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RANDOM FLUX REDISTRIBUTION OBSERVED FOR α-PARTICLES TRANSMITTED THROUGH Si, Ni AND Pt CRYSTALS

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1. Introduction

The last-decade experiments on the angular distributions of ions measured in the geometry of transmission through crystals [1,2] or of deep-layer emission [3-5] have changed significantly the phenomenology of the "channelography" images known since 1968 [6-9]. New and unexpected manifestations are evident today, and they do not allow to believe that the mechanism of the process is well understood.

However, it was clarified that the enhancement peaks along the crystallographic directions arise due to the charge-exchange process. Evidence of this effect was obtained in Refs. [1,2] when clear correlation of the enhancement with the final energy of transmitted ions was established. The enhancement is reduced and completely disappears at high energies corresponding to the ion equilibrium charge being close to the nuclear charge. At lower energies, multiple capture and loss events lead to the decrease of the transverse energy of the ions, and, respectively, to their concentration at small angles with respect to the axis or plane. Electron capture and loss provide this effect because of the different impact parameter dependences of the cross-sections of the two processes.

A Monte-Carlo code was also developed to simulate the trajectory of an ion with variable charge in the planar channel. The transverse momentum cooling was successfully demonstrated, and, finally, the dependences of electron capture and loss cross-sections on the impact parameter were estimated for some typical cases of experimentally observed cooling.

The reversal from cooling to heating observed at low enough heavy ion energies in Ref. [1] is not understood till now, and in general, a question arises whether the mechanisms based on the electron exchange processes may be effective when their cross-sections are extremely high.

2. Experimental

The crystallographic angular patterns have been observed using CR-39 solid state track detectors for α-particles transmitted through the thin crystals. The following single crystals were available in the form of self-supported foils: Si of 3 to 14 μm thickness, Ni – (1 to 4.5) μm and Pt – (1 to 6) μm. The foil was fixed onto the frame, and the window had a linear size of about 7 mm. The Si samples were prepared using a monocrystalline Si plate by the method of implantation and etching. Pt and Ni foils were grown by epitaxy with consequent removing them from the substrate and stretching onto a metal frame.

The radioisotope source of ^{241}Am or ^{252}Cf is placed at a distance of about 20 mm from the crystal, the diameter of the α-active material spot is 2 mm. The distance between the source and the plastic track detector, CR-39, is about 260 mm. This corresponds to the angular resolution of about 0.2°. All these assemblings were placed inside the vacuum chamber, and the activity of the source was enough to produce a track density at the detector surface as high as 500 mm^{-2} after a 5 days exposure.

Aluminum absorbers were installed between the source and the crystal in order to change the initial and final energies of the alphas. For particles transmitted through the crystal in random directions the final energy is varied from 0.5 to 4 MeV. At such energies α-particles are detected by CR-39 with 100% efficiency. The total flux of transmitted particles has been detected without any discrimination by energy losses, and thus, real flux (not energy) contrast might be observed.

After etching the CR-39 detectors, the crystallographic images could be visually seen on the detector surface. It is helpful for the selection of the region of interest in the scanning of the detector by the automatized system. The coordinate (angular) dependence of the track density was quantified using three variants of track counting: the laser automatic scanning system at Siegen University, the scanning microscope installed by a TV tube at FLNR, JINR Dubna, and visual counting in an optical microscope. The results of all three variants of detection are compared and they show good agreement.

After transmission through the crystal, the flux is no more isotropic, channeling maxima are clearly seen in the directions of crystallographic axes and planes. The orientation of the crystal foils allows detecting the <100> axial and (100), (110) planar maxima in the present experiment. The results of scanning are given in Figs.1-3 for Si and Pt crystals. The parameters corresponding to each pattern are listed in Table 1.

One can see in Figs.1-3 that only cooling-enhancement peaks and no heating dips are observed even at lowest final energies for all crystals. Cooling is effective both in Pt and Si, although the physical parameters of the crystals are very different, for instance, the volume electron density in Pt is by a factor of 7 larger than in Si. For understanding, let us discuss briefly the mechanism of the electron exchange processes. It is known, that the equilibrium charge of heavy ions in a medium at a certain velocity is defined by the balance of the electron capture and the loss probabilities. The projectile changes its charge state until the cross-sections of both processes become equal, then the charge fluctuates near the equilibrium value. Electron exchange processes are accompanied by the release of electrons from the target atoms and contribute to the ion energy-loss characterized by the dE/dx parameter. The main part of dE/dx arises due to another electron process: the target atom ionization in quasielastic collisions. However, all electron-interaction processes have universal nature, and the measurable parameter dE/dx reflects their inherent properties.

