Neutron reflection from fluorinated nanodiamonds for N-Nbar

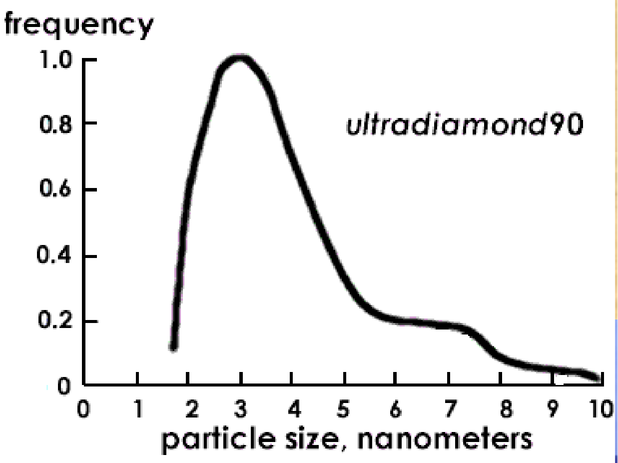
- Introduction to nano-diamond neutron reflectors
- The problem of hydrogen impurities
- The impact of hydrogen substitution by fluor
- Conclusions and prospects

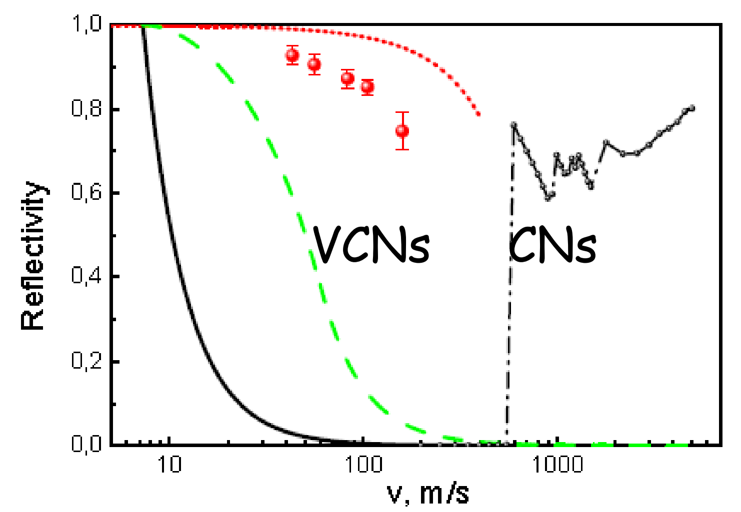
Introduction to nano-diamond reflectors (broader than particle physics)

Complementarity of neutron and synchrotron radiation.

Neutron radiation: particularly light elements and magnetic structures; the object of research in particle physics – in this sense, complementary to high-energy particle accelerators; mostly sensitive to nuclei in atoms.

Synchrotron radiation: mostly sensitive to electrons in atoms.





Introduction to nano-diamond reflectors

Slow neutrons are traditionally subdivided into several groups as a function of their energy/velocity:

- Cold neutrons (CNs): a typical velocity 1000 m/s; you have a lot of CNs from all typical neutron sources (nuclear reactors and spallation neutron sources), if they are equipped, for instance, with a cold liquid-deuterium or liquid-hydrogen source;
- Ultracold neutrons (UCNs): a typical velocity 5 m/s; in spite of all worldwide efforts, extremely low densities available for experiments, however a unique property of total reflection from material and magnetic walls thus storage of UCNs in traps;
- Very cold neutrons (VCNs): a typical velocity 100 m/s; limited fluxes and no efficient reflectors … until recently

Introduction to nano-diamond reflectors

Worldwide trend to increase the range of useful neutrons towards smaller energies (larger wavelengths), driven in particular by large-scale-structure diffractometers, reflectometers, time-of-flight and spin-echo techniques, by particle physics.

The progress is limited by low fluxes of slow neutrons (the wavelength larger than 0.5 nm, or the energy smaller than 3 meV).

