Dynamic magnetic response in intermediate-valence CeNi

E. S. Clementyev
Russian Research Centre ''Kurchatov Institute,'' 123182 Moscow, Russia
and Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, F-91191 Gif sur Yvette Cedex, France

J.-M. Mignot
Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, F-91191 Gif sur Yvette Cedex, France

P. A. Alekseev, V. N. Lazukov, E. V. Nefeodova, and I. P. Sadikov
Russian Research Centre ''Kurchatov Institute,'' 123182 Moscow, Russia

M. Braden
Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, F-91191 Gif sur Yvette Cedex, France
and Forschungszentrum Karlsruhe, INFP, Postfach 3640, D-76021 Karlsruhe, Germany

R. Kahn
Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, F-91191 Gif sur Yvette Cedex, France

G. Lapertot
DRFMC/SPSMS, CEA/Grenoble, F-38054 Grenoble Cedex, France
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The paramagnetic spectral response of the intermediate-valence compound CeNi has been studied by inelastic neutron scattering on isotopically (^{60}Ni) enriched single-crystal and powder samples. At low temperature ($T \sim 10$ K), no magnetic intensity was found up to an energy $E \approx 15$ meV, indicating a spin-gap-like response. The absence of detectable quasielastic scattering in high-resolution time-of-flight spectra provides clear evidence for the formation of a singlet ground state due to electron correlations. The magnetic response measured at $T = 11$ K on the single crystal consists of (i) a broad structureless contribution, extending beyond 60 meV, which is a characteristic feature of valence-fluctuating materials, and (ii) two extra narrow peaks at about 18 and 34 meV, which exist for practically all **Q** vectors investigated, and whose intensities vary as a function of both the reduced **q** vector and the direction in reciprocal space. This behavior is quite unusual among intermetallic intermediate-valence compounds. It implies that the mixed-valence state in CeNi cannot be described by a single-ion Anderson model, and that magnetic correlations should be taken into account. The extra peaks are tentatively related to crystal-field interactions, which are of the same order of magnitude here as the Kondo temperature.

I. INTRODUCTION

The paramagnetic spectral response in intermediate-valence (IV) rare-earth compounds exhibits pronounced anomalies, which reflect the unusual dynamics of their magnetic excitations.[1,2] Inelastic neutron scattering (INS) is a powerful tool for investigating these phenomena because it directly probes the energy dependence and **Q** dependence of the dynamical susceptibility. It is generally assumed that, in the IV state, the hybridization of $4f$ wave functions with conduction-band states (''*c-f* mixing'') is large, causing the suppression of sharp crystal-field (CF) excitations and the appearance of a strongly damped, featureless, magnetic spectral function. In particular, the low-temperature dynamic magnetic response of intermetallic Ce-based IV compounds ($CeSn_3$,[3] $CePd_3$,[4] etc.) is typically dominated by one broad inelastic peak centered at an energy of several tens to some hundreds of millielectronvolts. When temperature is increased, the spectral weight is gradually transferred to a quasielastic line. There is growing evidence, however, that this simple description does not exhaust the variety of situations occurring in real IV systems. One striking example of a magnetic response that is not amenable to the above picture is provided by the simple binary compound CeNi.

CeNi crystallizes in the orthorhombic CrB-type structure (space group *Cmcm*). Its bulk properties denote a rather conventional IV cerium compound similar to, e.g., $CePd_3$ or $CeSn_3$. The average Ce valence changes with temperature from approximately 3.15 at $T = 80$ K to 3.07 at $T = 300$ K.[5] The magnetic susceptibility is of enhanced Pauli type at low temperature and displays a maximum around $T = 140$ K,[6] which is ascribed the characteristic energy of magnetic fluctuations (i.e., the Kondo temperature). The magnetic component of the resistivity follows a T^2 law below 40 K, and also has a maximum near $T = 150$ K.[6] The dynamical properties, however, are rather unusual. A strong softening of acoustic phonon modes near the zone boundary has recently been found,[7] in comparison with the reference compound LaNi. According to recent neutron time-of-flight (TOF) experiments,[8] the magnetic excitation spectrum at low temperature ($T = 12$ K) extends to more than 200 meV,[9] with a broad peak centered at about 50 meV, in accordance with the typical behavior expected for an intermetallic IV compound.

