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Investigation of proton channeling using nuclear-reaction resonances

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Abstract. To investigate orientation effects, an approach based on the measurements of γ-ray yields following the excitation of "narrow" isolated resonances in the reactions occurring on the nuclei of interstitial impurity atoms, that occupy certain positions in a crystal, has been proposed. The carbon atoms were shown to be located in octahedral interstitial sites of the Re-0.4 at. % ^{13}C monocrystalline solution. The proton flux distribution in the (0001) channel was investigated via the 1.7476 MeV resonance of the ^{13}C(p,γ)^{14}N reaction. Some particular qualities of the reaction yield were found to be dependent upon the proton energy. The method of measurement of the electronic stopping power of channeled particles has been deduced. The γ-ray yield of the resonance reactions induced by the channeled protons was shown to be dependent on the amplitude of the thermal vibrations of carbon atoms.

PACS. 24.30.-v Resonance reactions – 61.85.+p Channeling phenomena (blocking, energy loss, etc.) – 68.55.Jk Structure and morphology; thickness; crystalline orientation and texture – 02.70.Uu Applications of Monte Carlo methods

1 Introduction

The overwhelming majority of data concerning orientation effects in crystals at a channeling of $(0.5, \ldots, 10)$ MeV charged particles were stored in the analysis of the yields of elastically scattered helium ions and protons which depend on energy and angle of the particle entrance into the channel, atomic charge of the crystal and the channeled particle, lattice type, crystallographic direction, etc. [1]. The trajectories of the particles which come to the crystal along low-index planes were investigated by several groups (see references in [2,3]). The scattered particle spectra show some special features (fine structure) at the high-energy edge. This is associated with the trajectory of ions, whose oscillation amplitude in the channel is close to half the distance between the planes. The probability of collision of these ions with nuclei considerably increases with their approach to the planes. The reliability of the results of measurements in this region is primarily determined by the energy resolution of the method. The resolution of the backscattering method is dependent on the intrinsical resolution of the spectrometer, the energy straggling during particle motion along the crystal, and the energy distribution of the probing incident beam. These causes limit energy scattering ion resolution and inevitably result in spectra smoothing and, following their structure smear, bring to difficulties and ambiguities in the interpretation of the results of measurements [2].

To investigate the fine structure, we offer an approach based on the use of radiation yields of isolated resonances excited with channeled particles on nuclei of interstitial (or substitutional) impurity atoms, which occupy certain positions in a crystal [4,5]. The proposed approach excludes the influence of the spectrometer resolution on the measured data. It allows to view the situation from a new perspective to research the peculiarity of channeled-particle flux formation. A new method of measurement of both electronic energy losses of channeled particles and amplitudes of thermal vibrations of interstitial impurity atoms in a crystal has been proposed on this approach basis.

A computer code simulating channeled-particle trajectories along the crystal and calculating nuclear-reaction yields was developed for the numerical analysis of this approach possibilities.

2 Excitation curve features of channeling-particle–induced resonance nuclear reactions

The characteristic property of many nuclear reactions is the presence of isolated resonances of radiation yield as a function of the incident energy.

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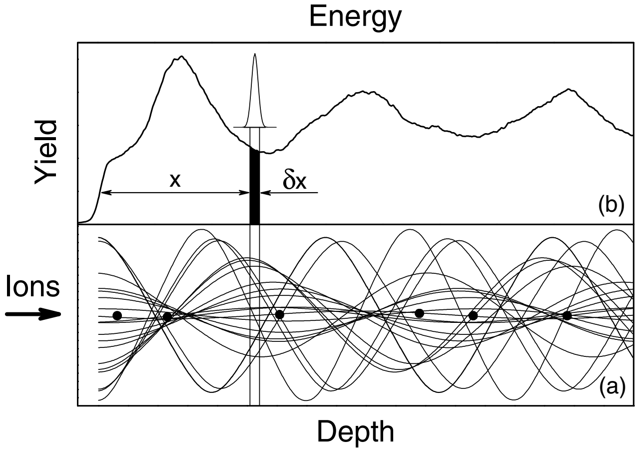

Fig. 1. Model approximation of the yield of the nuclear resonant reaction *versus* the channeled-ion energy: a) distribution of particle trajectories in the planar channel $\varphi_{in} = 0°$ (black circles show impurity atoms); b) yield of the resonant reaction on the nuclei of impurity atoms located in the center of the channel.

