Charged particle propagation through nanostructures

and associated radiation

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Abstract In this report, using computer simulations, we investigate the channeling of high-energy charged particles in nanotube ropes and fullerites and estimate the capability of bent nanocrystals to deflect a particle beam. We also discuss electromagnetic radiation arising both from the non-uniform motion of the particles in the electrostatic potential of aligned atoms and from the transient polarization of the medium caused by the particles.

Keywords Relativistic electron and positron beams, Nanocrystalline materials, Fullerenes, Nanotubes, Channeling, X-ray and gamma radiation

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1 Introduction

Channeling has found various applications in solid state physics (impurity localization or ion implantation, in crystals) and in high-energy particle physics (deflection of proton beams by bent crystals, measurement of the magnetic moments of short-living particles, etc.). Channeled electrons or positrons emit X- and gamma-rays with a much larger total intensity than bremsstrahlung at random orientation. This is used as a source of high-energy photons for particle physics or to enhance the efficiency of positron sources. The other interesting kinds of radiations emitted by charged particles in crystals are parametric X-rays (PXR), which is due to the diffraction of the electromagnetic field of a charged particle, and coherent bremsstrahlung. Their useful quasi-monochromatic spectra and linear polarizations come from interferences between the periodically spaced atomic rows or planes.

Fullerenes and nanotubes, recently discovered, are the building blocks of a new class of ordered materials.^[1,2] Spherical fullerene C_{60} molecules can assemble in fcc crystals called fullerites and single-wall nanotubes can gather in ropes whose cross section makes an hexagonal lattice. These structures differ from ordinary crystals in many points. In particular,

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they have spatial periods of the order of one nanometer, which is much larger than in ordinary crystals. In addition, they are very inhomogeneous in density, being made of hollow "cages" surrounded by a dense net of carbon atoms. In this report we discuss the peculiarities that the above mentioned phenomena may have in fullerites and nanotube ropes.

2 Geometrical structure of a nanotube

A carbon nanotube can be regarded as one gigantic carbon molecule (fullerene) which is obtained by folding graphite planes into a cylinder whose diameter is measured in nanometers and whose length can reach macroscopic dimensions.^[2] This linear structure determines the extremely high mechanical strength of nanotubes^[3] whereas their electrical conductivity depends strongly on the diameter and the helicity which is the angle between the most highly packed chains of atoms and the axis of the cylinder.^[4] There exist nanotubes whose walls contain a single layer of atoms (single-wall nanotubes, or SWNT)^[5] and nanotubes with walls consisting of several concentric or onion-like layers (multi-wall nanotubes, or MWNT).^[6] A remarkable feature of some single-layer nanotubes is their capability, as they grow from a plasma, to unite into a "rope" whose transverse cross section is a two-dimensional hexagonal superlattice.^[7] Since the

discovery of nanotubes in 1991 by Iijima,^[6] a large number of papers have appeared on the problems of synthesizing nanotubes, on their physical properties and on possible applications in nanoelectronics,^[8] catalysis^[9] and other fields. It should be noted that, besides nanotubes consisting of carbon atoms, similar structures based on boron nitride also exist. However, in this report we restrict ourselves to the case of single-wall carbon nanotubes.

Let *a* and *b* denote the basis vectors of a planar lattice of graphite, whose unit cell (dashed rhombus in Fig.1(A) contains two carbon atoms of coordinates (a+b)/3 and 2(a+b)/3, respectively. Taking account of the fact that the angle between the vectors **a** and **b** is $\pi/3$ and the vectors have the same modulus (a = b) and denoting by l the length of the bond between the carbon atoms (which is usually 0.14 nm), we obtain $a=b=l\sqrt{3}$. The roll-up vector \mathbf{r}_0 is determined as a linear combination $r_0 = na + mb$ of the basis vectors, where the pair (n,m) of integers is called the indices of the nanotube. The nanotube may be considered as a strip of width r_0 , marked up by dashed lines in Fig.1(A), cut out of the graphitic carbon plane perpendicularly to r_0 , rolled-up in a cylinder and closed at either end with caps containing carbon pentagons in a manner that conserves bonding length. The angle θ between r_0 and the lattice vector a defines the corkscrew symmetry of the nanotube and is called helicity or chiral angle. The helicity is also equal to the angle under which the most closely packed chains of carbon atoms are wound on the cylindrical surface of the tube. As follows from simple geometry, the nanotube indices(n,m) uniquely define the nanotube diameter d and helicity θ

$$d = (l\sqrt{3} / \pi)(n^2 + nm + m^2)^{1/2}, \qquad (1)$$

(2)

$$\theta = \arctan\left[\sqrt{3} m/(m+2n)\right]$$



Fig.1 Structure of an unrolled nanotube wall (A) and of the cross section of a nanotube rope (B).

Since the basis vectors **a** and **b** are equivalent, it can be assumed with no loss of generality that $n \ge m$ and therefore the helicity lies in the range $0 \le \theta \le \pi/6$. SWNTs with m=0 ($\theta=0^{\circ}$) are labeled as zigzag, tubes with m=n ($\theta=30^{\circ}$) are labeled as armchair, all other tubes (between $\theta=0^{\circ}$ and $\theta=30^{\circ}$) are chiral (helical). The structures of different SWNTs are presented in Fig.2.



Fig.2 Structure of (10, 10) armchair, (20, 0) zigzag and (16, 5) chiral nanotube.

It is easy to show^[10] that the spatial symmetry of a SWNT along its axis is determined by the following translational vector

$$t = q^{-1}[(2m+n)a - (2n+m)b],$$
(3)

which occurs to be orthogonal to r_0 (indeed, $tr_0=0$) and thus directed along the nanotube axis z. The integer $q=\gcd(2m+n, 2n+m)$ denotes the greatest common divisor of its arguments, therefore the modulus t of vector t represents the main spatial period of a nanotube along z. Thus in physics terms an isolated nanotube can be treated as a one-dimensional single crystal of a period t and with $2N = 4(n^2 + nm + m^2)/q$ atoms in the unit cell.

3 Nanotube continuum potential

It is noteworthy that channeling was primarily discovered as a result of computer simulations of ion beam propagation along atomic rows in a crystal, based on binary collisions of ions with atoms.^[11] After the experimental evidence of channeling effect^[12] Lindhard^[13] gave a simple theoretical explanation of it. He showed that if a fast charged particle enters a single crystal at small enough angle θ_0 with respect to an atomic row, it is governed by the continuum potential, i.e. the actual periodic potential of the rows averaged over the direction parallel to the rows. Then Erginsoy^[14] went further and in the case of a small enough angle with respect to a family of atomic planes (but far from directions of main crystallographic axes) he introduced the continuum potential of the planes.

Applying the concept of continuum potential to nanotubes it is important that a nanotube with indices (n,m)can always be represented as a collection of atomic rows parallel to the axis of the nanotube and arranged in a definite manner along the perimeter of the cylinder. In general, the distance d_R between the neighbouring atoms within such a row coincides with the modulus of the translational vector **t** and is given by

$$d_{\rm R} = (3l/q)(n^2 + nm + m^2)^{1/2} \tag{4}$$

Let us take into account the fact that the unit cell of a graphite plane shown in Fig.1(A) as a rhombus contains two carbon atoms and the surface density σ of atoms on the plane is $\sigma = 3^{-3/2} \times 4l^2$. After the rolling up of the plane the pair of the unit cell atoms generates two sequences of atomic rows parallel to the axis of the nanotube. In each of the sequences the rows are equally spaced around the cylinder. We choose a cylindrical coordinate system (r, φ, z) in which the radial coordinate *r* is measured from the axis of the tube, and we denote by $\varphi_k^{(\mu)}$ the azimuthal angle of the *k*th row (k=0,1,2,...,N-1) in the corresponding sequence of rows $(\mu = 1,2)$. Thus each sequence contains exactly

$$N = (2/q)(n^2 + nm + m^2)$$
(5)

atomic rows, whose azimuthal coordinates are determined by the relations

$$\varphi_k^{(1)} = k \delta \varphi, \quad \delta \varphi = \pi q / (n^2 + nm + m^2), \tag{6}$$

$$\varphi_k^{(2)} = \varphi_\kappa^{(1)} + \Delta \varphi, \quad \Delta \varphi = \pi (n+m)/(n^2 + nm + m^2).$$
 (7)

We note two limiting values of the helicity $\theta = 0$ and $\theta = \pi/6$ where the atomic rows, parallel to the axis of the nanotube, have the largest linear density and therefore the number of the rows is relatively small. The first case corresponds to zigzag nanotubes (n,0); here, q = n, N = 2n, $d_R = 3l$, $\delta \varphi = \Delta \varphi = \pi/n$, and the two row sequences overlap, i.e. there are actually 2n rows with a doubled linear atomic density $2/d_R$. This overlap does not occur with zigzag nanotubes exclusively, but is found, for instance, in chiral nanotubes with n=2m. The other case corresponds to armchair nanotubes (n, n); here q=3n, N=2n, $d_R=l\sqrt{3}$, $\delta \varphi=\pi/n$, $\Delta \varphi = 2\pi/(3n)$, i.e., there are 4n rows arranged in pairs.