On the other hand, it was observed that the magnetic intensity essentially vanishes in the energy interval $4 \leqslant E \leqslant 15$ meV, suggesting the existence of a gap or pseudogap in the magnetic response. This result is quite surprising because such a response was previously encountered only in IV materials that behave as narrow-gap semiconductors at low temperature, e.g., SmB_6,[10] $Ce_3Bi_4Pt_3$,[11] or CeNiSn.[12] The only notable exception to date was reported in the work of Murani[13] on $YbAl_3$, where the magnetic signal measured at $T=5$ K was found to vanish below approximately 30 meV. Magnetic intensity reappears rather sharply above the ''gap-like'' region, with a complicated line shape denoting at least two inelastic spectral components, at energies of 27 and 42 meV. No example of such a behavior has been found so far in intermetallic Ce compounds.

From the above discussion, it appears that IV phenomena in CeNi display unconventional characteristics that may be of great significance for our understanding of this class of materials. From neutron experiments performed on single crystals, one can determine the dispersion, anisotropy, and polarization of various types of lattice and magnetic excitations. In a previous report,[7] we have presented a detailed study of the phonon dispersion curves showing that quite anomalous electron-phonon interactions exist in this material. The present paper is mainly devoted to the magnetic response of CeNi, measured on a single crystal containing isotopically enriched ^{60}Ni. The latter point is essential because the use of ^{60}Ni isotope drastically reduces the contribution from nuclear scattering, making it possible to reliably single out the weak magnetic signal. New TOF results, with an energy resolution better than 0.2 meV, are also included to demonstrate the absence of magnetic signal in the spin-gap region down to very low energies. The results confirm that the magnetic response of CeNi is indeed quite anomalous, exhibiting not only a definite spin-gap-like behavior, but also additional Q-dependent narrow peaks just above the gap, which are reminiscent of the spectral structure observed in $YbAl_3$.

II. EXPERIMENTS

The determination of the magnetic component from the INS spectra of CeNi can be considerably improved by replacing natural Ni by an isotope with a low total nuclear scattering cross section σ_s. The isotope ^{60}Ni (enrichment $\sim$99%) was selected for this purpose because it has $\sigma_s = 1.0$ b, as compared to $\sigma_s = 18.5$ b for the natural isotopic composition. The reduction of the nuclear scattering from Ni atoms by almost a factor of 20 makes it possible to extract the weak magnetic contribution from the Ce atoms (magnetic cross section σ_m of only 3.7 b) with sufficient reliability and accuracy. The starting polycrystalline materials were tested by x-ray diffraction, and found to be single phase within the limit of a few percent. The single crystal of Ce^{60}Ni used in the present experiments was grown by the Czochralski technique and had a volume of approximately 0.8 cm^3. The lattice parameters obtained from the neutron-diffraction data were $a=3.78$ Å, $b=10.54$ Å, and $c=4.36$ Å, in reasonable agreement with Ref. 14. In order to ensure comparable purity, the powder used for the TOF measurements was produced by crushing fragments of single-crystal material with a large mosaicity.

Single-crystal INS experiments were performed on the triple-axis spectrometer 2T, located on a high-flux thermal beam in the reactor hall of *Orphée* (CEA Saclay). Constant-**Q** scans were measured at $T=11$ K for fixed final energies $E_f=30.5$ meV ($k_f=3.83$ Å^{-1}; energy range $0 \leqslant \hbar\omega \leqslant 60$ meV) and $E_f=14.7$ meV ($k_f=2.66$ Å^{-1}; $0 \leqslant \hbar\omega \leqslant 25$ meV). A pyrolytic-graphite, PG(002), analyzer was used in combination with either a Cu(111) or a PG(002) monochromator. In both cases, a PG filter was placed after the sample to suppress higher-order contaminations. The energy resolutions [full width at half maximum (FWHM)] at zero energy transfer were of order 2.5 and 0.9 meV, respectively. Let us mention that, because both Ce and ^{60}Ni have low neutron absorption cross sections ($\sigma_a=0.63$ b for Ce and 2.9 b for ^{60}Ni), leading to an estimated transmission of more than 95% for $E_i=60$ meV, absorption corrections were considered unnecessary.

