mentarily singly occupied donor sites, fluctuations which are expected to occur in the highly correlated electron gas.[15] In the CJ model, density fluctuations produce an inhomogeneous broadening of the valley-orbit line. Although the crude calculations described above are based on density fluctuations and therefore seem to favor the CJ model, the observed Raman line shapes might be fitted equally well by assuming the existence of only temporal fluctuations. Hopefully, our Raman data coupled with a more sophisticated theoretical interpretation will serve to distinquish between the models of Mott and CJ.

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Magnetic Excitations and Magnetic Ordering in Praseodymium

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The dispersion relations for magnetic excitons propagating on the hexagonal sites of double-hcp Pr provide clear evidence for a pronounced anisotropy in the exchange. The energy of the excitations decreases rapidly as the temperature is lowered, but becomes almost constant below about 7 K, in agreement with a random-phase-approximation calculation. No evidence of magnetic ordering has been observed above 0.4 K, although the exchange is close to the critical value necessary for an antiferromagnetic state.

For a number of years there has been a controversy about the existence of magnetic ordering in Pr, the only element to display the characteristic magnetic behavior of a singlet ground-state system with weak exchange interactions. An antiferromagnetic structure below a Néel tempera-

ture of 25 K was discovered by neutron diffraction on a polycrystalline sample by Cable *et al.*[1] but no traces of magnetic ordering were found in a single crystal at 4.2 K by Johansson *et al.*,[2] although a large moment was induced by a magnetic field in the basal plane. Andres *et al.*[3] interpreted their measurements on the nuclear heat capacity of a monocrystalline sample in terms of an antiferromagnetic state and proposed that such an ordering might not have been detected in the neutron experiments on single crystals. However, the more recent heat-capacity results of Gregers-Hansen and Pickett[4] on a single crystal showed no signs of magnetic ordering above 0.1 K, nor was any evidence for antiferromagnetism above 0.4 K detected in further neutron-diffraction studies by Lebech, McEwen, and Lindgård.[5] They did, however, observe an antiferromagnetic state in a monocrystalline alloy of 5.6% Nd in Pr with a T_N of 6.5 K. In this Letter, we report measurements of the magnetic excitations in Pr at low temperatures, discuss the information on the magnetic interactions which may be deduced from them, and comment on the implications for the possibility of magnetic ordering.

The first inelastic-neutron-scattering measurements in Pr were made by Rainford and Houmann,[6] who interpreted the bulk of their results in terms of magnetic excitons composed of linear combinations of transitions from the ground state $|0\rangle$ to the doubly degenerate first excited state $|\pm 1\rangle$ on those sites which have local hexagonal symmetry. This designation of the lowest-lying states is supported by neutron-diffraction[7] and magnetization[8] measurements. Some splittings in the energies of these modes were tentatively ascribed to anisotropic exchange by Lindgård and Houmann,[9] who considered a Hamiltonian of the form

$$\mathcal{H} = V_c - \sum_{\nu, i>i'} \mathcal{J}_{ii'}{}^{\nu\nu} J_i{}^{\nu} J_{i'}{}^{\nu} - \sum_{\nu, j>j'} \mathcal{J}_{jj'}{}^{\nu\nu} J_j{}^{\nu} J_{j'}{}^{\nu} - \sum_{\nu, ij} \mathcal{J}_{ij}{}^{\nu\nu} J_i{}^{\nu} J_j{}^{\nu}, \quad (1)$$

where V_c is the crystal-field potential, ν represents the Cartesian coordinates, and the indices i and j refer to sites on the two sublattices of the hcp structure of the ions on the hexagonal sites. At low temperatures, the excitation energies may be approximately deduced from this Hamiltonian as[9]

$$E_{\pm,\nu=x,y}{}^2 = \Delta^2 - \alpha^2 \Delta [\mathcal{J}^{\nu}(\vec{q}) \pm |\mathcal{J}'^{\nu}(\vec{q})|], \quad (2)$$

where Δ is the crystal-field splitting between $|0\rangle$ and $|\pm 1\rangle$, α is the matrix element of J^+ or J^- between these states, and $\mathcal{J}(\vec{q})$ and $\mathcal{J}'(\vec{q})$ are, respectively, the Fourier transforms of $\mathcal{J}_{ii'}$ and $\mathcal{J}_{ij}$. The four branches of the dispersion relation correspond to acoustic and optical modes (plus and minus, respectively), each with J_x and J_y excitations.