It is interesting to note that the total number of the rows 2N (sometimes overlapping) coincides with

the number of carbon atoms in the unit cell of a nanotube considered as a one-dimensional single crystal. The continuum potential of a nanotube can be written as

$$U(\mathbf{r}) = \frac{1}{d_R} \int_{-\infty}^{\infty} \sum_{i=1}^{2N} u(\mathbf{r} - \mathbf{r}_i, z - z_i) \,\mathrm{d} z \tag{8}$$

where $u(\mathbf{r} - \mathbf{r}_i, z - z_i)$ is the potential of a carbon atom number *i* with coordinates (\mathbf{r}_i, z_i) in the nanotube unit cell. Eq.(8) can be easily modified in order to take into account thermal displacements of carbon atoms from their equilibrium positions. The further analytical calculations of the nanotube potential (8) are based on the particular expression for the Fourier-transformed potential $f(\mathbf{k})$ given by Doyle and Turner:^[15]

$$f(\mathbf{k}) = 4\pi Z e \sum_{j=1}^{N=4} a_j \exp[-\mathbf{k}^2 / (4b_j^2)]$$
(9)

where

 $a_j = \{3.222, 5.270, 2.012, 0.5499\} \times 10^{-4} \text{ nm}^2$ $b_j = \{10.330, 18.694, 37.456, 106.88\} \text{ nm}^{-1}$

are dimensional parameters which were determined from the condition of the best fit of (9) to the most accurate calculations of $f(\mathbf{k})$ based on the Hartree-Fock method.^[16] The Doyle-Turner model (9) corresponds to the following representation of atomic potential in real space

$$\varphi(r_a) = \frac{4Ze}{\sqrt{\pi}} \sum_{j=1}^{4} a_j b_j^3 \exp(-b_j^2 r_a^2)$$
(10)

To obtain the continuum potential of a nanotube rope inside the superlattice unit cell (shown as hexagons in Fig.1(B)) it is necessary to add the continuum potentials of a sufficient number of neighbouring nanotubes.

The calculated continuum potentials of the ropes of (10, 10) armchair, (20, 0) zigzag and (16, 5) chiral nanotubes are illustrated in Figs.3~5, respectively. For positive particles there are relatively deep potential wells inside nanotubes, about 15.9 eV for (10, 10), 24 eV for (20, 0) and 60 eV for (16, 5), and very shallow (about 1 eV deep for all of the ropes) potential well between the nanotubes. It can be seen that chiral nanotubes like (16, 5) have the deepest and axially symmetrical potential well inside. Their continuum potential can be also considered as originated from the

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potential of carbon atoms of a graphitic plane averaged over the plane.



Fig.3 Continuum potentials of the ropes of (10, 10) armchair nanotubes.



Fig.4 Continuum potentials of the ropes of (20, 0) zigzag nanotubes.



Fig.5 Continuum potentials of the ropes of (16, 5) chiral nanotubes.

Continuum potentials of fullerites

Fullerenes are molecular cages consisting of carbon atoms. The arrangement of atoms is almost exclusively in the form of hexagons and pentagons. Nanotubes discussed in the previous subsection may be considered as a kind of fullerenes. Twelve pentagons and twenty hexagons make the famous C_{60} cage. The C_{60} molecule contains a carbon at each vertex of the "truncated icosahedron", C=C double bonds along the lines separating hexagons, C–C single bonds along the hexagon-pentagon boundaries, and has a diameter of roughly 7.1Å.

This cage was first identified experimentally in Ref.[17]. It might indeed impart unusual stability to the C₆₀ cluster because all valences are satisfied. The proposed molecule was dubbed buckminsterfullerene because its shape is reminiscent of the geodesic domes popularized by American architect Richard Buckminster Fuller. Bulk solid C₆₀ is sometimes referred to as "fullerite" in analogy to graphite, and X-ray powder diffraction has shown^[18] that fullerite adopts the face-centered cubic (fcc) structure with a relatively small Van der Waals cohesive energy and a lattice constant a = 14.17 Å. The molecules in fullerite freely rotate for temperatures above ≈ 257 K. At lower temperatures, they begin to "stick" at certain orientations and eventually, below \approx 90 K (the precise value depends on how fast the system is cooled), they become completely stuck. Besides, the low temperature structure is no longer fcc but simple cubic.

Another interesting feature of fullerite is the ability of the C₆₀ units to polymerize at high pressure (30~80 kbar) and high temperature (600~1000 K)^[19] or under intense exposure to visible or UV light. Three-dimensional polymerization can lead to ultra-hard material, a material which is harder than diamond.^[20] The density of C₆₀ formed as a fcc crystal at 298 K is 1.65 g·cm⁻³ while the density of a new solid formed by 3D polymerization is 2.6 g·cm⁻³.

Besides fullerites consisting of C_{60} fullerenes, there exist similar molecular crystals consisting of "non-spherical" C_{70} molecules.^[21] The fullerene derivatives (fullerides), such as compounds of fullerenes with alkaline metals, may also have a crystalline structure.^[22] From the practical point of view, fullerites have still the evident advantage of being presently grown in single crystals of macroscopic dimensions, while existing nanotube ropes are available only as microscopic objects as yet. The speci.c features of continuum potentials and channeling in C_{60} fullerites were first considered in Ref.[23] and [24].

It should be noted that in the case of nanotube ropes discussed above the existence of a regular ordering between nanotubes is not necessary, at least for the channeling along the ropes. By contrast, for the channeling in 3D crystals, particularly in fullerites, the existence of perfect enough crystalline structure is crucial.

There are 360 electrons in the C_{60} molecule, half of which are involved in typical covalent bonding between the atoms of the molecule. In a first approximation, neglecting the influence of the bonding on spatial distribution of the valence electrons, we may calculate the total potential of C₆₀ fullerene as a sum of atomic potentials centred at the vertices of the truncated icosahedron. We consider a normal fullerite crystal at room temperature in which fullerenes are not stuck at certain orientations or polymerized, but weakly bound with Van der Waals forces, and can rotate freely. When a fast charged particle travels along a fullerene row in a fullerite it collides with randomly oriented molecules and in the first approximation it is reasonable to average the potential of a fullerene row not only over the longitudinal direction, but also over all angular coordinates of carbon atoms. As a result of the averaging over the random orientations we find the fullerene potential (for a positron) in the form

$$U_1(r) = 60 \frac{Ze^2}{\pi^{1/2} rR} \sum_{j=1}^4 a_i b_j [\exp(-b_i^2 (r-R)^2) - \exp(-b_i^2 (r+R)^2)]$$
(11)

where *r* is the distance from the centre of the fullerene and R = 3.53 Å is the fullerene radius. Further averaging (11) along the row gives the continuum potential of the fullerene row

$$U_{R}(\rho) = 60 \frac{2Ze^{2}}{Rd_{R}} \sum_{j=1}^{4} a_{j} \int_{0}^{\infty} \exp(-k_{\perp}^{2} b_{j}^{-2} / 4) J_{0}(k_{\perp} \rho) \sin(k_{\perp} R) dk_{\perp}$$
(12)

where $d_{\rm R}$ is the distance between the centres of two neighbouring fullerenes in the row, ρ is the distance from the row, J_0 is the Bessel function and the channeling particle considered has charge +*e*. The effective potential $U(\mathbf{r})$ acting on an axially channeled particle is the sum of potentials of the form (12) of all rows parallel to the given crystallographic direction:

$$U(\mathbf{r}) = \sum_{n} U_{R}(|\mathbf{r} - \boldsymbol{\rho}_{n}|)$$
(13)

where *r* denotes the vector coordinate in the plane normal to the rows, and ρ_n is the vector coordinate of the *n*th row.

The calculated continuum potential of a fullerite at room temperature for positron channeling along the [100] direction is presented in Fig.6.

Cartesian coordinates x, y are in the plane normal to the rows and measured in units of fullerene radius Rwhile the *z*-coordinate of the plot represents the values of the continuum potential of a fullerite measured in eV. The potential wells inside the rows have almost axial symmetry and are about 7 eV deep. Other wells about 14 eV deep are located between the rows. The wells are separated by a relatively thin potential barrier. We come to the conclusion that two kinds of channeling of positive particles may exist in fullerites, one corresponding to the motion between the rows (like in the ordinary crystals), the other one corresponding to the motion inside the rows (impossible in the ordinary crystals). For negative particles (electrons) the sign of the potential must be changed to negative, the wells transform to the barriers and vice versa and as a result there are practically no potential wells for the channeling of electrons along the [100] direction.