To determine the distribution profile of an element concentration for some depth of a sample particularly by means of the (p,γ)-reaction, the γ-ray yield has to be measured with a small increment of the incident proton energy [6–8].

The situation is radically changed if the resonance is excited with channeled particles. Here, the yield depends not only on the atom concentration as in the case of a polycrystalline target, but also on the crystallographic direction, the arrangement of reacting atoms in the crystal, the angle at which the particles enter into the channel, the electronic density distribution in the channel, etc.

Figure 1(a) shows the calculated proton trajectories for a beam coming to the channel along the atomic planes. Under the action of repulsion forces on the part of atoms lying in the planes, the particles start to oscillate; the proton flux becomes redistributed, the trajectories come close together or apart. Let us assume that the impurity atoms on which the reaction occurs are uniformly distributed in the crystal and are placed in the center of the channel. Obviously, the probability of channeled particles colliding with the impurity atoms at the depth "x", where the resonance is realized, depends on the particle flux distribution in the transverse plane of the channel. The reaction yield will have the highest value when the particles coming into the channel and having the energy $E_p > E_{res}$ experience slowing-down and reach E_{res} at the depth where the flux density in the center of the channel is maximum (fig. 1(b)).

In this case it is enough to register the number of interactions resulting in radiation emission as opposed to the scattering method that needs reaction product energy measurement. The influence of the spectrometer on the resolution is excluded. The straggling makes the resolution worse only in the "*in*" part of the ion trajectory. So the resolution of this method is better in comparison with the scattering one.

So if the atoms on which the resonance reaction occurs are distributed uniformly and occupy known sites in the crystal lattice, the measurement of the excitation curve contributes to ascertain the distribution of the channeled-particle flux, to investigate its dependence on the lattice type, crystallographic direction, stopping power and on the angle of the particle entrance into the channel etc.

The possibilities of the resonant reaction method provide a new approach for the investigation of orientation effects; make it possible i) to reveal a fine structure in the channeled-particle flux near the crystal surface, ii) to estimate the amplitude of thermal vibrations of impurity atoms in the lattice, iii) to determine electronic energy losses of channeled particles, etc.

Up to now resonance reactions have not been used to study channeling-particle flux features.

In the present experiments we have used the Re-0.4 at. % ^{13}C monocrystalline solution as object of study. As mentioned above, when using the resonance method, it is necessary to know the sites of impurity atoms in the crystal lattice. Therefore, before demonstrating the possibilities of resonant nuclear reactions for investigating the orientation effects, it was decided to carry out measurements and to obtain reliable experimental data on the localization of carbon atoms in the rhenium lattice.

3 Localization of carbon atoms in the Re-0.4 at. % ^{13}C monocrystalline solution

Some metals and alloys dissolve carbon in relatively large quantities. This element forms interstitial solid solutions. At concentrations not exceeding the solubility limit, the interstitial atoms occupy certain energy-favorable sites in the lattice. Among them, octahedral and tetrahedral voids are the most well-defined sites in hexagonal close-packed metals which may be occupied by interstitial atoms. Nuclear-physics methods in combination with the use of channeled particles and orientation effects can provide direct data on the sites of interstitial atoms in the crystal [9].

Single-crystal rhenium samples were used in the experiments. Disks of 1.5 mm thickness were cut out by the electrospark method perpendicularly to the $\langle 0001 \rangle$, (0001), $(1\bar{1}20)$ and $(1\bar{2}1\bar{2})$ crystal directions. In order to remove the faulted and damaged layers, the sample surfaces were subjected to grinding and polishing. A subsequent electrochemical etching of the samples removed the remains of the faulted layer.

