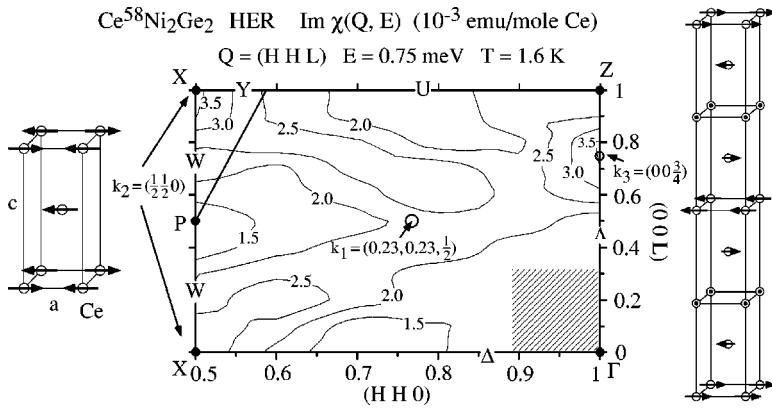


FIG. 1. A contour map of constant- E scans taken with $E = 0.75$ meV in the (HHL) scattering plane at $T = 1.6$ K. No data were taken in the hatched area, due to nonmagnetic background. Possible antiferromagnetic spin configurations, depicted on the left and right sides, illustrate low-energy spin fluctuations with wave vectors $\mathbf{k}_2 = (\frac{1}{2}, \frac{1}{2}, 0)$ (X point) and $\mathbf{k}_3 = (0, 0, \frac{3}{4})$, 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 ($\Gamma_Q \approx 4$ meV) shows maximum intensity (Ref. 18).

that there are two peak structures around $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, 1)$ and $(1, 1, \frac{3}{4})$, i.e., at reduced wave vectors of $\mathbf{k}_2 = (\frac{1}{2}, \frac{1}{2}, 0)$ and $\mathbf{k}_3 = (0, 0, \frac{3}{4})$. The wave vector \mathbf{k}_2 is the X point in the Brillouin zone, which also corresponds to $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, 0)$ in Fig. 1, where a smaller peak is seen. We note that strong intensities were observed only around \mathbf{k}_2 and \mathbf{k}_3 in the whole Brillouin zone, except for the vicinities of the Γ point, where the high background prohibited us from measuring the magnetic scattering. Possible antiferromagnetic spin configurations modulated by \mathbf{k}_2 and \mathbf{k}_3 are illustrated on the left and right sides of the contour map, respectively, assuming that the spins are parallel to the a axis.

The spin-fluctuation scattering at $E = 0.75$ meV is peaked at the wave vectors \mathbf{k}_2 and \mathbf{k}_3 , in contrast to that at ~ 4 meV, which is centered at \mathbf{k}_1 (see Fig. 1) and elongated in the $[110]$ direction.¹⁸ This feature cannot be accounted for by the spin-fluctuation theory of Ref. 3, since the product $\chi(\mathbf{Q})\Gamma_Q$ of Eq. (1) is predicted to be \mathbf{Q} independent. A constant $\chi(\mathbf{Q})\Gamma_Q$ implies that $\text{Im}\chi_L(\mathbf{Q}, E)$ peaks at a \mathbf{Q} vector where Γ_Q is minimum, which excludes the possibility to have other peaks in $\text{Im}\chi_L(\mathbf{Q}, E)$ at different energies. The archetypal heavy fermions CeRu_2Si_2 and CeCu_6 , on the other hand, are in agreement with the \mathbf{Q} independent product $\chi(\mathbf{Q})\Gamma_Q$.¹⁹ The failure of the description of CeNi_2Ge_2 by the spin-fluctuation theory will be a clue to clarify its NFL behavior.

To investigate the energy response around \mathbf{k}_2 , we performed constant- \mathbf{Q} scans at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, 1)$ and time-of-flight measurements with a locus approximately along the line $(\frac{1}{2}, \frac{1}{2}, L)$ for $0.8 < L < 2.2$. We note that \mathbf{k}_2 is close to the antiferromagnetic wave vectors of $\text{Ce}(\text{Ni}_{1-x}\text{M}_x)_2\text{Ge}_2$ with $M = \text{Pd}$ and Rh .²⁰ Figure 2(a) shows energy spectra in the temperature range $0.1 < T < 20$ K. One can see a pronounced enhancement of the low-energy spin fluctuations at low temperatures where NFL behavior in bulk properties become evident. In order to show the relevance of these low-energy spin fluctuations to the NFL behavior, we measured the magnetic-field dependence of the intensity at $E = 0.4$ meV. The inset of Fig. 2(a) shows a significant reduction in the intensity, in agreement with the recovery of the FL behavior with applied field.⁹ We conclude that the observed enhancement of the low-energy spectral weight is at the origin of the NFL behavior.