TOF measurements were carried out on the spectrometer MIBEMOL, located on the cold neutron guide G6. The incident energy was fixed at $E_i=3.27$ meV by means of a six-chopper system, providing a resolution of 0.16 meV (FWHM) at zero energy transfer. Measurements were done at different temperatures comprised between 30 and 120 K.

III. RESULTS

The main purpose of the present single-crystal study is to determine the **Q** dependence of the anomalous low-temperature spectral response revealed by the previous powder experiments. In particular, we are interested in the possible existence of dispersive excitations which might have been overlooked, owing to **Q**-space averaging, in the TOF results. Measurements as a function of the reduced **q** vector have thus been performed along the three main crystallographic axes [100], [010], and [001]. To test the anisotropy and polarization of the magnetic response, we have also recorded spectra corresponding to the same **q** vector in different Brillouin zones, in particular for different zone centers (Γ points).

Because the magnetic cross section is weak and the response is spread over a very broad energy range, a careful subtraction of the phonon part of the signal is prerequisite, despite the use of ^{60}Ni isotope, to extract the weak magnetic signal accurately. The method is based on the different Q dependences of the magnetic and phonon contributions, as in the standard treatment of TOF spectra, but its application is different here because the measurements are performed on a single crystal. Let us first emphasize that, from the results of Ref. 8, the phonon density of states measured on polycrystalline Ce^{60}Ni has an energy cutoff at about 24 meV. Indeed, our data confirm that, in the energy range $E>22$ meV, the magnetic contribution dominates even for rather large momentum transfers $Q>3-4$ Å^{-1}, whereas at lower energies the nonmagnetic component becomes significant. The latter contribution, which arises mainly from single-phonon processes, should vary like Q^2, whereas the former is expected to be proportional to the squared Ce^{3+} magnetic form factor $f_M^2(Q)$, as derived from polarized-neutron results.[15] Their separation can thus be achieved by combining data measured

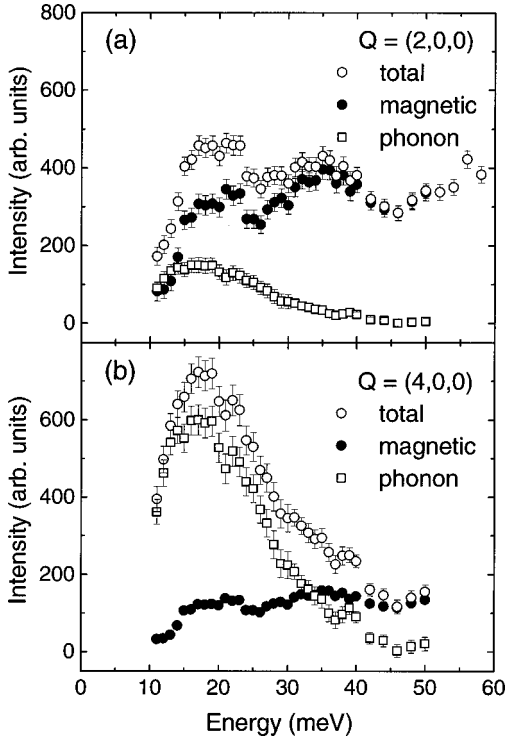

FIG. 1. Background-corrected energy spectra of single-crystal $Ce^{60}Ni$ at $T=11\,K$ for two scattering vectors at equivalent zone centers: (a) $\mathbf{Q}=(2,0,0)$, (b) $\mathbf{Q}=(4,0,0)$. ○: experimental data; ●: magnetic component; □: phonon component (for the separation procedure, see text).

