Enhancement of the Inelastic Nuclear Interaction Rate in Crystals via Antichanneling

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The interaction rate of a charged particle beam with the atomic nuclei of a target varies significantly if the target has a crystalline structure. In particular, under specific orientations of the target with respect to the incident beam, the probability of inelastic interaction with nuclei can be enhanced with respect to the unaligned case. This effect, which can be named antichanneling, can be advantageously used in the cases where the interaction between beam and target has to be maximized. Here we propose to use antichanneling to increase the radioisotope production yield via cyclotron. A dedicated set of experimental measurements was carried out at the INFN Legnaro Laboratories with the AN2000 and CN accelerators to prove the existence of the antichanneling effect. The variation of the interaction yield at hundreds of keV to MeV energies was observed by means of sapphire and indium phosphide crystals, achieving an enhancement of the interaction rate up to 73% and 25%, respectively. Such a result may pave the way to the development of a novel type of nozzle for the existing cyclotrons, which can exploit crystalline materials as targets for radioisotope production, especially to enhance the production rate for expensive prime materials with minor upgrades of the current instrumentation.

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Interactions between charged particle beams and solid media are studied and exploited throughout different scientific fields. If the target has a crystalline structure, a series of coherent phenomena may manifest. One of these effects is the channeling phenomenon [1], which may occur as a charged particle impinges a crystal nearly aligned with the crystal planes or axes, i.e., when the angle between the particle direction and the crystal planes or axes is lower than the critical angle θ_C. Under channeling conditions, the particle is captured by the potential well formed by two neighboring planes and is forced to bounce between them. The angle θ_C depends on the crystal potential well depth U_0 and the particle energy E, $\theta_C = \sqrt{U_0/E}$. A particle under channeling can traverse a longer distance into the medium with respect to a particle interacting with an amorphous medium with the same average material density. Indeed, a channeled positive particle interacts less frequently with nuclei and core electrons, losing its energy only because of the interaction with the valence electrons.

These effects can be exploited to modify the probability of a close encounter with nuclei [2]. The variation of the nuclear interaction rate was experimentally observed from hundreds of keV [2] to hundreds of GeV energies [3–5]. Since the reduction of the interaction frequency with nuclei induces a lowering of the rate of nuclear inelastic interactions within a crystal, channeling has been proposed to be used to collimate or extract the circulating beam at the LHC accelerator [6].

For instance, when fast protons impinge on a crystal near a crystal axis or plane, some of the protons will travel the crystal remaining above the potential barrier, thus approaching at close distances to the crystalline nuclei. An interesting case is when a beam enters a crystal with an angle close to θ_C. In such a case, almost all the beam particles, even those entering in the middle of two planes, undergo an over-barrier motion with respect to the potential wells of the crystal. These particles have the ability to hang above the maxima of the potential barriers, where the nuclei of the lattice atoms are located. The role of the above-barrier particles in the interaction of fast particles with a crystal was noted in the seminal article of Akhiezer [7] on the radiation process of fast electrons and positrons in a crystal. Subsequently, the interaction of over-barrier particles with crystals was reported, for example, in Ref. [8], where the orientation dependencies of the processes associated with small impact parameters were studied.

Here below, a simple case is presented. Figure 1 shows the simulated trajectories of charged particles that enter parallel (a) or at θ_C (b) with respect to the (110) planes of a Si crystal. The simulation refers to a 655-keV proton beam, $\theta_C = 0.33°$. In the first case, the particles enter parallel to the crystal planes—the red dashed lines—and bounce

