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Deuteron and Helium Ion Channeling in Uranium Carbide

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The channeling of 1 MeV He^+-ions, of 400 keV protons, and of deuterons of energies between 1.0 and 1.25 MeV in UC single crystals has been studied. Wide-angle scattering off uranium atoms and the nuclear reaction C-12 (d, p) C-13 have been used to investigate the interaction of the ion beam with uranium and carbon atoms separately. The observed critical angles for channeling along the ⟨100⟩ axis are about 25% smaller than the theoretical predictions for ψ_1. However, when considering the influence of lattice vibrations, agreement with theory is achieved.

1. Introduction

Channeling studies have so far mainly been performed with monatomic crystals. With increasing application of the channeling technique in different solid state investigations, a growing interest arises to study channeling in polyatomic substances as well. Following the early preliminary experiments on CaF_2, NaCl, KCl and UO_2 [1-7], studies on the compound semiconductors GaP, GaAs, GaSb [8-11] and SiC [12] were performed. Later, the channeling properties of UO_2 [13, 14], U_4O_9 [14], and CaF_2 [15] were studied more carefully, as well as those of some alkali halides [16-18], of MgO [18, 19], and of SiO_2 [20, 21]. The channeling technique was in addition used to study the formation of oxide layers on single crystals of uranium monocarbide, UC [22], and the position of rare gas atoms in UC single crystals [23].

The purpose of the present investigation is to present more channeling data on UC, a new material of nuclear interest of the NaCl type (cubic) structure with partly covalent and partly metallic bonds, and to measure independently the steering properties of each sublattice. For this purpose, UC single crystals were bombarded with high energy helium ions (to study the interaction with the U-atoms) and with deuterons (to study in addition the interaction with the C-atoms). Furthermore, results on the blocking of α-emission from nuclides embedded in UC single crystals were obtained.

The experimental results on critical angles and minimum aligned yields are compared with theoretical expectations.

2. Experimental

Most of the measurements were performed in the Chalk River Nuclear Laboratories using the 2 MeV Van de Graaff accelerator of the Research Chemistry Branch. A few additional experiments were done at the University of Aarhus using the heavy ion accelerator [24] of the Institute of Physics. The target chambers and the set-up of goniometers and detectors have been described in the literature [2, 25]. The target could be rotated and tilted with respect to the beam with an accuracy of 0.02°.

The target was always kept at room temperature. Beam currents were varied in the range 0.5 to 10 nAmps. The beam spread was $\leqslant 0.05°$. Surface barrier detectors were used to measure backscattered particles and protons of the C-12 (d, p) C-13 reaction. The detector used for measuring these protons was placed close to the target to obtain counting rates comparable to those of Rutherford scattering, and was shielded with an aluminised Mylar window against backscattered deuterons. Due to their longer range, the protons easily penetrated the Mylar foil. Counting and energy analysis were performed with a set of single-channel analysers (for measuring yield curves at various depths) and a 100-channel analyser (for measuring energy spectra). The energy resolution was $\approx$ 15 keV FWHM for deuterons and protons, and $\approx$ 20 keV FWHM for helium ions.

The targets used were stoichiometric single crystals of UC, grown using a modified zone-melting technique [26]. The crystals were cleaved and the {100} cleavage faces exposed to the ion beam. For α-blocking experiments, U-232 was diffused into UC single crystals, and the α-particles emitted from U-232, and from its daughter substances Th-228, Ra-224, Em-220, Po-216, Bi-212, and Po-212 were counted selectively.

3. Theory

According to Lindhard's theory [27], the critical angle for axial channeling is given by

$$\psi_c = C\psi_1 \text{ with } \psi_1 = (2\, Z_1 \bar{Z}_2 e^2/E\bar{d})^{1/2} \quad \text{provided } \psi_1 < a/d \tag{1}$$

Z_1 and E are the atomic number and the energy of the projectile, and $\bar{Z}_2$ and $\bar{d}$ are the mean values of the atomic number and of the atomic spacing along the chosen row of lattice atoms; a is the Thomas-Fermi screening distance (≈ 0.1 Å for U-atoms and ≈ 0.2 Å for C-atoms). The condition $\psi_1 < a/d$ is always satisfied under the experimental conditions of the present study.

The constant C can be estimated from Andersen's extension of the theory of channelling [28] and the data for thermal vibrations in UC of Colella, Merlini *et al* [29, 30]. Because of the relatively large mean square displacement, the constant C is smaller than unity (about 0.75). In a perfect, non-vibrating lattice, C would be ≈ 1.7.