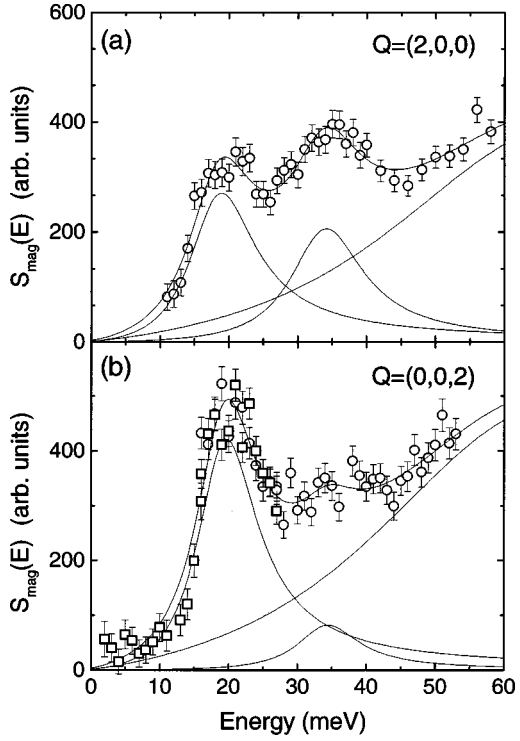

FIG. 2. Magnetic spectral response of single-crystal $Ce^{60}Ni$ at $T=11\,K$ for two different scattering vectors: (a) $\mathbf{Q}=(2,0,0)$, (b) $\mathbf{Q}=(0,0,2)$. Symbols: experimental data [○ $E_f=30.5\,meV$, Cu(111) monochromator; □ $E_f=14.7\,meV$, PG(002) monochromator]. Lines are fits of the magnetic spectra by three inelastic Lorentzian peaks (see text). Magnetic contributions have been normalized to $Q=2.9\,Å^{-1}$ [momentum transfer for $\mathbf{Q}=(0,0,2)$] by assuming a Ce^{3+} magnetic form factor.

at equivalent reciprocal-lattice points in different Brillouin zones, which have very different momentum transfers, provided their phonon structure factors are identical. This procedure is correct so long as the dependence of the energy resolution on the momentum transfer remains small, which was checked to be the case here. An example of the decomposition is shown in Fig. 1 for $\mathbf{Q}=(2,0,0)$ and (4,0,0). It should be emphasized that the large difference in the momentum transfers (3.4 and 6.8 $Å^{-1}$) provides an excellent ''contrast'' to separate the two types of components.[16] Qualitatively, one sees that, at low Q values ($Q<3\,Å^{-1}$), the magnetic contribution dominates as soon as the energy exceeds 15 meV. The residual phonon intensity that is observed above the energy cutoff may be due to multiphonon processes and/or spurious scattering from the Al sample holder (mass $m\approx 1$ g).

The magnetic excitation spectra measured at $T=11\,K$ for two different zone centers, $\mathbf{Q}=(2,0,0)$ and $\mathbf{Q}=(0,0,2)$, are plotted in Fig. 2. The intensities have been normalized to a momentum transfer $Q=2.9\,Å^{-1}$ [corresponding to $\mathbf{Q}=(0,0,2)$] using the magnetic form factor of Ce^{3+}. In Fig. 2(b), data points obtained with a better energy resolution ($E_f=14.7\,meV$) have been included (open squares) to reveal the intensity drop at low energies, which is otherwise obscured by incoherent elastic scattering. For $\mathbf{Q}=(0,4,0)$, the spectrum was found to be similar to that obtained for $\mathbf{Q}=(0,0,2)$. In all cases, two narrow peaks are observed on the low-energy side ($E<40\,meV$) of a broad contribution which seems to have its maximum at about 50–60 meV. Furthermore, essentially no significant magnetic intensity exists below 10 meV. A good fit to the data is obtained using the standard expression

$$S_{\mathrm{mag}}(E,T)\propto \frac{E}{1-\exp(-E/k_BT)} \times\sum_{i=1}^{3}\chi_i'(T)\frac{\Gamma_i(T)/2}{[\Gamma_i(T)/2]^2+(E-E_i)^2}, \quad (1)$$

consisting of three Lorentzian spectral components, with excitation energies $\hbar\omega$ of about 18, 34, and 46 meV, and half widths $\Gamma/2$ of about 4–5 meV for the former two peaks, and 24 meV for the latter one. As evidenced by Fig. 2, the low-energy components of the spectra are anisotropic: the peak at 18 meV is rather strong for $\mathbf{Q}=(0,0,2)$ and (0, 4, 0), and less pronounced for $\mathbf{Q}=(2,0,0)$. On the contrary, the excitation at 34 meV is stronger for $\mathbf{Q}=(2,0,0)$, and weaker for the other zone centers.