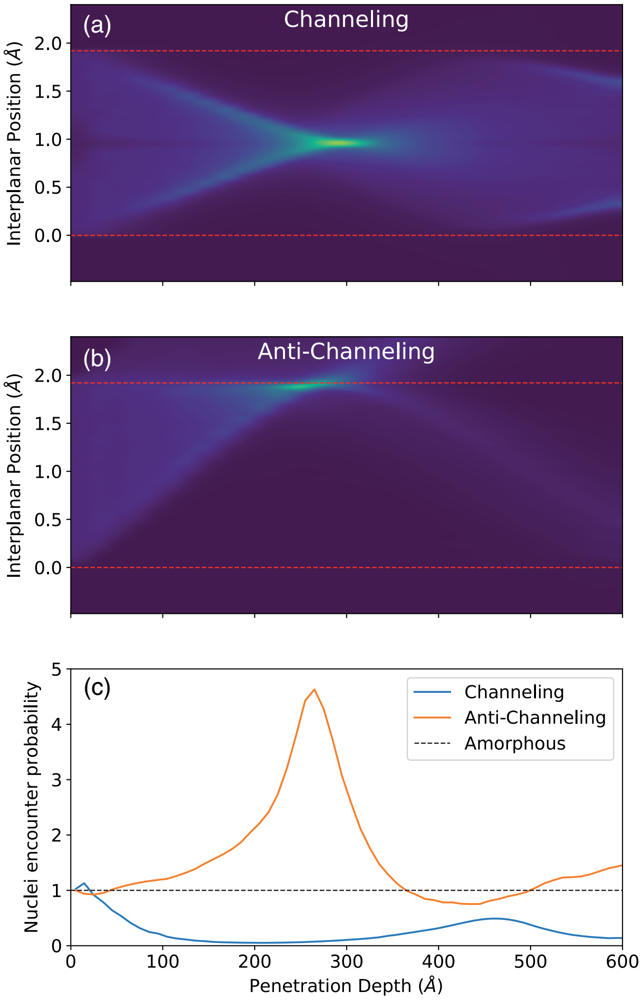

FIG. 1. GEANT4 simulation of the distribution of the particle trajectories for a 655 keV proton beam impinging on a (110) Si crystal under channeling condition (a) and antichanneling conditions (b). (c) The average nuclei encounter probability as a function of the penetration depth for the two cases.

between them, in a region without nuclei. Therefore, their probability of interaction with nuclei is lower with respect to the case of amorphous material or randomly oriented crystal. On the contrary, the particles entering the crystal with an angle close to θ_C result to be focused toward the atomic rows. As a consequence, their probability of interaction with the nuclei is higher with respect to the case of amorphous material or randomly oriented crystal [9,10].

The latter phenomenon can be called antichanneling, and can be extremely useful in the case where the interaction between the beam and the target has to be maximized, as for the production yield of radioisotopes for medical applications. Here, experimental measurements carried out at the INFN Legnaro Laboratories on the observation of the antichanneling effect at sub-MeV energies are shown. Finally, a series of GEANT4 simulations was used to validate the experimental data and to propose the usage of the antichanneling effect to increase the production yield of the radioisotope for medical applications.

In order to quantify the variation of the nuclear interaction rate of a charged particle beam with a target, let us define the nuclei encounter probability (NEP) as the average of the nuclear density experienced by the particle of a beam undergoing coherent interactions into a crystal, normalized by the nuclear density of an amorphous material. For a quantitative definition of NEP see Ref. [11]. The NEP can be computed by Monte Carlo calculation of an ensemble of beam particle trajectories and evolves with the penetration depth into the crystal. Figure 1(c) shows the GEANT4 simulation of the evolution of the NEP as a function of the penetration depth into the crystal for the channeled and the antichanneled particle beams of Figs. 1(a) and 1(b), respectively. Detailed information on the code is available below.

Most of the channeled particles oscillate far from the crystalline nuclei. As a consequence, after the first crystalline layers the NEP goes down almost to zero. On the contrary, the antichanneled particles are focused on the atomic rows of the crystal, resulting in a higher NEP, up to ~4.5.

A scheme of antichanneling is here shown for a sapphire (Al_2O_3) crystal. In particular, Fig. 2 shows the GEANT4 simulations of protons interacting with the Al_2O_3 crystal under channeling (a) and antichanneling (b) conditions.

