Anisotropic passage of high-energy protons through a quasicrystalline material

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Theoretical and experimental results of investigations of the passage of protons with energies in the 1–1000 MeV range through a quasicrystalline material have been presented. The quasicrystalline material consisted of specially oriented graphite crystallites measuring no more than 1000 Å. The computer experiment was based on recognized models of the passage of protons through matter and the Monte Carlo method. It took into account energy-loss processes and multiple scattering of the protons and microstructural features of the material. A quantitative dependence of the characteristics of the passage of protons on the microstructural parameters of the material and on the direction of propagation and energy of the protons has been discovered. In some cases, the mean range of protons in the material decreased to 80%, and the range straggling varied by more than two- to threefold at a constant density of the material $\rho \cong 2.25$ g/cm^3. The authenticity of the effect has been confirmed by experimental measurements. © *1995 American Institute of Physics.*

1. INTRODUCTION

Utilization of the microstructural properties of solids for controlling beams of charged particles has recently become a very crucial problem in connection with the broad use of high-energy charged particles in solving fundamental and practical problems. Numerous effects discovered in this area of research, such as the shadow effect, channeling by bent crystals, etc.,[1,2] have found practical application.

The passage of high-energy charged particles through ordered systems (crystals) differs significantly from their propagation in an amorphous medium. The processes of planar and axial channeling, as well as the dechanneling of charged particles in crystals, should be mentioned first. When a charged particle travels at a small angle to atomic planes or linear chains, it interacts with an array of crystal atoms. In the case of propagation along a channel, the transverse momentum of the particle increases owing to the periodic arrangement of the atoms and their thermal vibrations, and the particle undergoes transitions to a dechanneling regime and to a multiple scattering regime. In a channeling regime, a charged particle loses an extremely small amount of energy, since it interacts with atoms at large impact parameters, and the energy losses on free electrons are fairly small. However, collisions with atoms at small impact parameters predominate in the dechanneling regime, increasing the losses of the charged particles in the crystal per unit length of trajectory.

The intensive investigation of these processes has led to the discovery of several orientational effects in crystals. In addition, numerous models reflecting particular aspects of the interaction of charged particles with a crystal have been developed. Advances in this area have made it possible to solve a long list of problems in accelerator technology, crystallography, and nuclear physics.[2,3]

However, there has been little research into problems concerning the influence of the parameters of extended oriented polycrystalline solid-state structures on the passage of high-energy charged particles, where various orientational peculiarities of the propagation of the particles may be manifested more strongly, depending on the characteristic dimensions of the crystallites and their manner of orientation. These peculiarities may not be described by the widely used models of multiple scattering,[1] but may be of investigative and practical interest.

Therefore, the aim of the present work was to study the influence of the structural characteristics of oriented polycrystals (quasicrystals) on the passage of fast charged particles through extended barriers of such materials.

2. DESCRIPTION OF THE MATHEMATICAL–PHYSICAL MODEL

Let us consider a broad class of oriented polycrystalline solid-state structures having the form of systems of crystallites that are co-oriented relative to the surface of the macroscopic sample and have characteristic mean dimensions $\geqslant 500$ Å in the orientation plane, which is perpendicular to the principal crystallographic axis, and dimensions equal to approximately 50–300 Å in the direction perpendicular to that plane. The spread of the misorientation angles of the planes of the individual crystallites relative to the surface of the macroscopic sample amounts to less than 10°.

The placement of the crystallites in the orientation plane itself has a random character, i.e., the individual crystallites are randomly displaced relative to one another in layers parallel to the orientation plane. Figure 1 shows the schematic form of the typical quasicrystalline structure of graphite. The characteristic mean dimensions of the crystallites L_x, L_y, and L_z, the spread of the misorientation angles $\Delta\theta_d$, and the characteristics of the graphite crystal lattice were taken as the parameters which specify the features of the quasicrystalline structure.