Solid thermodiffusion in vacuum followed by quenching in helium atmosphere was used to saturate and homogenize the samples with carbon enriched in the ^{13}C isotope to concentrations of 0.4 at. %. Before saturation, the samples were annealed and degassed. After saturation, the samples were subjected to additional electropolishing. As a result of the operations, the samples had a mirror surface.

The samples were fixed on a goniometer head in the nuclear-reaction chamber. The goniometer provided for

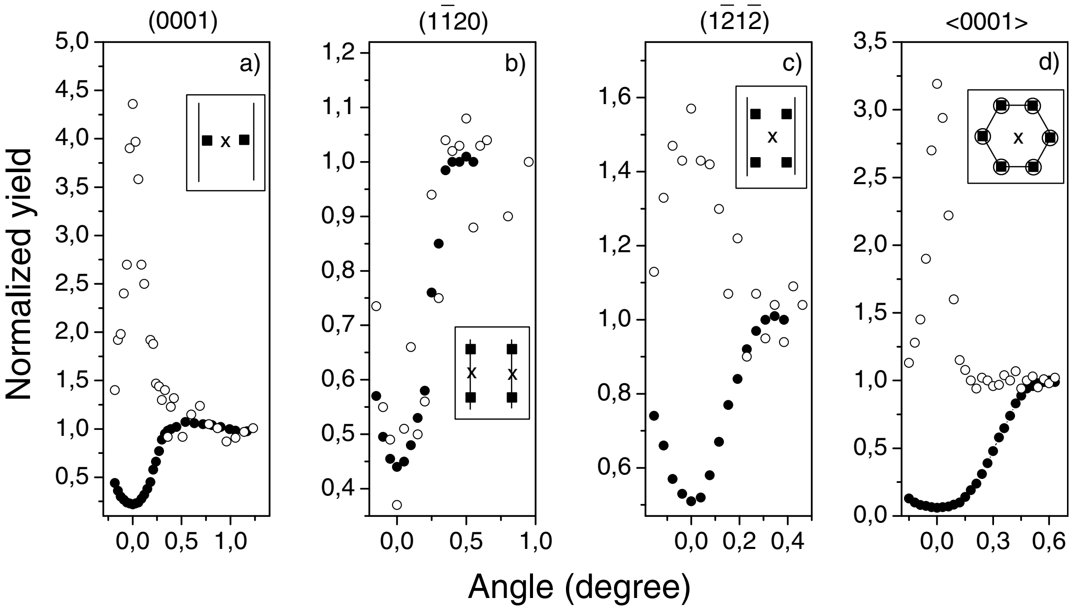

Fig. 2. The ^{13}C(p,γ)^{14}N reaction yield (white circles) and scattered protons (black circles) *versus* the angle relative to the chosen direction of close packing of the crystal: (0001), (1$\bar{1}$20), (1$\bar{2}$1$\bar{2}$), ⟨0001⟩.

crystal rotation about three mutually perpendicular axes, and also, for angular scanning at steps of 0.05° and 0.03°. The angular beam divergence was not more than 0.03°. The strong isolated resonance of the ^{13}C(p,γ)^{14}N reaction ($E_{res} = 1.7476$ MeV, $\Gamma_{res} = 135$ eV) [10] was used.

The spot of the incident proton beam has a size of 0.8 mm in diameter. The beam current equals 4 nA. The capture of the 1.7476 MeV proton with the ^{13}C nucleus results in the production of the ^{14}N* nucleus with an excitation energy of 9.1724 MeV [10]. The level is isolated, therefore the excitation curve of the reaction on the polycrystalline target should inevitably have a plateau. The γ-rays were counted by means of a $3'' \times 6''$ NaJ(Tl) detector. The detector was placed at a distance of 6 cm from the crystal at an angle of 90°. The optimum effect/background ratio is shown to be realized at a γ-ray detection in the energy range between 8.1 and 9.2 MeV. The background was equal to 32% in these conditions.

The protons were registered by means of a silicon surface barrier detector, mounted at a distance of 11 cm from the crystal at an angle of 158°. The target crystal was displaced after identification of the plane (0001) and the finishing measurements were performed at the non-irradiated area. To improve vacuum a screen, cooled down to liquid-nitrogen temperature, was installed close to the crystal. All the measurements were carried out at room temperature.