However, more favorable conditions for electron channeling exist in the [110] direction. The corresponding continuum potential for electrons is shown in Fig.7. There are wide and deep enough (about 40 eV) potential wells separated by relatively thick barriers. The interesting feature is the existence of the central bump inside the wells. On the contrary, for positrons in the [110] direction the central well is narrower and much shallower than in the [100] direction.



Fig.6 Continuum potential of a fullerite at room temperature for positron channeling along the [100] direction.



Fig.7 Continuum potential of a fullerite at room temperature for electron channeling along the [110] direction.

5 Motion equations

In channeling the characteristic quantum parameter is the de Broglie wavelength of the particle $\lambda_D = \hbar / m v \gamma$ divided by $\sin \theta_0$ where θ_0 is the incidence angle. The typical value of θ_0 is the Lindhard angle $\theta_{\rm L} = (2U_0/E)^{1/2} \ll 1$, where U_0 is the potential well depth for the transverse motion. The light speed c is equal to unity and m, E, v, γ are the mass, total energy, velocity and Lorentz factor of the particle, respectively. The typical transverse de Broglie wavelength $\lambda_{\perp} = \lambda_{\rm D}/\theta_{\rm L}$ is proportional to $E^{-1/2}$ and can be comparable to the width of the channels d_w up to relatively high energies $E \sim 10^2$ MeV of electrons and positrons. Hence, the transverse motion of particles in the channels may be considered either as classical or quantum-mechanical, depending on the ratio $d_{\rm W}/\lambda_{\perp}$. For the particles heavier than electrons this ratio is always high and the transverse motion may be considered in terms of classical trajectories. However, for electrons and positrons in the MeV energy range, numerical

estimates show that quantum effects are important. In order of magnitude the ratio $(d_w / \lambda_{\perp})^2$ represents the number of quantum bound states of the transverse motion in the two-dimensional well. Due to quantum tunneling through the potential barrier separating neighbouring channels, there are energy bands rather than discrete energy levels. Besides, incoherent scatterings of channeled particles on the electrons of the crystal and on atomic vibrations give rise to an additional broadening of the transverse energy levels. To neglect quantum tunneling through a potential barrier we must additionally assume that λ_{\perp} is small compared to the wall thickness. This may be the more severe condition for the applicability of the classical approach to channeled positrons (especially moving close to the top of the wall). Quantum channeling is a well-known phenomenon in ordinary crystals^[25] where in the axial case it is noticeable up to electron energies of 50 MeV.^[26] However, in nanocrystals the situation may be different due to the larger width of the potential well.

The theory of quantum channeling in nanotubes and fullerites can be briefly described as follows. The wave-functions of a channeling relativistic electron or positron are presented as a product of the plane-wave $\exp(ip_z z)$, corresponding to the uniform motion in the axial direction, and the transverse wave-function $\psi(r)$ describing the particle motion across the axial channel in the continuum potential and satisfying the following equation

$$[\Delta_r + p_\perp^2(\mathbf{r})]\psi(\mathbf{r}) = 0 \tag{14}$$

where Δ_r is the Laplacian in the plane of the transverse coordinates \mathbf{r} , $p_{\perp}^2(\mathbf{r}) = 2E[\varepsilon - U(\mathbf{r})]$, E is the total energy of the relativistic particle, $\varepsilon = E - (p_z^2 + 1)^{1/2}$ is the so-called transverse energy, $U(\mathbf{r})$ is the continuum potential (to simplify the formulae we use here and below $\hbar = m = c = 1$ as units). Eq.(14) looks like the Schroedinger equation, however E plays the role of the "relativistic mass" of the particle. Eq.(14) implies that spin effects are negligible.

It is well known that the number of transverse states in the axial potential wells grows approximately linearly with the particle energy E and for electron (positron) energies about 10 MeV it becomes possible to use classical mechanics for the description of the

transverse motion. As noted above, heavier particles, like π -mesons and protons, can be treated as classical irrespective of their kinetic energy. Let us first consider the case of classical motion of high-energy particles in a straight nanotube rope or a non-deformed fullerite. Since the continuum potential U(r) of the axial channels does not depend on time and on the longitudinal (with respect to the channels) coordinate z, there are two integrals of motion : the total energy of the particle E and the relativistic longitudinal momentum p_{\parallel} :

$$E = U(\mathbf{r}) + (1 - v_{\parallel}^2 - v_{\perp}^2)^{-1/2}$$
(15)

$$p_{\parallel} = v_{\parallel} (1 - v_{\parallel}^2 - v_{\perp}^2)^{-1/2}$$
(16)

where v_{\parallel} denotes the longitudinal velocity, $v_{\perp} = dr/dt$ is the two-dimensional vector of the transverse velocity, the system of units $\hbar = m = c = 1$ is used to simplify the expressions. The so-called transverse energy ε is defined as the following difference $\varepsilon = E - (1 + p_{\parallel}^2)^{1/2}$, which is an integral of motion too. On channeling or close to channeling conditions the inequality $\varepsilon \ll E$ is always satisfied, then using the definition of ε and Eqs.(15), (16) we may rewrite it as

$$\varepsilon = \frac{E \boldsymbol{v}_{\perp}^2}{2} + U(\boldsymbol{r})$$

from which we find the following well-known equation of motion in the transverse plane

$$\frac{\mathrm{d}^2 \boldsymbol{r}}{\mathrm{d} t^2} = -\frac{1}{E} \nabla U(\boldsymbol{r}) \tag{17}$$

This equation can be recognized as the Newton equation for non-relativistic transverse motion of a particle with "relativistic mass" E and potential energy $U(\mathbf{r})$. It is in full agreement with the corresponding quantum analog Eq.(14).

The bending of a nanotube or the crystallographic axis of a fullerite with a constant curvature radius R_b (large compared to the diameter of a nanotube rope or the thickness of a fullerite single crystal) can be taken into account in the analysis of the transverse motion by replacing the electrostatic potential $U(\mathbf{r})$ in Eq.(17) by the e^{\(\mathbf{e}\)} ective potential,^[27]

$$U_{\rm eff}(\mathbf{r}) = U(\mathbf{r}) - \frac{Ex}{R_b}$$
(18)

where x is the transverse coordinate in the plane of

bending. A particle which is initially in the middle of the channel and with zero transverse velocity can overcome the potential barrier if U_{eff} (d/2,0)<0, i.e., if the curvature radius is less than the so-called Tsyganov radius $R_c = Ed/(2U_0)$ determining the tolerance of the channeling to crystal bending.

6 Dechanneling and computer simulation method

The Lindhard continuum potential describes totally coherent scattering of particles by carbon atomic or fullerene rows conserving the transverse energy. We discuss now how the motion equations should be modified to take into account the incoherent scattering which violates the conservation of the transverse energy and eventually leads to the dechanneling.

For this purpose, following Ref.[28], let us consider with more details the scattering of the fast particle by the row of vibrating atoms. Let ρ_0 and z be the transverse radius-vector and the longitudinal coordinate of the particle, ρ_i the vector displacement of the ith atom from the axis of the row due to the thermal vibrations, $w(\rho_i)$ the probability of the displacement, $N = \Delta z/d_R$ the number of atoms with which the particle interacts on its path Δz along the row, and d_R the mean inter-atomic distance along the row. The potential acting on the fast particle from *i*th atom is $U_1(r_i)$, where $r_i = [|\rho_i - \rho_0|^2 + (z - id_R)^2]^{1/2}$ is the distance between the particle and the carbon atom (we can assume that the row is made of equidistant atoms). The transverse momentum q transferred to the particle due to the scatterings on N atoms of the row, on assumption that the scatterings are independent, may be presented as a sum

$$\boldsymbol{q} = \sum_{i=1}^{N} \boldsymbol{q}_i$$

where q_i refers to the momentum transfer by the *i*th individual atom. If we assume that the variations of the transverse coordinate of the particle $|\Delta \rho_0|$ over the path Δz along the row is small compared to the distance at which atomic potential changes significantly, we find the following expression

$$q_i = -\nabla \int_{-\infty}^{\infty} U_1 (|\rho_i - \rho_o|^2 + z^2)^{1/2} dz$$

where ΔU_1 denotes the gradient of atomic potential in

the transverse plane.

We may represent the mean (over thermal vibrations) momentum transfer $\langle q(\rho_0) \rangle$ as the sum of the corresponding values for the individual atoms:

(

$$\left\langle \boldsymbol{q} \right\rangle = \sum_{i=1}^{N} \left\langle \boldsymbol{q}_{i} \right\rangle$$

where

$$\left| \boldsymbol{q}_{i}(\rho_{0}) \right\rangle = -\nabla \int_{0}^{\infty} U_{1} \left(\left| \rho_{i} - \rho_{0} \right|^{2} + z^{2} \right)^{1/2} \omega(\rho_{i}) \, \mathrm{d} \, z \, \mathrm{d} \, \rho_{i}$$
⁽¹⁹⁾

Hence, the mean value of momentum transfer is defined by the atomic potential U_1 averaged along the nanotube axis and over the transverse thermal vibrations of carbon atoms. It is the basis of the existing channeling theory.