Salient features of the Q dependence of the magnetic signal are summarized in Fig. 3. The dispersion of the lower two excitations was found to be weak along the main symmetry directions [see Fig. 3(b) for $\mathbf{Q}=(0,\eta,0)$], but the *intensities* of the peaks at 18 and 34 meV are clearly modulated along b^* [Fig. 3(c)]. Significant intensity variations were also observed along the other directions. In particular, for $\mathbf{Q}=(0,0,\zeta)$, the intensity of the 34-meV peak appears to be stronger at the Z zone-boundary point, $\mathbf{Q}=(0,0,2.5)$, than at the zone center, $\mathbf{Q}=(0,0,2)$. The 18-meV peak also ex-

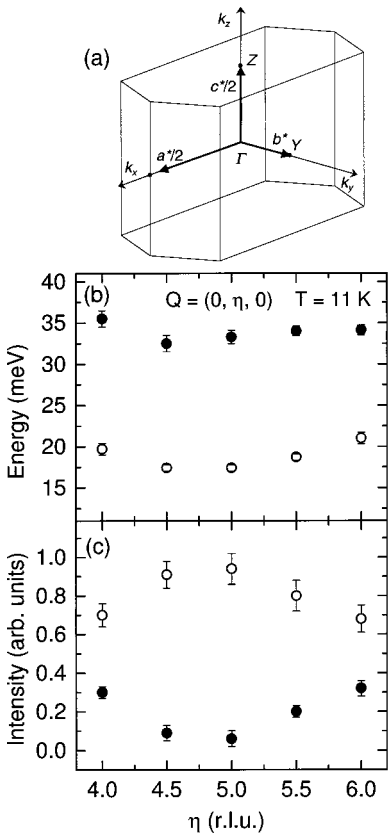

FIG. 3. **Q** dependence of the magnetic spectral components in CeNi at $T=11$ K. (a) schematic representation of the Brillouin zone and points of symmetry in the notation of C. J. Bradley and A. P. Cracknell, *The Mathematical Theory of Symmetry in Solids* (Clarendon, Oxford, 1972), p. 98; (b) excitation energies, and (c) corresponding integrated intensities of the low-energy magnetic excitations ($\hbar\omega \approx 18$ and 34 meV) for $\mathbf{Q}=(0,\eta,0)$; the values $\eta=4$ and $\eta=6$ correspond to zone centers, whereas $\eta=5$ is on the zone boundary.

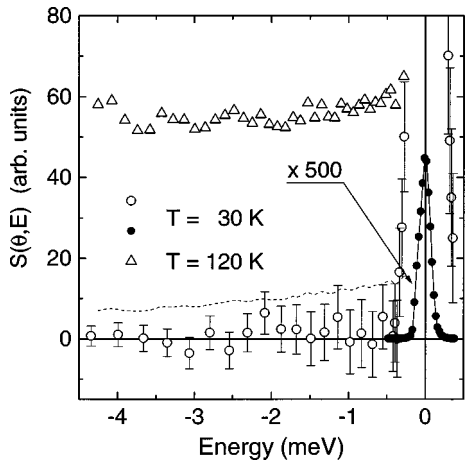

FIG. 4. Time-of-flight (background-corrected) energy spectra of polycrystalline $Ce^{60}Ni$ at $T=30$ K (○, ●) and 120 K (△), measured with an incident neutron energy $E_i=3.27$ meV (detector angles $5° < 2\theta < 70°$). Dashed line: calculated signal at $T=30$ K assuming the same spectral function as at 120 K (effect of the detailed-balance factor).

hibits a **q**-dependent intensity, but a quantitative determination for all directions was not possible because of difficulties with the phonon subtraction procedure. Finally, the data for the broader peak cannot be simply interpreted at the present stage because only the low-energy tail of the signal ($E \leqslant 60$ meV) was accessible with the existing experimental conditions.