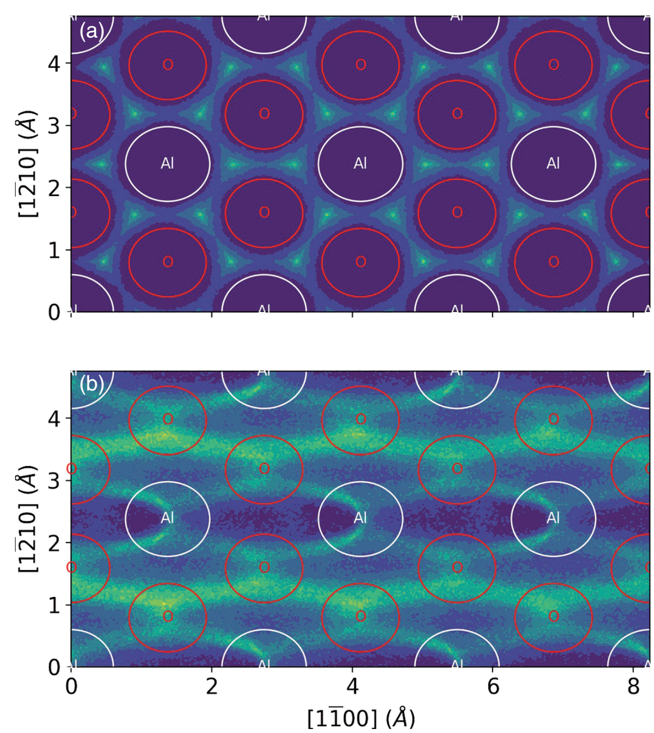

FIG. 2. GEANT4 simulations of the distribution of the protons interacting with an Al_2O_3 crystal under channeling (a) and antichanneling conditions (b). The potential of the Al_2O_3 crystal oriented along the ⟨0001⟩ axis is shown.

The particle trajectories are along the z axis; i.e., the direction of propagation is perpendicular to the figure, while the transversal movements of the particles are shown. In Fig. 2(a), the particles enter in the channeling condition. Thus, they follow a path far from the nuclei of the crystal, minimizing the probability of interaction with them. On the contrary, if the particles enter the Al_2O_3 crystal in antichanneling condition, they are forced to oscillate in a region with a high concentration of oxygen atoms, leading to an increase of the probability of interactions with the oxygen nuclei. In Fig. 2(b) the trajectories of particles under antichanneling are shown. In contrast to what happens in Fig. 2(a), here the particles are forced by the potential generated by aluminium atoms to pass closer to the oxygen nuclei.

In order to experimentally quantify the increase of the nuclear interaction, we studied the behavior of a proton beam as a function of the impinging angle with two samples, i.e., an Al_2O_3 and an InP crystal. Al_2O_3 and an InP crystals were chosen because they can be fabricated with good crystalline quality. The experiments were performed at the AN2000 and CN accelerators at the Laboratori Nazionali di Legnaro (INFN—LNL). Both the accelerators are Van de Graaf electrostatic with 2.2 and 6 MeV maximum energy, respectively. The output energy available by such compact-size accelerators makes them ideal to promote nuclear reaction events within the first layers of target materials. Indeed, O and P atoms composing the samples show nuclear resonances at available energies for the used accelerators; thus it was possible to measure the increase of the nuclear interaction.

The samples were installed on a goniometer with an angular resolution of 0.01° and a minimum pressure inside the chamber of approximately 10^{-7} mbar. This goniometer allowed the alignment of the crystal axis with respect to the incoming beam, to study the transition from a channeling to antichanneling condition. Beam divergence and goniometer resolution—both of the order of 0.01°—are perfectly suitable for channeling studies at low energy, taking into account that θ_C for 2 MeV protons is of the order of 0.2°.

