Observation of spin waves in Pd(1.5% Fe)

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Inelastic neutron scattering measurements have been carried out on the "giant-moment" alloy system Pd(1.5% Fe), which is in the dilute ferromagnetic regime. Below the Curie temperature of 67 K relatively well defined spin wave excitations have been observed in the small wave vector region ($Q \leq 0.14$ Å^{-1}). The dispersion of these excitations is consistent with the quadratic relation $E = DQ^2$ expected for an isotropic ferromagnet, with $D = 40$ meV Å^2 at a temperature of 40 K. With increasing temperature the spin waves are found to renormalize in energy and broaden rapidly with both increasing Q and increasing temperature.

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INTRODUCTION

Although pure palladium does not exhibit a magnetic phase transition, the high density of d-band states at the Fermi level renders the system especially susceptible to polarization by d-band magnetic impurity ions such as iron. This strong spin polarization of the palladium bands results in the formation of "giant-moment" clusters (~10 μ_B/iron atom).[1] In the range of concentration where individual clusters do not interact with each other the system is thought to form a spin glass type state at low temperatures,[2] while at high iron concentrations a conventional ferromagnetic state is obtained. Because the effective iron-iron interaction is large and of long range, the threshold for ferromagnetism occurs at a very low concentration,[3] $x = 0.0012$.

At relatively high iron concentrations the Pd lattice should be uniformly polarized, and thus linear spin wave theory should be valid. Indeed spin wave excitations have been directly observed by inelastic neutron scattering in the chemically ordered[4,5] Pd_3Fe and in disordered[6] Pd_3Fe. At lower iron concentrations the polarization of the Pd host will become nonuniform which may lead to overdamped excitations. Indeed no spin wave excitations have been previously observed directly at low concentration, although the dynamics have been probed by the "diffraction-technique."[7] The nature of the excitations near the threshold for ferromagnetism takes on added significance in light of the ferromagnetic to spin-glass "transition" recently observed in several systems and the consequent effects that have been observed on the spin dynamics[8,9] in these materials.

EXPERIMENTAL CONSIDERATIONS

The sample used in the present study was a 9 gm sample which was arc-melted, annealed and then powdered. The concentration was chosen to be 1.5% Fe, with a measured magnetic transition $T_c = 67$K. At this concentration the Pd lattice should be polarized relatively uniformly, and we then expect a dispersion relation at long wavelengths of the form[10]

$$E = Dq^2 \tag{1}$$

where D is a temperature and concentration dependent constant which is a measure of the effective exchange in the system. In addition to this uniform (acoustic) mode, an optic mode due to the out-of-phase precession of the iron and palladium moments may also exist, but single crystal specimens will be needed in order to observe this mode. In the present study we are interested in the acoustic mode, and for these studies powder specimens are adequate.

The measurements were carried out on the BT-4 triple-axis spectrometer at the National Bureau of Standards Research Reactor. A cold (77K) Be filter was placed before the pyrolytic graphite (PG) monochromator. The incident neutron energy was chosen to be 4.90 meV, with 40' FWHM collimation before and after the monochromator and (PG) analyzer. This gave an energy resolution of 0.15 meV (FWHM) at the elastic position. Because the experiments were all done around the forward direction (000), the flight-paths before and after the sample were isolated and filled with Ar gas to suppress the (inelastic) air scattering. Thin Aℓ windows were used on the cryostat vacuum jacket and radiation shields to reduce the small angle scattering from the cryostat.

RESULTS AND DISCUSSION

The observed inelastic scattering spectrum at a wave vector of 0.10 Å^{-1} is shown as a function of energy in the first figure. These data were taken at 40K, well below the ferromagnetic transition of 67K determined by bulk magnetization measurements. The transition was independently determined by measuring the intensity of the elastic neutron scattering as a function of temperature, which peaked at T_c in the customary fashion.