In accordance with published works, for instance [10,11], the major mechanism responsible for the stripping of electrons from the projectile should be the electron-impact ionization. Coulomb excitation of the ion by the nuclear potential has a lower probability. For near-channeling trajectories, when close impact collisions with the atom are eliminated, the electron interactions dominate even stronger than in a disordered solid. Consequently, the number of electron-loss events per unity of path-length is defined only by the ion velocity and by the target electron density. Not all electrons have to be taken into account. For instance, deeply bound electrons in the Pt atom are typically passive, because of both strong binding and location at small radii. Respectively, the Pt to Si ratio in the dE/dx parameter is only 3:1, while the electron density ratio is as high as 7:1. One may assume similar 3:1 ratio for the electron loss cross-sections in collisions of ions with Pt or Si atoms. Then, the mean free path-length ℓ_{loss} in Pt should be 3 times shorter than in Si.

As is discussed in Ref. [2], one important parameter that regulates the strength of cooling can be introduced as a ratio of ℓ_{loss} to the quarter of channeling trajectory wave-length, $\lambda/4$. When the inequality $4\ell_{loss}/\lambda<<1$ is fulfilled, cooling is reduced. And it seems interesting to find that λ in Pt is also 3 times shorter than in Si, because of the stronger potential of lattice and the 3 times larger critical angle ψ_1. Thus, the ratio between ℓ_{loss} and $\lambda/4$ remains almost the same both for Si and Pt, and this means the same good condition for the manifestation of the charge-exchange cooling mechanism, described in Refs. [1,2].

In addition, it is well-known in the literature that for ions in solids the equilibrium charge-state distribution shows weak Z_2-dependence. So, the process of equilibrium balance between electron capture and loss processes remains similar in both crystals, Si and Pt, despite the strongly different Z_2 and other parameters including the type of symmetry and lattice constant. These arguments are useful for general understanding of the data, but many details should be specified.

3. Properties of angular distributions

The observed properties of the angular distributions are systematically described below. The experimental data are not restricted by the set of the present results taken with α-particles; heavy-ion experiments [1,12] are also taken into account. The consequent theory of the process is not yet created, and the present discussion might be considered as an attempt of understanding, what physical reason defines a certain systematical manifestation. Some regularity can be, in principle, independent of the microscopic mechanism, and it would be useful to find such simple explanations, and then isolate those properties, which are really sensitive to elementary interactions. This would be a necessary step on the way to full-scale theory.

The qualitative approach was used also in Ref. [2], and there was found that the cooling enhancement is reduced both when charge-exchange cross-section is low and when it is extremely high. The latter conclusion is not trivial, and is confirmed recently in regular simulations, Ref. [13].

The angular distributions clearly demonstrate the variation with changing of the crystal and ion species, the final energy, crystal thickness, and Miller indexes of the direction. One can find some special manifestations, and among them it would be possible to distinguish:

1. Growth of the enhancement with the crystal thickness, t. That is understood as integration of the effect due to multiple electron capture-loss events. The decrease of the transverse energy is accumulated with the growth of the number of elementary events. The latter number can be estimated roughly as a ratio $t/\ell_{capt.}$, where $\ell_{capt.}$ is the free-path length of the ion for the electron capture process.
2. At large thicknesses, the enhancement peak becomes to be high in amplitude, and it is typically narrow, the angular width is lower than the Lindhard angle. It could be explained assuming that the total number of particles in the peak is conserved, and the growth of amplitude means decrease of the width.
3. Within some range around the axial direction when $a \le \psi \le b$, the axial intensity is already decreased, but the planar enhancements have not yet appeared. They are clearly seen at the region of $\psi > b$, i.e. far from the axis. An example of such a pattern is given in Fig. 4, taken with the selection the energy loss of the ions to be near the mean value between channeling and random values. Thus, particles really captured in the cooling process are selected, and the contrast of the picture is the best for illustration. The discussed property is evident even without selection, see for instance in Fig.6a.

   As is known, near an axis the plane looks like a consequence of atomic chains almost parallel to this selected direction. Thus, the particle has a few collisions with atoms per unity of length, much less than typically in the planar direction.
4. Oscillations in the angular dependence are often manifested, see in Figs.1-3, especially, in Figs.1b and 3b. When the central peak has low amplitude, the amplitude of the oscillations can be comparable with the maximum of the peak. One may try to explain them by the quantum levels of the transverse energy in the potential well, but the estimations show a high level density, and support a rather classical interpretation. The resonance dechanneling process, discussed in Refs. [14,15], is another possibility. However, it was proposed specially for the axial-peak scanning within some plane, while we observe the oscillations for the data taken in

the circular scanning or in the crossing of the axial peak by the diameter out of any plane. So, until now the origin of oscillations is not absolutely clear.