A few measurements have been performed as a function of temperature. There is clear indication that the low-energy excitations persist up to about 100–150 K, but a more detailed analysis of their intensities is hampered by the temperature dependence of the broad high-energy component. Experiments on a hot source would be useful to clarify this point.

The TOF spectra measured on MIBEMOL with a high energy resolution confirm that the magnetic signal definitely vanishes below 10 meV. The data plotted in Fig. 4 have been obtained for a polycrystalline sample of $Ce^{60}Ni$ with a mass of about 20 g. Our analysis is restricted to the energy-gain part of the spectra (negative energy transfers), because the energy-loss side contains a contamination around 0.3 meV. Whereas a rather large intensity, mainly magnetic in origin, exists at $T=120$ K between -4 and -0.5 meV, this signal disappears almost completely at $T=30$ K. Indeed, the integrated intensity found experimentally in this energy interval represents less than 10% of that estimated from the 120 K spectrum: the dashed line in the figure represents the intensity change expected from the detailed-balance factor alone (i.e., assuming the spectral function to be unchanged). It is obvious that this factor cannot account for the experimental temperature dependence. We are thus led to the conclusion that most of the spectral weight has been transferred to higher energies, in agreement with the previous TOF results,[9] and that no significant in-gap intensity (in particular no quasielastic scattering) remains at the lowest temperature. Consistent results have also been obtained using a cold-source triple-axis spectrometer operated in the constant-Q mode for a fixed final energy of 14.7 meV (energy resolution of 0.9 meV).

IV. DISCUSSION

The first salient result of the present work is the confirmation, with a greatly improved experimental resolution, that the inelastic magnetic signal in CeNi at low temperature essentially vanishes below 15 meV (at least down to 0.2 meV), giving rise to a spin-gap-like spectral response. It is important to note that no quasielastic line is observed with an energy resolution of better than 0.2 meV, even though the typical spin-fluctuation energy can be estimated from the bulk measurements to be of the order of 7–15 meV. Therefore the ground state cannot be a Kramers doublet, as would be expected for Ce^{3+} in a low-symmetry CF. We ascribe this behavior to the formation of a nondegenerate many-body eigenstate due to electronic correlations. Whereas the predominantly inelastic character of the magnetic response function at low enough temperature had been previously reported for other Ce-based intermetallic compounds, here the complete suppression of the quasielastic contribution is experimentally established.

For energies larger than the gap, the general structure of the magnetic response of single-crystalline CeNi is consistent with the earlier powder results: appreciable magnetic

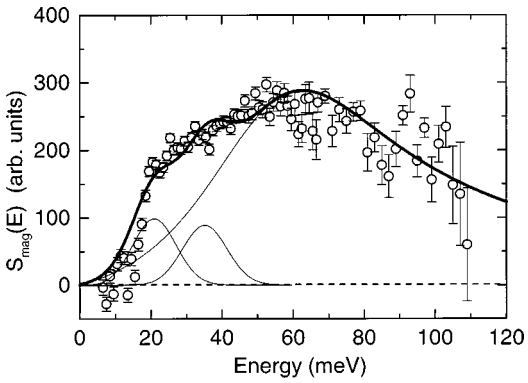

FIG. 5. Magnetic spectral response of polycrystalline $Ce^{60}Ni$ at $T=12$ K (time-of-flight data from Ref. 8). Lines: fit by three inelastic Lorentzian peaks (see text).

intensity appears above ~15 meV and a very broad peak, with its maximum located above 50 meV, is observed for all **Q** values. As noted above, this is indeed a rather common feature among IV cerium compounds at low temperature. For instance, it was shown in Ref. 4 that the magnetic excitation spectrum of $CePd_3$ at $T=10$ K can be fitted by a single Lorentzian with $\hbar\omega=53$ meV and $\Gamma/2=38$ meV, comparable to our values for CeNi. This type of behavior is consistent with the results obtained by various theories for a *single-ion* Anderson model using the noncrossing approximation.[17,18]