A model of the passage of high-energy protons through a quasicrystalline structure was developed with consideration of multiple Coulomb scattering, energy-loss fluctuations, and

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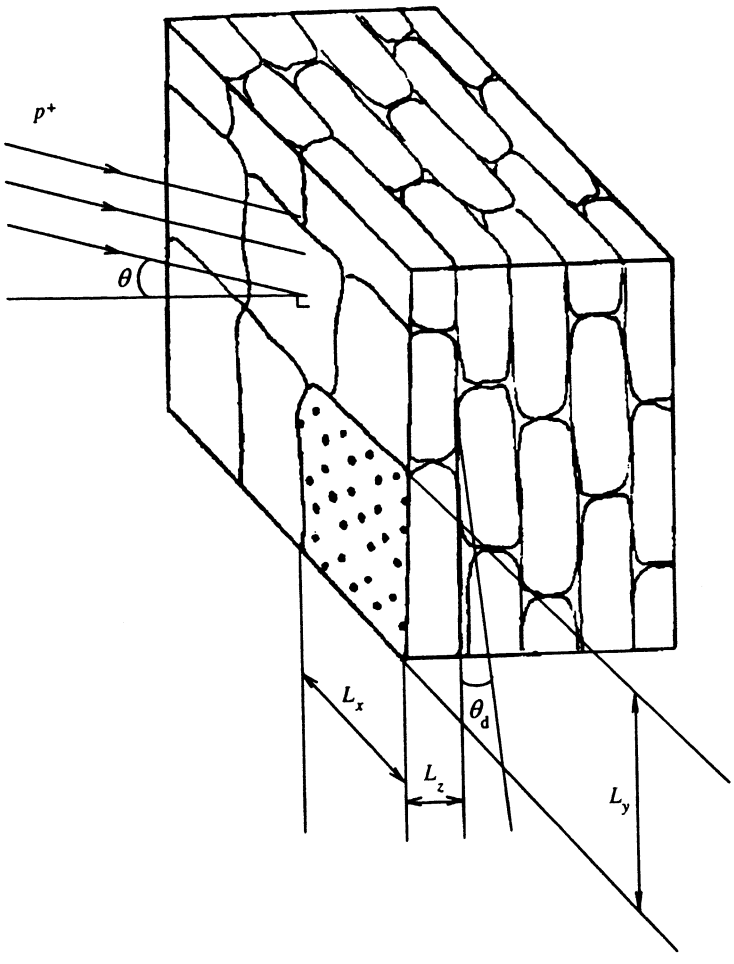

FIG. 1. Schematic representation of a quasicrystal and parameters specifying its microstructure.

the microstructural features of the medium, but without consideration of nuclear reactions.

The model of the passage of protons through such quasiordered structures is based on the following physical assumptions.

1. The contribution of channeling to passage through the structure under investigation is negligible for characteristic proton energies ranging from 1 to 100 MeV, since the random relative displacement of the crystallites in the orientation plane every 70–300 Å along the crystallographic axis results in the "disruption" of channeling and an increase in the probability of movement of the particles in the dechanneling regions of individual atomic chains.

2. For macroscopic thicknesses of the quasicrystal $L \cong 1$–100 μm, which satisfy the requirements of the Molière-Nigam theory of multiple scattering,[4,5] the angular distributions of the particles depend on the mean number of collisions with atoms of the structure and are equivalent to the angular distributions of the particles after passage through a layer of an amorphous substance in which the particles experienced the same mean number of collisions.

The model, which is based on the Monte Carlo method, is a two-level scheme, in which the anisotropic properties of the medium were simulated at the first level with consideration of the individual collisions of the particles with the atoms of an assigned microstructure. The passage of particles through extended quasicrystalline barriers was simulated at the second level using the results of the existing theories of the passage of fast ions in solids, as well as with consideration of the anisotropic properties of the medium, which depend on the direction in which each particle travels and its energy.

On this basis, the mean proton range in the quasicrystal $R_{sk}(\theta,E)$ as a function of the angle θ between the momentum vector of the proton and the normal to the macroscopic sample (see Fig. 1), and of the energy of the particle E, was taken as the natural functional parameter characterizing the anisotropy of the structure under investigation toward high-energy protons.