5. It was observed that different planes sometimes show very different behaviour. Not only the peak amplitude, but the type of pattern is changed being a peak for stronger plane and a dip for weaker. That was detected in some cases of heavy-ion transmission through Si, when (110) – strong and (100) – weak planes are compared. Within the model of charge-exchange, what is the ratio between mean radii of capture ρ_{capt}, and loss ρ_{loss} is important. The ratios of both radii to half the interplane spacing, $d_p/2$ are also important. For the weaker plane with more narrow planar channels, both capture and loss radii at some energy reach a value close to $d_p/2$, and in this situation the heating may appear easy. The observation of qualitatively different profiles for the (110) and (100) planes, probably, corresponds to such a case.
6. The angular width of the heating minimum is typically larger than the Lindhard angle, while the cooling peak is more narrow.

4. Discussion of possible mechanisms

Cooling enhancement peaks were explained in Refs. [1,2] by the regularized ion-charge variation with corresponding changes in the potential part of the transverse energy. However, for heavy ions of low energies in Ref. [1] the reversed pattern is detected with dips instead of peaks. The transition from "cooling" to "heating" happens at a definite ion energy E_{tr} as is shown in [12], and the latter energy is dependent on the crystal and ion species, and the crystallographic direction. The reliable reasons for the transition are not yet found. The direct extension of the charge-exchange mechanism leads to the conclusion that at an energy E_{tr}, the difference of the mean radii of electron loss and capture changes its sign, being negative at $E < E_{tr}$, and positive at $E > E_{tr}$. This is not impossible, but such transformation looks sophisticated and physically not well understood.

Another question remains, viz. what happens when both radii are comparable or larger than the channel width in the crystal. Large values of the radii and cross-sections are realistic for electronic processes in the cases of heavy ions with an energy near, or lower than 1 MeV/u. They may be valid for the atomic collisions in gas targets, but not in crystals. When an electron is located near the median plane of the channel, it is not definitely linked to some individual atom. Two, three, or more atoms can be found at almost the same distances from the electron. The ion interacts with electrons belonging to the lattice, not to some individual atom. It would thus be difficult to support the mechanism of impact-parameter dependent electron exchange processes in this case.

The described situation is independent of the microscopic details, it arises only due to the large cross-section, being some kind of saturation in the probability of the processes. Indeed, let us suppose that both capture and loss cross-sections are of the order of 10^{-15} cm^2. Such an area corresponds to a radius of 1.8 Å, even if we assume 100% probability inside the circle. Circles belonging to a few atoms are overlapping in the crystal, and there is no more difference between the radii of capture and loss, because the probability is saturated on a level of 100%. At larger distances, not actual in crystals, the probabilities of both processes can be different.

It would be important to know the real cross-section values for ion-charge exchange processes. The experimental data are fragmentary, Refs. [16-18], but being supported by theoretical calculations, for instance [19,20], they systematically show cross-sections above $5\cdot10^{-16}$ cm^2 at energies near and below 1 MeV/u. Thus, the saturation can be really the reason of some changes in the angular distributions. Obviously, the observation of different type patterns for weaker and stronger planes (see point 5 in section 3) indicates that mean radii of capture and loss ρ exceed $d_p/2$ for one plane, and not for another. This would be the way for estimation of cross-sections, if such interpretation is confirmed.

For the ionization of $He^+ \rightarrow He^{++}$, the cross-section has been estimated using a semiempirical formula from Ref. [21]. The cross-section of electron-impact ionization has been figured out as a function of the He-ion velocity, and then it is recalculated to an atomic cross-section. Deeply-bound electrons (at K- shell in Si atom, at K- and L- shells in Ni atom, and at K-, L-, and M- shells in Pt atom) have been excluded in an estimation of the number of interacting electrons. The numerical results are reduced in Table 1. Clearly, the cross-sections are not extremely high, even for Pt atom, and the probability saturation takes no place in the case of α-particles. One may conclude the evident correlation with the absence of heating for alphas in all crystals at different energies.

In general, any modulation of the wide-angular flux on the transmission through a crystal appears as a contradiction to the principle of reversibility of trajectories. J. Lindhard has shown theoretically that the reversibility should take place for the channeling trajectories governed by the lattice potential. It is also conserved in the presence of multiple scattering and weakly perturbed by energy losses. The large-angle single scattering has also to be in the group of processes, which provide finally reversible distributions.