We have studied these excitations by inelastic neutron scattering, using a triple-axis spectrometer at the DR3 reactor at Risø. The sample was an 0.67-cm^3 single crystal grown by an induction-zone-melting technique using a (99.9+)% pure praseodymium stock, with the highest impurity levels (in ppm by weight) being O 160, F 60, Nd 150, and Ta 160 (the remaining impurities were under 50 ppm). The results at 6.4 K are shown in Fig. 1, in which the four branches of the excitons propagating in the basal plane are seen

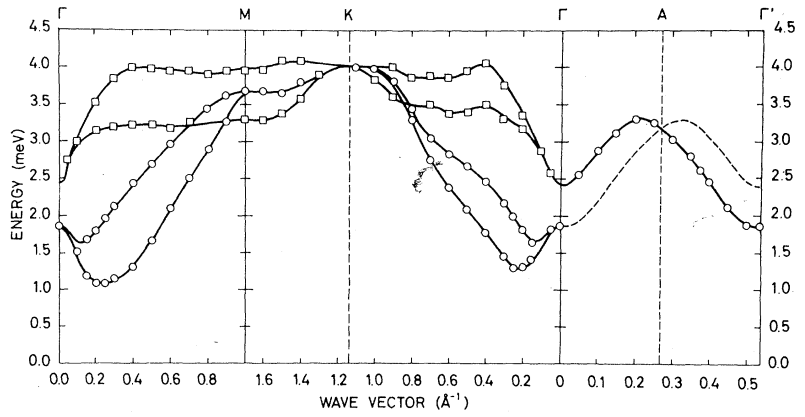

FIG. 1. Dispersion relations for magnetic excitons propagating on the hexagonal sites in Pr at 6.4 K. The double-zone representation is used in the ΓA direction.

generally to be clearly resolved. For those excitations propagating along the c axis, symmetry eliminates the splitting between the J_x and J_y modes, and the use of the double-zone representation further reduces the number of branches to one. From these dispersion relations and Eq. (2), we may immediately deduce that the anisotropic component of the exchange interaction is a substantial component of the total, as is also observed in the heavy rare earths.[10]

Of particular interest is the mode of lowest energy between Γ and M, whose wave vector corresponds very closely to that which characterizes the longitudinal-wave magnetic structure in dilute *Pr*Nd alloys.[5] The neutron-scattering cross section contains a term $(1-\kappa_y^2/\kappa^2)$, where $\vec{\kappa}$ is the scattering vector. Intensity measurements have thereby allowed us to deduce that this mode corresponds to a longitudinal fluctuation of the ionic moment, and may therefore be considered as the incipient magnetic soft mode; i.e., it is that mode which corresponds in wave vector and polarization with the magnetic structure formed when a small amount of Nd is added to Pr. The temperature dependences of this mode, the low-energy mode at Γ, and that of minimum energy between Γ and K are shown in Fig. 2. The incipient soft mode has indeed the most rapid temperature variation at higher temperatures, but below about 7 K neither it nor any other excitation decreases appreciably in energy as the temperature is reduced to 0.4 K. In a simple random-phase approximation (RPA), the temperature dependence of the excitations is given by multiplying the second term in Eq. (2) by a temperature renormalization factor $R(T)$ which is just the difference in Boltzmann population factors between the ground and excited states,[11] i.e.,

$$R(T)=n_0-n_1=[1-\exp(-\Delta/kT)]\times[1+2\exp(-\Delta/kT)]^{-1}. \quad (3)$$

The temperature dependence of the excitations predicted by this theory is plotted in Fig. 2 for a Δ of 3.2 meV and values of the exchange which give agreement with the low-temperature limit of the energies. In contrast with earlier measurements on polycrystalline Pr and Pr_3Tl,[12] the simple RPA is seen to give an excellent account of the temperature dependence of these modes. From Eq. (2), using this value of Δ, we can immediately deduce that the exchange is approximately 90% of that which would be required to drive the energy of the soft mode to zero. A