The alignment of the samples was done through a silicon detector for Rutherford backscattering analysis (RBS), i.e., by measuring the backscattered particles as a function of the angle between the beam and the samples, since the backscattering process is proportional to the NEP [11]. The RBS detector was placed at 110 mm and at an angle of 160° from the target. The surface of the detector was 25 mm^2 large.

The measurement of the reaction products was done via a silicon detector for nuclear resonance analysis (NRA). Starting with the beam entering the crystal as perfectly aligned to the lattice planes—the channeling case—we measured the nuclear reaction products as a function of the angle of incidence of the beam with respect to the crystal planes. The NRA detector was placed at 150° and at a distance of 40 mm from the target. It had a 300 mm^2 surface and was covered with a mylar foil to prevent backscattered primary particles from entering the detector. The experimental setup is schematized in Fig. 3.

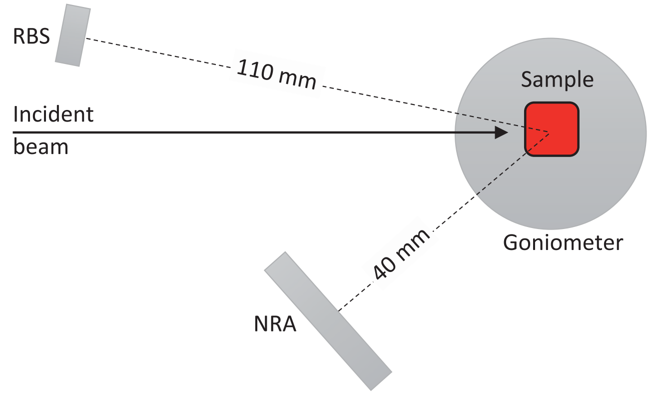

FIG. 3. Schematic representation of the experimental setup.

The reaction studied for the Al_2O_3 crystal was the production of ^{15}N atoms starting from ^{18}O with protons at 642.5 keV [$^{18}O(p,\alpha)^{15}N$], which has a sharp resonance at 628 keV. The reaction studied for the InP crystal was the production of ^{34}S starting from ^{31}P [$^{31}P(\alpha,p)^{34}S$] using α particles at 4.960 MeV, for which the nuclear reaction shows a sharp resonance [12].

Figure 4(a) shows the $^{18}O(p,\alpha)^{15}N$ reaction rate—which is proportional to the NEP with oxygen atoms—for the $\langle 0001 \rangle$ Al_2O_3 crystal as a function of the angle of incidence of the particles, with the trajectory being parallel to the $[1\bar{2}10]$ plane; an angle equal to zero corresponds to the $\langle 0001 \rangle$ axial channeling condition. The values in the plot are normalized to the reaction rate for random orientation, i.e., for a case comparable to that of an amorphous or randomly misaligned Al_2O_3 sample. The beam energy was 642.5 keV, about 14 keV above the resonance. One can see that when the beam is perfectly aligned with respect to the crystal $\langle 0001 \rangle$ axis, the interaction rate decreases to less than 40%. On the other hand, when the beam is aligned under the antichanneling condition, the interaction rate grows and reaches 1.2 times the rate under random orientation; i.e., the probability that the particles interact with the oxygen atoms increases of 20%.

The increase in the NEP is related to the energy of the outgoing α particles, which is a function of the penetration depth of the protons in the crystal. Indeed, the more a proton penetrates deep into the crystal, the more a generated α particle that exits the crystal loses energy, since it has to traverse a thicker portion of the crystal. By investigating the RBS spectra, it was possible to record an increased interaction rate of the proton beam with the oxygen atoms if the crystal was aligned in the antichanneling condition, reaching a NEP 73% higher with respect to the amorphous case, which indicates that the protons are actually focused on the oxygen atoms if the Al_2O_3 crystal is in an antichanneling condition.