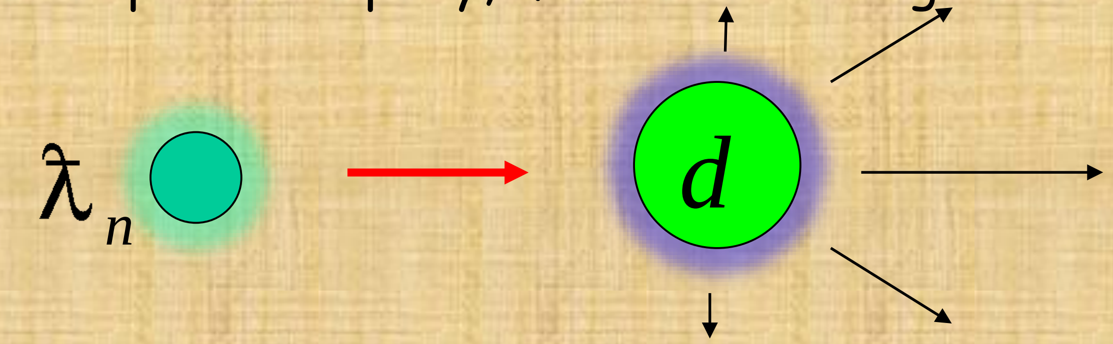
The drop of flux is due to a fundamental reason: independently of the choice of materials for neutron reflectors, they are composed of atoms, thus a finite interatomic distance, thus diffraction and thus simply mean neutron-nuclei optical potential.

The solution of this problem: “Mimicking” conventional reflectors with nanoparticles.

Introduction to nano-diamond reflectors

Neutron-atom: the neutron-electron interaction is usually a minor correction compared to the neutron-nuclei interaction; ***Neutron-nucleus***: as the wavelength of a slow neutron is larger than the size of a nucleus, we always deal with isotropic s-scattering and can characterize it with a single parameter: a scattering length; ***Neutron-matter***: as the wavelength of a slow neutron is also larger than a typical inter-atomic distance, we deal with coherent scattering of neutrons at many nuclei simultaneously; ***Optical potential***: as a result, any medium can be represented as a uniform effective optical potential; ***Potential strength***: a typical value of the optical neutron-nuclei potential is $\sim 10^{-7}$ eV (could be thought of as a typical nuclear potential of ~10 MeV diluted over volume).

Two contradicting tendencies: 1) Cross-section increases rapidly as a function of the nanoparticle size; 2) Angular divergence and cross-section drop down rapidly, if the size is large.



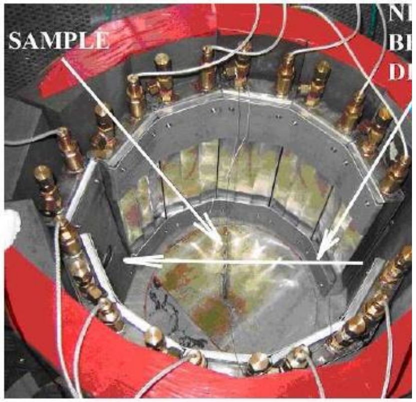
1) The optimum neutron wavelength is approximately equal to the nanoparticle size;

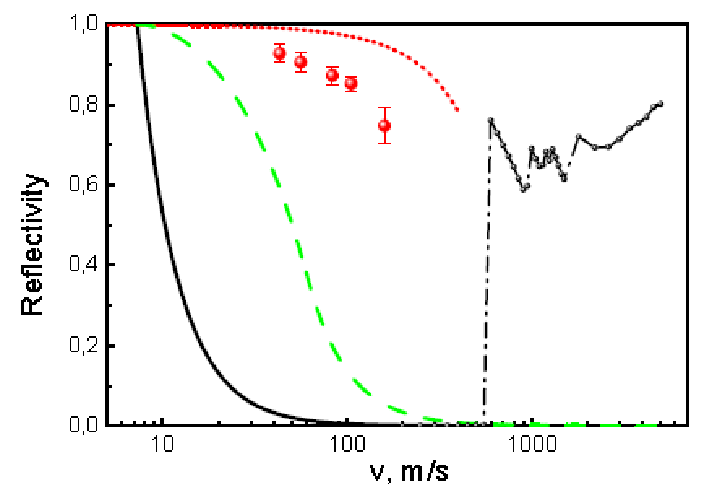
2) The cross-section is then *measured in square nanometers;*

3) The best material is diamond.

Introduction to nano-diamond reflectors

The (elastic) *reflection* of VCNs from powder of diamond nanoparticles, the *storage* of VCNs in closed volumes with nano-powder walls, and *quasi-specular* reflection of cold neutrons (CNs) from diamond nano-powder have been proven.





