# Low-Temperature Neutron Diffraction in the (Pb,Cd)Te Solid Solution Containing 2.2% of Cd

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The (Pb,Cd)Te solid solution forms a metastable single crystals for a solution containing more than 1% of CdTe due to its very low solubility in PbTe. A possible presence of a phase transitions at low temperatures (like, e.g., that occurring in the (Pb,Ge)Te solid solution starting from 1% of Ge) which could influence the physical properties of (Pb,Cd)Te system was never checked. In order to verify this possibility the neutron diffraction studies were performed in the temperature range from 21 K to 39 K in the single crystal of (Pb,Cd)Te solid solution with 2.2% of CdTe grown by self-selecting vapour growth method and characterized by powder X-ray diffraction. The (Pb,Cd)Te sample was measured using the four-circle diffractometer installed at a hot neutron source ($\lambda = 0.83$ Å). The possible transition does not take place to the lowest investigated temperature and the FWHM of Bragg peaks remains fixed in all temperature range under studies.

## 1. Introduction and motivation

The PbTe is a well-known IV–VI type semiconductor, serving e.g. as a basis for thermoelectric devices or optoelectronic applications in the infrared spectral range. The first information on its phonon dispersion appeared more than fifty years ago [1], the softening of the TO-mode with decrease of temperature in this compound was also reported a long time ago [2]. The phonon dispersion presented in [1] was commonly accepted for several dozen years. Over the last few years studies devoted to the PbTe lattice dynamics attract again a lot of attention due to a strong anharmonicity of this compound and possible modifications of its phonon dispersion [3, 4]. This semiconductor is an incipient ferroelectric which can easily be transformed into a real one by the doping with Ge. The transition temperature corresponding to the cubic to trigonal phase transformation of (Pb,Ge)Te decreases along with decrease of Ge content in the solid solution [5]. According to the estimates given in [5] the transition temperature reaches a value of 0 K when only about 1% of Pb atoms is replaced by Ge atoms. What should be pointed out is that the decrease above mentioned depends on the crystal composition in a non-monotonous manner. The phase transition temperature decreases with decrease of Ge content in the solid solution and is much more rapid in the composition range corresponding to small amount of Ge than that observed for a higher crystal compositions. Roughly speaking the slope of the relevant curve at

the phase diagram changes when about 10% of Pb is replaced by Ge. The temperature dependence of the chemical composition corresponding to the phase transition observed in the (Pb,Ge)Te differs qualitatively from that determined for the (Sn,Ge)Te, where the dependence under discussion is almost linear (see [5] and the references).

A PbTe alloying with other binary telluride is a well-known method to improve selected properties of a crystal (like, e.g., to modify such mechanical property as a hardness, the parameter important for various applications). Apart from the (Pb,Ge)Te solid solution a few other materials grown on the basis of PbTe are known and intensively investigated, among them the (Pb,Cd)Te system (see, e.g., [6–11] and references). Due to a low CdTe solubility limit in the PbTe (about 1% under the thermal equilibrium conditions at room temperature) this solid solution can be considered as a metastable crystal for a higher CdTe content. This limit results from quite different symmetry of crystal lattice corresponding to two compounds forming the solution (PbTe and CdTe). Moreover, it should be mentioned that the length of Pb–Te and Cd–Te bonds is also quite different in PbTe and CdTe, respectively, so it can be expected that Cd in the rock salt type (Pb,Cd)Te solid solution is not in its "natural" position. Taking into account a particular character of the (Pb,Cd)Te solid solution it is not clear: is a symmetry lowering possible at some temperature or the crystal structure of this system remains of the rock salt type independently of its chemical composition and temperature? According to our former results [7] the system containing a few percent of CdTe exhibits a rock salt structure in a wide temperature range, but to our best knowledge a low-temperature area of the phase diagram has not been explored till now except the PbTe

case [3, 12]. A possible phase transition taking place in the (Pb,Cd)Te could clearly modify a lot of physical properties of this system.

The goal of the present investigations was to look for possible (Pb,Cd)Te structure phase transition at helium temperatures for a solid solution containing only a few percent of CdTe. The neutron diffraction has been selected as an experimental method serving for that purpose.

## 2. Experimental details

The (Pb,Cd)Te solid solution ingot containing about 2.2% of CdTe has been grown at the Institute of Physics, PAS, by SSVG method ([6, 13], see also [14]). After the growth a part of the crystal was powdered and its structure quality was checked by X-ray diffraction (XRD) measurements with the help of a laboratory diffractometer (X'Pert PRO Alpha-1 MPD, PANalytical) and Cu $K_{\alpha_1}$ radiation. The chemical composition of the solid solution was determined using the lattice parameter dependence on the composition given in [6]. The small single crystal in the form of a cube and the volume of about 2 mm$^3$ selected for neutron diffraction studies was carefully oriented and placed in a closed cycle helium cryostat. The neutron diffraction measurements were performed at Laboratoire Léon Brillouin with the use of 5C2 four-circle diffractometer installed at the hot neutron source. This particular diffractometer is dedicated to measurements of the Bragg reflections in single crystals in order to evaluate the structure factors. The short wavelength of incident neutron beam ($\lambda = 0.83$ Å) used in the present studies and a small (Pb,Cd)Te unit cell allows to obtain a very precise information on possible thermal displacement parameters corresponding to investigated sample. The maximum resolution of the experimental setup was better than 0.2 deg. The neutron diffraction measurements were performed at few temperatures in the low temperature range from 21 K to 39 K, a $^3$He position sensitive neutron detector was applied.

