CHANNELING EFFECT AND IMPROVEMENT OF THE EFFICIENCY OF CHARGED PARTICLE REGISTRATION WITH CRYSTAL SCINTILLATORS

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Energy of ions (recoil nuclei) channeled along definite directions in crystals is transferred to the lattice electrons mainly. In NaI(Tl)-type scintillators, this leads to increasing the light yield from $\sim 10\%$ to $\sim 100\%$ when compared with the yield for electrons of the same energy. Taking into account this effect at processing data of DAMA/NaI experiments in Gran Sasso, which had demonstrated the year modulation of number of signals in a range of 2-6 keV of electron equivalent, reveals that DAMA/NaI results could be caused by $\sim 6 \times 10^{-7} \text{ cm}^{-2} \text{ s}^{-1}$ flux of daemons (Dark Electric Matter Objects – presumably Planckian relic particles) falling out from strongly elongated heliocentric orbits with velocities of 30-50 km/s. The flux value and the 2-6 keV signal intensity agree rather well with values emerging from our former estimates and interpretations of ground-level and underground measurements.

Keywords: DM scintillation detection; recoil nucleus channeling; scintillation light yield

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1. Introduction

Experiments aimed at the detection of dark matter (DM) objects and the determination of their properties by investigating the recoil ions (nuclei) with scintillation detectors rest essentially on the knowledge of the efficiency parameter $q$ (quenching factor), the light yield ratio for the recoil ion and the electron of the same energy.\textsuperscript{1} Our interest in this parameter stems from the possibility that the year modulation in the number of signals revealed with a high confidence in the DAMA/NaI(Tl) and DAMA/LIBRA search for WIMPs (Weakly Interacting Massive Particles, hypothetical neutral particles $\sim 10^2$ GeV in mass considered to be candidates for DM)\textsuperscript{2,3} can be assigned to registration of a flux of electrically negative daemons (Dark Electric Matter Objects, presumably the elementary Planckian black holes, $\sim 10^{19}$ GeV in mass and with an electric charge $Ze \approx 10e$) captured out of the galactic disk by combined action of the moving Sun and of the Earth into strongly elongated, Earth-crossing heliocentric orbits (SEECHOs). These orbits crowd in the antapex zone beyond the Sun, which is crossed by the Earth sometime in early June. It is essential that for $q \approx 1$ the amplitude range of the significant 2-6-keV signals in the DAMA/NaI(Tl) and DAMA/LIBRA experiments coincides with the energy of the...
iodine recoil ions if the latter are knocked out elastically by daemons falling on the Earth from SEECHOs with velocities of 30-50 km/s.

The parameter $q$ for the I (and Na) ions is determined traditionally in neutron scattering experiments. These are either direct measurements of the amplitude of the scintillations correlating with monoenergetic neutrons scattered to a preset angle or statistical treatment (by Monte-Carlo simulation) of the scintillation spectrum of a NaI(Tl) crystal irradiated by neutrons with an average energy of 2.35 MeV from a $^{252}$Cf source. In the first case, $q_{Na} = 25-30\%$ for Na ions with a recoil energy $E_r > 4$ keV and $q_I = 8-8.6\%$ for $E_r > 10$ keV. In the second case, $q_{Na} = 30-40\%$ for $E_r > 5$ keV and $q_I = 5-9\%$ for $E_r > 22$ keV (see Table 1). We see that the data obtained by different groups fit fairly well.

Such calibrations of NaI(Tl) scintillators are usually assumed to mean that $q$ does not depend on the energy of the recoil ion (i.e., $\partial q / \partial E_r = 0$, see Refs. 7,12). Actually, while this conclusion does not follow from anything else, it is buttressed by the observation that the light energy yield of an inorganic scintillator (for NaI(Tl) it amounts to about 15%\(^1\)) depends only weakly on electron energy, varying by less than 20% from 5 to 1000 keV,\(^1,13,14\) just as the light energy yield for heavy particles with $E > 1$ MeV. On the other hand, as Birks\(^1\) pointed out long ago, $q > 1$ in heavy metal iodides for protons and deuterons with $E \approx 5$ MeV. Davis et al.\(^15\) (see also Ref. 8) observed $q$ to grow for Ca and F ions with decreasing $E_r$ in the CaF\(_2\)(Eu) crystal. On the contrary, in liquid Xe the quenching relative to gamma rays was found by Aprile et al.\(^16\) to grow almost two-fold with increasing $E_r$ from 10.4 to 56.5 keV. So one cannot rule out the possibility that the conclusion of the constancy of $q$ in the particular case of NaI(Tl) is simply a consequence of the difficulties encountered in measurements in the keV range (even for electrons, measurements on NaI(Tl) were performed only down to 0.87 keV\(^17\)).

