

THE INFLUENCE OF Y DILUTION ON THE
CRYSTALL-ELECTRIC-FIELD PART OF THE POINT-CONTACT
SPECTRA OF VAN VLECK PARAMAGNET $\text{Pr}_{1-x}\text{Ni}_x$ ¹

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We report on the study of the influence of Y dilution on the Crystalline Electric Field (CEF) interaction in the Van Vleck paramagnet PrNi_5 using the point-contact (PC) spectroscopy. PC spectra of $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ compounds ($x = 0, 0.02, 0.05, 0.1$ and 0.2) in the ballistic regime have been measured. A shift of the main CEF peak ($1_4 \rightarrow 1_5^2$) situated at about 4.2 meV for $x = 0$ and 5.1 meV for $x = 0.2$ is observed. This shifting is in agreement with that obtained from susceptibility measurements in this system. The asymmetry in the dV/dI dependences of all heterocontacts could be explained by the model taking into account the difference in the effective masses of conduction electrons.

The physical properties of the hexagonal Van Vleck paramagnet PrNi_5 (CaCu₂-structure type) are well understood by consideration of the Crystalline Electric Field (CEF) splitting of the $3H_4$ multiplet of the Pr^{3+} ions, which leads to a non-magnetic singlet as ground state [1]. This and the fact that only very small exchange interaction coupling of the Pr-ions is present, prevent the apparition of magnetic order to be established among Pr-4f moments. Because of this feature, PrNi_5 is currently one of the best adiabatic nuclear cooling materials [2]. Recently, the influence of Y "dilution" on the electronic magnetic properties has been studied, and in particular, the change of the nuclear ordering temperature [3] and the decrease of the magnetic Van Vleck susceptibility with increasing Y content were analyzed.

In this paper, the influence of Y dilution on the CEF interaction in $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ compounds by the point-contact (PC) spectroscopy is investigated and a comparison with the results obtained from susceptibility measurements on the same samples is made.

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PC spectroscopy has been shown to be a very useful experimental probe for the direct study of the energy dependence of the interaction between conduction electrons and quasiparticle excitations in metals [4, 5]. Moreover, in PrNi_5 the Zeeman splitting of CEF levels has been measured [6]. As is well-known, small constrictions between two metals show deviations from Ohm's law. For clean metallic contacts where both the inelastic l_1 and elastic l_e mean free paths of the electrons are larger than the contact diameter d , the transport of electrons through the constriction is ballistic. With a voltage applied across the contact, therefore the electrons are accelerated when passing through the contact area. The applied voltage directly defines the energy scale for the scattering processes of the electrons. The relaxation of accelerated electrons results in voltage-dependent corrections to the current and these deviations are most clearly visible in a measurement of the second derivative d^2V/dI^2 of the I-V characteristic.

The d^2V/dI^2 curve is proportional to the spectral function for the interaction process under investigation. However, it is often problematic to fulfil the ballistic condition (i.e., $d \ll l_1, l_e$) for intermetallic compounds and alloys. In this case, the relation between l_1 and l_e is important. Then the spectroscopic regime is still possible if the condition $l_e \ll d \ll l_1$ is valid (diffusional regime) [7]. The experimental resolution is the same as in ballistic regime.

Moreover, the resistances of PCs between different materials frequently reveal an asymmetry with respect to the polarity of the applied voltage [8]. In the case of simple metals and for the ballistic regime the asymmetry was explained by considering the phonon-drag effect inside PC. If $d \gg l_1$ (thermal regime [5]) the dissipation in the contact region heats the contact above bath temperature which leads to an asymmetry in the differential resistance dV/dI of heterocontacts due to the thermoelectric effects. An enhancement of the electronic density of states at the Fermi energy, e.g. in the heavy fermion systems, can also result in an asymmetry effect [9]. The evidence for EMP in a PCs between two metals (PrNi_5 , CeNi_5 with Cu, Ni, Fe) with different effective electron masses was observed [10].

We have studied the polycrystalline samples of $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ with $x = 0, 0.02, 0.05, 0.1$ and 0.2 . The details about the sample preparation are in reference [3]. The hetero-PCs $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ -Cu were made at 4.2 K using a Cu needle. Before measurement the sample surface was electrochemically polished. The derivatives dV/dI and d^2V/dI^2 of the I-V characteristics were measured by means a phase-sensitive technique [5]. The electrical resistances of the PCs have varied from 1 Ω to about 7 Ω .