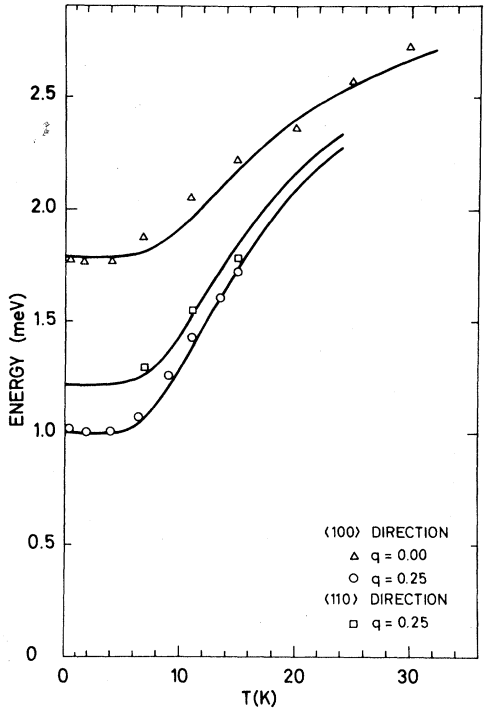

FIG. 2. Temperature dependence of selected magnetic excitations in Pr. The full lines are calculated in the random-phase approximation, as described in the text.

more sophisticated RPA calculation by Lindgård,[13] in which the dispersion of the excitons is explicitly taken into account, yields a somewhat greater value of Δ and an exchange which is even closer to the critical value for magnetic ordering.

At low temperatures, the observed neutron peaks have an energy width corresponding to the experimental resolution but, as the temperature is increased, the lifetime of the excitations decreases so that, for example, the low-energy mode at Γ has a natural width of about 1.3 meV at 30 K. The dispersion relations are also strongly field dependent and we have observed exciton-phonon interactions in the basal plane whose strength increases rapidly with the applied magnetic field.

Our neutron measurements are therefore consistent with the hypothesis that the exchange interactions in Pr are not sufficient to produce magnetic ordering above 0.4 K in a monocrystalline sample. However, the exchange is close to the critical value for an antiferromagnetic state and the recent heat-capacity measurements of Gregers-Hansen and Pickett[4] suggest that a combined nuclear-electronic ordering of the type

proposed by Murao[14] may be formed at a temperature below 0.1 K. We plan to attempt to detect this ordering by neutron diffraction at lower temperatures and to investigate the mechanism of magnetic ordering in dilute PrNd alloys by inelastic scattering measurements on single crystals. The behavior of polycrystalline samples is not yet fully understood; presumably their magnetic ordering at relatively high temperatures is associated with strains and defects in the crystallites.

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Tetracritical Points in Mixed Magnetic Crystals*

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Recently developed renormalization-group techniques are used to study the tetracritical (or bicritical) point of quenched mixed magnetic crystals (e.g., A_pB_{1-p}, where A and B have ferromagnetic and antiferromagnetic phase transitions). The tetracritical (bicritical) exponents are possibly given by the $n=0$ value of their ϵ expansions. In particular, the crossover exponent φ, describing the shape of the p-T phase diagram near the tetracritical point, is *exactly* equal to 1. This explains various experiments.

Recent developments in the theory of critical phenomena led to a better understanding of phase diagrams which exhibit bicritical and tetracritical points.[1-4] Such points usually occur when a system has two order parameters, with competing interactions. A large class of such systems is that of quenched mixed magnetic crystals, of the type A_pB_{1-p}, when pure A and pure B have different types of magnetic ordering (e.g., ferromagnetic and antiferromagnetic). Examples are $Fe(Pd_pPt_{1-p})_3$, where $FePd_3$ is ferromagnetic and $FePt_3$ is antiferromagnetic,[5] $(Mn_{1-p}Fe_p)WO_4$, where pure $MnWO_4$ and $FeWO_4$ have distinct antiferromagnetic orderings,[6,7] $NH_4Cl_{1-p}Br_p$, where pure NH_4Cl and NH_4Br have parallel and antiparallel ordering of the ammonium tetrahedra,[8] UAs_pSe_{1-p}, where USe orders ferromagnetically while UAs orders antiferromagnetically,[9] and many others.

Unlike the anisotropic antiferromagnets in a uniform field, which tend to exhibit a *bicritical* spin-flop point and a first-order spin-flop line,[1] most of the mixed magnetic crystals exhibit a *tetracritical* point, and a mixed "intermediate"