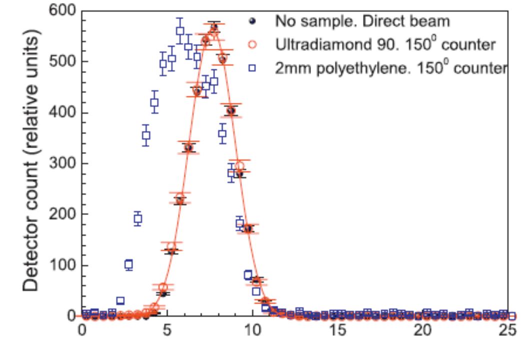
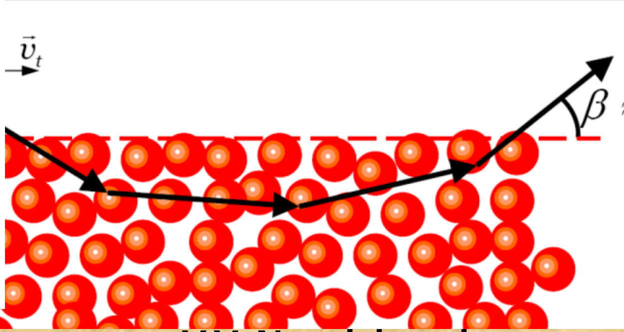


Fig. 6. The neutron count rate is presented as a function of the time of flight of the neutrons with an average initial velocity of 60 m/s. The zero time is synchronized with opening the chopper. The black circles correspond to the initial neutron spectrum. The empty circles indicate the data for the spectrum of neutrons scattered to an angle of 150°. The thickness of the ultradiamond90 powder sample is equal to 2 mm. The squares show results for the scattering of neutrons at a polyethylene sample with a thickness of 2 mm, measured at the same counter.



The danger of hydrogen impurities

An important limitation for further improving the reflectivity consisted of the presence of hydrogen in the amorphous shells of diamond nanoparticles.

σ_{abs}^{C} = 3.5 mb

σ_{abs}^{H} = 333 mb

σ_{abs}^{F} = 9.6 mb

b^{C} = 6.65 fm

b^{H} = -3.74 fm

b^{F} = 5.65 fm

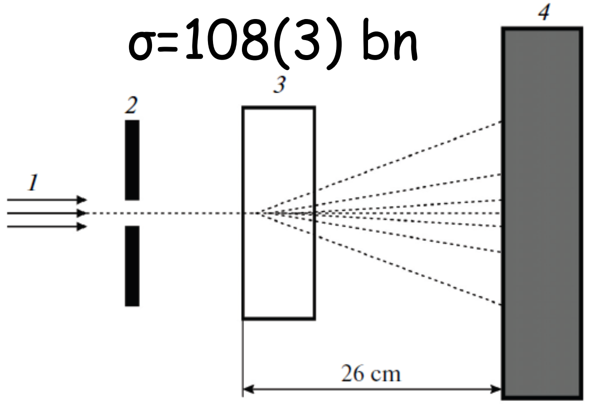
1% of mass but !! $\sigma_{in.sc.}^{H}$ = 108(3) b

We have explored three methods to remove hydrogen:
- Thermal treatment;
- Isotopic substitution;
- Chemical treatment.

Fluorination of nanodiamonds

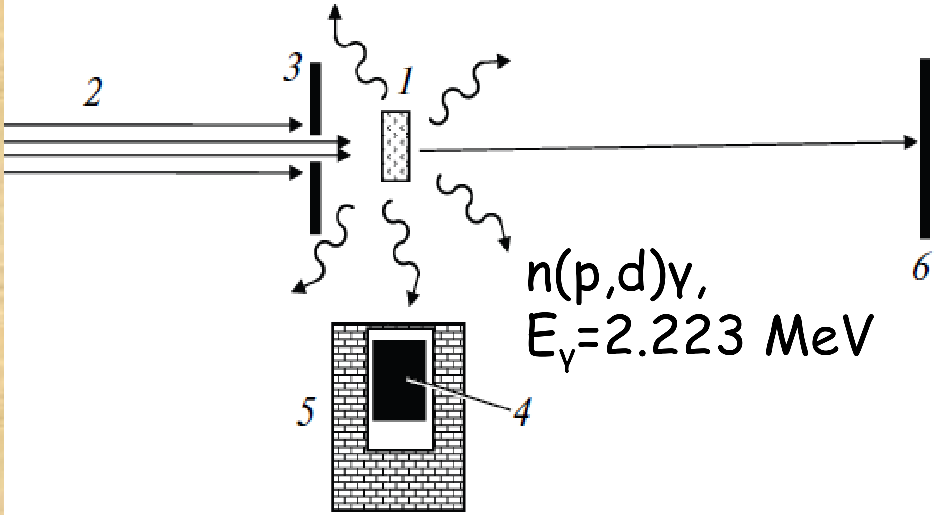
An important limitation for further improving the reflectivity consisted of the presence of hydrogen in the amorphous shells of diamond nanoparticles

