# Papers

## B10

### Spectroscopic Properties of $Nd^{3+}$ in $Cs_2Na(La,Nd)Cl_6$

W. STRĘK, C. SZAFRAŃSKI, J. HANUZA, Z. MAZURAK
and B. JEŻOWSKA-TRZEBIATOWSKA*

*Institute for Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 937, Wrocław 50950, Poland*

The results of the absorption, infrared, Raman and fluorescence spectra are reported. Owing the analysis of vibrational levels based upon the IR and Raman spectra the assignment of vibronic peaks of fluorescence spectra is proposed. The anomalous concentration dependence of fluorescence decay rates is found. A discussion of the possible mechanism responsible for concentration quenching of $Nd^{3+}$ fluorescence in the $Cs_2Na(La,Nd)Cl_6$ crystal indicates that at low temperature the non-resonant cross relaxation occurs whereas at room temperature both the non-resonant cross relaxation and the anti-Stokes resonant cross relaxation contribute in the Process.

## B11

### Magnetic Circular Dichroism of Rare Earths and $UO_2^{++}$ Complexes

C. GÖRLLER-WALRAND

*Lab. voor anal. en anorg. scheikunde-Celestijnenlaan 200F, 3030 Heverlee, Belgium*

## B12

### Quasi-relativistic SCF-Xα Scattered-wave Calculation of Uranocene, Thorocene and Cerocene

N. RÖSCH

*Lehrstuhl für Theoretische Chemie der Technischen, Universität München, Munich, F.R.G.*

## B13

### Evidence for 1D Coupling in the Neutron Spectroscopy Study of Uranium Trihalides

A. MURASIK

*Institute of Atomic Energy Swierk, 05-400 Otwock, Poland*

Published data of magnetic susceptibilities for $UBr_3$ and $UCl_3$ show apparent but rather broad maxima occurring at 15 and 22 K respectively. They were interpreted as due to cooperative magnetic transitions [1]. However, recent neutron scattering experiments performed at low temperatures on polycrystalline samples of $UBr_3$ and $UCl_3$ have proved unambiguously that the long range 3D antiferromagnetic ordering occurs at temperatures ($UBr_3$: $T_N < 4.2$ K, $UCl_3$: $T_N = 6.5$ K) which lie substantially below the values anticipated from magnetic susceptibility measurements [2].

This highly unusual behaviour can be explained by the suggestion that the magnetic exchange interaction is strong within linear chains of uranium ions lying parallel to the *c* axis of the crystal, and much weaker between uranium ions lying in different chains. The above hypothesis has interesting consequences in the case when intrachain coupling between nearest neighbours is antiferromagnetic, since the dominance of the nearest-neighbour magnetic interaction will result in the ions ordering in antiferromagnetic linear chains along *c* axis. This by itself does not lead directly to a long-range-ordered structure, and by symmetry argument appears to preclude any simple magnetic ordering.

Consequently, we may expect these compounds to behave as antiferromagnetic linear chains over a considerable range of temperature. The magnetic susceptibility maxima at 15 K and 22 K observed for $UBr_3$ and $UCl_3$ may be interpreted as due to exchange interactions within the chains, whereas at sufficiently low temperature, interchain interactions become important. They are expected to be largely, if not entirely dipolar in character, and tend to align the ions which lie in planes perpendicular to the *c* axis, into what is equivalent to the highly degenerate antiferromagnetic triangular net arrangement [3]. In fact, neutron experiments performed at $T < T_N$ revealed in both compounds a rather complex non collinear magnetic structure. However, the theoretical interpretation of the ordered state is found to be complicated by the unusual symmetry of the lattice.

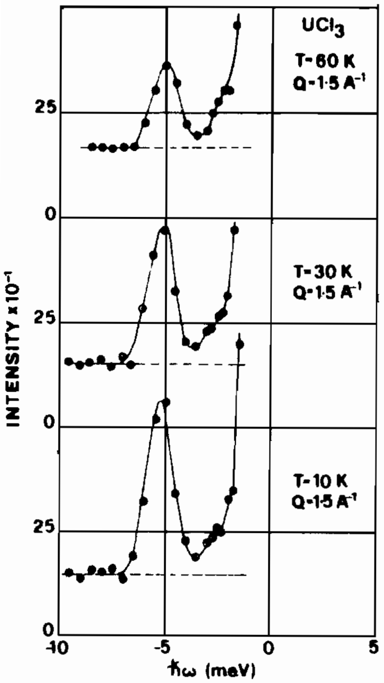

Fig. 1. Energy spectra of neutrons scattered from 1D antiferromagnet $UCl_3$ at various temperatures. The curve is drawn as a guide to the eye.

In spite of the fact that both substances appear to be a good example of 1D system, the hypothesis of the existence of linear antiferromagnetic chains in $UBr_3$ and $UCl_3$ needs direct experimental confirmation. Of course, for powdered sample the diffraction method is useless but the neutron spectroscopy can provide some valuable informations even if they are masked due to polycrystalline nature of the sample.

The measurements to be presented here were performed on a triple axis spectrometer at Wurenlingen working in the constant Q mode and scanning the incident neutron energy. The outgoing energy was fixed at 15 meV. Various energy loss configurations at different temperatures and with different incoming neutron energies were realized to cover energy transfers ħω up to 65 meV. To a first sight all observed lines of magnetic origin might be attributed to crystal-field transitions from the ground state to the excited states since the intensities of ground state transitions decrease with increasing temperature. On the other hand, recently published optical crystal spectra for $U_3$: in $LaCl_3$ preclude up to ~25 meV any CF excitation from the ground state [4]. Therefore the inelastic scattering of neutrons corresponding to energy transfers of ~5 meV and ~4 meV in $UCl_3$ and $UBr_3$ respectively, appears to have now a very simple explanation: most likely it corresponds to the excitation of 1D system. Such excitations are quasi 1D spin waves which propagate along individual chains.

Theory predicts that for the momentum components perpendicular to the chains these excitations are dispersionless, whereas along the chain direction, dispersion relation should follow a perfect sine curve. In powder measurements possible dispersion effects are most likely smeared out due to the polycrystalline nature of the sample. From Fig. 1 it is seen that as the temperature is raised, the 5 meV excitation in $UCl_3$ decreases rapidly, but there is no appreciable change either in peak position or in its linewidth. The same feature was noticed in earlier measurements on $UBr_3$ where damping of the 4 meV excitation was even more severe, but also no apparent change in its halfwidth was detected even at 100 K. This result is not quite surprising as might first appear, since as a consequence of the low dimensionality, the length over which the spins are correlated will be larger than in 3D systems at the same temperature relative to $T_N$ or $T_c$. As a result, in Ising systems well defined excitations were detected at temperatures as high as 10 $T_N$ [5].

The spin wave linewidth in 1D system is mainly due to the finite correlation length and the excitations which are very sharp for $T < T_N$ and usually limited only by the instrumental resolution, broaden rapidly with increasing temperature. This fact is in sharp contrast with our measurements and represents a major puzzle that is not understood. Clearly more systematic study, possibly on the single crystals would be most welcome.

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## B14

# The Quantum Chemical Interpretation for the Cation–Cation Interactions

G. V. IONOVA*, V. G. PERSHINA and V. I. SPITSYN

*Institution of the Physical Chemistry of the U.S.S.R. Academy, 117071 Moscow, Leninsky prospect 31, U.S.S.R.*

The pentavalent actinides $AnO_2^+$ where An = U, Np, Pu, and Am show the interaction with some active cations like $UO_2^{2+}$, $Fe^{3+}$, $Th^{4+}$, $In^{3+}$ etc. [1–5].