The minimum aligned yield, χ_{min}, *near the surface*, i.e. the counting yield for the aligned crystal, can be expressed, for clean and undamaged crystals, as [27]

$$\chi_{min} = \pi\, N\, d\, (r_{min})^2 \tag{2}$$

where N is the atomic density, d as above is the spacing along the chosen rows of atoms, and r_{min} the closest distance of approach of the channelled beam to the aligned row. A rough estimate for χ_{min} can be obtained by substituting the mean square vibrational amplitude ρ^2 for r^2_{min} in equation 2.

According to Davies *et al* [10], a correlation between χ_{min} and ψ_c exists

$$\chi_{min} \approx \pi\, N\, d \left\{ \frac{\pi a}{4} \left(\frac{\psi_1}{\psi_2} \right)^2 \right\}^2 \tag{2a}$$

Equations 2 and 2a yield similar values for χ_{min}: for equation 2, a value of 0.023 is obtained, whereas for equation 2a and e.g. 1 MeV helium ions, a value of 0.022 results.

4. Results and Discussion

All experiments were performed with the crystals either randomly orientated to the beam, or else the beam aligned with the ⟨100⟩ direction. Fig. 1 shows that the ⟨100⟩ channel in the rocksalt-structure, and hence in UC, consists of identical rows that contain both U and C atoms, alternately.

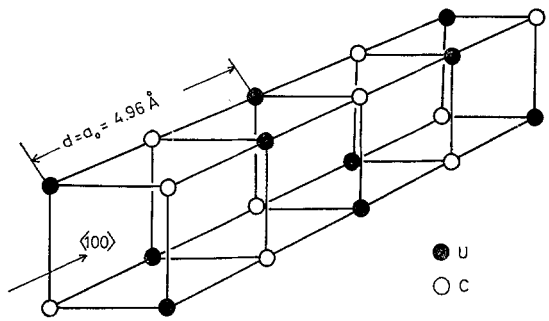


Figure 1 Schematic representation of a ⟨100⟩ channel in UC. The strings of atoms consist alternately of U and C atoms.

Fig. 2 shows energy spectra of backscattered helium ions and deuterons, with the beam incident along random and aligned directions. The figure illustrates the large reduction in yield that occurs whenever the ⟨100⟩ is aligned with the direction of the beam. Particles with the highest energy (i.e. at the spectrum edge) correspond to scattering from the U-atoms in the surface region. The small peaks at the spectrum edge are due to random scattering from a (polycrystalline or amorphous) thin oxide layer of about 30 Å thickness. (The growth of oxide layers on UC as studied with channeling techniques is discussed in detail in reference [22].) The edge of the spectrum for deuterons is at slightly higher energies than the edge for helium, since the energy loss in the scattering event increases with the mass of the projectiles: 1 MeV He^+-ions loose about 60 keV, 1 MeV deuterons loose about 30 keV while being scattered off the uranium atoms of the surface. The continuum at lower energies corresponds to scattering off U-atoms at progressively larger depths, due to a gradual loss of energy of the projectiles as they penetrate the crystal. With the known or estimated stopping power, one can convert the

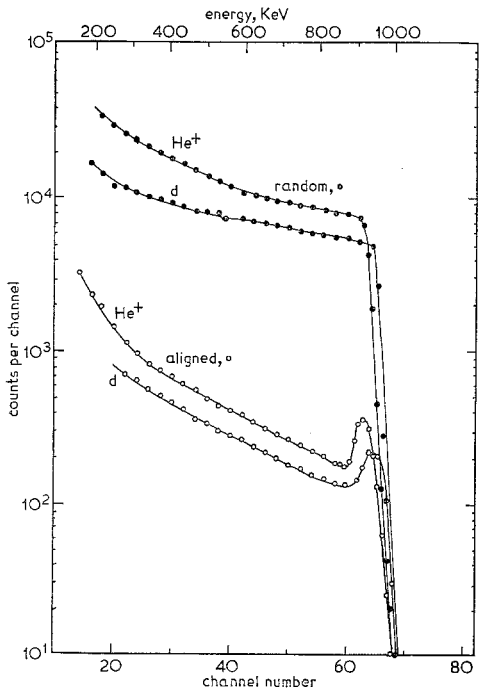


Figure 2 Energy spectra for 1 MeV He^+-ions and 1 MeV deuterons backscattered from UC single crystals, the beam being either incident along the ⟨100⟩ (aligned case, circles) or along a direction that does not coincide with any low index crystal axis or plane (random case, full dots). The small surface peaks in the aligned spectra indicate the existence of thin oxide layers on the UC surfaces.