As-prepared (detonation technology)	$C_{7.4 \pm 0.15}H$
After thermal treatment	$C_{12.4 \pm 0.2}H$
After fluorination	$C_{430 \pm 30}H$



σ=108(3) bn

Fig. 3. A scheme of the measurement of the total cross-section of neutron scattering. 1—neutron beam; 2—diaphragm; 3—sample; 4—position-sensitive neutron detector.



n(p,d)γ,
E_γ=2.223 MeV

Fig. 1. A scheme of the measurement of a fraction of hydrogen in powder. 1—sample; 2—neutron beam; 3—collimator; 4—γ-detector; 5—detector shielding; 6—neutron detector.

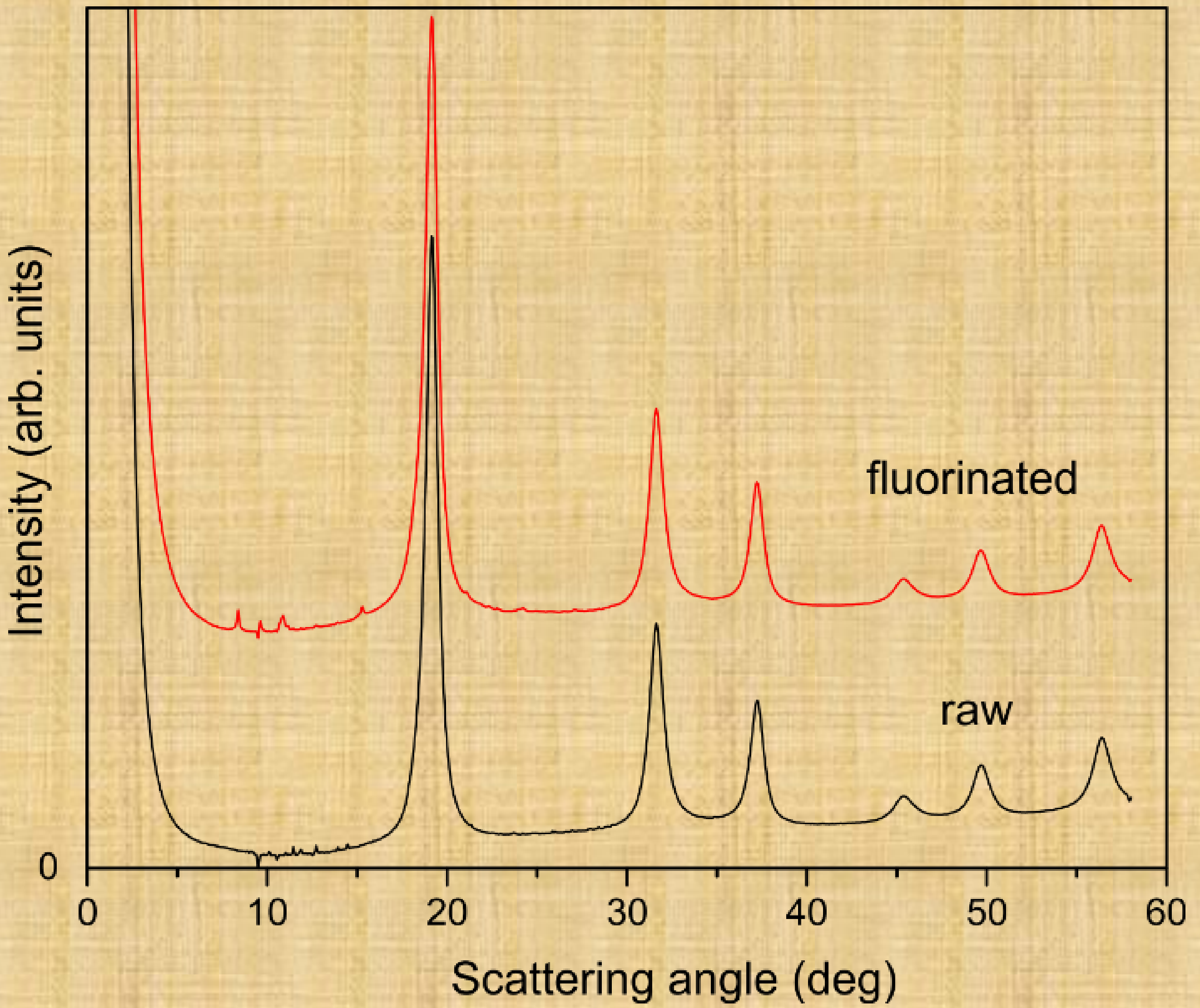
Energy dependence of neutron inelastic scattering showed that C-H bond is responsible for neutron losses

Fluorination of nanodiamonds

X-ray diffraction patterns of raw and fluorinated nano-diamond powder:

- Diamond sp^3 cores remain unaffected;
- Destruction of sp^2 carbon shells.