The data of Fig. 1 show spin wave peaks in neutron energy gain ($E<0$) and energy loss ($E>0$). The peak at $E=0$ is due primarily to residual scattering from the cryostat as well as incoherent scattering from the sample. The scattering cannot be explored to larger energy transfers because momentum and energy cannot be conserved. The solid curves in the figure are the result of the convolution of the instrumental resolution function with Eq. (1), plus a Gaussian peak centered at $E=0$ with a width fixed to the measured energy resolution. Note that since $kT/E \gg 1$, the spin wave creation and annihilation peaks are almost of equal intensity. The value of D found to best fit the data is 40 meV-Å^2 at 40K, which is considerably smaller than the value at this concentration inferred from the diffraction technique.[7]

Figure 2 shows the temperature dependence of the scattering on the energy gain side. At the higher

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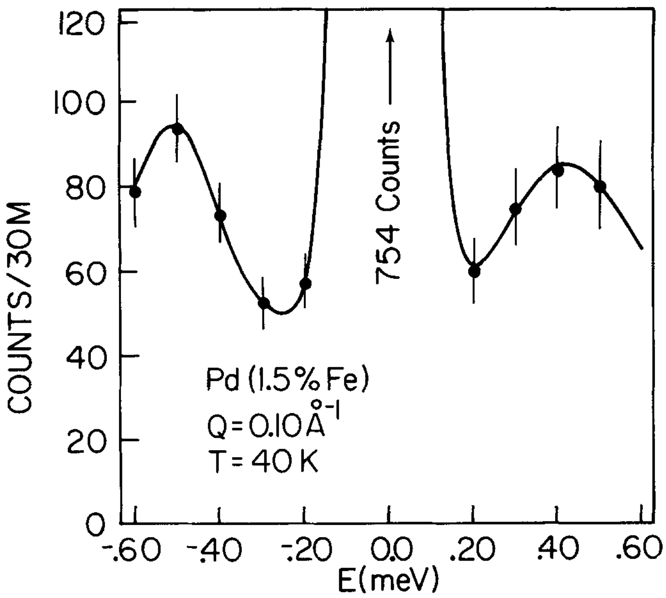

Fig. 1. Observed scattering at a momentum transfer of 0.10 Å^{-1} below the ferromagnetic transition temperature of 67K. The spin waves are observed in energy gain (E>0) and energy loss (E<0). The peak in the center is due to incoherent nuclear scattering and residual scattering from the cryostat. Data cannot be taken at larger energy transfers because momentum and energy cannot be conserved.

temperature the scattering peaks at a smaller energy, which is the expected behavior as the spin waves renormalize to smaller energies as the transition temperature is approached.

The magnetic scattering for 40 and 50K is shown in Figure 3 for a somewhat larger momentum transfer of 0.12 Å^{-1}. There data have been counted for an hour per point to improve the statistical accuracy. In addition a flat background value, determined at low temperatures, and the peak at the elastic position determined at both low and high temperatures, have been subtracted. The remaining scattering is clearly due to spin waves, and the solid curves are again a convolution of the instrumental resolution with 3 Eq. (1). In these calculations the intrinsic linewidth was assumed to be negligible.

It is clear from the data that well defined spin waves exist below T_c even at the relatively low concentration of 1.5% Fe. Indeed the solid curves in Fig. 1 and 3 were generated assuming that the intrinsic spin wave linewidths were small in comparison with the instrumental resolution. The data are consistent with a quadratic dispersion for the spin waves, but it should be noted that the present set of data is rather limited. The temperature and wavevector dependence of the spin waves and their intrinsic linewidths will be investigated in more detail when larger samples are available. Further measurements of the dynamics of the Pd(Fe) system as a function of temperature and concentration are in progress.

