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Crystal-field effects in NdP-UP solid solution studied by neutron spectroscopy

A. Murasik (*), J. Leciejewicz (*), R. Niedzielski (†) and R. Troć (†)

(*) Institute of Nuclear Research, Świerk Research Establishment, 05-400 Otwock, Poland
(†) Institute for Low Temperature and Structural Research, 50-950 Wroclaw, Poland

Résumé. — On a réalisé des expériences de diffusion inélastique des neutrons pour le système NdP-UP. Les paramètres de champ cristallin ont été déterminés pour les composés Nd₃U₀.2P et Nd₇U₀.3P.

Abstract. — Neutron inelastic scattering experiments on the NdP-UP system have been performed. The crystal-field parameters were determined for the compositions Nd₃U₀.2P and Nd₇U₀.3P.

1. Introduction. — In spite of many efforts undertaken at various laboratories with the aim to observe the crystal-field or spin-wave spectrum in metallic actinide systems (with the possible exception of few compounds where single transitions were observed [1]), experimental results are not very encouraging. On the other hand, there is still current interest in determining the crystal-field parameters directly from the experiment, since apart from their intrinsic interest one may interpret with them other physical data such as magnetic susceptibility, spontaneous magnetization, resistivity, etc.

The present work is an attempt to obtain information on the magnitude of crystal field in a system, where the actinide ions are successively replaced by rare-earth ions. To perform such a study using the neutron inelastic scattering technique, we selected the mixed uranium-neodymium monophosphides. The boundary compounds NdP and UP have metallic conductivity and form a complete range of solid solutions [2]. Both compounds have the rock-salt type crystal structure and order antiferromagnetically at 11 and 125 K respectively. Uranium and neodymium ions in their monophosphides have the same 3+ oxidation states with the 4f⁹ multiplet as ground state. The crystal field of octahedral symmetry splits ten-fold degenerate ground state multiplet into a doublet Γ₄ and two quartets Γ⁹⁴ and Γ⁹⁶. In NdP well defined crystal-field transitions were observed [3]. Experiments led to identification of the quartet Γ⁹⁴ as ground state and determination of levels sequence with an overall splitting of 14.6 meV. In isostructural UP, no evidence for crystal-field transitions was reported [4]. An attempt to deduce the crystal-field splitting scheme from susceptibility data was made by Troć and Lam [5]. They found that 5f⁶ configuration of U³⁺ ion, with a model involving a mixed J ground state gives a resonable fit to the measured susceptibility of UP. The crystal-field level scheme for uranium ion as derived from the high-temperature susceptibility results, indicates that the doublet Γ₄ and quartet Γ⁹⁴ are the excited states at 79 and 310 meV above the Γ⁹⁴ ground state.

Our interest in NdP-UP system arises from the possibility of studying of 4f-5f magnetic cooperative effects. On the other hand, exchange effects are known to cause the serious disturbance in observing crystal-field transitions. Thus, their influence should be readily seen in a set of experiments on samples with progressive increase of uranium content.

2. Experimental. — The mixed uranium-neodymium monophosphides were prepared by sintering the pellets of NdP and UP in desired proportions. The final products were examined by X-ray. The following compositions of NdₙUₓP solid solution were synthetized and investigated : y = 0.2, 0.3, 0.5, and 0.6. The experiments were performed on the triple-axis spectrometer KSN-2 at Świerk. The incident neutrons had a constant energy of 33.96 meV. The measurements were carried out in energy loss configuration with the scattering angle kept constant. To confirm the magnetic origin of observed intensities the measurements have been performed at several temperatures : 5 K, 80 K, and 293 K.

3. Results and discussion. — Results obtained can be summarized as follows. For samples with low uranium concentration (y = 0.2, 0.3) energy spectra at 5 K and 80 K exhibit well shaped maximum at about 12-13 meV which can be attributed to magnetic transitions from lower to higher states. When uranium concentration increases to y = 0.5 and y = 0.6, the observed maximum apparently broadens and shifts towards the lower energy transfers (see Fig. 1). The temperature variation of energy spectra for a fixed composition shows, typical for magnetic scattering behaviour ; i.e. transitions from ground to excited levels increase as the temperature is decreased (Fig. 2).
At the present stage of experiment, we have analysed the data only for low uranium content and within the classical formalism; i.e. we used the cross-section formula derived by de Gennes [6] and single-ion CF Hamiltonian for $^{4}P_{1}$ configuration with $^4p_{1/2}$ ground state multiplet [7]. In order to determine the crystal-field schemes, a least-squares fitting procedure was applied to the observed spectra. In this procedure, the three fitting parameters were the crystal-field parameters $W$, $x$, and a linewidth parameter $\gamma$ which has been assumed to be the same for all crystal-field transition peaks at a given temperature. The elastic peak and the crystal-field transition peaks were approximated by Gaussians. The scale factors have been determined self-consistently.

The resulting crystal-field parameters are: for $y = 0.2$, $T = 80\, \text{K}; \quad W = -0.231\, \text{meV}, \quad x = 0.734, \quad \gamma = 2.8\, \text{meV}$, whereas for $y = 0.3$, $T = 293\, \text{K}$, and $T = 80\, \text{K}; \quad W = -0.197\, \text{meV}, \quad x = 0.72, \quad \gamma = 6.0\, \text{meV}$, and $W = -0.207\, \text{meV}, \quad x = 0.71, \quad \gamma = 4.1\, \text{meV}$ respectively. Owing to the large line broadening at $293\, \text{K}$ spectrum, obtained results seem to be less reliable as compared with those for $80\, \text{K}$ spectrum. Judging from the crystal-field parameters listed above, the $F_{g}^{0}$ quartet is the ground state for both samples. The overall splitting is 13 and 11.7 meV for $y = 0.2$ and 0.3 respectively. However, it is of interest to note, that from energy spectra for Nd$_{0.7}$E$_{0.3}$P, another set of $W$, $x$, and $\gamma$ parameters, with nearly the same quality of fit but with the doublet $F_{g}$ as ground state, can be derived. Namely: $W = -0.207\, \text{meV}, \quad x = 0.861, \quad \gamma = 5.0\, \text{meV}$ ($T = 80\, \text{K}$), and for $293\, \text{K}$ spectrum:

$W = -0.194\, \text{meV}, \quad x = 0.848, \quad \gamma = 6.8\, \text{meV}$.

For the analysis of energy spectra measured in the ordered state (5 K), we used single ion Hamiltonian describing the CF + exchange in a molecular field approximation. For the model with the $F_{g}^{0}$ quartet as ground state, the resulting fit parameters $W$, $x$, $\gamma$, and $H_m$ were found to be for $y = 0.2$:

$W = -0.23\, \text{meV}, \quad x = 0.73, \quad \gamma = 3.0\, \text{meV}, \quad H_m = 100\, \text{kG},$

and for $y = 0.3$:

$W = -0.206\, \text{meV}, \quad x = 0.721, \quad \gamma = 3.1\, \text{meV}, \quad H_m = 170\, \text{kG}.$

With these parameters in mind, the peak observed at both 5 K spectra can be interpreted as a transverse $J$-transition from the ground to sixth excited state. Comparing present results with those obtained for pure NdP [3], we notice that the overall splitting decreases slightly when Nd ions are partially substituted by uranium ions. This is an unexpected result. Another striking fact is strong temperature dependence of the linewidth observed for the composition Nd$_{0.7}$E$_{0.3}$P. Clearly further neutron scattering measurements are needed for a fuller understanding of this very interesting system.
References