The uniform spectrum of transverse energy at entering the crystal should remain uniform at the exit. This is a concrete manifestation of the reversibility principle for our transmission experiment. However, the flux redistribution is observed in the form of cooling or heating almost in all cases, except for a narrow window near E_{tr}, and large energies, when ions are completely bare. Thus, one has to find some reasons for the violation of reversibility. Regularized charge variation is one of the mechanisms, and it is effective for the explanation of cooling, as is discussed above. The situation with heating is not so clear.

high, then it would be possible to assume that blocking overwhelms the channeling, and the heating dip arises as a result. The probability of scattering is calculated for the real geometry of the present experiment, illustrated in Fig. 5a.

At any point on the detector surface, the directly transmitted particles are accompanied by the ones scattered on the way through the crystal. The thickness of the crystal is finite, and the contribution of the scattered component is not negligible. As is clear from Fig.5a, the scattering angle θ_s is almost equal to the deviation of the initial α-particle from the axis, and the latter angle is restricted to be ≤10° by the frame of the crystal foil. Thus, all scattering angles from 0 to θ_{max}=10° are allowed. It is easy to calculate the probability of scattering "p" within the range of angles $\theta_{min} \le \theta \le \theta_{max}$ by integrating the scattering cross-section:

$$p = 1 - e^{-W}; \qquad (1)$$

$$W = 2\pi \int_{E_{max}}^{E_{min}} \int_{\theta_{max}}^{\theta_{min}} \frac{d\sigma}{d\Omega_L}(E_L, \theta_L) \frac{\sin\theta_L}{dE/dx} d\theta_L dE_L . \qquad (2)$$

Assuming in first approximation dE/dx= const, one can deduce a quantitative result:

$$W = \frac{3.92 \cdot 10^{-5} Z_1^2 Z_2^2}{(dE/dx) A_2} \left(\frac{1}{E_{min}} - \frac{1}{E_{max}} \right) \left(\frac{1}{\theta_{min}^2} - \frac{1}{\theta_{max}^2} \right), \qquad (3)$$

where E_{min} and E_{max} are expressed in MeV and correspond to the exit and entrance energies of the particles, dE/dx is in MeV·cm^2/mg, and θ_{min}, θ_{max} – in rad. The assumption of small angles, when θ=sinθ=tgθ, and the Rutherford formula for the differential cross-section are used. From eqs. (1-3), one deduces immediately, that the probability of scattering to angles above the critical angle is large, comparable with 50 % at real thickness of the crystal and at the final ion energy of almost E_{tr}. Presence of multiple scattering does not eliminate the large-angle events. Among multiple deflections, scattering to a large angle $\theta > \psi_1$ is also possible. This may influence the final angular distribution due to the blocking effect, independently on the exit angle, being it larger or within multiple scattering half-width. The probability is high, and it seems, that the origin of heating due to the blocking effect is supported. But yet, the blocking dip might be compensated by the channeling peak, at least, the reversibility requires that.

In order to make evident the compensation, one more experiment was performed for S ions transmitted through the 13.6 μm Si crystal with final energy of 48 MeV, using the scheme shown in Fig. 5b. This was an experiment at the Tandem beam, but the geometry schematically was the same as in the case of the alpha-source. The additional screen was installed on the line connecting the beam spot and the detector centers. The size of the screen was the same as the source diameter. The screen stops the direct transmission of particles along the crystallographic axis, and allows the particle to reach the central area of the detector with scattering in the crystal. The selection of the scattering angles is not perfect, because of the finite size of the source, but all scattering angles near 0 are suppressed.

We may expect that the blocking minima appear immediately in the presence of the screen, and their depth would be enough to provide the heating due to scattering. However, the experiment revealed another result. The screen, really, suppressed the direct transmission, the particle flux in the central area of the detector was decreased by a factor of about 0.3, and no clear blocking appeared. In the axial and planar directions weak cooling maxima are still observed. It means that cooling is effective even for scattered particles. The comparison of images detected with and without the screen is given in Fig.6. Strong cooling peaks are seen without the screen and weak ones in presence of it. Thus, the idea of heating due to scattering is not confirmed by this experiment.