As we are going to show, there are grounds to believe that the value of $q$ for a keV-range iodine recoil ions in the NaI(Tl) crystalline scintillator may approach 1, and that under certain conditions the efficiency of scintillation excitation by ions may even exceed somewhat that by electrons. From this follows the conclusion that DAMA/NaI results are quantitatively explicable within the framework of the daemon paradigm.

### 2. On Interaction of keV-range Ions with a Solid Body

#### 2.1. Amorphous solids

When an ion enters a solid, it passes without stopping a certain path. This path length is determined by interaction with nuclei (ions) and the electronic component, with the major contribution at low energies coming from slowing down by nuclei, and at high energies, by electrons. The stopping power due to electrons is proportional to the ion velocity.

The first self-consistent theory of ion slowing down in monoatomic amorphous materials (including gases, but disregarding the ionization and dissociation of
Table 1. Comparison of the experimentally determined quenching factors $q$ for Na and I recoil ions in NaI(Tl) scintillator ($q_{\text{exp}}$ and $q_{\text{exp}+MC}$ -- the both determined with making use the neutron scattering technique, but the $q_{\text{exp}+MC}$ is a result of statistical treatment; see text) with the SRIM-code calculated values of $q$ (and $\lambda_a$ -- the mean path length to rest) in amorphous NaI.

| $E_r$, keV | 2   | 4   | 10  | 20  | 50  |
|------------|-----|-----|-----|-----|-----|
| Na         |     |     |     |     |     |
| $q_{\text{exp}}$ | 0.25-0.30 (for $E_r > 4$ keV) |     |     |     |     |
| $q_{\text{exp}+MC}$ | 0.3-0.4 (for $E_r > 5$ keV) |     |     |     |     |
| $\lambda_a$, Å | 71  | 115 | 233 | 421 | 985 |
| I          |     |     |     |     |     |
| $q_{\text{exp}}$ | 0.05-0.086 (for $E_r > 10$ keV) |     |     |     |     |
| $q_{\text{exp}+MC}$ | 0.05-0.09 (for $E_r > 22$ keV) |     |     |     |     |
| $\lambda_a$, Å | 50  | 70  | 114 | 170 | 307 |

molecules) was developed by Lindhard, Scharff, and Schiott (LSS). They divided the stopping power into two components associated, accordingly, with nuclei and electrons. Then the scintillator efficiency $q$ is determined simply as the ratio of the energy imparted to the electronic component to the total kinetic energy of the ion (i.e., to the sum of the nuclear and electronic stopping powers). It is this theory that is frequently compared with measurements associated with the motion of recoil ions in crystalline scintillators. The theoretical LSS values of $q$ were shown to exceed its experimental values by approximately a factor 2-2.5.

Firsov took into account the ionization resulting from overlap of the electronic shells of the moving and the target atoms, an approach that provides in some cases a better agreement with experiment than LSS theory. Further progress in the field of theory involved primarily a closer analysis of the effective charge of the projectile ion through a consecutive refinement of the quantum-mechanical description of ion interaction with the target atoms, a point of particular importance below the Bohr velocity. Ziegler outlined the history and present status of the problem involving the motion of ions with $E \approx 1$ keV-1000 MeV in amorphous solids, and developed a computer code SRIM, which permits calculation of the ion range and scattering, as well as the ion and electron stopping powers for any combination of substances, including nonmonatomic targets. Compounds are analyzed using the Bragg rule that states that the stopping power of an ion in a compound can be approximated by a combination of stopping powers of the constituent target ions. This rule was shown to hold the better, the higher are the atomic numbers of the elements. SRIM calculations suggest that the mean path length to rest in amorphous NaI of a 2-keV iodine ion projected on the initial direction of motion is $\lambda_a = 50$ Å, a comparison of the ion with electron stopping powers yields $q = 0.068$, and further on (see Table 1). We readily see that the values of $q$ following from the nowaday theory agree fairly well with experiment (see Sec. 1 above and Refs. 6-11).
2.2. The channeling effect in crystals