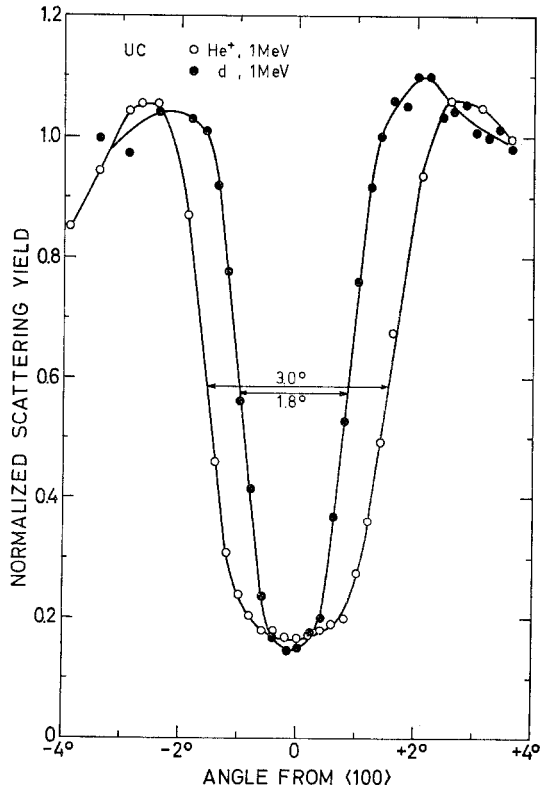


Figure 3 Orientation dependence of the normalised Rutherford scattering yield of 1 MeV deuterons and 1 MeV He^+-ions using the same crystal for both runs.

energy scale into a depth scale and hence study the channeling behaviour as a function of depth.

Since the energy loss during scattering for a given projectile decreases with the mass of the scattering atoms, and since the scattering cross section is proportional to Z_2^2, Rutherford scattering mainly reflects heavy atoms. The energy of the 1 MeV He^+-ions (deuterons) backscattered from the light carbon atoms is only 0.28 (0.54) MeV. Simultaneously, the cross section for scattering off carbon atoms is only 0.8 (1.2)% of that for scattering off uranium atoms. Therefore, the energy spectra for the backscattered He^+-ions and deuterons do not show any clear indication of a contribution of the carbon atoms.*

Fig. 3 shows a scan through ⟨100⟩, i.e. the orientation dependence of backscattering of 1 MeV deuterons and He^+-ions around ⟨100⟩. To measure the yield curves, the crystals were titled in a plane that did not coincide with any low index plane. A single channel analyser was used to measure the backscattered particles in the energy region 0.80 to 0.85 MeV, hence the region just behind the surface peaks of fig. 2 (channels 55-58). Due to the charge difference of 2 between deuterons and He^+-ions, the angular width of the yield curve for deuterons should, according to equation 1, be smaller than that for He^+-ions by a factor of $\sqrt{2}$. Experimentally, a slightly bigger factor of 1.66 was found. The crystal had been exposed to the ambient for about 1 d before the channeling experiment and therefore had an (amorphous or polycrystalline) oxide skin of about 30 Å. Such a disordered layer is known to increase the counting yield for the aligned crystal, the minimum aligned yield, χ_{min}, and helps to explain the fairly high values of 0.145 and 0.165 observed with this crystal.

Fig. 4 shows similar yield curves for a freshly cleaved crystal that did not have such a disordered surface layer. The χ_{min} for backscattered He^+-ions of 0.027 only is, therefore, smaller than the χ_{min} values in fig. 3. As mentioned above, backscattering and hence the two curves of fig. 3 and the curve for He in fig. 4

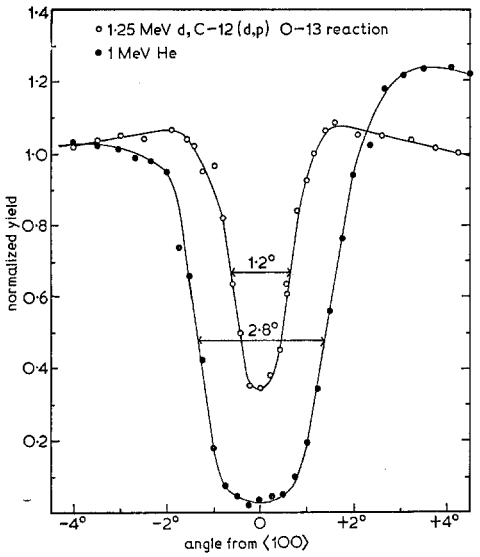


Figure 4 Orientation dependence of the normalised Rutherford scattering yield of 1 MeV He^+-ions and normalised proton yield from the C-12 (d, p) C-13 reaction induced with 1.25 MeV deuterons.