The violation of reversibility in the form of cooling or heating remains still intriguing, and it would be important to verify, what are the predictions of the numerical simulations for the transmission of a wide-angular flux. Existing Monte-Carlo codes do not operate with the collective potential of the lattice in continuos approximation, they evaluate the trajectory of a particle as a consequence of binary collisions with ordered atoms. Calculations using the "Crystal TRIM" code did not demonstrate any significant modulation of the flux, Ref. [1]. Code "UPIC", Ref. [22], also shows the absence of the crystallographic structure at the transmission images for the wide-angular flux, even with account of energy losses and realistic electron distributions. Only electron capture and loss processes may change the situation.

In addition to the charge-exchange model, it would be interesting to find some other possibilities for the explanation. Among them, one has to consider the role of second-order effects, when the ion is no more like a probe charge, and the medium has not anymore static properties, as is known since 1963, Ref. [23].

Collective processes may also be important, for instance, two-electron exchange events. Their significance is growing when the cross-section of ion-electron interaction is increased, means at low velocities. A possibility of ion interaction with electrons belonging to the lattice, not necessary to any individual atom, is described above. Spontaneous events of a multiple Auger ionization can lead to the strong deviation of ion charge from equilibrium value, increasing effectively the transverse energy of ion. In general, the variety of interaction mechanisms in solids provides the possibilities for understanding of the heating effect and its conversion to cooling. An extensive theoretical analysis supported by calculations is necessary to clarify the details of processes, while experimental results are already available in Refs. [1,2,12] and in the present work.

To summarize, a new set of transmission patterns has been obtained using α-radioactive sources and thin crystals of Si, Ni and Pt. The general properties of the observed crystallographic images, as well as some special details are now clarified. Possible mechanisms responsible for the cooling or heating of ion trajectories in the transverse momentum are discussed. The cooling is attributed to the different impact-parameter dependences of the electron capture and loss processes combined with the changes in the potential transverse energy due to variation of the ion charge. The transition from cooling to heating at low energies in the case of heavy ions is not yet well understood.

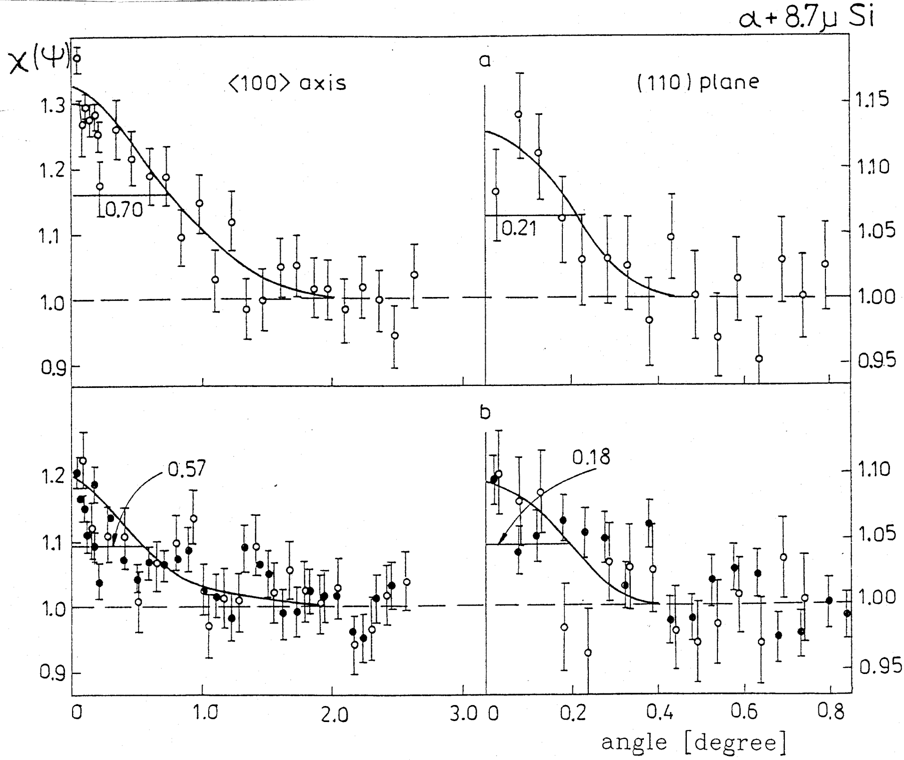

Fig.1. Cooling peaks observed in axial and planar cases for α-particles transmitted through Si crystal of 8.7 μm thickness: a) at final energy E_f=1.7 MeV; b) at E_f=3.1 MeV.