In the present case, however, additional narrow lines are observed on the low-energy side of the above broad peak. Furthermore, we have found that the magnetic spectral response in CeNi is **q** dependent and anisotropic (i.e., polarization dependent). Indeed, the peculiar shape of the powder spectrum reported in Ref. 8, with a steep rise of the intensity just above the upper limit of the gap, can now be traced back to the reciprocal-space averaging of the narrow spectral components at 18 and 34 meV. This is illustrated in Fig. 5, where the TOF data are well reproduced by a phenomenological fit using the same three spectral features as for the single crystal (the energy positions, as well as the half width of the broad Lorentzian were kept constant). Physically, the observed **q** dependence indicates that single-ion processes alone cannot account for the magnetic excitation spectrum of CeNi at low temperature, and that coherence and/or magnetic correlation effects should be taken into account. This assumption is supported by the previous observation that the gaplike behavior in the inelastic response disappears when 20% of the Ce is substituted by La.[8] On the other hand, the quasielastic spectral response observed in Ref. 9 at $T=200$ K suggests that a single-ion behavior is practically recovered at this temperature. It is worthwhile to mention, in this connection, that the electronic component of the thermal expansion coefficient $\alpha(T)$ varies nonmonotonically as a function of the lattice constant in solid solutions containing either La (lattice expansion) or Y (lattice compression): the peak which exists at $T=100$ K in pure CeNi, is strongly depressed and shifted to lower temperatures in both types of alloys ($Ce_{0.8}La_{0.2}Ni$ and $Ce_{0.8}Y_{0.2}Ni$),[19] again indicating that coherence effects occur in pure CeNi.

To describe such a situation, one can start from a periodic Anderson model, in which *sd-f* hybridization takes place coherently at each Ce site. It is well known that this Hamiltonian eventually produces a ''hybridization gap'' in the electron density of states. In the case of the orthorhombic compound CeNiSn, Ikeda and Miyake[20] have further shown that, owing to the **q** dependence of the hybridization matrix elements, this gap can be strongly anisotropic, and the results of their model calculation seem to account for a number of anomalous physical properties of the material. Experimentally, CeNiSn has been reported to exhibit an anisotropic and **q**-dependent magnetic excitation spectrum,[12] with a spin gap ($E_g\sim 1$ meV) and two peaks at $E=2$ and 4 meV. These excitations, which have attracted considerable attention, have been claimed to reflect the coherent origin of the ground state. In view of the similarities with the situation in CeNi, one might be tempted to apply a similar description to the latter compound. However, the underlying physics is basically that of a ''Kondo insulator'' (the existence of a pseudogap in the magnetic response is associated with a semiconducting character of the electron density of states), which certainly does not correspond to the case of CeNi, and possibly not even to that of CeNiSn which is now believed to stay metallic at $T=0$. Furthermore, the existence of narrow extra peaks in the magnetic response of CeNi provides some evidence that other mechanisms, in particular the crystal field and electron-phonon interactions, might play a part, in addition to the usual *sd-f* hybridization.

In trivalent Ce compounds with localized $4f$ states and a point-group symmetry lower than cubic, the CF interaction splits the ground-state multiplet (total angular momentum $J=\frac{5}{2}$) into three Kramers doublets, which produce narrow excitations in the neutron scattering spectra. In the IV regime, these excitations are normally smeared out by magnetic fluctuations. However, the idea that the CF might play a role in the formation of the excitation spectra in IV materials, even in the case of relatively high fluctuation temperatures, has been substantiated by the observation of several peaks in the magnetic spectra of $YbPd_2Si_2$ and $YbCu_2Si_2$.[21] For CeNi, the splitting that would exist without the valence fluctuations can be estimated in two different ways. First, one can extrapolate the energy positions of the CF levels determined experimentally in the solid solutions $Ce_{1-x}La_xNi$ ($x=0.5,0.8$),[19] in which the Ce $4f$ state is trivalent and localized (Kondo regime), by taking into account the change in the lattice parameter. The starting energy values are 0-7-15 meV for $x=0.8$ and 0-11-20 meV for $x=0.5$. Alternatively, one can calculate the Ce^{3+} energy level scheme by using the parameters of the CF potential derived from the measurements of CF effects of other rare-earth impurities in the CeNi matrix.[19] Both methods were found to give consistent results, and the ''nominal'' CF scheme of Ce^{3+} in pure CeNi is thus estimated to be approximately 0-14-25 meV.