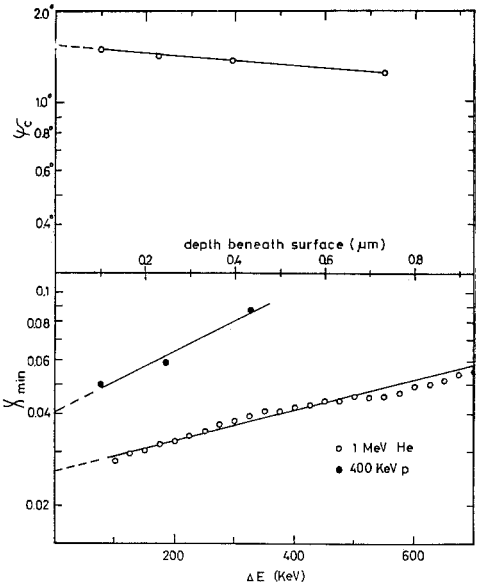


Figure 5 Depth dependence of χ_{min} and ψ_c for 1 MeV He^+-ions and 400 keV protons, expressed in terms of energy loss, ΔE that the beam has undergone in the crystal (for He^+-ions, lower scale) or in terms of depth of the scattering zone in μm (for He^+-ions and for protons, upper scale).

reflect the heavy ions and therefore the *uranium sublattice*. The C-12 (d, p) C-13 reaction, however, gives specific information on the *carbon sublattice*. For the (d, p) reaction, χ_{min} is 0.35 and hence much bigger than χ_{min} for backscattering off uranium atoms, measured on the same crystal.

The critical angle, ψ_c, for the (d, p) reaction of 0.6° is smaller than that for backscattering of deuterons (fig. 3, $\psi_c = 0.9°$), even if corrected for the higher energy used: 0.6° at 1.25 MeV energy would correspond to about 0.7° at 1 MeV energy (equation 1). Since in the ⟨100⟩ axis, all the atomic rows are identical, there should be only one single value of r_{min} involved in equation 2 and, therefore, both backscattering and the (d, p) reaction should exhibit the same χ_{min} and ψ_c values. There are two possible explanations for the observed discrepancy:
(*i*) The thickness of the analysed layer and its distance from the surface is not as clearly defined for the (d, p) reaction as for scattering because of the energy dependence of the cross section of the C-12 (d, p) C-13 reaction. Therefore, the reaction may have occurred in greater depth than backscattering. However, a yield curve very similar to that shown in fig. 4 was obtained for deuterons of 1.15 MeV energy.

(*ii*) The disorder in the carbon sublattice is higher by some orders of magnitude and is much easier to quench than the disorder in the uranium sublattice [31, 32]. Most probably, both facts contributed to the observed discrepancy. Similar observations have been made with deuteron backscattering and the O-16 (d, p) O-17 reaction in UO_2 and U_4O_9 [13, 14].

Fig. 5 shows the effect of the depth of the scattering zone (or of the energy loss of the particle) on ψ_c and χ_{min}. The depth scale was calculated from the measured energy loss using values of the stopping power, estimated as shown in reference [22]. Since the stopping power is different for helium and protons, the depth scale refers to both types of particles, whereas the energy scale is shown for He^+-ions only. ψ_c decreases and χ_{min} increases with increasing depth due to dechanneling of some of the particles along their trajectory. However, even deep in the crystal, χ_{min} is still $\lesssim 0.1$. Extrapolation to the surface yields values that are not affected by depth effects: for 1 MeV He^+-ions, ψ_c is 1.55° and χ_{min} is 0.026. The experimentally observed χ_{min} is therefore in

*If higher energy particles were used, the scattering intensity of carbon would be much higher due to resonance scattering; for e.g. 6 MeV, the intensity could be similar to the scattering intensity of uranium.

