Dispersive Magnetic Density Fluctuations in Ni_3Ga

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(Received 5 February 1988)

We report the first successful measurements of the magnetic fluctuation, or paramagnon, spectrum of an exchange-enhanced paramagnet. The spectrum measured in pure specimens of Ni_3Ga by inelastic neutron scattering can be described by a spin-density relaxation rate $\Gamma(\mathbf{q})$ which vanishes at small wave vectors and is strongly dispersive. The experimental findings are analyzed in terms of the theory of a Fermi liquid in the collisionless quasiparticle regime.

PACS numbers: 75.30.Et

Low-energy excitations of strongly interacting itinerant electron systems may be expected to be reflected particularly strongly in the spectrum of *magnetic* fluctuations, or in the imaginary part of the magnetic response function $\tilde{\chi}(\mathbf{q},\omega)$, as a function of frequency ω and wave vector $\mathbf{q}$, probed by inelastic neutron scattering. The low-frequency spectrum in the paramagnetic state (or in a weakly ordered state for $\mathbf{q}$ far away from the ordering wave vector) should be diffusive in nature, characteristic of that of strongly damped or overdamped fluctuations.[1-3] When this low-frequency spectrum can be characterized in terms of a single relaxation rate $\Gamma(\mathbf{q})$ which is appreciably suppressed from its value in a noninteracting system by particle interactions, a mathematically tractable relationship between some low-temperature properties and the magnetic fluctuation spectrum may exist.[4-14] This relationship has *some formal* similarities to that between the normal-mode frequencies and the entropy, for example, in the conventional lattice vibrations problem.

The bulk of the information on $\tilde{\chi}(\mathbf{q},\omega)$ has thus far been obtained in magnetically ordered materials (both below T_c, where magnetic fluctuations of propagating character exist, and above T_c) and in a number of nonmagnetic (or weakly magnetic) rare-earth and actinide compounds.[15] In the heavy electron members of the latter class of materials such as UPt_3 the nature of $\tilde{\chi}(\mathbf{q},\omega)$ at very low ω remains uncertain, especially at low $\mathbf{q}$ where the characteristic behavior anticipated in conventional Fermi-liquid theory has not yet been reported[16] despite the explicit observation of a well defined quasiparticle Fermi surface.[17]

Thus far measurements of the spectral composition of magnetic fluctuations in exchange-enhanced *paramagnetic* transition metals such as Pd, Ni_3Ga, and $TiBe_2$, in which the spin-orbit interaction is weak and the quasiparticle component of $\tilde{\chi}(\mathbf{q},\omega)$ at low $\mathbf{q}$ and ω is expected to be most readily identified, have not been presented, despite intensive experimental effort on this problem for many years.[18,19]

Here we report a first investigation of $\tilde{\chi}(\mathbf{q},\omega)$ a low $\mathbf{q}$ and ω in one of these paramagnetic systems, namely Ni_3Ga, carried out by means of a procedure which we have developed to investigate the magnetic fluctuation spectrum of the low-temperature ferromagnets Ni_3Al[19] and $ZrZn_2$.[20] In the latter systems the inelastic paramagnetic scattering cross section, though very weak, is typically an order of magnitude greater than in Ni_3Ga.

Ni_3Ga has been selected for this study because it crystallizes in a simple cubic (Cu_3Au) structure, it can be purified to the high level required, for example, for de Haas–van Alphen experiments,[21] and it exhibits a strongly temperature-dependent susceptibility which is approximately 1 order of magnitude greater than in Pd and 3 times larger than in UPt_3 at low temperatures.[22] Moreover, the quasiparticle band structure of this system has been extensively characterized[21] and its observed simplicity suggests that model calculations of $\tilde{\chi}(\mathbf{q},\omega)$ for detailed comparison with experiment are practical with existing band-structure algorithms.

Samples were prepared from a high-purity melt of zone refined starting materials by a radio frequency heating technique.[21] To reduce neutron scattering in the long-wavelength regime of interest, arising from chemical and structural disorder, attention was focused on attaining purity, homogeneity, and stoichiometry over the large, 5-cm^3 dimension required for optimum signal-to-noise ratios. The best specimens fulfilling these requirements were polycrystalline with grains of 1 to 2 mm in diameter and were ideally suited for the study of the magnetic fluctuation spectrum or $\mathrm{Im}\tilde{\chi}(\mathbf{q},\omega)$ at low $\mathbf{q}$ which, in a simple cubic lattice, is expected to be diagonal and dependent only on the magnitude of $\mathbf{q}$. Samples prepared by this procedure, and characterized by the methods discussed elsewhere,[19,21] have electronic mean free paths at low temperatures in excess of 2000 Å and were pure enough to reveal, in a parallel experiment, unusually strong quantum oscillations in the magnetization at low temperatures and in high magnetic fields requiring low-range coherence throughout the bulk of the conduction-electron system.[21]