## 3. Results and discussion

The analysis of powder X-ray diffraction pattern taken at room temperature has shown that measured material exhibits a rock salt type crystallographic structure, possible supplementary phases or precipitates have not been found. With the aim of determination of the absolute lattice parameter and the chemical composition of the solid solution alumina internal standard (SRM 676, NIST) was used. Figure 1 shows the result of XRD analysis by the Rietveld refinement method (fitting was performed with the FullProf.2k program) [15]. The experimental data are indicated by dots and the calculated ones by the solid line. The short vertical ticks mark the positions of the Bragg reflections (upper line — (Pb,Cd)Te crystal, lower line — Al$_2$O$_3$ standard). The lower curve shows the difference between the observed and calculated powder diffraction patterns. The refined lattice parameter is 6.45684(6) Å.
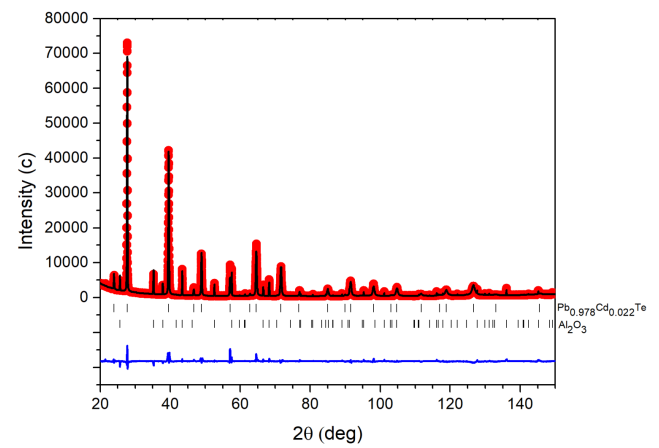


Fig. 1. The powder X-ray diffraction pattern accumulated in (Pb,Cd)Te solid solution with 2.2% of CdTe at room temperature. The results of the Rietveld refinement demonstrated a perfect rock salt type structure of investigated crystal.
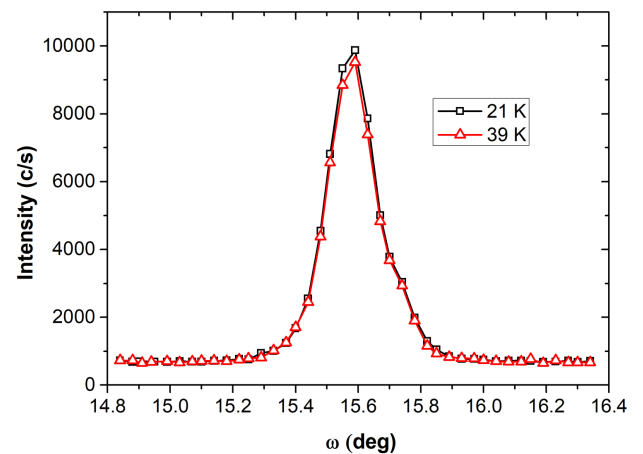


Fig. 2. The form of the 331 Bragg peak determined by the neutron diffraction at 21 K and 39 K.

As the result of neutron diffraction measurements a piece of information on a scattered neutron intensity distribution around selected Bragg reflections in the reciprocal space at given temperature and on possible temperature evolution of the form of such peaks was obtained. Figure 2 presents the form of 331 Bragg peak, determined in such a manner in the single (Pb,Cd)Te crystal by the neutron diffraction performed at low temperatures. This form does not depend on temperature, at least in the temperature range under studies and the full-width at half maximum (FWHM) of this peak remains constant (the results for only two selected temperatures are shown in this figure). A lack of the temperature evolution of the form of peak was observed in a few other analyzed Bragg peaks as well. Under the circumstances a possibility of the phase transitions, which could take place at low temperatures in the (Pb,Cd)Te solid solution containing 2.2% of CdTe is excluded. More experimental data of

the same kind, to be taken in solid solution crystals with a similar CdTe content and at still lower temperatures are clearly required for the confirmation of present findings and for more general conclusions. This work is in a progress now and the results will be soon send for a publication.

## 4. Conclusions

The (Pb,Cd)Te solid solution crystal with 2.2% of CdTe grown by self-selecting vapour growth method was measured by the neutron diffraction at low temperatures (below 40 K). The analysis of the form of a few Bragg reflections demonstrated a lack of temperature modifications of their FWHM up to 21 K. The obtained results suggest that possible phase transition, e.g. similar to that observed in the (Pb,Ge)Te does not occur in the (Pb,Cd)Te and the latter solid solution exhibits a rock salt type structure also at low temperatures.

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