From this analysis, it appears that the CF interaction might account qualitatively for the extra peaks observed near 18 and 34 meV. However, the application of such a picture to the present system poses several problems. First, the absence of quasielastic signal (see above) implies a drastic change in the nature of the ground state, incompatible with the assumption of a normal CF Kramers doublet. Second, the peaks measured in IV CeNi are significantly narrower ($\Gamma/2=4$ meV) than the CF excitation observed in the Kondo compound $Ce_{0.5}La_{0.5}Ni$ ($\Gamma/2=7$ meV).[8] This seems to con-

tradict the general observation that, in unstable *f*-electron systems, CF excitations gradually broaden in the neutron spectra as the hybridization energy increases. Finally, as already noted, the modulation of the intensities in reciprocal space indicates that correlations exist from site to site, even though no long-range magnetic order is observed.

It is important to keep in mind that, in CeNi (possibly also in $YbCu_2Si_2$), the spin fluctuation (k_BT_K) and CF (Δ_{CF}) energies are of comparable magnitude. The interplay between spin fluctuations and CF excitations was recently investigated in Ref. 22, with emphasis on the case when the condition $k_BT_K \ll \Delta_{CF}$ is *not* fulfilled. The theory considers spinon (fermion) excitations in a resonance-valence-bond (RVB) spin liquid, coupled by Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interactions. In the specific case of CeNiSn, a pronounced minimum is predicted to occur in the spin density of states near the spinon Fermi level, leading to an anisotropic pseudogap in the magnetic spectral response.[22] Various low-temperature properties (specific heat, magnetic susceptibility, thermal expansion, and magnetostriction) are also explained by assuming that the thermodynamics is dominated by the spin-liquid-type excitations.[23] At the same time, because charge and spin degrees of freedom are decoupled in the model, the charge carrier spectrum can be gapless, and the system retain a metallic conductivity as observed in the best CeNiSn single crystals.

In CeNi, the metallic character is unequivocal, and a number of spectral features reported in the present work closely resemble those of CeNiSn: existence of a low-energy gap at low temperature; pronounced narrow peaks for well defined momentum transfers; anisotropic **Q** dependence. Furthermore, the basic assumption of the model is also realized in CeNi ($\Delta_{CF} \lesssim k_BT_K$), despite the fact that both the Kondo temperature and the CF splitting are about one order of magnitude larger than in CeNiSn. Therefore it seems that the above approach could also be applied to the case of CeNi.

In principle, electron-phonon interactions are another possible source of anomalies in the magnetic excitation spectra. It was already mentioned that the cutoff of the phonon spectrum in CeNi is situated at about 24 meV, close to the energy of the extra magnetic components. In recent INS experiments, it has been shown that the phonon dispersion curves in CeNi exhibit considerable softening in comparison with the nonmagnetic reference compound LaNi.[7] This strongly suggests that anomalous electron-phonon interactions (coupling of charge fluctuations with lattice vibrations) exist in CeNi. Whereas it is likely that this mechanism could produce anomalies in the magnetic excitation spectra, the nature and magnitude of these anomalies has not yet been determined theoretically for IV compounds.

V. CONCLUSION

In summary, the present work provides novel experimental information on the dynamic magnetic response of the IV compound CeNi. The low-energy powder data clearly indicate that the ground state is a nonmagnetic singlet. The existence of a spin-gap-like response ($E_g \sim 15$ meV) in this metallic compound is confirmed by the single-crystal results. Above the gap energy, the structure of the excitation spectrum is rather complex. Several components could be singled out, in particular two relatively narrow peaks at energies of 18 and 34 meV, superimposed on the usual broad inelastic Lorentzian, which have no counterpart in any of the Ce IV compounds studied previously. These anomalies may be due to modified CF effects or resonant electron-phonon interactions, both of which have energy scales comparable to the Kondo temperature of the material.

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