The energy and momentum resolved neutron-scattering cross section has been measured by means of a time-of-flight technique on the IN5 multidetector spectrometer at the European High Flux Reactor, the Institut Laue-Langevin in Grenoble. The high available flux at long wavelengths, the low background at small scattering angles near the forward direction, the simultaneous detection over a wide angle and energy range, and the high resolution, made this spectrometer ideally suited for the present investigation. Here we report on measurements at the discrete incident neutron wavelengths λ_N of 7.9 ± 0.1 Å ($E_N=1.30\pm0.03$ meV) and 5.0 ± 0.1 Å ($E_N=3.3\pm0.1$ meV), and with the detectors placed near the forward direction. The larger incident wavelength is beyond the Bragg cutoff and hence precludes contamination of spectra by elastic nuclear multiple-scattering processes. The shorter incident wavelength utilizes the greater available flux and resolution volume to extend the range of the study.

A wide tailed aluminum cryostat with carefully shielded walls and an evacuated sample space was used to minimize temperature-dependent background scattering at the sample position, while the systematic division of data collection minimized the effects of time-dependent noise. Intensity calibrations were made independently by means of the elastic nuclear incoherent scattering of a vanadium standard sample, of the $\mathrm{Ni_2Ga}$ sample itself, and by Plexiglas. The results were consistent with one another within an experimental error of 15% over the temperature range $1.5\leq T\leq 45$ K studied. The temperature was monitored by means of carbon and platinum resistors, in good thermal contact with the sample, to an accuracy of better than ±0.3 K.

To further reduce interference from spurious signals we have focused attention on the change $I(\theta,E)$ in the scattering cross section $d^2\sigma/d\Omega\,dE$, as a function of scattering angle θ and energy transfer E, relative to the 1.5-K background level (see Fig. 1). Plotted in this figure is the scattering intensity $I(\theta,E)$ at 22 K as a function of neutron energy transfer E (-1 to 2 meV) and scattering angles θ near the forward direction, averaged over the azimuthal angle ϕ. The scattering wave vector $q=|\mathbf{q}|$ may be evaluated for each θ and E via the simultaneous energy and momentum conservation condition

$$q(\theta,E)^2=k_N^2[2+E/E_N-2\cos\theta(1+E/E_N)^{1/2}]\,,$$

where E_N and $\hbar k_N$ are the energy and momentum of the incident neutrons, respectively. The scattering wave vector at zero energy transfer $q_0(\theta)=q(\theta,E=0)$ was, in our investigations, in the range 0.04–0.26 $\mathrm{Å}^{-1}$. The resolution of the elastic wave vector q_0 was in the range 0.01–0.05 $\mathrm{Å}^{-1}$ FWHM, and the resolution of the energy transfer E between 0.05 and 0.2 meV FWHM at the incident neutron wavelengths of 7.9 and 5.0 Å, respectively.

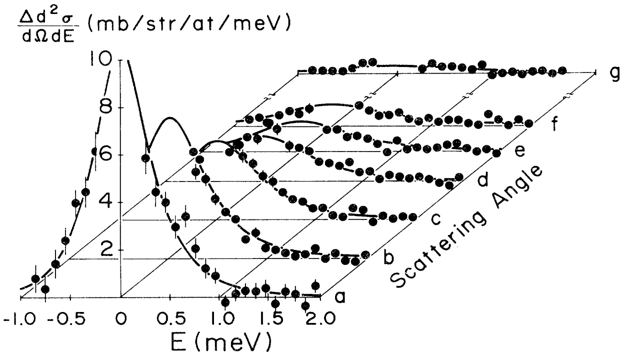

FIG. 1. Energy dependence of the scattering intensity at 22 K relative to the 1.5-K background, measured by the time-of-flight technique for incident wavelength $\lambda=5.0\pm0.1$ Å and for mean scattering angles θ of 3.4°, 4.7°, 6.0°, 7.2°, 8.2°, 9.0°, and 12.0°, as shown in frames a–g, respectively. The scattering intensity has been averaged over the azimuthal angle ϕ and over a range of θ approximately equal to $\pm\frac{1}{2}°$ in frames a–f and ±1° in frame g. The solid lines represent the scattering intensity calculated in terms of the single-pole model for $\chi(q,\omega)$ given in the text with a dispersive relaxation rate of the form $\Gamma(q)=\gamma q(\chi^{-1}+cq^2)$ with $\hbar\gamma=3.0$ μeV Å, $c=1.0\times10^5$ $\mathrm{Å}^2$, and $\chi(T=22\text{ K})$ as given in Ref. 22. The calculations are based on an absolute calibration of the intensity including the effects of instrumental resolution and the energy dependence of the scattering wave vector. (The uncertainties in the values of $\hbar\gamma$ and c are discussed in the text.)