The function $R_{sk}(\theta,E)$ was simulated by the Monte Carlo method, approximating interactions of the particles with effective atomic spheres in individual collisions. The atoms were represented by interaction spheres with an effective radius $r_{eff}=(\sigma_{eff}/\pi)^{1/2}$, where

$$\sigma_{eff}=0.7\cdot 10^5 \pi Z^2 Z^{1/2}(Z+1)r_e^2(E+1)^2[E(E+2)]^{-1}$$

is the total elastic scattering cross section for hydrogen ions in the Thomas–Fermi approximation,[6] r_e is the classical radius of the electron, and Z is the atomic number of the target element. The structure of the quasicrystal was simulated by introducing a set of unit cells of the crystal lattice that completely covered the volume of the structure investigated. The

trajectory of a proton between two collisions was constructed by translating the unit cell with consideration of the dimensions and relative placement of the individual crystallites. The function $R_{sk}(\theta,E)$ was calculated by averaging the distances between collisions for all possible parameters of proton collisions in the structure: azimuthal angles, impact parameters, arrangement of the atoms in a crystallite, and relative placement of the crystallites themselves.

The passage of high-energy protons through extended graphite quasicrystals was thus simulated by the Monte Carlo method using the calculated functional parameter of the anisotropy of the structure $R_{sk}(\theta,E)$, but in a scheme of combined collisions.[1] The anisotropy of the medium was taken into account by introducing the variable effective number of target atoms per unit volume $N(\theta,E)$, which actually reflected the mean number of binary collisions of a proton with atoms of the quasicrystal as a function of the direction in which the particle travels and its energy:

$$N(\theta,E)=(R_{sk}(\theta,E)/R_{sa}(E))N_a,$$

where $R_{sa}(E)=1/(\sigma_{eff}N_a)$ and N_a is the number of atoms per unit volume in disordered graphite of the same density.

The scattering parameters were determined using Molière's theory of multiple scattering[4] as refined by Nigam *et al.*[5] in the simplified representation of Marion and Zimmerman,[5] where for nonrelativistic protons that have passed through a layer of matter of thickness t, which corresponds to a collision multiplicity of more than 20 and an energy loss from a particle equal to 1–3% of its initial energy, the angular distribution function of the particles is represented in the form

$$F(\vartheta)=[1/(\chi^2B)](F^{(0)}+(1/B)F^{(1)}+(1/B^2)F^{(2)}),$$

where $\vartheta=\theta/(\chi^2B)^{0.5}$, $\chi^2=(4\pi Nte^4Z(Z+1))/(p^2c^2\beta^2)$, θ is the scattering angle, N is the number of target atoms per unit volume, e is the charge of the electron, t is the thickness of the target, Z is the atomic number of the target atoms, p is the momentum of an impinging particle, c is the speed of light in vacuum, $\beta=v/c$, v is the velocity of an impinging particle, and $F^{(0)}$, $F^{(1)}$, and $F^{(2)}$ are Molière's universal functions as refined by Bethe.[7]

The parameter B was found from the equation

$$B-\ln B=b,$$

where $b=\ln[(2730(Z+1)Z^{1/3}t)/(A\beta)]-0.1544$ and A is the atomic weight of the target element. In the passage of high-energy protons through a heterogeneous medium, Z_c and A_c—the mean effective atomic number and mean atomic weight of the mixture—were used.

The energy loss of a particle between lattice points at which a "combined collision" takes place was calculated on the basis of the data in Ziegler and Andersen's monograph.[8] The energy-loss fluctuations were approximated by a Gaussian distribution in the Bohr approximation.[9] Chu showed[10] that this dependence exhibits good agreement with the Landau distribution for proton energies above 1 MeV.

Thus, the proposed approach makes it possible to simulate the passage of fast ions in extended barriers having a polycrystalline microstructure with consideration of the features of the crystal lattice, the dimensions of the crystallites, and their relative placement in the barrier material.