The yields of protons scattered by rhenium nuclei and the ^{13}C(p,γ)^{14}N reaction γ-ray were measured simultaneously at angle scanning near to the close-packed row ⟨0001⟩ and planes (0001), (1$\bar{1}$20), (1$\bar{2}$1$\bar{2}$) of the Re atoms. The yields of scattered protons and reaction γ-rays were normalized to the results of measurements at a complete disorientation of the crystal relative to the proton beam.

The statistical equilibrium of the flux appears at depths exceeding 1000 Å from the surface [11]. For this reason the angular scanning in the vicinity of crystal axis and planes as well as the measurements of the γ-ray yield and the yield of protons scattered by rhenium nuclei were carried out at an energy of 1.758 MeV, *i.e.*, above the energy values at which a fine structure of the scattered proton yield is observed.

Figure 2 shows the measured angular dependences of the γ-ray and scattered proton yields. If the beam pulse direction is coincident with the ⟨0001⟩ crystallographic direction, the proton yield gives only $\approx 3.0\%$ with respect to the normalized yield. This witnesses the high quality of the crystals used and a fair preparation of the specimens.

As the beam pulse direction becomes coincident with any of the directions shown in fig. 2, the yields of both γ-rays and scattered protons substantially change. This is explained firstly by the specific location of the ^{13}C atoms in rhenium, secondly, by the strong angular dependence of the distribution of protons channeled within the chosen direction. The inserts given in the figures show the projections of the rows (circles) and planes (lines) of the rhenium atoms, and also the projections of the octahedral (crosses) and tetrahedral (black squares) interstitials onto the plane normal to the corresponding axial or planar direction. The appearance of a strong narrow peak in the γ-ray yield, which is discovered when the beam direction is coincident with either the (0001) plane or the ⟨0001⟩ axis (see fig. 2 a, d), testifies that the impurity (^{13}C atoms) is located in the interstitial void, if carbon concentrations are lower than its solubility limit in rhenium. Indeed, if the carbon atoms occupy tetrahedral voids, the angular scanning in the vicinity of the mentioned axis and plane would show a decrease of the γ-ray yield at decreasing angle [1]. For the (1$\bar{1}$20) plane the angular dependence of the γ-ray yield is close to the angular dependence of the scattered proton yield. This directly testifies that the majority of the carbon atoms in the Re-0.4 at. % ^{13}C monocrystalline solution lies in the planes of octahedral interstitials.

4 Simulation of proton trajectories in the (0001) channel of the Re-0.4 at. % ^{13}C single crystal

The computer simulation code [12] and the experimental data on the location of carbon atoms in rhenium crystal were used to calculate the evolution of proton trajectories in the (0001) channel.

The energy losses for one step of the proton trajectory calculation in the transverse plane of the channel were simulated using the function

$$\Delta E_{(0001)}(y)=\begin{cases}\Delta E_{rand}-2h\dfrac{y}{D}, & \text{for } 0<y<\dfrac{D}{2},\\ \Delta E_{rand}-h, & \text{for } y=\dfrac{D}{2},\\ \Delta E_{rand}-2h\left(1-\dfrac{y}{D}\right), & \text{for } \dfrac{D}{2}<y<D,\end{cases}$$

where y is the particle entrance coordinate in the transverse plane of the channel, ΔE_{rand} is the energy loss in the amorphous media, D is the interplanar spacing, h is the energy loss decrease in the centre of the planar channel. The function $\Delta E_{(0001)}(y)$ is drawn as the solid thick line in fig. 3, where the two neighbouring planes of the channel are coincident with the left and right vertical axes of the plot.

Input data for calculations are the characteristics of the beam and the nuclear resonance of the $^{13}C(p,\gamma)^{14}N$ reaction at an energy of 1.7476 MeV, and also the parameters of the (0001) channel of the rhenium crystal and the ^{13}C atom data.

The angle between the beam pulse direction and the (0001) plane remained unchanged and was taken to be $\varphi_{in} = 0°$. The angular beam divergence was $\Delta\varphi_{in} = 0.03°$. The energy distribution of the probing incident beam did not exceed 200 eV.