In the next approximation we should take into account the deviations Δq of q from the mean value $\langle q \rangle$ caused by atomic vibrations and the presence of electron gas. We add the mean-squared fluctuations of the corresponding vector q_i :

$$\left\langle \left(\Delta \boldsymbol{q}\right)^{2}\right\rangle = \sum_{i=1}^{N} \left(\left\langle \boldsymbol{q}_{i}^{2}\right\rangle - \left\langle \boldsymbol{q}_{i}\right\rangle^{2}\right)$$
 (20)

where $\langle q_i^2 \rangle$ is the mean-squared momentum transfer from a single atom. The mean deviation by the angle q/E corresponds to coherent scattering by the *N* atoms, whereas the fluctuating part corresponds to incoherent scattering. We can assume that the intensity of incoherent scattering on nuclei depends on the local density $n_a(\mathbf{r})$ of the nuclei. A similar assumption will be done for incoherent scattering on the electrons of the material with local density $n_e(\mathbf{r})$. On these assumptions the mean-squared momentum transfer during a longitudinal step Δz (containing many atomic distances, but such that $n_a(\mathbf{r})$ and $n_e(\mathbf{r})$ do not vary too much) may be found similarly to the case of multiple scattering in amorphous media. In terms of the multiple scattering angle $\theta_s = \Delta q/E$, we have then

$$\left\langle \theta_s^2(\boldsymbol{r}) \right\rangle = 16\pi e^4 \left(\Delta z / E^2 \right) [n_a(\boldsymbol{r}) Z^2 L_n + n_e(\boldsymbol{r}) L_e]$$
(21)

with Coulomb logarithms $L_n = \ln (191Z^{-1/3})$, $L_e = \ln(1194Z^{-2/3})$ for electron-nucleous and electron-electron scattering, respectively. Unlike in amorphous media,^[29] here $n_a(\mathbf{r})$ and $n_e(\mathbf{r})$ are the local number density of carbon nuclei and medium electrons as a function of the transverse coordinates \mathbf{r} .

These quantities, therefore $\langle (\Delta q)^2 \rangle$ also, vary strongly with r and are concentrated near the axis. From the equality $q = \langle q \rangle + \Delta q$ we may consider the transverse motion of a fast particle in a nanotube rope as a relatively smooth motion in the continuum potential on which stochastic perturbations are superimposed due to the incoherent scattering on atomic vibrations and electrons of the medium. Similar considerations can be performed in the case of axial channeling in fullerites^[24] where additional averaging over the fullerene angular coordinates should be included.

The stochastic character of the perturbations of the particle trajectories prevents us from formulating exact equations of motion. However we may apply the following calculation algorithm. Since the entrance points \mathbf{r}_0 of the particles into the channels are randomly distributed over the transverse plane, we can use the Monte-Carlo method to determine the initial transverse coordinate of the particle. For the numerical calculations it is more convenient to transfer from motion Eq.(17) to the equation system

$$d \mathbf{p}_{\perp} / dt = -\nabla U(\mathbf{r})$$
$$d \mathbf{r} / dt = \mathbf{p}_{\perp} / E$$
(22)

Then we integrate numerically the equations of motion (22) in the averaged potential of the nanotube rope $U(\mathbf{r})$ over time interval Δt and obtain the values of the transverse coordinate \mathbf{r}_1 and momentum $\mathbf{p}_{\perp 1}$ at time $t_1 = \Delta t$. The integration interval Δt_1 should be chosen so short that the nuclear number density $n_a(\mathbf{r})$ as well as the electron number density $n_e(\mathbf{r})$ are practically constant in this interval. At the same time the number of collisions should be large enough for the validity of the Gaussian distribution of $\Delta \theta$ (central limiting theorem of the probability theory). Then, using the Monte-Carlo method we generate a random value of the incoherent multiple scattering angle $\Delta \theta = \left\{ \Delta \theta_x, \Delta \theta_y \right\}$ due to incoherent scattering on path $\Delta z \approx \Delta t$ according to the probability distribution

$$P(\Delta\theta) = 2 \frac{\Delta\theta \exp(-(\Delta\theta)^2 / \langle \theta_s^2 \rangle)}{\langle \theta_s^2 \rangle}$$
(23)

being given above. The random value of $\Delta q = E \Delta \theta$ is then added to $p_{\perp 1}$, and the sum is considered as the actual value of the transverse momentum at time t_1 . The above procedure with the transverse momentum is repeated for the next time interval from t_1 to t_2 (= $t_1+\Delta t_2$) and so on.

This method of taking into account incoherent scattering (sometimes referred to as the method of aggregated collisions^[30]) is much faster than other ones, e.g. the method of kinetic equations^[25] or the binary collisions method,^[11] at least in the case of axial channeling, where the phase space for the transverse motion is four-dimensional and kinetic equations are too complicated, and in the case of high-energy particles where an extremely large number of binary collisions must be taken into consideration to reach the penetration depths comparable to the dechanneling length.

7 Computer simulation results

The method of aggregated collisions outlined above was applied in Ref.[28] to the computer simulations of the propagation of ultra-relativistic positively charged 150 GeV particles (e.g. positrons, protons or π^+ -mesons) through a (11, 9) nanotube rope made of a hexagonal 2D lattice of nanotubes. The simulated beam of particles consists of $N=10^3$ particles, has zero initial angular spread and enters into the nanotube rope at an angle $\theta_0 = \theta_L/2$ equal to the half of the Lindhard angle $\theta_L = 2.83 \times 10^{-5}$. The penetration depths of the particles into the rope are measured in units of $L_0 = R/\theta_L = 24 \ \mu m$ which represents the typical path of the particle in the rope between two subsequent encounters with nanotube wall. The angular distribution of the beam at various depths is illustrated in Figs.8~10 by series of scattering plots. The values of the penetration depth are shown in the upper-right corner of the corresponding plot, θ_x and θ_y are the angular coordinates of particles in the transverse (with respect to the nanotube axis) plane.

Three stages of the transformation of the angular distribution may be distinguished. At the first stage from z = 0 to $z \approx L_0$ the initial distribution quickly broadens due to the strong coherent scattering of the beam by the atomic rows. The influence of the incoherent scattering at this stage is relatively small. At

the second stage from $z \sim L_0$ to $z \sim 10L_0$ the angular distribution tends to the ring-like pattern, the radius of which corresponds to the angle of incidence (equal to the half of θ_L) and the thickness of the ring is determined by the thickness of the nanotube wall. The ring



Fig.8 Angular distribution of the beam at various depths (the first stage).



Fig.9 Angular distribution of the beam at various depths (the second stage).



Fig.10 Angular distribution of the beam at various depths (the third stage).

appears as a result of the multiple reflections of the beam on nanotube walls which lead to rotations of the transverse momentum of the particles at different azimuthal angles. At the third stage due to the increasing influence of the incoherent scattering the ring-like pattern broadens and at very large depth $z \approx 10^4 L_0$ it is hardly noticeable.

The variation of relative number $N_{\rm ch}/N$ of particles bound inside the nanotubes as a function of *z* is shown in Fig.11 for 150 GeV positive particles. At entrance about 60% of the beam is captured into the nanotubes, but after a few decimetres only a small part of them remains bound inside the channels. The dechanneling length in nanotube ropes estimated from this figure is close to 250 mm for 150 GeV positive particles, which is one order of magnitude higher than in ordinary crystals under similar conditions.



Fig.11 Relative number of particles remaining inside the nanotubes at different penetration depths into the nanotube rope.

The spatial distribution of the beam over the transverse coordinates strongly varies with the penetration depth as well. The transformation of the spatial distribution of the beam in the hexagonal two-dimensional lattice cell of the rope is illustrated in Fig.12. Initially at $z\approx 0$ the beam is uniformly distributed over the unit cell. A significant redistribution of the spatial distribution may be noticed already at $z\approx L_0$. It is noteworthy that, like in ordinary crystals, this effect may serve for the location of impurity atoms inside relatively short $(z \sim L_0)$ nanotubes, using secondary processes sensitive to the close collisions of the particles with the impurity atoms, i.e. backward Coulomb scattering, characteristic X-ray production or nuclear reactions.



Fig.12 Transformations of spatial distribution of the beam over the transverse coordinates at various depths.

The simulated angular distribution of the beam passing through (11, 9) nanotubes bent with a curvature radius R_b equal to Tsyganov critical radius $R_c=1.70$ m, at various penetration depths, is shown in Fig.13. The point $\theta_{0x} = \theta_{0y} = 0$ is the direction of the initial beam, supposed to be extremely collimated. It



Fig.13 Angular distribution of the beam passing through (11, 9) nanotubes bent with the Tsyganov curvature radius at various penetration depths.

can be seen that a noticeable part of the beam follows the curved channel and thus is deflected at an angle $\alpha = z/R_b$ which can be equal to several times the Lindhard angle. This part has an angular divergence close to the Lindhard angle. The remaining particles include non-deflected and partially deflected particles, the latter traveling preferably along the major superlattice planes.