Numerical implementation of the model described above permits simulation of the passage of protons with energies up to 100 MeV in heterogeneous polycrystalline materials without taking nuclear reactions into account. The fraction of particles participating in nuclear reactions over the length of their ionization path does not exceed 20–25% for light nuclei ($Z\leqslant 30$) and an initial proton energy of 100 MeV, while the value for 100-MeV protons decreases to 5–7%. This permits the use of the model described for solving many practical problems.

The statistical error in the calculations of the integrated parameters of the propagation of protons in unoriented amorphous or polycrystalline materials was determined either on the basis of biased estimates of the dispersion of the respective quantities (when the central limit theorem holds) or by directly tracing the straggling of the random quantities during the counting at an assigned probability level of 0.95. In actual calculations with $N\cong 10\,000$ trajectories, the error for the mean proton range R_p did not exceed 0.7%, and the value for the range straggling of the particles (the width of the particle range distribution in the sample) ΔR_p was 10%. The program was tested with numerous experimental data,[8,9] and was in good agreement with the sources cited. The error in R_p did not exceed 2%, and the value for ΔR_p was no greater than 10–15%.

3. NUMERICAL SIMULATION OF THE PROPAGATION OF PROTONS IN A QUASICRYSTALLINE GRAPHITE STRUCTURE

To be specific, let us consider the following oriented polycrystalline structure of graphite. The structure consists of single crystals that are co-oriented relative to a planar surface of the macroscopic sample and have characteristic mean dimensions $L_x\cong L_y\cong 1000$ Å in the orientation plane and $L_z\cong 70$–100 Å in the direction perpendicular to that plane, the (001) plane of the graphite crystal lattice being parallel to the L_xL_y plane to within the misorientation angle θ_d. The spread of the misorientation angles of the individual crystallites relative to the surface of the macroscopic sample was $\Delta\theta_d\cong 1$–$3°$. The relative placement of the crystallites in the orientation plane itself had a random character, i.e., the individual crystallites were randomly displaced and tilted in layers parallel to this plane (Fig. 1).

Such graphite structures are accessible to existing technologies for obtaining pyrographite, and there is a real possibility of varying the parameters of the microstructure over broad ranges by technological means.

Using the previously described mathematical–physical model and the programs written to implement it, we obtained results on the passage of protons with energies up to 100 MeV through extended pyrographite quasicrystals with various statistical parameters for the structure. The results of the simulation of $R_{sk}(\theta,E)$ for a quasicrystal with $L_z\cong 70$ Å and $\Delta\theta_d\cong 3°$ (structure A) and various proton energies are presented in Fig. 2, where the averaged range of the protons was normalized to $R_{sa}(E)$: $R_s(\theta,E)=R_{sk}(\theta,E)/R_{sa}(E)$. It can be seen from the figure that $R_s\Rightarrow 1$ by the time $\theta\cong 7$–$8°$, i.e., at

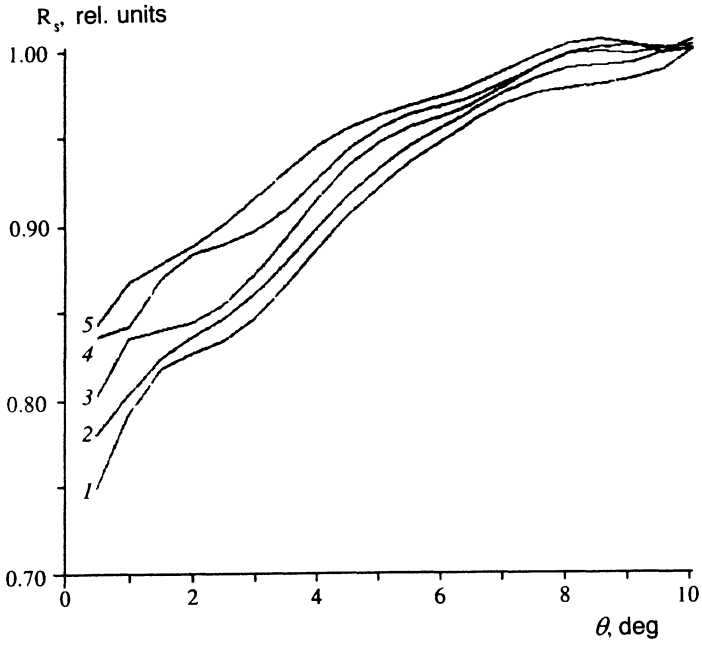

FIG. 2. Calculated plots of the range of protons versus θ in a quasicrystal of structure A: *1*) 4 MeV; *2*) 12 MeV; *3*) 30 MeV; *4*) 50 MeV; *5*) 80 MeV.