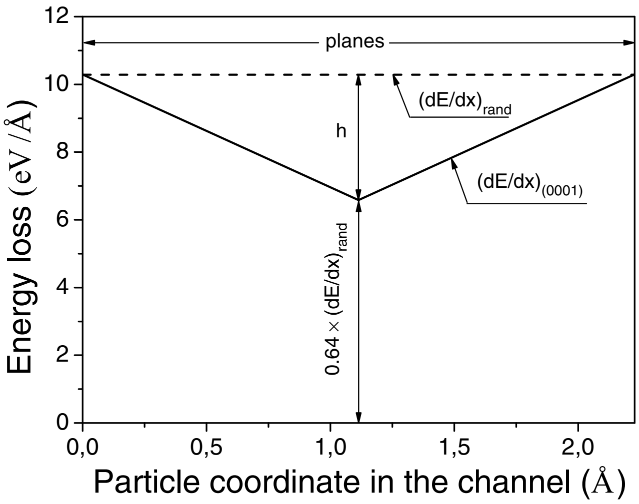

Fig. 3. Sketch of channeled-proton energy losses.

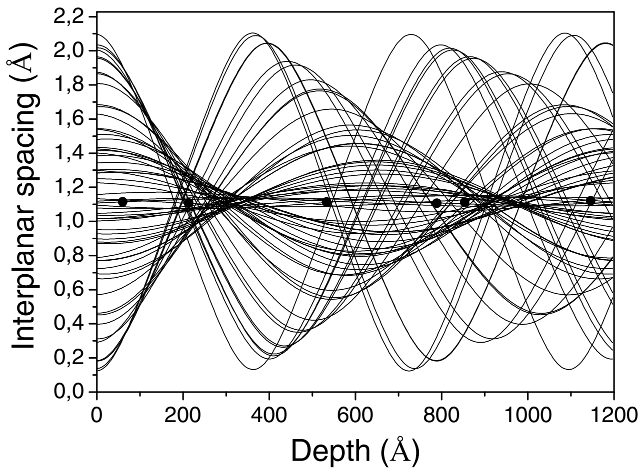

Fig. 4. Trajectory oscillations of 1.7476 MeV protons in the (0001) channel of the Re-0.4 at. % ^{13}C crystal. $\varphi_{in} = 0°$. The ^{13}C atoms in the center of the interstitial plane channel are shown as black points.

To calculate the forces acting on the proton in the channel, we used a thermally modified approximation of the averaged potential in the Molliere terms [13]. The potential of four planes nearest to the channeled particle was taken into account.

We have demonstrated above that the carbon atoms in rhenium occupy octahedral sites. In the hcp crystals (rhenium being among them) there are as many octainterstitials as atoms in the matrix. In the (0001) direction, the planes of rhenium atoms alternate with the planes of octainterstitials. The planes of interstitials where carbon atoms are found are arranged in the center between the atom planes.

Figure 4 shows the trajectories of a proton flux in the (0001) channel. The trajectory wavelength decreases with increasing amplitude. The slope of the trajectories is dependent on the distance between the proton entrance point into the channel and the nearest plane. Nothing can prevent the channeled protons from colliding with carbon atoms arranged in the octainterstitials (black circles in the center of the channel). Moreover, the probability of proton collisions with the atoms increases at the expense of flux redistribution in the channel [9]. Figure 5 illustrates some peculiarities of the dynamics of the proton flux distribution in the (0001) channel up to the first focus of trajectories for a variety of depth values. As a result of a series of soft correlated collisions with the atoms of the planes, protons having a large trajectory amplitudes give rise to peaks along the edges of the flux distribution. As the depth grows, the peaks approach to the center of the channel and form the maximum in the region of the first focus of the trajectories. As can be seen from fig. 5 in the middle of the channel, up to the maximum, the flux of channeled protons is uniform in the transverse plane, and is close to a constant value irrespective of the depth value.