In Fig. 1 the characteristic PC spectra of $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ -Cu ($x = 0, 0.02, 0.05, 0.1$ and 0.2) heterocontacts in ballistic regime are shown. All of them show a typical first peak at energies ranging between 4 to 5 meV for $x = 0$ to $x = 0.2$. This peak is related to the CEF excitation of the Pr^{3+} ion from the ground state Γ_4 to one of the excited states Γ_2^2 by the accelerated electrons [6]. A small shoulder or plateau at 9 meV is ascribed to the phonons contribution in PrNi_5 . The broad maximum between 16 and 20 meV corresponds to the energy of transverse Cu and PrNi_5 phonons. The 13.5 meV CEF ($\Gamma_4 \rightarrow \Gamma_3$) transition, observed previously in PrNi_5 [6], occurs in the spectra as a shoulder and it remains practically at the same position with increasing the Y content. The maximum at around 30 meV is related to the longitudinal phonons in Cu. Because

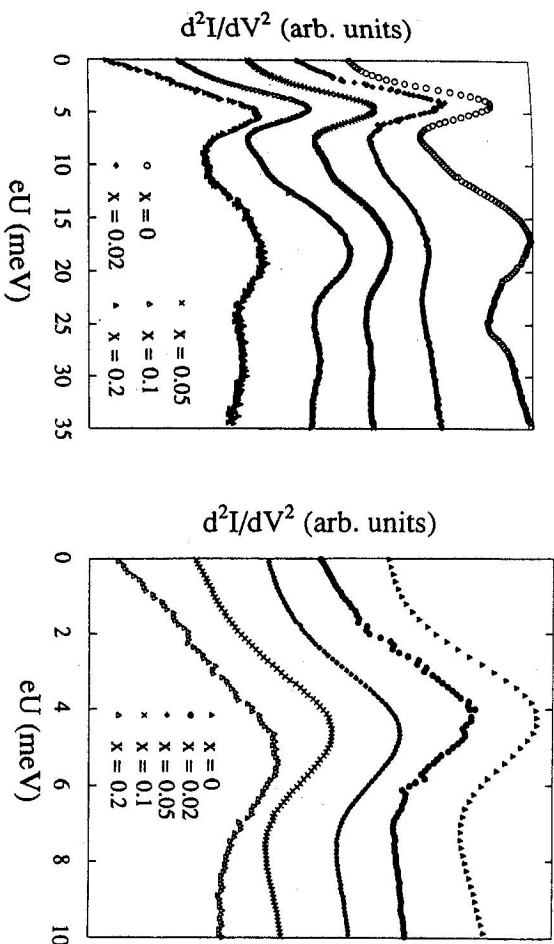


Fig. 1 - Characteristic PC spectra of $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ -Cu ($x = 0, 0.02, 0.05, 0.1$ and 0.2) heterocontacts in ballistic regime.

Fig. 2 - Detail of the first CEF peak for $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ -Cu ($x = 0, 0.02, 0.05, 0.1$ and 0.2) heterocontacts in ballistic regime.

of the small probability of transitions to higher CEF levels it is possible to study the shift of the first allowed transition which gives the dominant contribution to the CEF part of PC spectra. Therefore, the analysis is concentrated on the first peak.

As shown in Fig. 2, the characteristic energy shift of the first peak with the yttrium amount is about 1 meV for $x = 0.2$ with regard to the position of the peak associated with $x = 0$. This shift is reproducible and is larger than the spreading of the measured position of the first peak observed in PC spectrum of PrNi_5 . We explain this shift taking into account that Y-ions are nonmagnetic and by introducing them into a PrNi_5 hexagonal lattice the mean distance between Pr-ions increases. From this feature it follows that the remaining Pr-ions are more localized (the overlap of 4f wave functions effectively decreases) and one needs more energy in order to excite them to higher CEF states. This modification of the CEF is directly observable only by PC spectroscopy or via inelastic neutron scattering. Indirect confirmation can also be obtained by measuring the shift of the low temperature maximum which appears in the magnetic susceptibility [3].