large angles of incidence of the particles to the first-order atomic chains there is virtually no orientational effect. It follows from the calculations that $R_s(\theta,E)$ has a dip at $\theta \leqslant 6-8°$, which narrows with respect to θ as the energy of the incident particle increases, but R_s continues to increase. The weak dependence of $R_s(\theta,E)$ on E allowed us to restrict ourselves to eight energy groups in the 0.5–100 MeV range in the calculations.

Figure 3 presents the results of the calculation of $R_s(\theta,E)=R_{sk}(\theta,E)/R_{sa}(E)$ for a graphite quasicrystal with $I_z \cong 70$ Å and $\Delta\theta_d \cong 1°$ (structure B) for various energies of the incident protons. It can be seen from the figure that for the structure with a smaller value of $\Delta\theta_d$, the dip in $R_s(\theta)$ deepens and narrows, if the values of I_z for the two structures are identical. As the energy increases, $R_s(\theta,E)$ also experiences decreases in the depth and width of the dip. In addition, this dependence shows up more strongly in the structure with the smaller value of $\Delta\theta_d$.

The trends just noted in the behavior of the anisotropy function $R_s(\theta,E)$ of the quasicrystals in response to the variation of $\Delta\theta_d$ and I_z have an adequately substantiated qualitative explanation. As E increases, the total interaction cross section σ_{eff} decreases, with a resultant decrease in the number of collisions over distances $R^i_{sk}<R_{sa}$ at any fixed θ. As the misorientation increases, the number of "short-range"

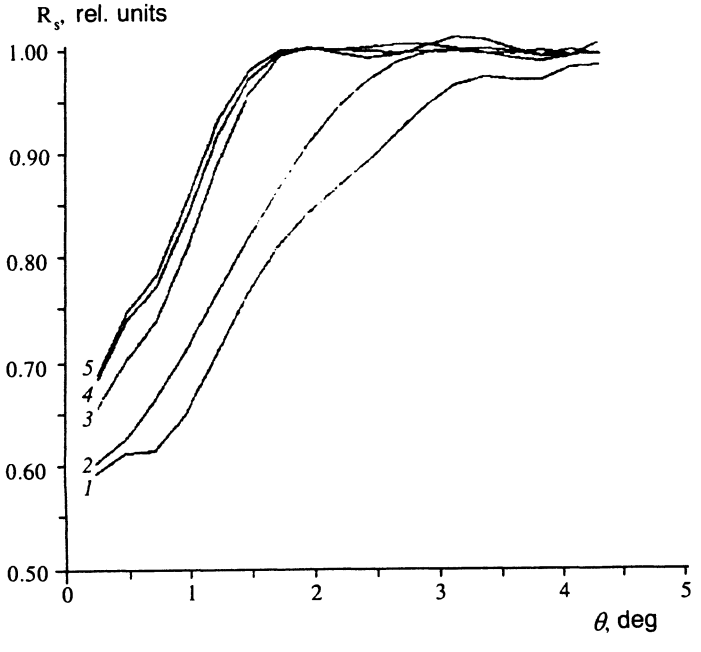

FIG. 3. Calculated plots of the range of protons versus θ in a quasicrystal of structure B: *1*) 4 MeV; 2) 12 MeV; 3) 30 MeV; 4) 50 MeV; 5) 80 MeV.