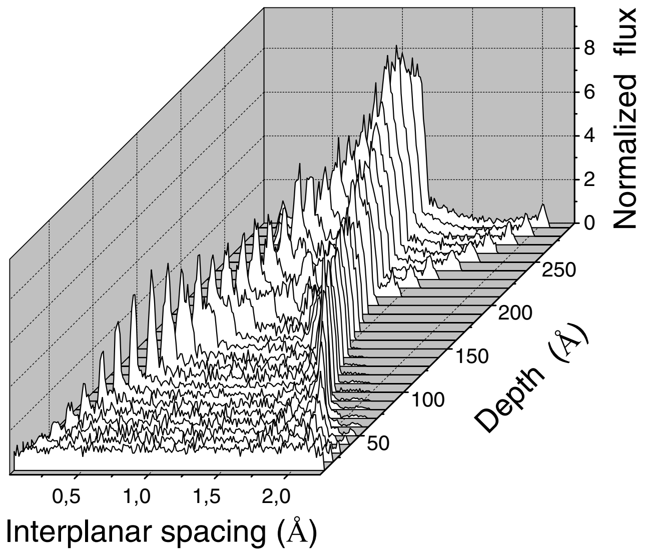

Fig. 5. Proton flux distributions in the transverse plane and along the (0001) channel.

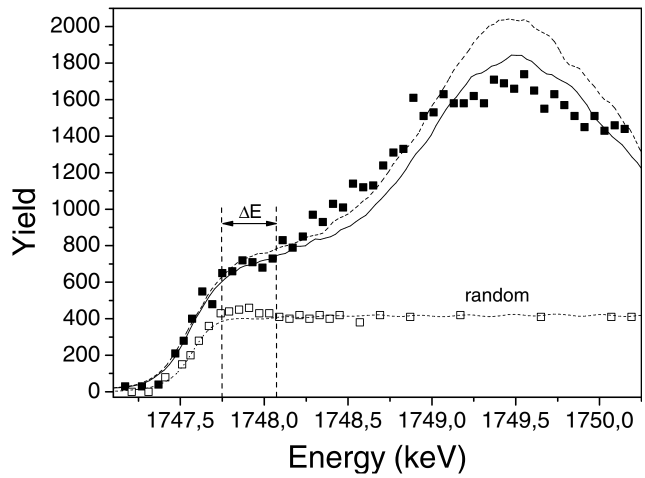

Fig. 6. Excitation curves of the ^{13}C(p,γ)^{14}N reaction. The black and white squares are experimental points: respectively, yields from the protons channeled along the (0001) plane and from the beam randomly oriented with respect to the crystal. The solid and dashed lines stand for calculations: protons are channeling along the (0001) plane, the r.m.s. amplitudes of thermal vibrations of carbon atoms are 0.102 Å and 0.09 Å, respectively. The background is subtracted. The incident proton energy step equals 72 eV.

The weak potential dependence on the distance to the planes in the central part of the channel where ^{13}C atoms are situated condition the small oscillation amplitudes of the protons and so they practically remain within the ^{13}C atom zone. The proton motion in this zone is determined by energy losses by valence electrons. Near the surface, before forming the first focus, the trajectories form a peculiar "cone". The proton flux is constant in the center of the cone. Therefore, one may expect the existence of a "step" in the γ-ray yield as a function of the initial proton energy in this region. As a result, there arises the possibility to determine electronic energy losses of channeled particles from the reaction yield value in the region of the "step".

5 Energy losses of channeled protons

Figure 6 shows the measured (black squares) and calculated (solid and dashed lines) excitation curves of the ^{13}C(p,γ)^{14}N reaction at proton channeling along the (0001) plane. The same figure shows the excitation curve for unchanneled protons (the white squares are experimental data, the dotted line stands for calculation).