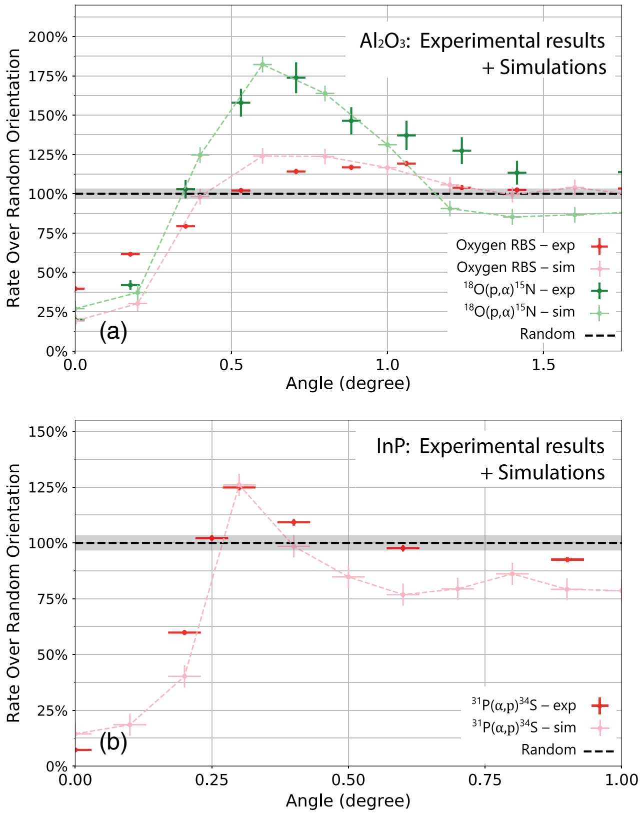

FIG. 4. Experimental and simulated interaction rates for Al_2O_3 (a) and InP (b) crystals as a function of the particle incoming angle. (a) The $^{18}O(p,\alpha)^{15}N$ reaction and RBS spectra for the $\langle 0001 \rangle$ axis to the $[1\bar{2}10]$ plane in an Al_2O_3 crystal with protons at 642.5 keV. (b) The $^{31}P(\alpha,p)^{34}S$ reaction for the $\langle 001 \rangle$ axis to the [010] plane in an InP crystal with α at 5.057 MeV.

Figure 4(b) shows the interaction rate of the $^{31}P(\alpha,p)^{34}S$ reaction for the $\langle 001 \rangle$ InP crystal as a function of the angle in the [010] plane. The data were collected at the CN accelerator with α particles at 4.968 MeV. A 1.25 increase in the nuclear interaction rate was recorded.

A series of *ad hoc* Monte Carlo simulations was worked out using the GEANT4 toolkit [13,14] in order to give an insight into the mechanism and to evaluate the effective enhancement rate of the nuclear interaction due to the antichanneling effect for the experimental data of this manuscript. The simulations are shown in Fig. 4. These simulations are similar to the numerical evaluations worked out by Chesnokov *et al.* for protons at higher energies traveling in bent crystals [15].

The channeling process is implemented by including DYNECHARM++ [16] and ECHARM [17] into the GEANT4 channeling package [18]. DYNECHARM++ allows the tracking of a relativistic charged particle inside a crystalline medium via the numerical integration of the classical equations of motion. The continuum potential approximation proposed by Lindhard is used [1]. ECHARM allows the computation of the electric field of the crystal within this approximation. The GEANT4 application was developed on top of the 10.3 version of the toolkit, which allows particle-trajectory simulations through crystalline structures [19]. All physical phenomena occurring for a channeled particle are strongly affected by the encountered number of nuclei and electrons, which depends on the particle trajectory [1]. Therefore, the probability of a physics process to occur in the simulation has been weighted as a function of the density of material experienced by a channeled particle.

The simulations show a good agreement with the experimental data. Indeed, the measured enhancement ratios turned out to be 75% and 25% for Al_2O_3 and InP materials, respectively, while the simulated enhancement ratios are $81 \pm 5\%$ and $26 \pm 5\%$

In this work, we show the possibility of increasing the nuclear interaction between a particle beam and a properly oriented crystal via antichanneling. Here, the antichanneling effect was experimentally observed for two different crystalline samples, i.e., Al_2O_3 and InP crystals, at various energy, as proof of concept.