No.2

The relative number of particles following the curved nanotubes is shown in Fig.14 as a function of the deflection angle $\alpha = z/R_b$. This figure clearly indicates that about 20% of the beam may be deflected at angles $\alpha \approx 10$ mrad which are two orders higher than the Lindhard angle. As pointed out above, the channeling of high-energy particles in curved nanotube ropes is of special interest due to the possibilities of beam deflection at comparatively large angles.



Fig.14 Relative number of particles following the curved nanotubes as a function of the deflection angle.

The method of aggregated collisions was also applied in Ref.[24] to the case of the propagation of high-energy particles along [100] axial channels in fullerites. The results for particles with different energies *E* are given below. For each value of total energy *E*, the trajectories of $N=10^3$ particles with random entrance positions were calculated. The incident beam was supposed without divergence and parallel to the fullerite crystallographic axis. The continuum potential governing the particle motion is shown in Fig.6. Fig.15 illustrates the decrease of the relative number N_{ch}/N of ultrarelativistic positively charged particles remaining in [100] axial channels as a function of the depth *z*. About 80% of the beam is initially captured into the channels both between and inside fullerene rows. Curves 1 to 3 correspond to particle energies E=150, 50 and 10 GeV respectively. The comparison of their slopes indicates that the dechanneling length is proportional to E. There is a slowing down of dechanneling at relatively large depths, apparently associated with the fact that at such depths channeling predominantly occurs between the fullerene rows ("square" channels of Fig.6), where the effect of incoherent scattering is weaker than inside the rows (circular channels of Fig.6). However, in average, the dechanneling length in fullerites occurs to be about one decimal order shorter than in nanotubes (at the same particle energies).



Fig.15 Dependence of the relative number N_{ch}/N of ultrarelativistic positively charged particles remaining in [100] axial channels of a fullerite at the depth *z*.

The comparison of channeling of positive and negative particles in fullerites is illustrated in Fig.16. Curve 1 shows the relative number of 10 GeV ultrarelativistic negatively charged particles remaining in [110] axial channels (Fig.7) as a function of the depth z while curve 2 shows the similar results for 10 GeV positively charged particles in [100] axial channels. About 80% of negative particle can be captured in [110] channels. The dechanneling length for negative particles is about 0.2 mm which is about two times less than for positive particles (in [110] channels).

The angular and spatial distributions of 10GeV positron beam at various depths in a fullerite is illustrated in Fig.17 by a series of scattering plots. Initially the beam has zero angular spread and zero entrance angle to [100] axial channels. The penetration depths z

of the particles into the fullerite (indicated by numbers in the upper side of the figures) are measured in units of $L_0 = R/\theta_L = 6.786 \ \mu\text{m}$, where R is the fullerene radius and θ_L is the Lindhard angle corresponding to the channeling in the deepest "square" well. The circle in all the left figures corresponds to the mentioned above Lindhard angle. Similarly to the case of a nanotube rope discussed above, at the first stage from z=0 to $z \approx L_0$ the initial distribution, which is represented by the point (0, 0) in the transverse momentum space, quickly broadens due to the strong coherent scattering of the beam by the molecular rows. The central almost axially symmetrical core of the angular distribution is probably formed by the coherent scattering of particles moving inside the more shallow ($U_0 \simeq 7 \text{ eV}$) circular channels while the broader star-like distribution is due to the scattering of particles moving between the fullerene rows in almost twice deeper $(U_0 \simeq 13.5 \text{ eV})$ "square" channels. At larger z the dechanneling brings some part of the beam outside the ring. It is interesting to note that at $z \simeq 4L_0$ and $z \simeq 8L_0$ the re-focusing of the beam is observed. This is due to the closeness of the potential of the circular channels to the parabolic one. Indeed, it is known that in a two-dimensional parabolic potential oscillation periods of the particles with zero angular momentum do not depend on their oscillation amplitudes, thus after one oscillation period the beam can become parallel again. However, with the increasing penetration depth the focusing effect becomes weaker both due to the inharmoniousness of the potential and the influence of the incoherent scattering giving non-zero angular momenta to the particles.



Fig.16 Dependence of the relative number N_{ch}/N of ultrarelativistic 10 GeV charged particles remaining in axial channels of a fullerite at the depth *z*. Curve 1 for electrons and [110] chan-

nels, curve 2 for positrons and [100] channels.



Fig.17 Angular (left of a pair) and spatial (right) distributions of the positron beam at different depths of channeling in a fullerite.

The spatial distribution of the beam, shown in the limits of the the square unit cell of the transverse lattice, at the entrance to a fullerite is homogeneous, but it rapidly transforms to extremely inhomogeneous already at $z \approx 0.6L_0$. Due to the approximate conservation of the phase-space volume of the beam one can see strong correlations between the angular and the spatial distributions.

Compared to ordinary crystals bent fullerites can effectively deflect both positive and negative particles. The angular distribution of 10 GeV positron beam (with negligibly small initial angular divergence) passing along [100] axial channels of a fullerite with thickness $z = 10L_0$ curved with radius R_b equal to Tsyganov critical radius R_c is shown in Fig.18 as the scattering plot.

The similar distribution for the case of 10 GeV

electrons and [110] channels is shown in Fig.19. It can be seen that a noticeable part of the both beams can be deflected at angles about five Lindhard angles.

No.2



Fig.18 Angular distribution of the positron beam passing through the [100] channels of the fullerite bent with the Tsy-ganov curvature radius at penetration depth $z = 10L_0$.



Fig.19 Similar to Fig.18, but for electrons and [110] channels.

8 Emission of radiation by charged particles passing through nanocrystals

Propagation of electrons and positrons through ordinary crystals as well as nanocrystals is accompanied by various types of electromagnetic radiation. As

$$\frac{\mathrm{d}^{2}\omega}{\mathrm{d}\omega\mathrm{d}\Omega} = \frac{e^{2}\omega^{3}}{8\pi} \sum_{f} \left[\left(\theta^{2} + E^{-2}\right)^{2} \left| \boldsymbol{n}_{\perp} \times \boldsymbol{r}_{if} \right|^{2} + \left(\theta^{2} - E^{-2}\right)^{2} \left| \boldsymbol{n}_{\perp} \cdot \boldsymbol{r}_{if} \right|^{2} \right] \cdot \delta \left[\frac{\omega}{2} \left(\theta^{2} + E^{-2} - \chi_{0}^{'}(\omega)\right) - \omega_{if} \right]$$
(24)

Here $(\omega, \theta, \varphi)$, with $\theta \ll 1$, represent the photon momentum in polar coordinates about the crystallographic axis; $d\Omega \simeq \theta d\theta d\varphi$ is the differential of the solid angle of emission; $\mathbf{n}_{\perp} = \{\cos\varphi, \sin\varphi\}$ is the unit vector collinear to the transverse momentum of the photon, $\varepsilon_n(\kappa)$ is the transverse energy of the particle with quasi-momentum κ in the *n*th band, noted above, they arise both from the non-uniform motion of the charged particles in the electrostatic potential of aligned atoms and from the transient polarization of the medium by the particles. Below we present general properties of these radiations and their particular features in the nanotube and fullerite cases.

8.1 Radiative transitions (channeling radiation) between the transverse energy levels of MeV electrons and positrons

Spontaneous radiative transitions between the transverse states of channeled particles give rise to the so-called channeling radiation (ChR). As mentioned above, at relatively low energies of positrons and electrons, there are transverse energy bands $\varepsilon_n(\kappa)$, which, in the limit of infinitely small transmittance of the potential barrier separating neighbouring nanotubes, degenerate into discrete levels. In this case the electromagnetic radiation can be treated as being due to dipole radiative transitions between the bands (levels). Due to the Doppler effect caused by the relativistic longitudinal motion of the particles the energy of an emitted photon is high compared to the transverse energy difference, but it occurs to be small compared to the energy of the particle ($\omega \ll E$) in the region of E under the consideration. Electromagnetic radiation at quantum channeling of electrons and positrons is well known phenomena in ordinary crystals. There are review articles, e.g. Ref.[25], describing various theoretical approaches, corresponding experimental results and the usefulness of ChR for the solid state physics as a new spectroscopic method.