As expected, a sharp increase in the γ-ray yield is observed at the resonance energy, irrespective of the beam orientation relative to the crystal. The increase in the reaction yield for unchanneled protons attains saturation and further remains unchanged irrespective of energy. This means that within the energy range shown, one resonance is excited, and the ^{13}C atoms are homogeneously distributed in the crystal. In the case of channeling (black squares) the reaction yield reaches its maximum in the focus of the proton trajectories. The channeled-proton flux in the center of the channel (fig. 5) is distributed uniformly down to a depth of ≈140 Å. Carbon is found in the center of the channel and is distributed uniformly in the crystal. The resonance is isolated. Therefore, the excitation curve for channeled protons shows a small plateau (labeled by ΔE in fig. 6). The extension of the plateau is several times greater than the resonance width Γ_{res}. The reaction yield in this region is substantially higher than in the case of unchanneled protons. The channeled-proton flux in the center of the channel, where valence electrons are found (fig. 4), gives the main contribution to the reaction excitation curve in the plateau part. In the center of the (0001) channel the electron density is lower than the average electron density in the crystal. Therefore, the path of channeled protons is longer on the length ΔE and more ^{13}C nuclei participate in the reaction thus increasing the yield in comparison with that from unchanneled protons. In other words, smaller electronic energy losses of the channeled protons, the trajectories of which are situated in the valence electron zone, call for the yield increase observed in the plateau region. The dechanneled protons contribute an insignificant part to the exitation curve. The ratio of γ-ray yields corresponding to the white and black squares on the ΔE area of fig. 6 is dependent on the energy losses of unchanneled and channeled protons. Estimations give the ratio $(\mathrm{d}E/\mathrm{d}x)_{(0001)}/(\mathrm{d}E/\mathrm{d}x)_{rand}$ as 0.64 ± 0.12. This result is consistent with the experimental and theoretical data [13,14] of other authors.

The difference between the excitation curve growths for the cases of channeled and unchanneled protons (figs. 6 and 7) is explained by different stopping-power values.

Figure 7 shows the calculated excitation curves of the reaction for several thermal vibration amplitude values of the ^{13}C atoms. The yield in the focus of the trajectories is substantially dependent on the vibration amplitude of carbon atoms. Owing to the Coulomb force action, a

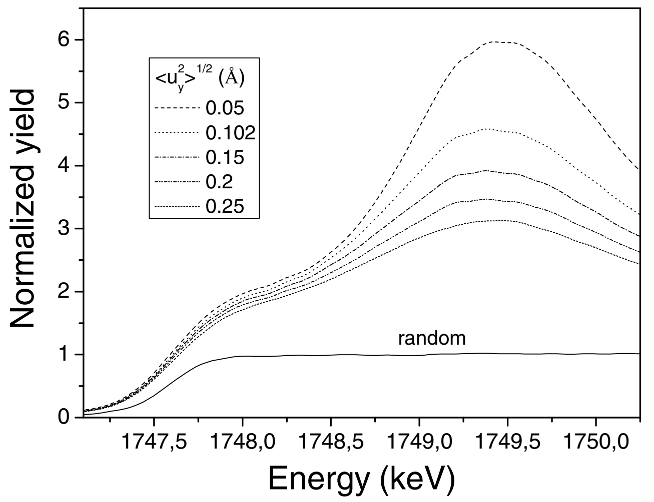

Fig. 7. Calculated excitation curve of the $^{13}C(p,\gamma)^{14}N$ reaction *versus* the thermal vibration amplitudes of ^{13}C atoms. Protons are channeling along the (0001) plane.

considerable part of channeled protons is focused in the crystal region where the ^{13}C atoms are located. In this case the probability of the reaction is governed by the probability of presence of ^{13}C atoms in the region of the trajectory focus. As the vibration amplitude increases, the ^{13}C atoms stay for most of the time beyond this region. As a result the probability of proton collision with ^{13}C nuclei decreases and hence the reaction yield also decreases. For the experimental data shown in fig. 6, the best agreement with the calculations has been obtained at a r.m.s. thermal vibration amplitude of carbon atoms equal to 0.102 Å.

The excitation curve of the reaction was also calculated for protons which are channeling along the ⟨0001⟩ axis. In this case, as in the planar channeling case (fig. 7), a plateau was observed.