We proceed to examine the main features of the scattering observed and then place it in the framework of a generalized spin-fluctuation model. First, within the experimental resolution over the ranges of wave vectors and energy transfers measured, the thermally induced scattering is diffusive, i.e., $I(\theta,E)$ is peaked around the origin in E. Secondly, the scattering intensity has a rapid θ dependence, and the peak height of $I(\theta,E)$ falls by approximately an order of magnitude between the scattering angles of 3° to 10°. This behavior is inconsistent with that expected for incoherent or multiple-phonon scattering which should increase in intensity with increasing scattering angle. The intensity of scattering for such processes has been estimated from the observed phonon spectrum of $\mathrm{Ni_3Al}$,[23] which has the same crystal structure and similar electronic structure as $\mathrm{Ni_3Ga}$, and

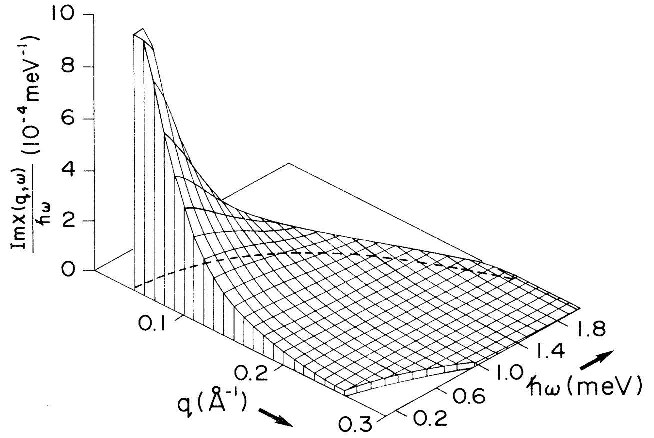

FIG. 2. The imaginary part of the dynamical wave vector dependent susceptibility $\chi(q,\omega)$, divided by $\hbar\omega$, in the experimental q-ω range, consistent with the scattering intensity presented in Fig. 1 at 22 K. The sharp rise of $\mathrm{Im}\chi(q,\omega)/\omega$ at low q and ω is a consequence of a strong dispersion of the relaxation frequency $\Gamma(q)$ in Ni_3Ga.

from the measured phonon density of states of Ni_3Ga inferred from neutron scattering at higher temperatures and wave vectors than reported here. These estimates suggest that the observed scattering intensity in Fig. 1 is 2 orders of magnitude stronger than expected for any phonon scattering mechanism, and hence is unambiguously attributed to scattering from thermally excited fluctuations in the magnetization. A similar conclusion was obtained for the companion metal Ni_3Al in the paramagnetic state, above its Curie temperature of 41 K.[19]

The magnitude of the scattering is also much too large to be attributed, for example, to magnetic polarization clouds around impurity iron atoms which are estimated by mass spectroscopic analysis to be present at levels of less than 10 ppm in our samples.

From the known resolution function and absolute calibration of the spectrometer, the imaginary part of the dynamical wave vector susceptibility $\chi[q=q(\theta,E),\ \omega=E/\hbar]$, which is diagonal and isotropic for Ni_3Ga at low q, may be calculated in absolute magnitude.[24] The spectrum $\mathrm{Im}\chi(q,\omega)/\omega$, which [like $I(\theta,\omega)$] is peaked at the origin in ω, may be characterized within our experimental accuracy at low ω by a one-pole model of the form

$$\mathrm{Im}\chi(q,\omega)/\omega=\frac{z(q)\chi(q)\Gamma(q)}{\omega^2+\Gamma(q)^2}\,, \tag{1}$$

where $\chi(q)=\chi(q,\omega=0)$, $z(q)$ measures the effective weight of the low-lying pole in $\mathrm{Im}\chi(q,\omega)$, and $\Gamma(q)$ is a relaxation rate, or the paramagnon spectrum.