The energy spectrum at $T = 20$ K is well described by the Lorentzian form of Eq. (1) with $\Gamma_Q = 4$ meV, as reported in

Refs. 11, 18. To fit the additional peak structure below 1.5 meV at $T < 8$ K, we parametrize the data by adding an ad hoc Gaussian

$$\text{Im}\chi_G(\mathbf{Q}, E) = \delta\chi(\mathbf{Q}) \frac{\sqrt{\pi}E}{\gamma_Q} e^{-(E/\gamma_Q)^2} \quad (2)$$

to Eq. (1). The resulting fits of Eqs. (1) and (2) give an excellent description of the data [solid lines in Fig. 2(a)]. It is also possible to describe the data by two Lorentzians for $E < 1.5$ meV, but the long tail of the second Lorentzian disagrees with the data at higher energies. Below $T < 1.6$ K, the energy width [half width at half maximum (HWHM)] of the low-energy Gaussian (Lorentzian) term is 0.7 (0.45) meV. We note that the necessity to include Eq. (2) expresses the failure of the spin-fluctuation theory³ in another way.

Figure 2(b) shows that antiferromagnetic correlations, peaked at integer values of L along $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, L)$ at $E = 0.75$ meV, develop only at low temperatures. The slow \mathbf{Q} variation of the intensity can be described by an orientation factor $(1 + \hat{Q}_c^2)$ [see dashed curve in Fig. 2(b)], which implies that the spins fluctuate predominantly in the ab plane. This spin anisotropy is consistent with the antiferromagnetic structures of the Pd-doped compounds.²⁰ However it disagrees with the susceptibility measurements,¹² which indicate an Ising-like anisotropy along the c axis at $T \sim 50$ K.

To characterize the spin fluctuation at \mathbf{k}_3 , constant- E scans along $(11L)$ and $(00L)$ are shown in Fig. 3. The similar intensities between the $(11L)$ and $(00L)$ scans indicate that the spin fluctuations are isotropic. The energy spectrum at the peak position $\mathbf{Q} = (1, 1, 0.7)$ (see the inset of Fig. 3) was also parametrized using Eqs. (1) and (2) assuming $\Gamma_Q = 4$ meV for the Lorentzian. The energy width of the Gaussian is 0.9 meV (HWHM) at 1.6 K. This is slightly larger than that of \mathbf{k}_2 , suggesting that the spin fluctuations at \mathbf{k}_3 have a smaller importance for the NFL behavior.

Finally, we compare the present neutron data with other measurements. The wave-vector dependent susceptibilities $\chi(\mathbf{Q})$ at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, 1)$ (\mathbf{k}_2), $(1, 1, 0.7)$ (\mathbf{k}_3), and $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ (Ref. 23) were calculated using the Kramers-Kronig relation from $\text{Im}\chi(\mathbf{Q}, E)$, and are shown in Fig. 4(a) together with the uniform susceptibilities χ^c and χ^a . While the susceptibility $\chi(\mathbf{Q})$ at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ shows a T independent FL behavior, $\chi(\mathbf{k}_2)$ reproduces the upturn at low temperatures of χ^c and χ^a .