In Fig. 3 the measured [3] and fitted thermal dependences of the magnetic susceptibilities of polycrystalline $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ compounds are presented. The dilution with yttrium depresses the amplitude of the maximum and shifts its position to higher temperatures. As is known [6], the origin of this maximum is related to the non-magnetic character of the groundstate and its position depends mainly on the relative difference between energy levels of the states associated with Γ_4 and Γ_2^2 . For the Pr^{3+} ion, the 3H_4 multiplet is split in 3 doublets and 3 singlets, being described in the CEF hamiltonian

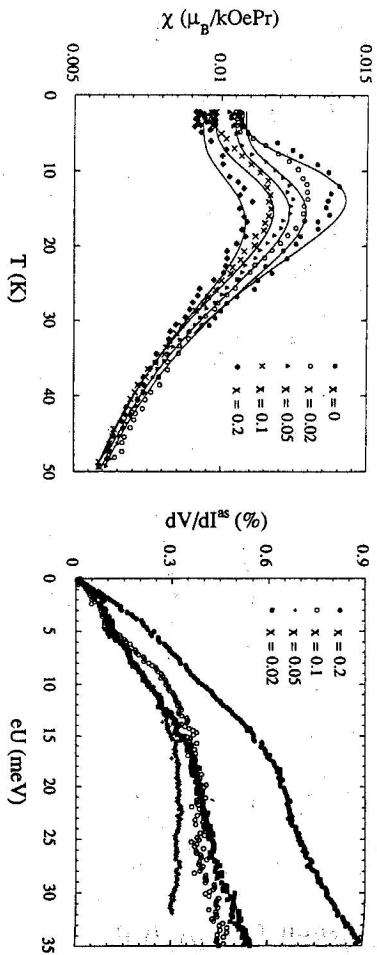


Fig. 3 - Measured [3] and calculated (solid lines) magnetic susceptibilities per mole of $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ versus temperature ($x = 0, 0.02, 0.05, 0.1$ and 0.2).

Fig. 4 - Characteristic asymmetry of the dV/dI^{as} (see in text) for $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ -Cu ($x = 0.02, 0.05, 0.1$ and 0.2) heterocontacts.

by 4 parameters (B_2^0, B_4^0, B_6^0 and B_6^2). Taking into account that B_2^0 and B_4^0 are responsible for the high temperature variation of the susceptibility, they are expected to be quite similar in $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ and they have been fixed to the values obtained for PrNi_5 in reference [6]. We can then fit our experimental curves using B_6^0 and B_6^2 as adjustable parameters, assuming an isotropic distribution of the polycrystalline grains (in this case $\chi = 1/3 \chi_{\parallel} + 2/3 \chi_{\perp}$ where parallel means along c direction and perpendicular inside the basal plane). The solid lines in Fig. 3 correspond to the best fit. From this analysis it was concluded that the difference in energy between Γ_4 and Γ_2^2 is increased about 1 meV for $x = 0.2$ with respect to $x = 0$. The position of the Γ_2^2 CEF level was found to be 3.9; 4.3; 4.3 and 5 meV for $x = 0, 0.02, 0.05, 0.1$ and 0.2 respectively. This result is in a good agreement with the experimental determination obtained above from the PC spectra. More precise measurements using inelastic neutron scattering could confirm this conclusion.

In general, all PC spectra showed an asymmetry of PC resistance with respect to the applied voltage polarity. Fig. 4 represents the characteristic asymmetric part $dV/dI^{as} = [dV/dI(+eU) - dV/dI(-eU)]/2$ of the PC resistance (in $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ -Cu heterocontacts). The +/- sign of the eU energy marks the polarity on the $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$. The magnitudes of dV/dI^{as} at 30 meV are less than about 2. There is no observable correlation of the change of this magnitude with changing of Y content. These dependences are in agreement with the previous measurements on RENi_5 ($\text{RE} = \text{Pr, Ce}$) compounds [10]. We believe, therefore, that Y substitutions can not produce substantial changes in the

effective mass of conduction electrons when compared to PrNi_5 . We suppose, then, that this asymmetry is due to the addition of electrons when compared EMF in heterocontacts between the two metals with different effective mass of conduction electrons.

In conclusion, we have observed the change in the energy position of the CEF Γ_2^2 CEF excited level of $\text{Pr}_{1-x}\text{Y}_x\text{Ni}_5$ compounds with the increasing Y content by PC spectroscopy, and shown that the dilution can create significant modifications in the low-energy CEF level scheme. Moreover, we have detected the same kind of asymmetry of the PC resistance as in PrNi_5 . This feature seems to be connected with the different effective masses of the conduction electrons in the metals forming the PCs.

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