6 Summary and conclusions

The possibilities of isolated nuclear-reaction resonances to study the distribution of the channeled-particle flux in crystals have been demonstrated. The resonance nuclear reaction $^{13}C(p,\gamma)^{14}N$ was used to establish and investigate the peculiarities of the flux distribution in the (0001) channel of the Re-0.4 at.% ^{13}C single-crystal solution.

The location of the ^{13}C atoms in this crystal was determined. The reaction yields of γ-rays were measured at angular scanning in the vicinity of the ⟨0001⟩ axis and of the (0001), $(1\bar{1}20)$ and $(1\bar{2}1\bar{2})$ planes. It was shown that the carbon atoms occupy octahedral interstitial positions.

The program was developed for computer simulation of channeled proton trajectories in the (0001) channel. The excitation curve of the reaction was measured and calculated for the protons channeled along the (0001) plane. The excitation curve exhibits a segment shaped as a plateau. It was shown that γ-ray emission of this segment is due to the protons moving along the center of the (0001) channel. This peculiarity of the excitation curve was used to determine the electronic energy losses of channeled protons. The value of $(\mathrm{d}E/\mathrm{d}x)_{(0001)}/(\mathrm{d}E/\mathrm{d}x)_{rand} = 0.64 \pm 0.12$ was determined.

It has been shown that the yield of resonance γ-rays excited by channeled particles is dependent on the thermal vibration amplitude of interstitial atoms.

Model excitation curves similar to those shown in fig. 7 were calculated for deuterons, and ^{3}He and ^{4}He ions. As in the proton case, a plateau was observed for the mentioned ions. The length of the plateau region is dependent on ion energy losses.

The use of isolated narrow resonance reactions induced by high-energy-resolution channeled ions in nuclei of impurity or matrix atoms opens new perspectives in the investigation of orientation effects. The elements (hydrogen, oxygen, nitrogen and carbon) are dissolved in relatively great amounts in some metals. The atoms of these elements are the interstitial impurities which occupy certain (octahedral as a rule) voids. Some of the resonances of the isotopes of these and other light elements can be used to investigate the angular and energy dependences in the particle flux distribution to determine energy losses, the Lewis effect [15] at channeling, etc. The fine structure that is observed in the channeled-particle scattering has been used more than once to choose the potential that most adequately describes the ion-atom interaction [2,3]. It can be expected that the use of high-resolution beams and narrow resonances of nuclear reactions will provide new ways for solving this problem.

References

1. D.S. Gemmell, Rev. Mod. Phys. **46**, 129 (1974).
2. J.H. Barrett, Phys. Rev. B **20**, 3535 (1979).
3. B. Schmiedeskamp, P. Jonk *et al.*, Nucl. Instrum. Methods B **17**, 309 (1986).
4. P.A. Svetashov, N.A. Skakun, Poverchnost: Fiz. Khim. Mekh. **5**, 113 (1995).
5. N.A. Skakun, V.M. Shershnev, Probl. At. Sci. Technol., Series: Physika Radiatsionnych Povrezhdenij **6**, 3 (2003).
6. N.A. Skakun, O.N. Kharkov, At. Energiya **27**, 351 (1969).
7. B. Maurel, G. Amsel, Nucl. Instrum. Methods **197**, 1 (1982).
8. G. Amsel, G. Battistig, Nucl. Instrum. Methods B **240**, 1 (2005).
9. L.C. Feldman, J.W. Mayer, S.T. Picraux, *Materials Analysis by Ion Channeling* (Academic Press, New York, 1982) p. 79.
10. F. Ajsenberg-Selove, Nucl. Phys. A **449**, 89 (1986).
11. J.H. Barrett, Nucl. Instrum. Methods B **44**, 367 (1990).
12. N.A. Skakun, V.M. Shershnev, N.A, Shlyakhov, Probl. At. Sci. Technol., Series: Nuclear Physics Investigation **5**, 43 (2004).
13. B.R. Appleton, C. Erginsoy, V.M. Gibson, Phys. Rev. **161**, 330 (1967).
14. D.V. Morgan, *Channeling: Theory, Observation, and Applications* (John Wiley and Sons, Inc., London, 1973) p. 131.
15. H.W. Lewis, Phys. Rev. **125**, 937 (1962).