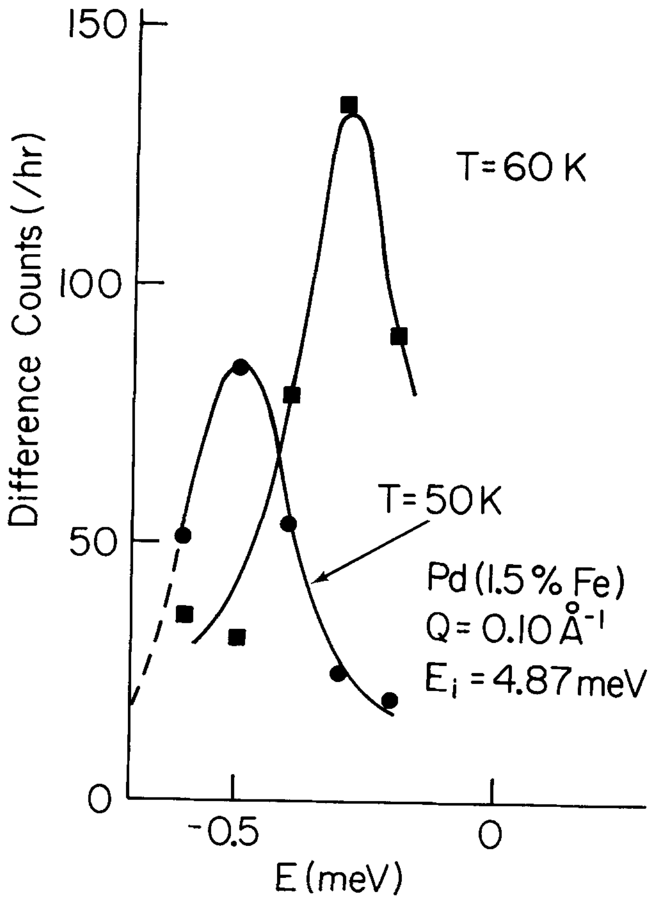

Fig. 2. Temperature dependence of the scattering in energy gain, showing that the spin waves renormalize to lower energy as T_c is approached.

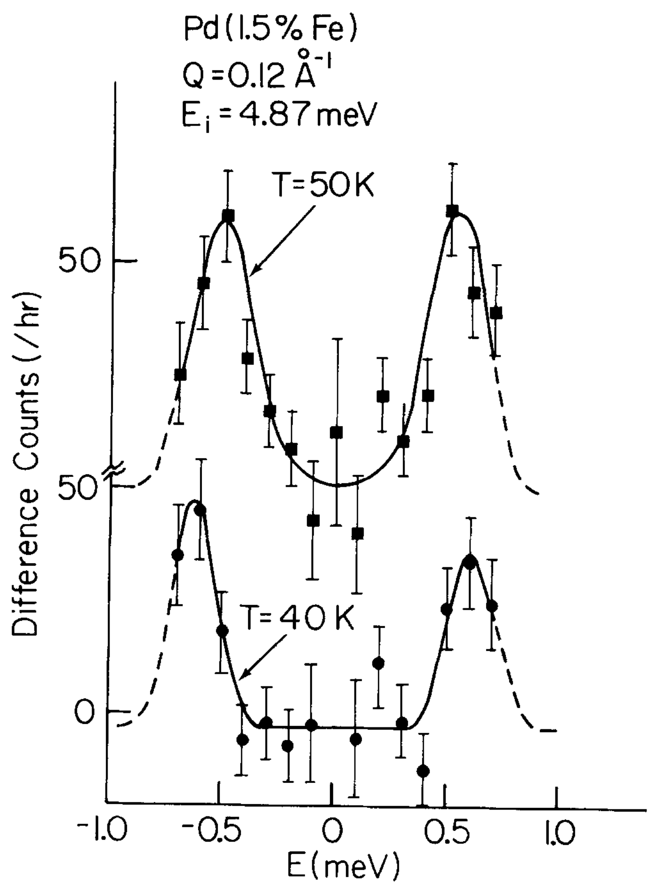

Fig. 3. Net scattering at a momentum transfer of 0.12 Å^{-1}, after subtraction of nonmagnetic background. The spin waves are seen to shift to smaller energies with increasing temperature as expected.

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