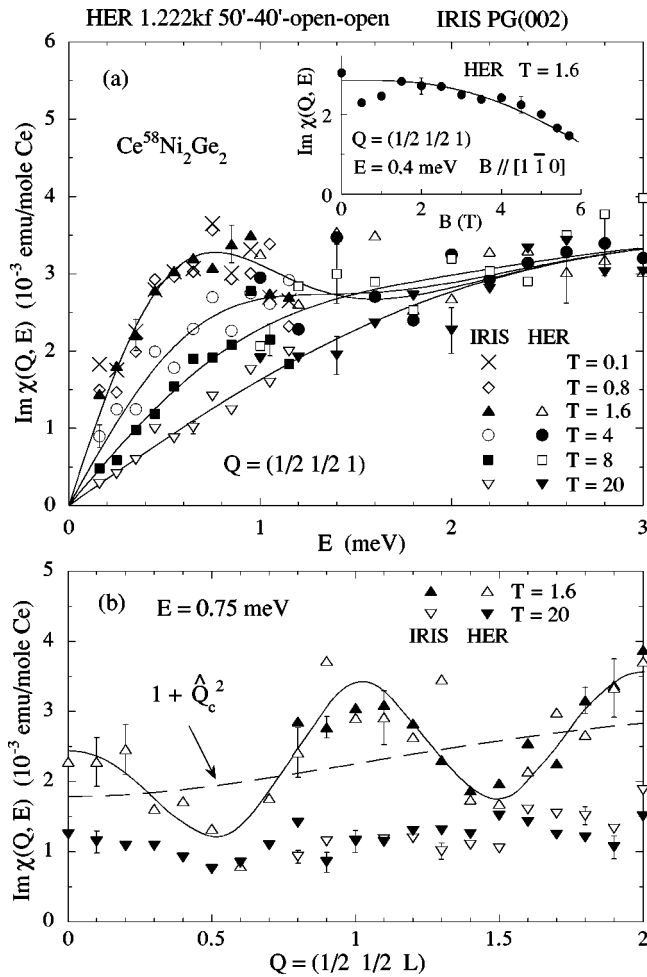


FIG. 2. (a) Constant- Q scans at $Q = (\frac{1}{2}\frac{1}{2}1)$. Solid lines are fits to the Lorentzian of Eq. (1) with $\Gamma_Q = 4$ meV with an additional Gaussian [cf. Eq. (2)] for data below 8 K. The inset shows the magnetic-field dependence of the intensity at $E = 0.4$ meV. (b) Constant- E scans along the line $Q = (\frac{1}{2}\frac{1}{2}L)$ taken with $E = 0.75$ meV. Solid line is a guide to the eye and dashed line is the orientation factor of an anisotropic spin fluctuation in the ab plane, for data at 1.6 K.

Since spin fluctuations dominate the specific heat at low temperatures, one would like to ask to what extent the observed low-energy spin fluctuations account for the NFL behavior of C/T . This can be answered semiquantitatively by using the self-consistent renormalization (SCR) theory of spin fluctuations,³ which were applied to several heavy fermions.²⁴ Since C is theoretically calculated from $\text{Im}\chi(Q, E)$ approximated by the Lorentzian form, a contribution from the 4 meV spin fluctuation can be calculated using the SCR technique. Figure 4(b) shows this part of C/T , evaluated assuming a Q independent $\Gamma_Q = 4$ meV, by a solid line together with the observed C/T .²² These show reasonable agreement above $T > 5$ K. An estimate of C/T including the low-energy spin fluctuations was obtained by replacing the Lorentzian spectral weight with the observed data.²⁵ It is plotted by a dashed curve in Fig. 4(b), showing a NFL upturn below $T < 5$ K with almost the same magnitude as the observed C/T . We conclude that the NFL behaviors

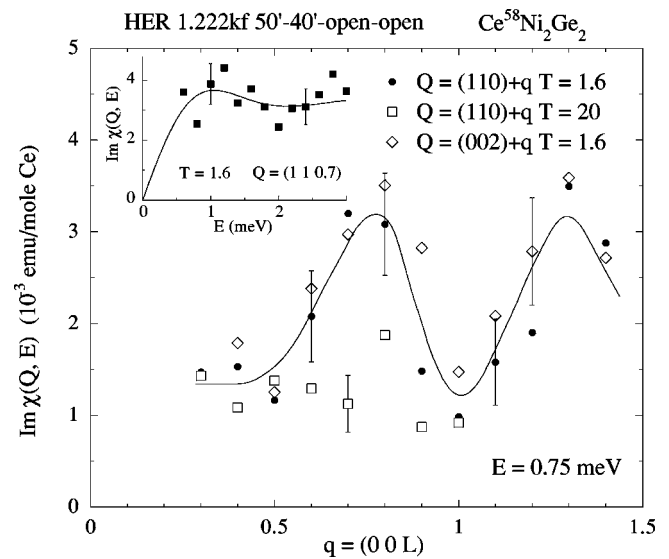


FIG. 3. Constant- E scans along lines $Q = (11L)$ and $(00L)$ taken with $E = 0.75$ meV at $T = 1.6$ and 20 K. The solid line is a guide to the eye. The inset shows a constant- Q scan at the peak position $Q = (1, 1, 0.7)$ and a fit to Eqs. (1) and (2).

observed in bulk properties are crossover effects due to the antiferromagnetic low-energy spin fluctuations. From the nondivergent behavior of $\chi(k_2)$ [see Fig. 4(a)] and $1/\gamma_{k_2}$ [see Fig. 2(a)] in the limit $T \rightarrow 0$, we also conclude that the location of CeNi_2Ge_2 is slightly off the QCP. This is supported by the fact that the E/T scaling is not observed in CeNi_2Ge_2 ; most easily seen from that the fact the peak position of the low-energy response is independent of T [see Fig. 2(a)]. In agreement with this interpretation, the recovery of the FL behavior, i.e., $C/T = \text{const}$ has been reported²² for some stoichiometric samples at the lowest temperatures $T < 0.3$ K.