The agreement between the simulations and the experimental results allows us to use GEANT4 as a tool for predicting the occurrence of antichanneling and its effects for various crystalline materials. A case of interest could be the radioisotope production with a charged beam, at the typical energies of cyclotrons, impinging on a properly oriented crystal. Indeed, the antichanneling effect would allow us to produce radio isotopes with higher efficiency; i.e., it would be possible to increase the production yield of radioisotopes consuming less time and/or using a smaller amount of prime material. In particular, antichanneling could be exploited to improve the performance for future cyclotrons using almost the same setup that already exists for solid targets, provided that the target is an aligned crystalline sample.

Modern medical applications, such as diagnosis and therapy of tumors, may benefit from an increased production yield of radioisotopes. Indeed, the spreading of these techniques is mainly inhibited by the limited availability of radioisotopes because they are costly and/or they have to be fast produced and transported. Cyclotrons represent the present and future source for radioisotopes for many hospitals, due to the possibility of producing radionuclides in loco.

In this context, antichanneling can be used for the enhancement of the production of zirconium 89 from yttrium crystals, i.e., to enhance the $^{89}Y(p,n)^{89}Zr$ reaction. Zirconium-89 is utilized in specialized diagnostic applications using positron emission tomography imaging, for example, with zirconium-89 labeled antibodies (immuno-PET) [20]. Figure 5 shows the enhancement of the nuclei encounter probability for a 17 MeV collimated proton beam impinging on the first layers of an yttrium crystal. In particular, the figure shows the transition from the $[1\bar{2}10]$ plane to random orientation. A $\sim 50\%$ enhancement of the nuclear interaction rate can be reached in the first crystalline layers by properly aligning the crystal under antichanneling conditions.

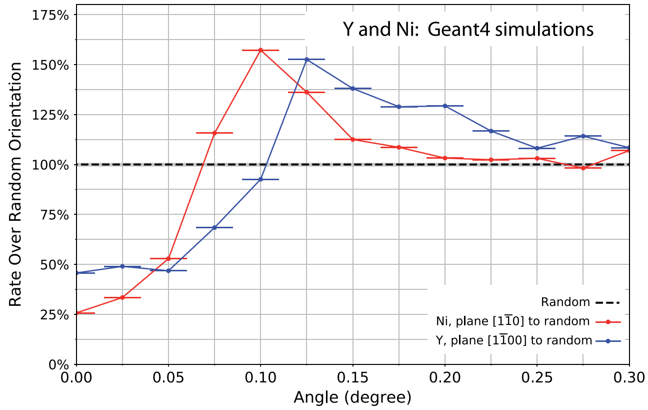

FIG. 5. GEANT4 simulations of interaction rates for Y and Ni crystals as a function of the particle incoming angle. The values are normalized to the average density ratio in the crystal. The transition from the [1$\bar{1}$0] plane to random for 13 MeV protons impinging on a Ni crystal and the transition from the [1$\bar{1}$00] plane to random for 17 MeV protons impinging on a Y crystal are shown.

The antichanneling effect could also be used for enriched crystalline material, i.e., for ^{64}Ni. A ^{64}Ni target is used to produce ^{64}Cu through the ^{64}Ni$(p,n)^{64}$Cu reaction, for which the cross section is maximum in an energy range between 10 and 14 MeV [21]. Figure 5 shows a simulation carried out with a 13-MeV collimated proton beam impinging on the first crystalline layers of a $\langle 111 \rangle$ Ni crystal. Also for this case, a ~55% enhancement of the nuclear interaction rate can be reached.

On the other hand, the importance of having a crystalline target for the production of radioisotopes via cyclotrons could encourage the industries of crystalline material, for the production of increasingly defect-free crystal samples and the development of new techniques for the manufacturing of new crystalline materials. Indeed, the simulations were worked out by considering the samples as ideal monocrystals.

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