In the low-q and ω range of interest $\Gamma(q)$ is taken to be isotropic and independent of ω with the general form

$$\Gamma(q)=u(q)\chi^{-1}(q)\,, \tag{2}$$

where the function $u(q)$ is expected to vanish at low q for a Fermi liquid or, in general, if the total magnetization is a conserved quantity. The factor $\chi^{-1}(q)$ reflects the fact that $\Gamma(q)$ softens near a magnetic instability where $\chi(q)$ is large. Expanding $\chi^{-1}(q)$ in leading order for a cubic lattice, one obtains $\chi^{-1}(q)=\chi^{-1}+cq^2+\cdots$, where χ^{-1} is $\chi^{-1}(q=0)$ and c is a constant which can be identified with the coefficient of the gradient squared term in a Ginzburg-Landau free-energy functional. We note that the range of validity of the latter expansion, which implies a Lorentzian form for $\chi(q)$, extends up to the Fermi wave vector for the electron gas model, and up to an appreciable fraction of the Brillouin zone in calculations in Pd (Ref. 25) and in measurements in several low-temperature ferromagnets.[18-20]

The form of the function $u(q)$ is more difficult to characterize because of the effect of nonconservation of the magnetization and the effects of real (quasiparticle) scattering processes. In this paper we explore the consequences of assuming a linear dispersion $u(q)=\gamma q$ at low q as motivated by a calculation of the quasiparticle part of $\chi(q,\omega)$ for a Fermi liquid in which any frequency dependences of the Landau molecular-field parameters are neglected, i.e., in the collisionless regime in pure specimens at low temperatures.[1-3,14] In the limit where the antisymmetric molecular field is zero the quantity $\gamma\chi^{-1}$ for a single isotropic band is, to within a numerical constant, the Fermi velocity. More generally, the parameter γ is some appropriate average over the quasiparticle Fermi surface. Within this model, if the magnetization is conserved and if the exchange enhancement of the susceptibility in the Landau Fermi-liquid description is large, then the factor $z(q)$ tends to unity as q tends to zero. For simplicity, the factor $z(q)$ will be absorbed in our definition of $\chi(q)$ at low q, which is therefore only the true static susceptibility in the $q=0$ limit and if the average magnetization is conserved.

The above model for $\mathrm{Im}\chi(q,\omega)/\omega$, in terms of only two temperature-independent parameters γ and c and the measured bulk susceptibility $\chi(T)$, provides a ready interpretation of the observed scattering cross section throughout the wave vector, energy, and temperature ranges investigated. The solid lines in Figs. 1 and 2 are the absolute intensities $I(\theta,E)$ calculated from the neutron-scattering cross section, based on the above model of $\chi(q,\omega)$, convoluted with the calibrated instrumental resolution function assuming unpolarized incident neutrons and setting the Debye-Waller and form factors to unity.[23] The temperature dependence of the calculated $I(\theta,E)$ is due to the thermal population factor in the scattering cross section and the measured temperature dependence of $\chi(T)$. The consistency between $\chi(T)$ and the integral of the observed $\mathrm{Im}\chi(q,\omega)/\pi\omega$ in the experimental ω range to an accuracy of 20% in the limit of zero q, indicates that contributions to $\mathrm{Im}\chi(q=0,\omega)$ from transitions of higher frequency than explored are small.

The values for the model parameter $\hbar\gamma$ and c obtained by fitting *all* available data (and using the known bulk susceptibility) are given in the caption of Fig. 1. A least-squares fitting procedure shows that the uncertainty in the product $\hbar\gamma c$ is small and approximately 10%. However, the uncertainties in the two parameters separately is as high as 30%. A plot of $\mathrm{Im}\chi(q,\omega)/\omega$, which is consistent within experimental error with the neutron data in the experimental q and ω range, is shown in Fig. 2.

We remark that the magnetic fluctuation spectrum of $\mathrm{Ni_3Ga}$ at low temperatures has close parallels to that of the ferromagnet $\mathrm{Ni_3Al}$, well above its Curie temperature ($T_C = 41$ K).[26] This close correspondence (which is not self-evident) and in particular the relationship between the dynamical susceptibility and the thermodynamic properties at low temperatures are the subjects of a more detailed report.

In conclusion, we have measured the magnetic fluctuation or paramagnon spectrum of an exchange-enhanced paramagnetic metal for the first time. We find that $\mathrm{Im}\chi(q,\omega)/\omega$ at low q and ω is diffusive and characterized by a strongly dispersive relaxation rate which can be conveniently parametrized as $\Gamma(q)=\gamma q(\chi^{-1}+cq^2)$. This form is consistent with that expected for the quasiparticle part of $\chi(q,\omega)$ in Fermi-liquid theory, if the frequency dependences of the molecular fields are ignored, i.e., in the collisionless regime. This behavior is in sharp contrast with that observed in the heavy electron rare-earth and actinide compounds, in which dominant parts of the spectrum $\mathrm{Im}\tilde{\chi}(\mathbf{q},\omega)/\omega$ seem to extend over a wide frequency range even in the limit $q=0$. Because of limited scattering intensity and resolution, the precise character of this broad spectrum is in most cases still not well understood. It is possible, however, that a component in the fluctuation spectrum analogous to that reported here may exist even in these systems in the pure state at low temperatures.

We wish to acknowledge the support of the Science and Engineering Research Council of the United Kingdom and the assistance of the staff at the Institut Laue-Langevin.

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