An aspect of the antiferromagnetic low-energy spin fluctuations that cannot be explained by the spin-fluctuation

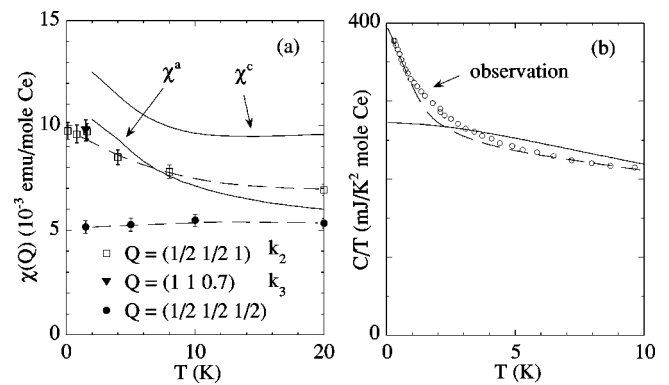


FIG. 4. (a) Temperature dependence of the wave-vector dependent susceptibility $\chi(Q)$ and uniform susceptibilities χ^c and χ^a (from Ref. 21). Error bars of $\chi(Q)$ include only statistical errors; the systematic uncertainty in the absolute normalization can be up to a factor of 1.5. Dashed lines are guides to the eye. (b) Observed specific-heat coefficient C/T (from Ref. 22) compared with model calculations using the SCR spin-fluctuation theory (Ref. 3). The solid and dashed lines are the SCR evaluations without and with the low-energy spin fluctuations, respectively.

theory³ was addressed in the itinerant-localized duality theory.²⁶ The dynamical susceptibility $\chi(\mathbf{Q}, E)$ was derived in the theory as $\chi(\mathbf{Q}, E)^{-1} = \chi_0(E)^{-1} - \Pi(\mathbf{Q}, E) - J(\mathbf{Q})$, where $\chi_0(E)$ is a local spin susceptibility and $J(\mathbf{Q})$ the Fourier transform of the RKKY interactions. The function $\Pi(\mathbf{Q}, E)$ reflects properties of the quasiparticle bands, and is usually absorbed into $J(\mathbf{Q})$ by neglecting its E dependence. The resulting $\chi(\mathbf{Q}, E)^{-1} = \chi_0(E)^{-1} - J(\mathbf{Q})$ was used as the starting assumption in the spin-fluctuation theory.³ However, the development of a particular quasiparticle band can bring about a non-negligible E dependence of $\Pi(\mathbf{Q}, E)$, which was discussed in Ref. 26 in connection with two kinds of spin fluctuations with energy scales of 5 meV and 0.2 meV in the heavy-fermion superconductor UPt₃.²⁷ We may speculate that the enhanced low-energy spin fluctuations and the deviation from the standard spin-fluctuation description of CeNi₂Ge₂ can be explained in this fashion. At present, however, other theoretical scenarios will have to be pursued.

It is interesting to compare the present results with two other compounds that are close to QCP and which have been studied in detail by single-crystal neutron scattering: CeCu_{6-x}Au_x with $x_c = 0.1$ (Ref. 6) and Ce_{1-x}La_xRu₂Si₂

with $x_c = 0.075$.²⁸ The dynamical susceptibilities of these systems can be, at least approximately, described by a single Lorentzian [cf. Eq. (1)] with $\Gamma_{\mathbf{Q}} \rightarrow 0$ at the antiferromagnetic \mathbf{Q} , in agreement with the spin-fluctuation theories.^{3,4} The essential problem of QCP is to determine the singularity, which may be different from the mean-field-type solutions of the spin-fluctuation theories. For CeCu_{6-x}Au_x,⁶ a detailed study of the divergence revealed a significant deviation from the single Lorentzian, which led them to propose an extended functional form with a nonstandard exponent of $\alpha \sim 0.75$. On the other hand, for Ce_{1-x}La_xRu₂Si₂ (Ref. 28) $\Gamma_{\mathbf{Q}}$ stays finite in the limit $T \rightarrow 0$. In this context, an exactly tuned system, e.g., CeNi_{2-x}Pd_xGe₂ (Ref. 16,17) with $x_c \sim 0.09$, is a promising candidate for studying divergent behavior of the Gaussian term of Eq. (2).

In conclusion, we have identified the low-energy spin fluctuations that lead to the NFL behavior in CeNi₂Ge₂. They are antiferromagnetic correlations around wave vectors $(\frac{1}{2}, \frac{1}{2}, 0)$ and $(0, 0, \frac{3}{4})$ with a characteristic energy scale of 0.6 meV.

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