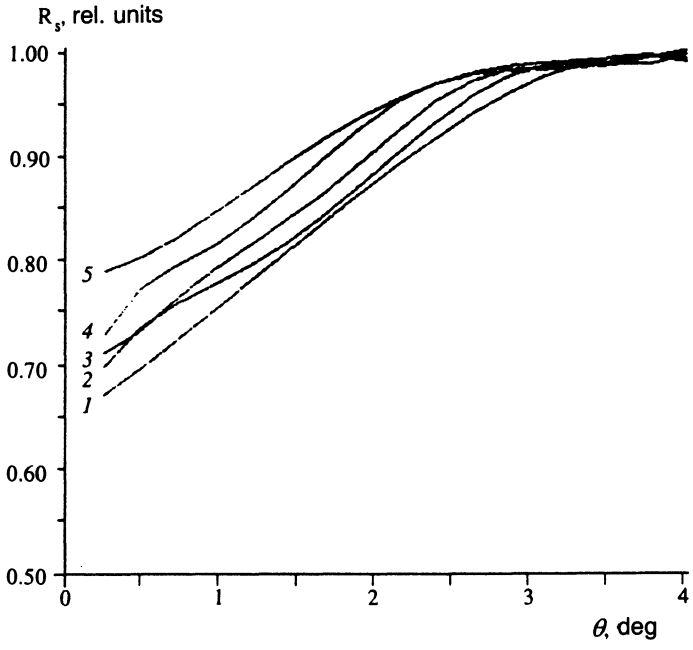

FIG. 4. Calculated plots of the range of protons versus θ in a quasicrystal of structure C: *1*) 4 MeV; *2*) 12 MeV; *3*) 30 MeV; *4*) 50 MeV; *5*) 80 MeV.

collisions ($R^i_{sk}<R_{sa}$) decreases, since the fraction of the collisions of a proton with neighboring atoms of a more inclined chain becomes smaller.

Figure 4 presents the results of the calculations of $R_s(\theta,E)$ for a structure with $L_z\cong 70$ Å and $\Delta\theta_d\cong 3°$ (structure C), which has a greater mean crystallite thickness L_z than structure B. As we see, a decrease in the degree of suppression of the anisotropy function $R_s(\theta,E)$ is observed for the quasicrystal structure with an increase in L_z. In addition, the dip has an insignificant depth, but is broader.

The trends discovered in the behavior of $R_s(\theta,E)$ as L_z increases are qualitatively attributable to the fact that at small θ the function $R_{sk}(\theta,E)$ in single crystals is significantly stronger than $R_{sa}(E)$ in an amorphous body, since channeling processes begin to play a significant role. In the case of $L_z\leqslant 100$ Å, "disruption" of channeling occurs often enough for the relation $R_{sk}<R_{sa}$ to be realized. An increase in L_z reduces the frequency of such random "disruptions," and a particle traverses greater distances without encountering effective interaction spheres.

The dependence of $R_s(\theta,E)$ calculated with consideration of the thermal vibrations of the graphite lattice atoms is interesting. The thermal vibrations of the atoms were simulated by random displacement of the interaction spheres within the range of the amplitude of the thermal vibrations of the lattice atoms, for which an estimated value was taken

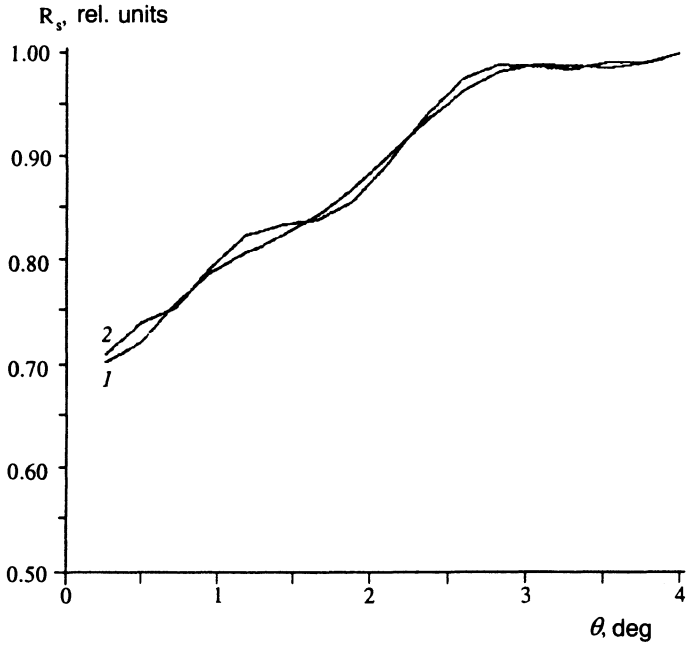

FIG. 5. Calculated plots of the range of protons with an energy of 12 MeV versus θ in a quasicrystal of structure C: *1*) without consideration of the thermal lattice vibrations; *2*) with consideration of the thermal vibrations at 300 K.