The spectral-angular probability density of photon emission in unit time due to a spontaneous transition of a particle from band *i* into band *f* with lower transverse energy can be represented in the form^[31]

 $\omega_{if} = \varepsilon_n(\kappa) - \varepsilon_f(\kappa)$ is the transition frequency, and δ is the Dirac function. The matrix element of a dipole transition between the bands has the form

$$\boldsymbol{r}_{if} = \int_{S} \boldsymbol{\phi}_{f}^{*}(\boldsymbol{r}) \boldsymbol{r} \boldsymbol{\phi}_{i\kappa}(\boldsymbol{r}) \mathrm{d}^{2} r$$

where $\phi_{i\kappa}$ denotes the periodical part of the transverse wave-function, *S* is the area of the transverse lattice cell. The quasi-momentum of the particle is conserved (up to a reciprocal lattice vector) during the dipole transitions. Wavefunctions $\phi_{i\kappa}$ are normalized according to the relation

$$\int_{S} \phi_{f\kappa}^{*}(\boldsymbol{r}) \phi_{i\kappa}(\boldsymbol{r}) \mathrm{d}^{2} \boldsymbol{r} = \delta_{if}$$

where δ_{if} is the Kronecker symbol. Following Ref.[32], we have taken into account the complex Doppler effect through the volume-averaged real part of the dielectric susceptibility of the medium $\chi'_0(\omega) \ll 1$. Formula (24) corresponds (at each *f*) to a quantum dipole moving with a relativistic speed in the dielectric medium. In contrast to ordinary crystals, the polarization can indeed have a large effect on the spectrum of emitted frequencies via the complex Doppler effect determined by zeros of the Dirac function in Eq.(24).

The characteristic channeling radiation frequencies can be estimated as $\omega \sim \omega_{if} E^2$. If ω is substantially higher than the K-shell ionization threshold of carbon $(\hbar \omega_{\rm K} \approx 284 \text{ eV})$, we can use the "plasma" formula $\chi'_0(\omega) = -\omega_p^2 / \omega^2$ where $\omega_p^2 = 4\pi n_e r_e$ is called plasma frequency of the medium. In this case the relation between the radiation frequency and the polar angle of the radiation takes on the form

$$\theta^2 = \frac{2\omega_{if}}{\omega} - E^{-2} - \frac{\omega_p^2}{\omega^2}$$
(25)

In ordinary crystals the influence of the last term in Eq.(25) is negligibly small due to relatively high values of the energy level difference ω_{if} at any reasonable for channeling particle energy *E* (below ~1 MeV only ordinary electron diffraction exists). However, the situation can change in nanocrystals where ω_{if} is by the order of magnitude smaller than in ordinary crystals. In general case the allowed frequencies are determined by the condition that the right-hand side of Eq.(25) is positive, which gives the following limitations

$$\omega_{\min} \leq \omega \leq \omega_{\max}$$
(26)
$$\omega_{\min} = E^2 [\omega_{if} - (\omega_{if}^2 - \omega_p^2 E^{-2})^{1/2}],$$

$$\omega_{\max} = E^2 [\omega_{if} + (\omega_{if}^2 - \omega_p^2 E^{-2})^{1/2}]$$

Radiation with frequencies ω_{\min} or ω_{\max} is emitted at zero angle θ . It is important that the expression in the brackets of Eq.(26) must be positive, otherwise radiation becomes impossible at all, in other words, the particle energy E must be high enough to satisfy the condition

$$E\omega_{if} \ge \omega_p$$
 (27)

Note that $\omega_{\rm ff}$ depends on E via the parametric dependence of the transverse energy levels on E. It should be taken into account that, if ω_{\min} and ω_{\max} happen to be outside the validity of plasma formula for the real part of the dielectric susceptibility, the dependence of the latter on ω can be more complicated giving rise to a more complex Doppler effect. Moreover, below the K-edge the absorption of radiation inside nanocrystals may be of importance. For example, the analysis based on energy level scheme calculated in Ref.[10] show that 1 MeV electrons and positrons channeled in a nanotube rope can emit only in the ultraviolet range, where radiation is almost completely absorbed in the matter itself and it is hardly possible to observe the radiation. Electrons with higher energy (3 MeV) can emit several lines in the range of 290~311 eV at zero angle of observation and increasing again the electron energy (9 MeV), radiation at even higher frequencies (1.3~2.2 keV) becomes possible. As for the intensities of the lines, they depend, besides the dipole matrix elements, on the life-time and the populations of the levels. The initial populations depend on the entrance angle of the beam with respect to the channels while the redistribution between the levels occurs due to the incoherent scatterings of the beam on substance electrons, fluctuations of the potential and other reasons which are superimposed on the radiative transitions. More detailed information on this problem can be found in review articles [25] and references therein.

Expression (24) describes radiative transitions between the above-barrier transverse energy bands of low energy particles as well. If the transverse energy is high above the barrier, i.e. the entrance angle of the particles substantially exceeds the Lindhard critical angle, we can consider the continuum potential as a source of a small perturbation of the incident wave function (plane wave) of the particle and use the first Born approximation to calculate such a perturbation. This regime of the radiation mechanism is called coherent bremsstrahlung (CB). In the first Born approximation the radiation probability is proportional to the squared charge of the particle, therefore in contrast to ChR discussed above, there is no difference in the radiation spectra from electrons and positrons. Historically CB was predicted and investigated experimentally in ordinary crystals^[33] early before the channeling, however only with the discovering of the latter the unified approach to the problem of radiation of fast particles in crystals was developed enabling to define the limits for the old theory of CB based on the Born approximation. We discuss this theory below in conjunction with CB and ChR from ultra-relativistic electrons and positrons in nanocrystals.

8.2 Coherent bremsstrahlung and channeling radiation from ultra-relativistic electrons and positrons

The standard theory of CB and coherent electron-positron pair production by γ -photons in crystals is based on the Born approximation. This approach implicitly assumes that the change in the direction of the charged particle momentum between the successive collisions with atomic rows or planes (or the change of the e⁺e⁻angle) is small in comparison with the effective angle $\theta_{\rm eff} \sim E^{-1}$ for the emission of radiation by ultra-relativistic particles. On the other hand, it is well known^[34] that the character of the electromagnetic radiation from a charged particle in an external electromagnetic field substantially depends upon the relation between the angular deviation \mathcal{G}_{dev} of the particle from the initial direction of motion and the effective angle of radiation $\theta_{\rm eff}$. If the deviation is relatively small ($\mathcal{G}_{dev} \ll E^{-1}$), the radiation spectrum has a dipole character, i.e. it is determined by the Fourier time expansion of particle velocity. In this case the radiation is formed over a relatively long path $l_{coh}=2\pi/(\omega - kv)$ called the coherence length^[33] (or the formation zone) of radiation. For radiation in vacuum ($k = \omega$) and in the direction close of particle velocity we may estimate the coherence length as $l_{coh} \sim \lambda E^2$ which substantially exceeds the wavelength of radiation λ . The CB arises if the coherence length coincides with the path between the successive collisions of the particle with atomic rows or planes. For ultrarelativistic particles the CB condition can be satisfied if they move at small angle to the crystallographic plane or axis, however one should keep in mind that in this case the angular deviation \mathcal{G}_{dev} of the particle will increase due to the channeling effect.

In the opposite case of large deviation angles $(\mathcal{G}_{dev} \gg E^{-1})$ the analysis of the problem can be simplified significantly. Indeed, since during this relatively short time interval the electric field of the crystal can be considered as constant in space, the trajectory details are not important for the radiation spectrum and it occurs to be similar to the synchrotron radiation spectrum, i.e. it is totally determined by the local value of particle acceleration, but since the latter varies along a trajectory, the synchrotron-like spectrum must be averaged over time along particle trajectories. Between these limiting cases there is an intermediate region $\mathcal{G}_{dev} \sim E^{-1}$ where the details of the particle trajectory are of importance and it is the most difficult case for the theoretical analysis of the radiation spectrum.

Let us consider the most simple case of channeled particles with relatively low ($E \leq E_{cr}^{(1)}$) energies (but high enough to use classical mechanics). In the frame of the dipole approximation the ChR spectrum from particles moving inside the chiral nanotubes was calculated.^[35] The energy $I(\omega)$ radiated by a single channeled particle per unit path in a nanotube and per unit frequency interval can be written as

$$I(\omega) = (e^{2}\omega/2)\sum_{n=0}^{\infty} \{\omega_{n}^{(+)2} \left| \rho_{n}^{(+)} \right|^{2} f[\omega/(2\gamma^{2}\omega_{n}^{(+)})] + \omega_{n}^{(-)2} \left| \rho_{n}^{(-)} \right|^{2} f[\omega/2\gamma^{2}\omega_{n}^{(-)}]\},$$
(28)

where $\omega_n^{\pm} = n\omega_{\rho} \pm \Omega$, $f(\xi) = (1-2\xi+2\xi^2) \eta(\xi)$,

$$\rho_n^{(\pm)} = \left[2/2\pi n \pm \Delta \varphi\right] \operatorname{Im} \int_{0}^{T_{\rho}/2} \exp\left[-i(n\omega_{\rho} \pm \Omega)t\right] (d/dt) \{\rho(t) \exp\left[\pm i\varphi(t)\right]\} dt$$

originate from the Fourier expansion of the linear combinations $v_x \pm i v_y$ of the Cartesian components of the transverse velocity of the particle.^[36]

The radiation spectra from channeled positrons and electrons are shown in Fig.20 in the case of a beam with zero angular divergence entering the (11, 9)

 η is the Heaviside step-function and

nanotubes at half of the Lindhard angle. The target thickness is considered to be small compared to the dechanneling length and the spectrum (28) was averaged over the random values of the initial coordinates of the particles. To emphasize the scaling of the dipole spectrum we introduced the characteristic frequency $\omega_c = 4E^2 \theta_L / d \sim E^{3/2}$ and the spectral density $I_c = Pe^2 \omega_c \theta_L^2 / 2 \sim E^{1/2}$, where *P* is the probability of capturing particle into the potential well.