Motion of an ion in crystals, in particular, in inorganic scintillators, differs substantially in many respects from that in amorphous solids. The ordered arrangement of nuclei in crystals makes possible the so-called channeling of ions propagating along certain directions (crystallographic axes and planes). Channeling becomes manifest in an anomalously deep penetration of ions into a target, an effect discovered half a century ago by Bredov and Okuneva. They observed penetration of 4-keV $^{134}\text{Cs}^+$ ions into a Ge crystal to a depth $\lambda_c \sim 10^3 \text{Å}$ (to feel the difference, a 4-keV Cs$^+$ ion would penetrate into amorphous Ge, according to SRIM calculations, only to a depth $\lambda_a = 44 \text{Å}$ for the ion to electron stopping power ratio in an amorphous solid $\kappa = 32$). The explanation for such a deep penetration, as was found 6 years after the discovery of Bredov and Okuneva, lies in the ion stopping power decreasing strongly along some directions in a crystal (see review Ref. 22). Within a channel, stopping is dominated by electrons. This is seen already from the above comparison of the data of Bredov and Okuneva with calculations made for amorphous Ge (whence it follows, in particular, that the mean path length of an ion in the channel, $\lambda_c$, exceeds that in an amorphous solid by a factor $\sim (\kappa + 1)$, i.e., $\lambda_c \approx (\kappa + 1)\lambda_a = \lambda_a/q$).

Measurements of the mean range of different ions, say, in W crystal likewise produce remarkable results. For instance, in the case of Xe$^+$ non-channeled ions, the ion stopping power, which originally is higher than that of the electronic one, decreases to become equal to the electronic one (which remains proportional all the time to the ion velocity) only at 2.7-MeV ion energy, whereas the ion stopping power observed under channeling conditions becomes equal to the electronic one already at 4 keV.\textsuperscript{21}

The energy of a channeled ion is transferred primarily to electrons, both single and to their continuum. The electrons transform eventually a part of this energy into light, as if the scintillator was irradiated by electrons from the outset. So it is obvious that even this one point leads to $q \rightarrow 1$ for the channeled ion case.

This could be a convenient time to stop discussion of the channeling effect and of its contribution to high-efficiency detection of low-energy recoil ions, were it not for some implications which we shall consider below.

2.3. An influence of making and breaking of quasi-molecular bonds of channeled ions with ions in the channel walls

Another process capable of improving the efficiency of scintillation radiation generated by the channeled recoil ions is the consecutive formation by them of quasi-molecular bonds with channel wall atoms resulting from the overlap of their electronic shells (while the corresponding theory is still lacking, recalling the concepts of Firsov\textsuperscript{20} would be here appropriate). The energy of dissociation of a single NaI ionic molecule is 3.16 eV, and the electron affinity of the iodine atom is 3.06 eV, which suggests that the particles knocked out from the NaI lattice are negative I$^-$.
recoil ions (recall that the iodine ionization energy is 10.45 eV, and the average energy required to remove an ion from the crystal lattice is 20-30 eV). Therefore, it appears reasonable to assume that consecutive ruptures of these bonds initiated by continuous motion of the ion would ideally release in the form of photons an energy of \( \sim 2.5-3 \) eV per each 2 Å (the distance between like ions along the channel in the NaI lattice). If there were no other channels and mechanisms of energy loss, a 4-keV ion would expend its energy in a distance of \( \sim 3000 \) Å. The existence of other ways of energy loss in interaction with the electronic component reduces naturally this path to one third, so that this mechanism would boost the efficiency of transformation of the ion’s kinetic energy to light by 30%, to result in \( q > 1 \). We thus see that the above process, even if it were realized only partially, would give rise to a marked increase in the efficiency of ion energy transformation to the scintillation radiation of the crystal.