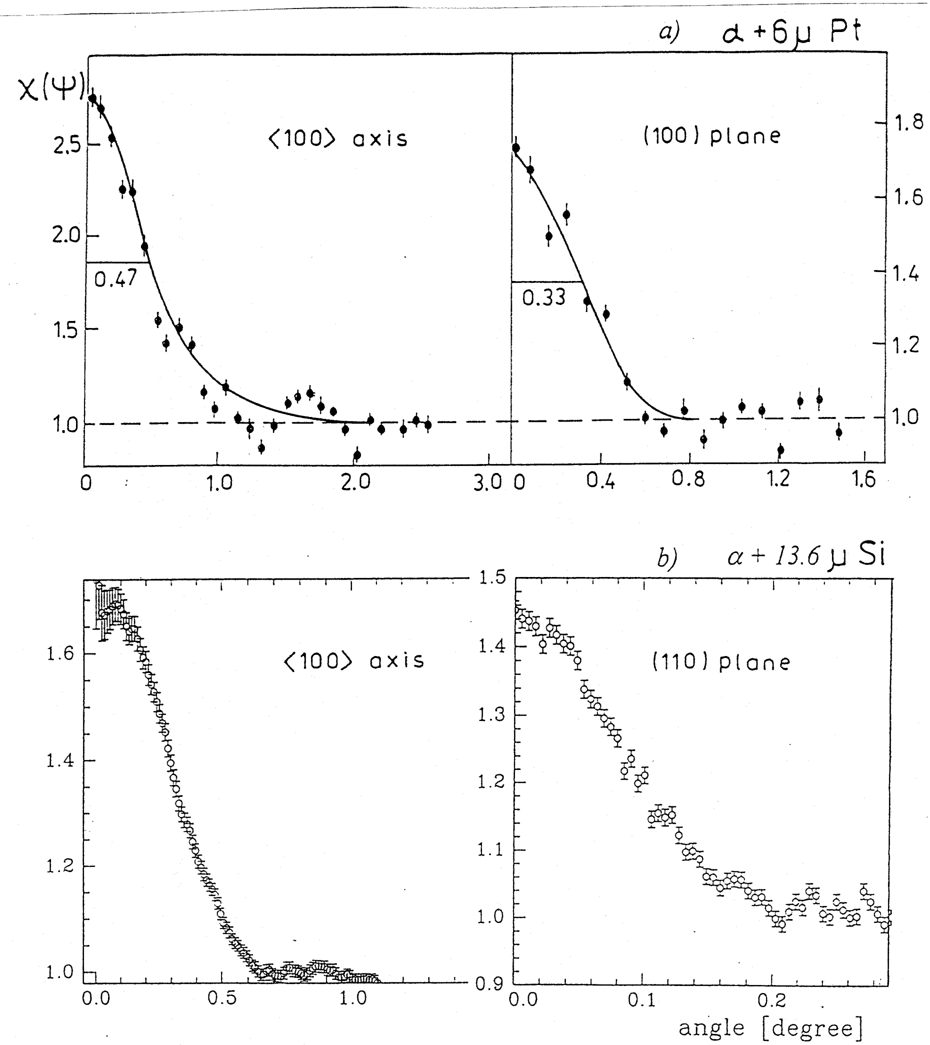

Fig.2. The same as in Fig.1 taken in following cases:

a) 13.6 μm Si and E_f=0.6 MeV;

b) 6 μm Pt and E_f=0.6 MeV.