Fig.20 The radiation spectra from positrons (e^+) and electrons (e^-) channeled in (11, 9) chiral nanotubes.

Compared to positrons, channeled electrons along with the peak near $\omega \approx \omega_c$ associated with the nutations of the orbits back and forth the fullerene spheres, produce more intense and hard radiation at $\omega \approx 8\omega_c$ due to the relatively fast radial oscillations. The calculated spectra does not take into account the input from positrons moving between the nanotubes as well as from above-barrier particles, however it shows a substantial difference in the shape and intensity of ChR spectra from electrons and positrons, in other words, the spectra are strongly charge-dependent.

Coherent bremsstrahlung (CB) in nanocrystals has a number of peculiarities. Electrons or positrons are scattered by a nanotube as a whole and since nanotubes in a rope make a two-dimensional superlattice, they can act coherently in the formation of CB, similarly to the atomic rows in ordinary crystals. It should be noted that if the entrance angle is above but still comparable to the Lindhard angle $(\theta_1 \le \theta_0 \le 2\theta_1)$, then particles may deflect significantly in the transverse plane destroying the coherence, similarly to the case of the ordinary crystals.^[35] The significant coherence between nanotubes (or atomic rows in ordinary crystals) may exist only at higher entrance angles $\theta_0 \ge 2\theta_L$ where standard theory of CB is applicable. As far as the CB in fullerites are concerned, it is quite similar to the case of ordinary crystals with the exception that instead of collisions of charged particles with atoms there are collisions with C₆₀ fullerenes.

According to the standard theory of CB in ordinary crystals, the emission probability per unit path of electron (positron) moving in a crystal at small angle to atomic rows or planes, differential with respect to the photon energy ω , can be related to the Fourier component of the continuum potential of the atomic row V_H

$$\frac{1}{1\omega} = \frac{e^2}{E^2} \sum_{H} \frac{H^2}{(H\nu)^2} |V_H|^2 \left[1 + \frac{u}{2(1+u)} - 2\Omega_H + 2\Omega_H^2\right] \eta (1 - \Omega_H).$$
(29)

Here we use the following notations: \mathbf{v} is the unit vector along the direction of the initial momentum of the charged particle, $u = \omega/(E - \omega)$, $\Omega_H = u/(2E \cdot H \mathbf{v})$, η is the Heaviside step function. The summation in Eq.(29) is performed over all reciprocal lattice vectors $H = \{H_x, H_y\}$ orthogonal to the chosen family of the rows (directed along z). In its turn, in monatomic crystal V_H is connected with the Fourier expansion of the atomic potential $f(\mathbf{k})$ (Eq.(9)) by the simple relation

$$V_{\boldsymbol{H}} = \mathcal{D}^{-1} \exp(-H^2 u^2) f(H_x, H_y, 0)$$

where \mathcal{D} is the unit cell volume of the crystal and u^2 is the squared amplitude of the thermal vibrations. If atomic rows are chaotically spaced, or the direction of ν is far from the direction of closely packed atomic planes, we may substitute the summation in Eq.(29) by an integration according to the rule

$$\sum_{\boldsymbol{H}} \Rightarrow \frac{S \cdot d^2 \boldsymbol{H}}{\left(2\pi\right)^2}$$

In order to calculate CB from nanotube superlattices we must generalize Eq.(29) according to Ref.[10]. Since different nanotubes in a rope have different azimuthal orientations we may first average the nanotube potential (Eq.(8)) over azimuthal positions of the nanotube rows (cf. Eqs.(6) and (7)). Then considering the continuum potential of a nanotube as azimuthally symmetric, the generalization reduces to the substitution of the Fourier expansion of the string potential V_H by the corresponding expansion of the continuum potential of the nanotubes over the reciprocal superlattice vectors H. The difference between the exact potential (8) and the averaged over azimuthal angles gives rise to a semi-coherent radiation (incoherent between nanotubes, but coherent between atoms of the same row), due to scatterings on separate atomic rows. As shown in Fig.4, this semi-coherent radiation may be significant for zigzag or armchair nanotubes, but it should be relatively small for chiral nanotubes due to the relatively weak dependence of the continuum potential on the azimuthal angle. Similarly, in a fullerite CB is described by Eq.(29) where V_H must be replaced by the corresponding Fourier expansion of the fullerene potential (Eq.(11)).

No.2

The distribution of the coherent bremsstrahlung energy per unit photon energy $\hbar \omega$ and per unit path $I(\omega) = \omega d^2 \omega / d(\hbar w) dz$ is shown in Fig.21 for 1 GeV electron (positron) entering at $\theta_0 = 2 \theta_L^{(s)}$, where $\theta_L^{(s)}$ is the Lindhard angle for the channeling, to the rows of nanotubes laying in (1, 0) planes of the rope. The dashed line represents the bremsstrahlung in case of randomized positions of carbon atoms (amorphous medium).

The similar CB spectrum from 1 GeV electron (positron) entering at $\theta_0 = 5 \theta_L^{(s)}$ to [111] fullerite axis is plotted in Fig.22. Compared to the case of ordinary crystals the maximum of the first radiation harmonic is at much lower photon energy. Besides, all harmonics have a fine structure associated with the hollow structure of nanotubes and fullerenes.



Fig.21 The frequency distribution of the coherent bremsstrahlung power from 1 GeV electron (positron) entering at $\theta_0=2\theta_L^{(s)}$ to the rows of nanotubes laying in (1, 0) planes of the rope.



Fig.22 The frequency distribution of the coherent bremsstrahlung power from 1 GeV electron (positron) entering at $\theta = 5 \theta_L^{(s)}$ to [111] fullerite axis.

8.3 Radiation due to the Bragg diffraction of the own electromagnetic field of fast charged particles

At last let us discuss the X-ray radiation from relativistic charged particle arising from the Bragg diffraction of the own electromagnetic field of a particle in crystals or MLS, which is called parametric X-ray radiation (PXR). It was predicted by Ter-Mikhaelyan^[33] who developed the theory based on the kinematical diffraction of virtual photons in infinite nonabsorptive media. The influence of absorption and extinction of X-rays inside crystals was taken into account later^[37] using the two-wave dynamic diffraction method. PXR in ordinary crystals was discovered and investigated experimentally.^[38~40] The modern state of the theory and experiments in this field was presented recently in review article^[41] where a comprehensive list of works devoted to PXR can be found. It should be noted that PXR is often referred to as quasi-Cherenkov, resonance transition or even diffraction X-ray radiation. The peculiarities of PXR from nanocrystal were investigated quite recentlv.^[42,43]

General approach to the problem is based on the multi-wave diffraction theory.^[37,42] We expand the electric field in a superlattice in series of Bloch waves and from Maxwell equations find the following infinite system of algebraic equations for the expansion coefficients $E_{K}(k, \omega)$

$$(\mathbf{k} + \mathbf{K}) \times (\mathbf{k} + \mathbf{K}) \times \mathbf{E}_{\mathbf{K}}(\mathbf{r}, \omega) + \omega^{2} \mathbf{E}_{\mathbf{K}}(\mathbf{r}, \omega) + \omega^{2} \sum_{\mathbf{H}} \chi_{\mathbf{H}}(\omega) \mathbf{E}_{\mathbf{K}-\mathbf{H}}(\mathbf{k}, \omega) = 8\pi^{2} i e \omega \mathbf{v} \delta(w - \mathbf{k} \cdot \mathbf{v}) \delta_{\mathbf{K}0}$$
(30)

In this equation

$$\chi_{H}(\omega) = \frac{1}{V} \int_{V} \chi(\mathbf{r}, \omega) e^{iH\mathbf{r}} d^{3} \mu$$

are Fourier coefficients of the periodic complex dielectric susceptibility $\chi(\mathbf{r}, \omega)$ of a crystal, \mathbf{H} and \mathbf{K} are the reciprocal lattice vectors and V is the volume of the crystal unit cell. It differs from usual equations describing X-ray diffraction only by the term in the right-hand side which represents the electric current of the uniformly moving electron. In most practical cases only two Bloch waves are of importance and the infinite system of Eq.(30) turns to the following two pairs of equations corresponding to the different (σ or π) polarization of the electromagnetic field

$$[\omega^{2}(1+\chi_{0})-k^{2}]E_{0}+C_{s}\omega^{2}\chi_{H}E_{H}=8\pi^{2}ie\omega\theta P_{s}\delta(\omega-k\cdot\nu)$$

$$C_{s}\omega^{2}\chi_{H}^{*}E_{0}+[\omega^{2}(1+\chi_{0})-(k+H)^{2}]E_{H}=0$$
(31)