2.4. Dechanneling due to collisions with Tl\(^+\) luminescent centers in NaI(Tl)

Massive NaI crystals used as scintillators are not perfect, if only because the conditions of their growth are not ideal. Their lattice contains inevitably various defects in the form of dislocations and so on. As it is well known,\(^{21,22,25}\) presence of defects and thermal vibrations in lattice are among the main causes responsible for the ion dechanneling. The main source of defects in the NaI(Tl) lattice are thallium atoms intentionally doped into the crystal to create luminescent Tl\(^+\) centers, which increase substantially the light yield of the phosphor.\(^1\) The optimum molar concentration of Tl in NaI(Tl) is 0.0013.\(^1\) Assuming the radius of Tl\(^+\) ions to be \( \sim 1.5 \) Å, and that of I\(^-\) ions, \( \sim 2.2 \) Å, we obtain \( \sim 1200 \) Å for the mean path length of an I\(^-\) ion between consecutive collisions with the ions of the Tl\(^+\) centers. This length corresponds to \( \lambda_\text{n} = 86 \) Å for the iodine ion with \( E_r = 6 \) keV, with \( q = 0.072 \), which yields exactly \( \lambda_c = \lambda_\text{n}/q = 1200 \) Å. Thus, starting already from an energy \( \sim 6-10 \) keV the iodine recoil ions will undergo dechanneling in NaI(Tl) because of their interaction with the heavy (\( A = 204.4, Z_n = 81 \)) Tl\(^+\) ions mainly (note that ion dechanneling is frequently used to probe the position of foreign atoms in crystals\(^{25}\)). It is conceivable that dechanneling of this kind in NaI(Tl) in collisions with Tl\(^+\) luminescent centers is accompanied by an excess scintillation light yield (i.e., again by an increase of \( q \) above 1). The dechanneling of ions due to their scattering on other type defects has a lower probability owing to their significantly lower concentration.

3. Probability of the Recoil Ion Channeling and the Near-Earth Daemon Flux as Observed by DAMA/NaI

The recoil ion (nucleus) knocked out of a lattice site by a heavy (DM) particle moves, generally speaking, in an arbitrary direction. The probability of channeling for an ion crossing a crystallographic channel (axial or planar) depends on the angle between its trajectory and the channel direction and is inversely proportional
to the quadruple root of its energy for an axial channel and to the square root (i.e. to its velocity) for a planar one. We used recommendations of Appleton et al.\textsuperscript{25} to calculate the angles of entry into the principal accessible channels, axial and planar, for the NaI crystal (NaCl-type cubic crystal, lattice constant 6.473 Å). For singly charged, 4-keV iodine ions the critical angle for the \(<100>\) axial channels is 6.4°, for the \(<110>\) channels it is 4.9°, and for the \(<111>\) channels, 2.8°; for the planar \{100\} channels it is 4.1°, for \{110\} it is 3.2°, and for \{111\}, 2.7°. Summing up the solid angles formed by these channels we obtain that the channeling probability for a 4-keV iodine recoil ion ejected within a crystal in an arbitrary direction is in this particular case as high as nearly 20%. Because, as we have seen, for channeling recoil ions \(q \approx 1\), it thus follows that the efficiency of detection of these ions, while being naturally somewhat lower, will nevertheless amount to about 20%.

Assuming now that the DAMA/NaI and DAMA/LIBRA experiments\textsuperscript{2,3} detect with an efficiency \(\eta = 20\%\) (for the remaining 80\% unchanneled ions \(q \approx 0.1\), because for the 2-keV detection threshold in this experiment these ions pass undetected) the flux of daemons crossing the Earth with \(V = 30-50\) km/s in the antapex relative to the Sun region,\textsuperscript{5} it is of interest to estimate this flux from available measurements (some possible processes of daemon interaction with matter were discussed somewhere earlier, see Refs. 4,5,26-28 for examples).