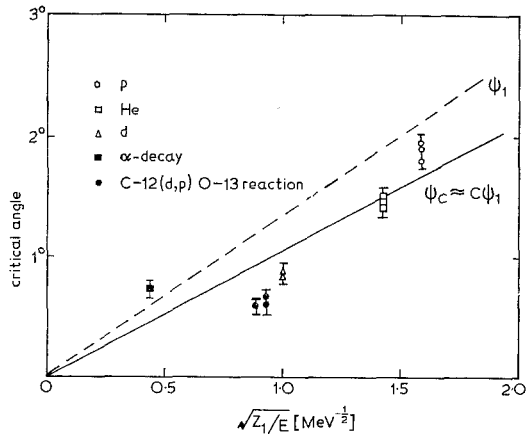


Figure 6 Comparison of experimental values of ψ_c with the values of ψ_1 predicted according to Lindhard, for various projectiles and energies. The orientation of the UC crystals was always ⟨100⟩.

good agreement with the expected value of 0.022 to 0.023 (section 3).

Fig. 6 gives a comparison between the experimentally determined values of ψ_c and the predictions for ψ_1 according to Lindhard [27], shown as dashed line. The experimental data are for backscattering of 1 MeV deuterons and He^+-ions and of 400 keV protons, for the C-12 (d, p) C-13 reaction with 1.15 and 1.25 MeV deuterons, and for the emission of 5.32 MeV α-particles during the decay of U-232 that had been diffused into a UC single crystal. Despite scatter, the full line is thought to represent the experimental data which generally are smaller than the predicted ψ_1 values. However, applying Andersen's extended theory of channeling [28] to allow for thermal vibrations, i.e. by introducing the constant C of section 3 ($C \approx 0.75$), good agreement is achieved between experiment and theory. A similar agreement within about 10% between theory and experiment has previously been reported for *monatomic* face-centred-cubic and body-centred-cubic crystals [2, 25] whereas large discrepancies of 20 to 50% ($\psi_c \ll \psi_1$) have been observed in UO_2, U_4O_9 [13, 14], and in several diamond-type substances – namely in diamond, Si, Ge, GaP, GaAs, GaSb [10] as well as in SiC of the 6 H type [12]. Eriksson and Davies suggested [13] that this discrepancy might be associated with the non-symmetrical atomic spacing that exists in these substances. Such non-symmetry might introduce anisotropy into the various modes of lattice vibration, and this in turn might significantly affect the critical channeling angle. The present study seems to confirm this suggestion, since it shows that the presence of two different kinds of atoms alone does not necessarily lead to unexpectedly small ψ_c-values.

Fig. 7 finally shows a blocking pattern obtained using a plastic nuclear particle track detector (cellulose nitrate, $H_6C_{12}O_{18}N_4$, see e.g. [33]. The plastic film was mounted on a brass plate and placed in the target chamber. Both the film and the plate had a central hole to allow the beam of 1 MeV He^+-ions to pass. During the exposure of 15 min, the ⟨100⟩ direction of the UC target was nearly parallel to the beam. Following exposure, the blocking pattern was processed by etching the film in a hot solution of NaOH. Fig. 7a shows the pattern as observed on the film, fig. 7b explains the main features. For a better presentation, the latter pattern was tilted by about 45° with respect to fig. 7a. The images of the various more important crystal directions and planes have been indexed. Higher index directions and planes can easily be seen as well, but were not indexed for clarity.

Conclusions and Summary

The present study provides channeling data for a cubic compound material of the rocksalt structure, i.e. uranium monocarbide, UC, a substance of nuclear interest. Three different methods were employed:

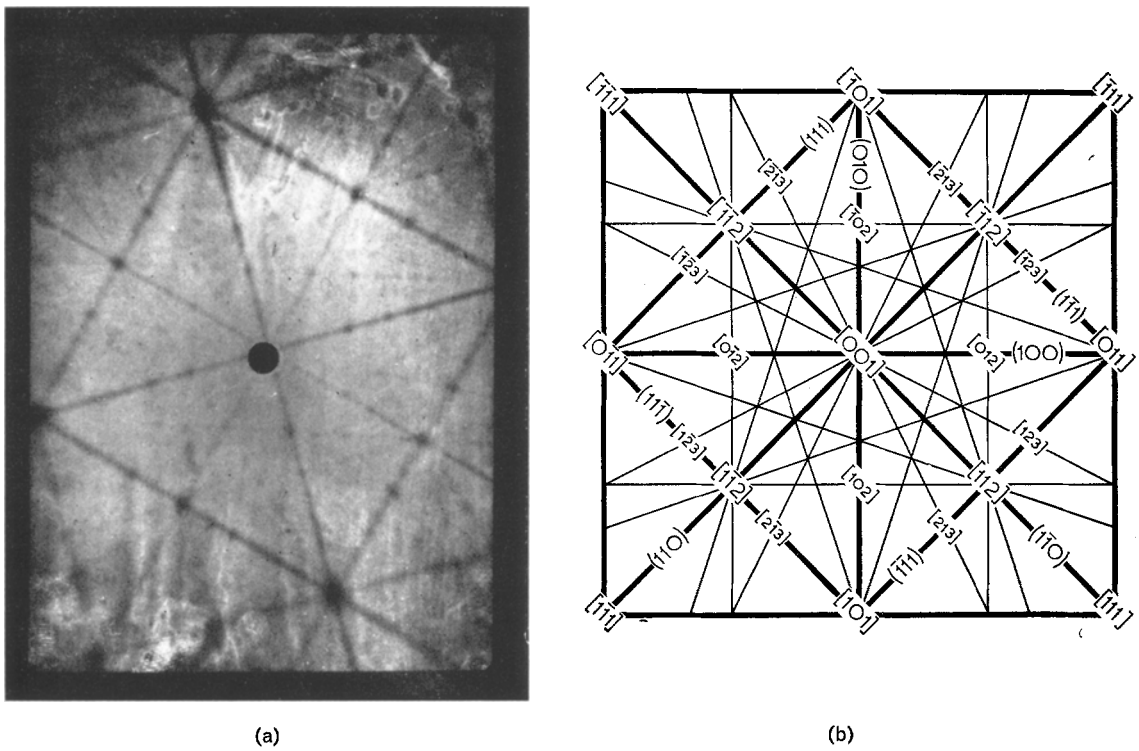
(a) Rutherford backscattering of protons, deuterons, and helium-ions was used to investigate the heavy component, i.e. the uranium atoms.

(b) The light carbon atoms and their sublattice was studied with the aid of the C-12 (d, p) C-13 reaction.

(c) Blocking patterns were obtained with the α-emission of U-232 following diffusion of U-232 into the UC single crystals.

The values for the minimum aligned yield, χ_{min}, for freshly cleaved crystals, Rutherford backscattering and extrapolated to zero thickness ($\chi_{min}^{exp} = 0.026$) were in agreement with predictions ($\chi_{min}^{pred} = 0.022$). The critical angles for backscattering, ψ_c, were smaller by about 25% than the ψ_1-values calculated from Lindhard's theory. Good agreement, however, was achieved when corrections were made for thermal vibrations.

The agreement with theory, besides proving the good quality of the single crystals used, indicates that the large discrepancy between ψ_c and ψ_1 observed so far for the compound materials UO_2, U_4O_9, SiC, Si, Ge, GaP, GaAs



(a) (b)

Figure 7 Detection of 1 MeV He^+-ions backscattered from a UC single crystal using a plastic nuclear track detector for observation. Part (a) shows the blocking pattern for incidence of the beam near the ⟨100⟩ direction as obtained on the detector. Part (b) gives a schematic representation of the pattern together with the indices for the major directions and planes.

and GaSb might be due to the non-symmetrical atom distribution along the rows that cause channeling, and hence to anisotropy in lattice vibrations, as suggested by Eriksson and Davies [13]. Evidently, the presence of more than one kind of atom in the atomic rows alone does not lead to a similar discrepancy.

The χ_{min} and ψ_c-values determined for the carbon sublattice were essentially different from those for the uranium sublattice. χ_{min} was much bigger for carbon than it was for uranium and ψ_c tended to be small. Since in the ⟨100⟩ direction studied, the rows of atoms consist of alternately C- and U-atoms, identical values of ψ_c and χ_{min} would have been expected for the C and U sublattices. Therefore, the disorder in the carbon sublattice seems to be much more pronounced than the disorder in the uranium sublattice. A certain additional contribution to the observed difference will be due to the badly defined depth and thickness of the layer analysed by the (d, p) reaction, but this alone can probably not explain all of the difference since (*i*) variations in deuteron energy did not yield essential changes in χ_{min} and ψ_c (*ii*) both ψ_c and especially χ_{min} for the uranium sublattice did not vary very much with depth. In fact, even deep in the crystal, the χ_{min} for the uranium sublattice was much less than the χ_{min} determined for the carbon sublattice near to the surface. Therefore, most of the discrepancy is thought to be due to a higher disorder of the carbon atoms.

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