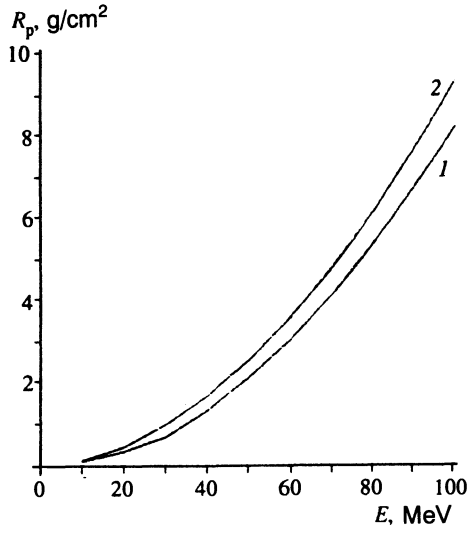

FIG. 6. Dependence of the mean range of protons on their initial energy: *1*) in a quasicrystal of structure *A*; *2*) in amorphous graphite of the same density.

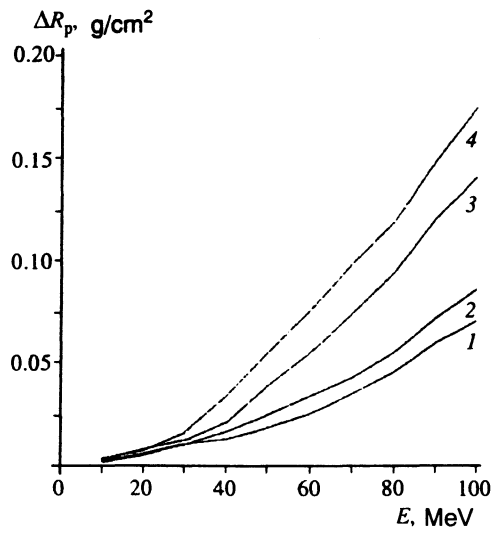

FIG. 7. Dependence of the range straggling of protons on their initial energy: *1*) in amorphous graphite; *2*) in a quasicrystal of structure *B*; *3*) in structure *C*; *4*) in structure *A*.

from Ref. 1 for $\simeq$20 °C. Figure 5 presents the results of the calculations of $R_s(\theta)$ in structure *C* for a proton energy of 12 MeV with and without consideration of the thermal vibrations. As is seen from the figure, the differences between these two cases are negligible and fall within the statistical error.

The dependences of $R_s(\theta,E)$ obtained for different structures of quasicrystalline graphite were used to simulate the passage of high-energy protons over distances of the order of the mean range in them according to the combined collision scheme.

Figure 6 presents calculated plots of the mean free path of the protons R_p as a function of the initial energy of the particles E in a graphite quasicrystal of structure *A* and in unoriented graphite of the same density. The figure reveals a