The double modulation amplitude (swing) of the number of events with a one-year period measured in these experiments is about 0.04 cpd/kg/keV in the 2-6-keV interval.\textsuperscript{2,3} In the DAMA/NaI case the NaI(Tl) crystal mass was 96 kg, the crystal packing density was about 50\%, which yields for the effective cross section of the system about 1500 cm\(^2\). Whence for the average 4-keV signal we obtain that the flux of SEECHO daemons falling on the Earth varies during a year from zero to \(f_\odot \approx (0.04 \times 96 \times 4)/(86400 \times 1500 \times \eta) = 6 \times 10^{-7}\) cm\(^{-2}\) s\(^{-1}\) in June, a figure in good agreement with our early estimates of the SEECHO flux of 3\(\times 10^{-7}\) cm\(^{-2}\) s\(^{-1}\) (Ref. 26), which, in its turn, is not at odds with our surface-level\textsuperscript{27} and underground\textsuperscript{28} measurements of the flux of daemons with \(V = 10-15\) km/s, which fall on the Earth near equinoxes from near-Earth, almost circular heliocentric orbits. Similar flux of daemons is needed to explain “Troitsk anomaly”, viz. a periodic drift of the tritium \(\beta\)-spectrum tail position in experiments on direct neutrino mass measurement exploiting the extended T\(_2\) gas source with Nb-containing superconducting magnetic coils (for more details see Ref. 29 and references therein).

4. Conclusions
One may thus conclude that channeling should play a dominant role in measurement of low-energy (keV-range) recoil ions with crystal detectors of the NaI(Tl) type. This effect reduces strongly nuclear stopping, to make interaction with valence electrons of the crystal lattice, including formation and rupture of quasi-molecular bonds with crystal atoms, the dominant factor in stopping of recoil ions. As a result of this highly efficient interaction, the light yield of the crystal may reach and even
exceed that observed when the crystal is acted upon by electrons of the same (keV-range) energies; indeed, in the latter case electrons expend part of their energy also in excitation of the inner-shell atomic electrons.

At high energies (above, say, ten keV), besides the above-mentioned lower probability of entering into a channel due to the acceptance angle diminution, the channeled ion moving in the NaI(Tl) crystal leaves eventually the channel, if for no other reason than as a result of interaction with a Ti$^+$ center, well before exhausting its energy owing to the electron stopping power action (which, in particular, stresses the need of reducing and optimizing the activator content in the scintillator). The ion scattered out of the channel loses its energy primarily through nuclear stopping, and this is what accounts for quenching of the light yield. This is why, in particular, the efficiency of scintillation excitation in NaI(Tl) by iodine recoils with an energy of tens of keV is an order of magnitude lower than that by electrons, a point substantiated by neutron beam calibrations.$^{6-11}$

The above stresses the need for developing methods to directly calibrate crystal scintillators with keV-range ion beams, as this was done by Bredov and Okuneva$^{24}$ in the first semiconductor implantation experiments.

A recent publication by the EDELWEISS Collaboration$^{30}$ reports on neutron scattering measurements of the dependence of heat and ionization quenching factors in Ge on $E_r$ performed in the 10-100-keV range. As pointed out by the authors, as of today these are the most precise absolute measurements for any detectors employed in the direct DM search. In the above energy range, the ionization $q$ increases with increasing $E_r$ by a factor ~1.7. On the other hand, as far back as 1971 Jones and Kraner$^{31}$ pointed out that for $E_r \leq 1.8$ keV, $q$ does increase too. An increase of $q$ for $2.7 \leq E_r < 7-8$ keV was reported by Messous et al.$^{32}$ (see also Fig. 2 in Ref. 30). The latter authors ascribed the effect to the poor resolution of the detectors, whereas Jones and Kraner$^{31}$ explicitly suggest the possibility of strong channeling (sic!) of recoil nuclei in the Ge crystal. Unfortunately, $E_r = 10$ keV for elastically knocked out Ge nuclei in the EDELWEISS experiment corresponds to the supermassive projectile velocity of $V \approx 80$ km/s, a figure substantially in excess of the SEECHO daemon velocity of 50 km/s. Thus, we have to state once more regretfully that it is apparently only the DAMA/NaI and DAMA/LIBRA, besides our experiment, that are presently capable of detecting the fall of daemons from Earth-crossing orbits.

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