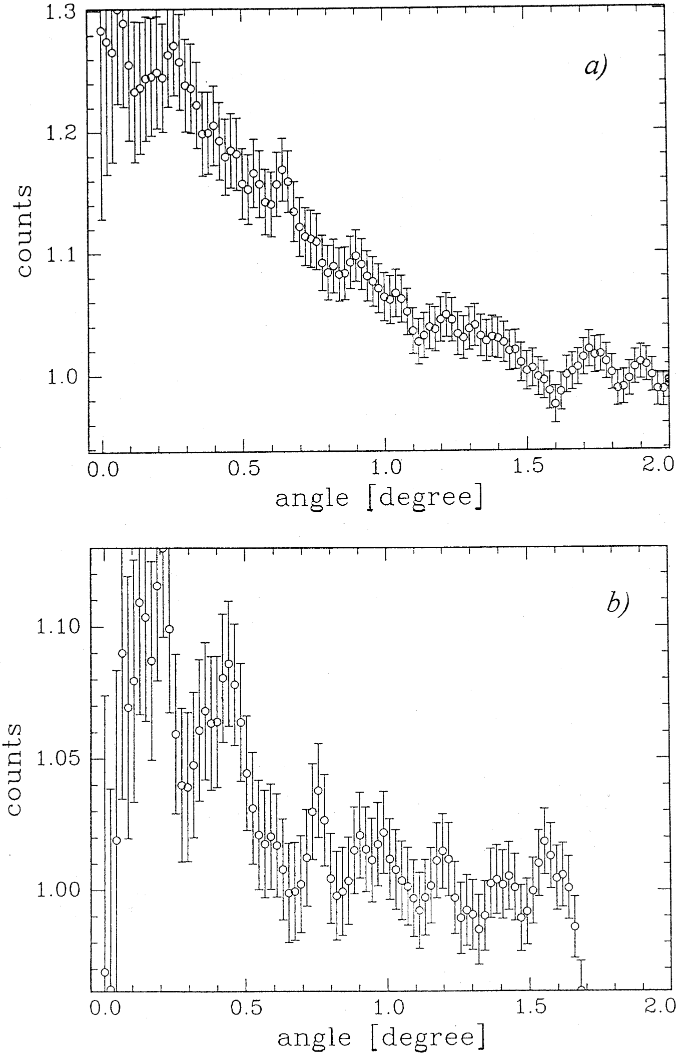

Fig.3. Axial <100> peaks obtained with 1.1 μm Pt crystal at final energy values: a) 2.0 MeV and b) 4.9 MeV.

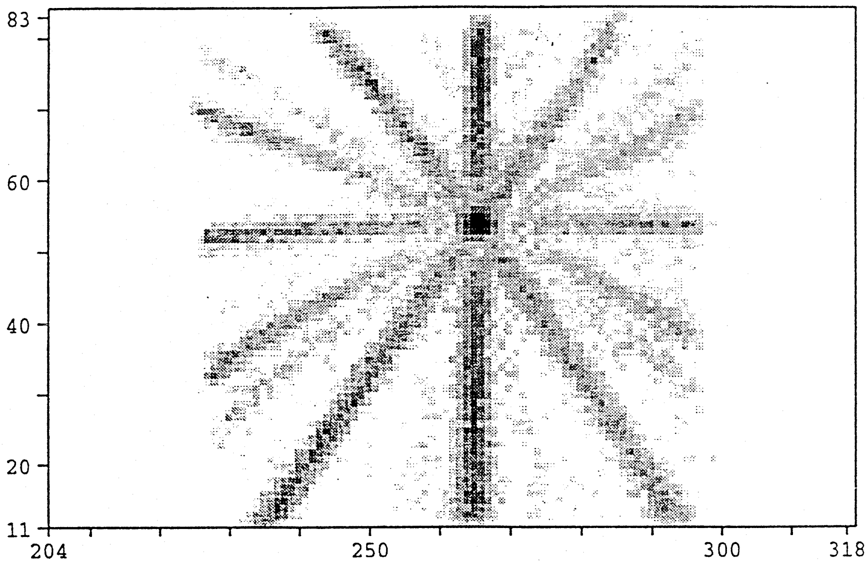

Fig.4. Transmission image of Ni ions through Si of 4 μm thickness near the axis <110>. Ions are selected by energy losses, the range corresponds to mean values between channeling and random losses.

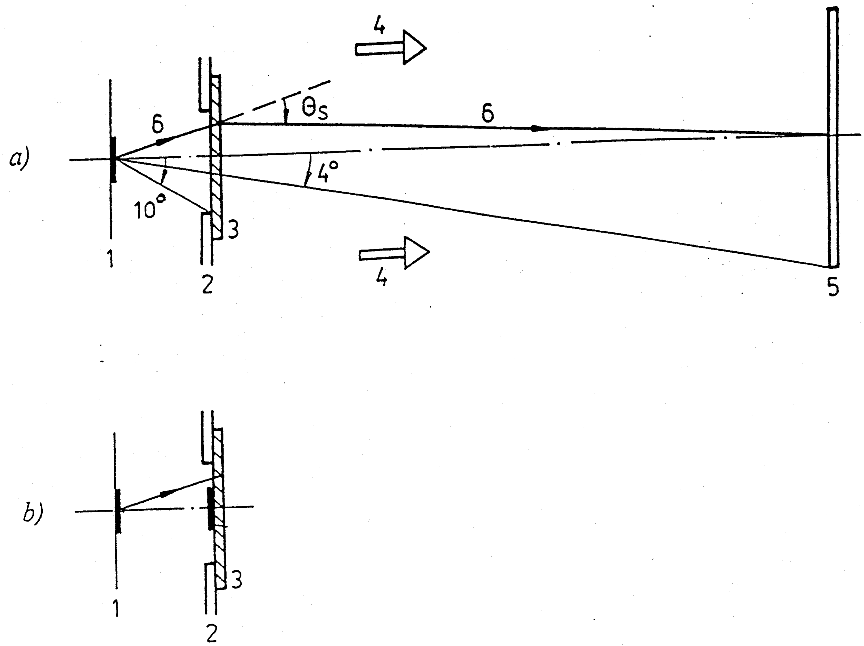

Fig.5. Details of the geometry in the present experiment on the detection of transmission crystallographic patterns for α-particles:

