

Coherent neutron analysis of diffuse scattering in an alkane–urea composite

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Abstract

Dynamics of the guest alkane sublattice of the urea–alkane incommensurate composite is studied via its coherent diffuse scattering. © 2000 Elsevier Science B.V. All rights reserved.

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Incommensurate composite crystals are made of at least two interpenetrating lattices with irrational periodicities in at least one direction [1,2]. Urea inclusion compounds constitute a very interesting family of molecular composite structure [3–5]. Urea molecules build up a host substructure of hexagonal infinite and parallel channels, the diameter of which (ca. 5.25 Å) can accommodate linear guest molecules such as n-alkanes. Their incommensurability lies only along the channel direction. Intermodulation features appear in diffraction measurements [6,7] via the existence of satellites in addition to the common reflections and the two sets of main reflections associated with each sublattice. Urea inclusion compounds present a structural phase transition on going from a high symmetry to a low symmetry superspace group. Considering both sublattices independently, a simple description has been given for this phase transition [3–5,8]: whereas the urea hexagonal sublattice distorts towards an orthorhombic structure, the alkane molecules choose a preferentially orientational order.

The topic presented here concerns the ordering process of the alkane guest sublattice through the analysis of the

diffuse scattering phenomena in the n-nonadecane/urea inclusion compound. Two types of diffuse scattering are associated with the n-alkane guest subsystem [3,4]. The first one, called s-type, forms sharp reciprocal planes perpendicular to the channel axis c^* . This s-diffuse scattering is assigned to the one-dimensional ordering of the guest molecules along c^* . The second one, called d-type, is generated by rotational ($\delta\varphi$) and translational (δz) decorrelations of the guest molecules in adjacent channels. With coherent neutron scattering on a fully deuterated single crystal, we have shown that this scattering is totally inelastic at room temperature [9]. The elastic scattering comes only from the incoherent scattering process.

A temperature study of these diffuse plans was performed using the high-resolution time-of-flight spectrometer NEAT at HMI. Results obtained at room temperature and in the low-temperature phase are presented in Fig. 1. At room temperature, four features can be identified: a quasielastic coherent scattering component described here by a single Lorentzian function, two inelastic coherent modes and the elastic incoherent scattering. The first feature reveals the dynamical nature of the diffuse scattering. The second one is an evidence of low-energy damped modes associated with the alkane molecules. These modes were previously reported by

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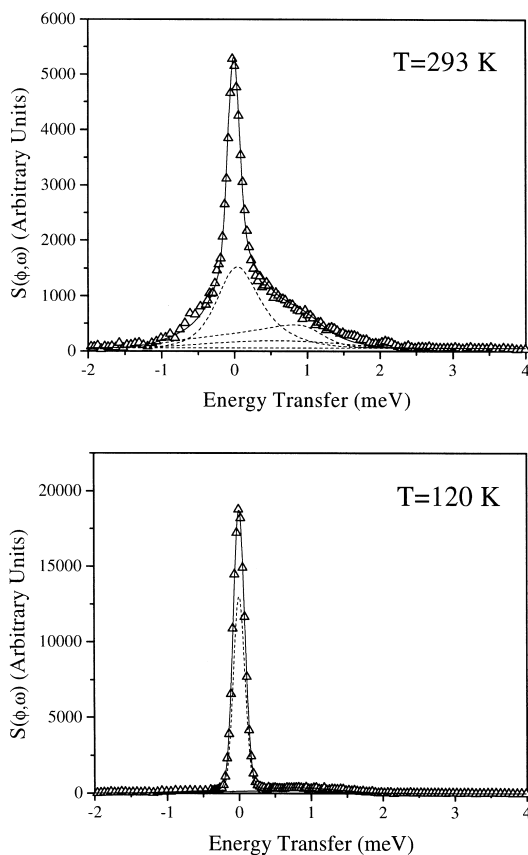


Fig. 1. Coherent neutron scattering energy analysis of the d-diffuse scattering of deuterated urea-nonadecane, at 293 and 120 K (NEAT, H.M.I.).

incoherent neutron scattering measurements as an excess in the phonon DOS [10]. These measurements performed on crystals clearly attributed the polarisation of these modes as being along the channel direction. Our

coherent neutron measurements demonstrate that these modes are located on the broad diffuse d-plane in reciprocal space. At low temperature in the orthorhombic phase, the d-diffuse scattering is elastic, but the inelastic modes are still observed. It follows that the guest subsystem is still disordered even in the low-symmetry phase.

In conclusion, our coherent neutron scattering measurements give a unique opportunity to study the diffuse scattering previously reported by X-ray diffraction in inclusion compounds. Further measurements and calculations are in progress, in order to characterise this disorder and these excitations in the frame of such an incommensurate composite.

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