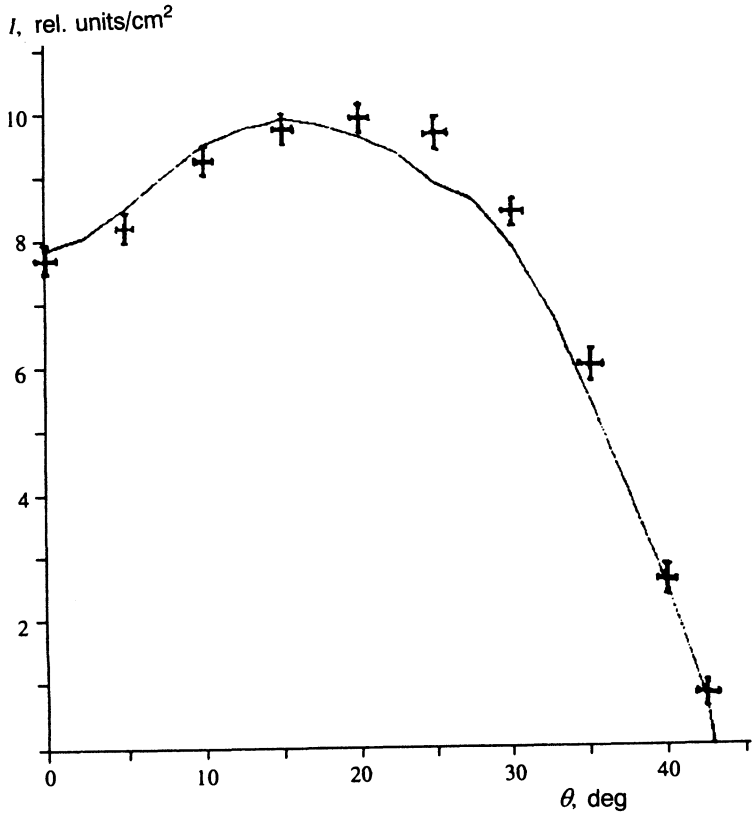

FIG. 8. Calculated and experimental values of the intensity of a beam of protons with an energy of 30 MeV scattered by a quasicrystalline barrier as a function of the angle of inclination of the barrier relative to the beam axis.

significant decrease in the mean range of the particles in the quasicrystal relative to amorphous graphite over the entire range of energies investigated. This model result may be of practical value in solving some specific problems of radiation shielding from high-energy charged particles.

Figure 7 presents calculated plots of the range straggling of protons versus their initial energy in amorphous graphite with a density of ≈ 2.25 g/cm^3, as well as in various quasicrystalline pyrographite structures. The figure demonstrates a strong dependence of the range straggling of the protons on the microstructural features of the medium. The results may be of interest in problems concerning implantation and the doping of materials by fast ions.

To test the proposed model and the programs implementing it in the two-level Monte Carlo scheme, as well as to experimentally confirm the anisotropy effects discovered, we performed a special experiment on a cyclotron of the U-150M synchrotron type (Institute of Nuclear Physics of the Kazakhstan Academy of Sciences, Alma-Ata). The experimental scheme was as follows. A beam of protons with an energy of 30 MeV, an energy straggling $\Delta E \simeq 1.5$ MeV, and an angular straggling of the particles $\Delta\theta \simeq 3°$ was incident upon a sample of quasicrystalline carbon (pyrographite) placed in a frame that is impermeable to the beam protons. A copper plate covering the aperture for protons scattered relative to the beam axis by more than 70° was placed behind the sample. The proton current by the quasicrystal was recorded from the plate. The thickness of the sample was 3.6 mm, which corresponds approximately to 60% of the mean range of protons with that energy in unoriented graphite of the same density ($\rho \simeq 2.25$ g/cm^2). A sample of a quasicrystal having the structural parameters $L_x \cong L_y \cong 1000$ Å, $L_z \cong 70$ Å, and $\Delta\theta_d \cong 3°$ (structure A) was oriented at an angle θ relative to the beam axis. Figure 8 presents the experimentally measured intensity of the scattered proton current $I(\theta)$ [rel. units/cm^2] versus the angle θ between the beam propagation axis and the normal to the surface of the sample. The figure also presents a calculated plot of $I(\theta)$ obtained on the basis of the models and programs described above for the conditions of the experimental setup. As is seen from Fig. 8, the characteristic dip on the plot of $I(\theta)$ at $\theta \leqslant 2°$ corresponds to as much as 15–20% of the maximum current intensity at $\theta \simeq 15°$. Such behavior of $I(\theta)$ was not observed when samples of unoriented types of graphite of the same density were used.

The good agreement between the calculated and experimental data points out the faithfulness of the proposed model and the authenticity of the effect discovered.

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