1) radioisotope source of α-particles,
2) frame,
3) crystal,
4) <100> axial direction,
5) CR-39 plastic track detector,
6) trajectory of a particle scattered in crystal.

a) b)

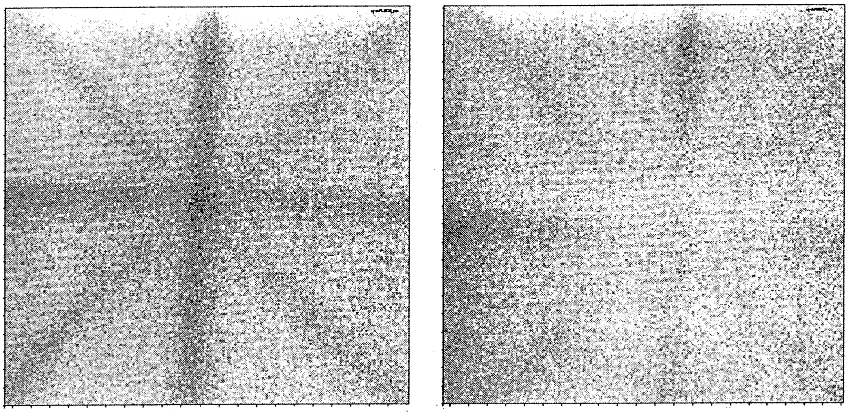

Fig.6. Comparison of the two-dimensional images taken for S ions in the geometry shown in Fig. 5a and 5b: a) without screen; b) with screen.

Table 1
List of measured angular distributions for He-ions in Si, Ni and Pt crystals of (100) orientation, the calculated cross- sections of ionization and relative yield χ_0 along <100> axis.

Crystal	Thickness, μm	E_f, MeV	σ_{loss}, 10^{-16} cm^2/atom	χ_0	Shown in Fig.
Si	13.6	0.6	0.63	1.74±0.06	2a
Si	13.6	4.1	0.45	≈1.0	-
Si	8.7	1.7	0.70	1.34±0.04	1a
Si	8.7	3.1	0.53	1.20±0.04	1b
Si	6.3	1.9	0.67	1.33±0.04	-
Ni	0.9	0.8	1.2	2.7±0.2	-
Pt	6	0.7	2.9	2.8±0.2	2b
Pt	1.1	2.0	2.7	1.30±0.04	3a
Pt	1.1	4.9	1.7	1.13±0.05	3b

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Карамян С. А. и др. E14-2001-247
Перераспределение потока α-частиц
при прохождении через кристаллы Si, Ni и Pt

Ранее было обнаружено перераспределение потока протонов и тяжелых ионов в монокристалле Si с образованием пиков каналирования, теперь тот же эффект изучен для α-частиц, прошедших через монокристаллические фольги Pt, Ni и Si. При энергии α-частиц меньше 4 МэВ (после прохождения) во всех случаях зарегистрированы пики каналирования («охлаждения»). В отличие от тяжелых ионов, минимумы «нагрева» не видны даже при самой малой энергии $\approx$ 0,1 МэВ/нукл. Зафиксированные особенности обсуждаются с целью качественного понимания. Механизм перераспределения потока включает регуляризованную вариацию заряда иона с соответствующим изменением потенциальной части поперечной энергии в присутствии потерь энергии и многократного рассеяния.

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Random Flux Redistribution Observed
for α-Particles Transmitted Through Si, Ni and Pt Crystals

Flux redistribution with formation of channeling peaks, observed earlier for protons and heavy ions in Si, has now been studied for α-particles transmitted through thin Pt, Ni and Si crystals. The channeling peaks are detected in all cases when the final energy of the alphas is below 4 MeV. Even at very low energies $\approx$ 0.1 MeV/amu, the «heating» minima are not visible unlike to heavy ions. Detected peculiarities are discussed, and qualitatively understood. The flux-redistribution mechanism includes the regularized charge variation, with corresponding changes in the potential part of transverse energy, in the presence of energy losses and multiple scattering.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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