The π and σ polarizations are respectively parallel and perpendicular to the incidence plane of the particle, $P_s = \cos\phi$, $C_s = \cos\psi$ for $s = \pi$ and $P_s = \sin\phi$, $C_s = 1$ for $s = \sigma$. The exact solution of Eq.(31) corresponds to PXR of the two-wave dynamical diffraction theory. More simple kinematical theory assumes $|E_H|$ to be small compared to $|E_0|$. As shown in Ref.[43] for nanocrystals the kinematical theory is quite sufficient. It is also reasonable to consider the case of relatively thick $(T \gg l_a)$ medium where the output of PXR is maximum. In this case the solution of Eq.(31), using the relative smallness of the diffracted field $|E_{H}|$, leads to the following spectral-angular distribution of PXR photons both for the symmetrical Bragg case and Laue (radiation through the backward crystal interface) case:*

$$\frac{\mathrm{d}^2 N_s}{\mathrm{d}\,\omega\,\mathrm{d}\,\Omega} = \frac{e^2\theta^2}{4\pi\chi_0^{"}(\omega_B)\sin^2\psi} \cdot \frac{\left|P_s C_s\chi_H^{'}(\omega)\right|^2}{\xi^2(\theta)}\Lambda(\omega,\theta)$$
(32)

Here $(\pi/2+\psi)$ is the angle between the particle velocity v and the reciprocal vector H; $\omega_{\rm B} = H/(2 \sin \psi)$ (it is supposed that is much higher than the "Fresnel angle" $\sim |X'_0(\omega)|^{1/2}$ to the reflecting crystal planes); the polar angle θ of radiation is counted from the specular direction of the particle velocity and the azimuth angle ϕ from the incidence plane. The line shape of the spectrum (32) is determined by the normalized Lorentz curve

$$\Lambda(\omega,\theta) = \frac{\Gamma}{2\pi} \cdot \frac{1}{\left[\omega - \omega_B (1 - \theta \cos \phi \cot \psi)\right]^2 + (\Gamma/2)^2}$$
(33)

The line width Γ of PXR radiated at sufficiently small solid angle interval around θ , ϕ is determined by the imaginary part of the volume-averaged dielectric susceptibility $\chi_0''(\omega)$:

$$\frac{\Gamma}{2} = \frac{\omega_B}{4\sin^2\psi} \chi_0''(\omega)$$

Integrating (32) over ω , azimuth angles ϕ , and polar angles from zero to some $\theta_m \sim [E^{-2} - \chi'_0(\omega)]^{1/2}$, corresponding to the detector aperture (of the order of the virtual photon angular spread), we find the total number of PXR photons N_s per electron

$$N_{s} = \frac{e^{2}\omega_{B}^{2}}{2H^{2}} \frac{\left|C_{s}\chi_{H}'(\omega_{B})\right|^{2}}{\chi_{0}''(\omega_{B})} \left[\ln\frac{\xi_{m}}{\xi_{0}} - \frac{\theta_{m}^{2}}{\xi_{m}}\right]$$
(34)

where $\xi_m = \xi(\theta_m), \xi_0 = \xi(0).$

Assuming that the modulation depth of the crystal electron density does not depend substantially on the modulation period the number of photons with energies $\approx \omega_B$ is proportional to the squared interplanar space $d_p^2 \sim H^{-2}$. According to Eq.(33), the maximum shift $\Delta \omega_{\text{max}}$ of the photon energy ω from ω_B corresponding to $\phi = \pm \pi$ depends on the detector acceptance angle θ_m and the incidence angle of electrons ψ through the relation

$$\frac{\Delta\omega_{\max}}{\omega_{B}} = \theta_{m} \cot \psi \tag{35}$$

This relation determines the width $\Delta \omega$ of PXR spectrum captured by a detector with relatively large (compared to the rocking curve width) acceptance

^{*} In Ref.[43] the PXR photon number was underestimated by factor 2.

angle.

No.2

The total number of PXR photons (Eq.(34)) with frequencies around ω_B radiated by one electron with Lorentz factor $\gamma = 10^3$ into the solid angle $0 \le \theta \le 2[E^{-2} - \chi'_0(\omega)]^{1/2}$ and $0 \le \phi \le 2\pi$ from nanocrystals as a function of the peak radiation frequency ω_B (determined by the incidence angle ψ to the crystal planes) is plotted in Fig.23, where (a) refers to the perpendicular polarization and (b) to the parallel polarization of photons.^[43]

For comparison there are also the corresponding results obtained for the carbon-tungsten multi-layered structure (MLS) and some ordinary crystals. Curves N correspond to the reflection from (1, 0) planes of (10, 10)10) nanotube ropes, curves F to (111) planes of fullerite, and curves C-W to the carbon-tungsten multilayered structure with period $d=16\text{\AA}$ and equal thickness of the adjacent layers. Curves D and Ge correspond to the reflection from (220) planes of diamond and germanium crystal at room temperature, respectively (in diamond-like crystals (110) reflection is suppressed due to the structure factor). Solid curves represent the results based on the dynamic diffraction theory^[43] and broken ones on the kinematical theory (Eq.(34)). For the nanocrystals and diamond the results of the dynamic and kinematical theory coincide with the accuracy corresponding to the curve thickness. Some difference may be noticed in case of C-WMLS and for germanium (and probably other high Zcrystals) the difference is substantial. The dips on all parallel polarization curves at some photon energies show that at such energies (incidence angles close to the Brewster angle $\approx 45^{\circ}$ of X-rays) PXR is linearly polarized perpendicular to the incidence plane.

Inside the photon energy interval from 1 keV to 30 keV both nanocrystals and MLS are more efficient, by two orders of magnitude in photon yield, than ordinary crystals. At photon energies higher than 10 keV the nanotube ropes dominate over fullerite and carbon-tungsten MLS. In the vicinity of 3 keV photon energy the total number of photons (in the above-defined solid angle) per electron from a nanotube superlattice is about 10⁻⁵, which is comparable to that from the carbon-tungsten MLS, while ordinary crystals like diamond or germanium are unable to emit PXR at such low frequencies at all.



Fig.23 The total number of polarized quasi-monochromatic PXR photons with a frequency close to the Bragg frequency, radiated by electron with Lorentz factor $\gamma = 10^3$ into the effective solid angle from various periodic media. (a) Perpendicular polarization, (b) Parallel polarization.



Fig.24 The relative width of PXR photon spectra from various media as a function of the radiation frequency.

The relative widths of the photon spectra from various media calculated using Eq.(32) are illustrated in Fig.24. One can see that PXR from ordinary crystals have much narrower photon energy distributions than from superlattices. However, it should be noted that for some applications, such as X-ray lithography, the whole number of photons is more important than high monochromaticity. Thus PXR from nanotube ropes and fullerene crystals represent relatively bright,

quasi-monochromatic and frequency-tunable (through the choice of electron incidence angle ψ) source of X-rays for those applications which do not demand very high radiation monochromaticity. It should be noted that PXR-based source can operate at much lower energies *E* of the emitting electrons than synchrotron radiation at compatible radiation frequencies.

9 Conclusions

Fullerite single crystals and aligned nanotubes can find various applications in such fields as: production of X-rays with high-energy particle beams, Bragg mirrors for X-rays down to nanometre wavelength which are probably more resistant to extreme irradiation and heating than other soft X-ray mirrors, steering of charged and neutral particle beams, etc.

Our theoretical calculations and computer simulations clearly indicate that nanocrystals are comparable to or even go beyond the regular targets (crystals) for the production of intense quasi-monochromatic X-ray photons of parametric X-ray radiation (PXR), coherent bremsstrahlung (CB), channelling radiation (ChR), and other types of emission. The intensity of PXR from nanocrystals can be up to three orders higher than in ordinary crystals like diamond; moreover, it becomes possible to extend the PXR spectrum to the soft X-ray region where ordinary crystals cannot emit this type of radiation at all. Using channelling of high-energy particles in bent nanotubes or fullerites it is possible to deflect and focus both positively and negatively charged particles more effectively than in ordinary single crystals.

Other interesting phenomena like X-ray and thermal neutron channeling in multi-wall nanotubes, Bragg diffraction of soft X-rays, channeling in spirally bent nanotubes and channeling radiation at very high particle energies stayed beyond the scope of the present report. The comprehensive review of all mentioned phenomena will be published soon elsewhere.

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