Destruction of sp^2 carbon will lead to significantly higher efficiency of neutron scattering!

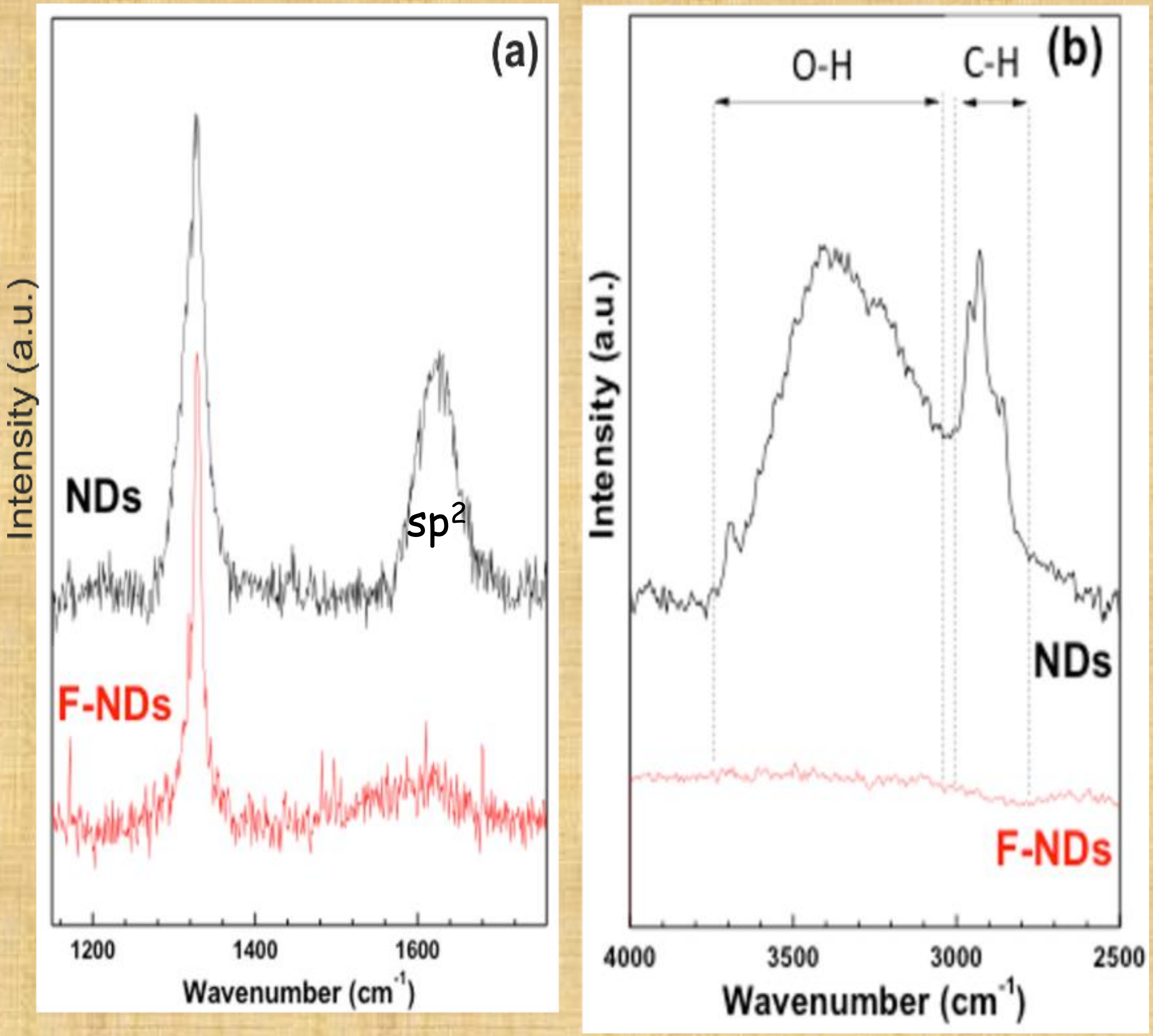


Fluorination of nanodiamonds

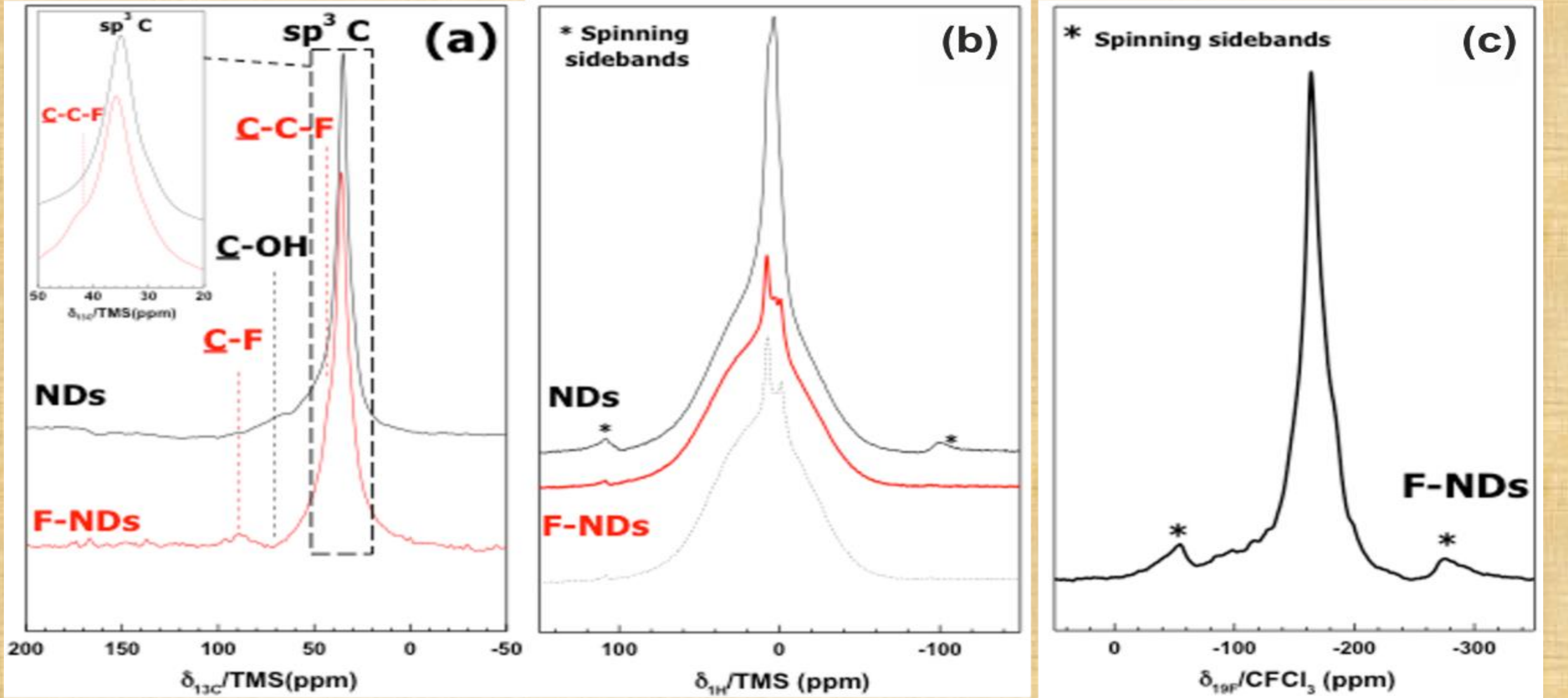
A) Raman spectra (inelastic scattering of monochromatic light) of raw and fluorinated nano-diamond;

B) FTIR (Fourier-transform infrared spectroscopy) spectra of raw and fluorinated nano-diamond.

Disappearance of sp^2 carbon, C-H and O-H.



Fluorination of nanodiamonds



^{13}C (a), ^{1}H (b) and ^{19}F (c) MAS NMR spectra of raw and fluorinated nano-diamonds

- The departure of –OH groups and their replacement by –F groups;
- Minor residual H content upon fluorination;
- C-F bonds with sp^3 carbons, very low amount of C-F bonds with sp^2 carbons.

Chemical cleaning from isotopes posing the neutron activation danger

Fluorination does not affect most disturbing radioactive chemical elements(isotopes).

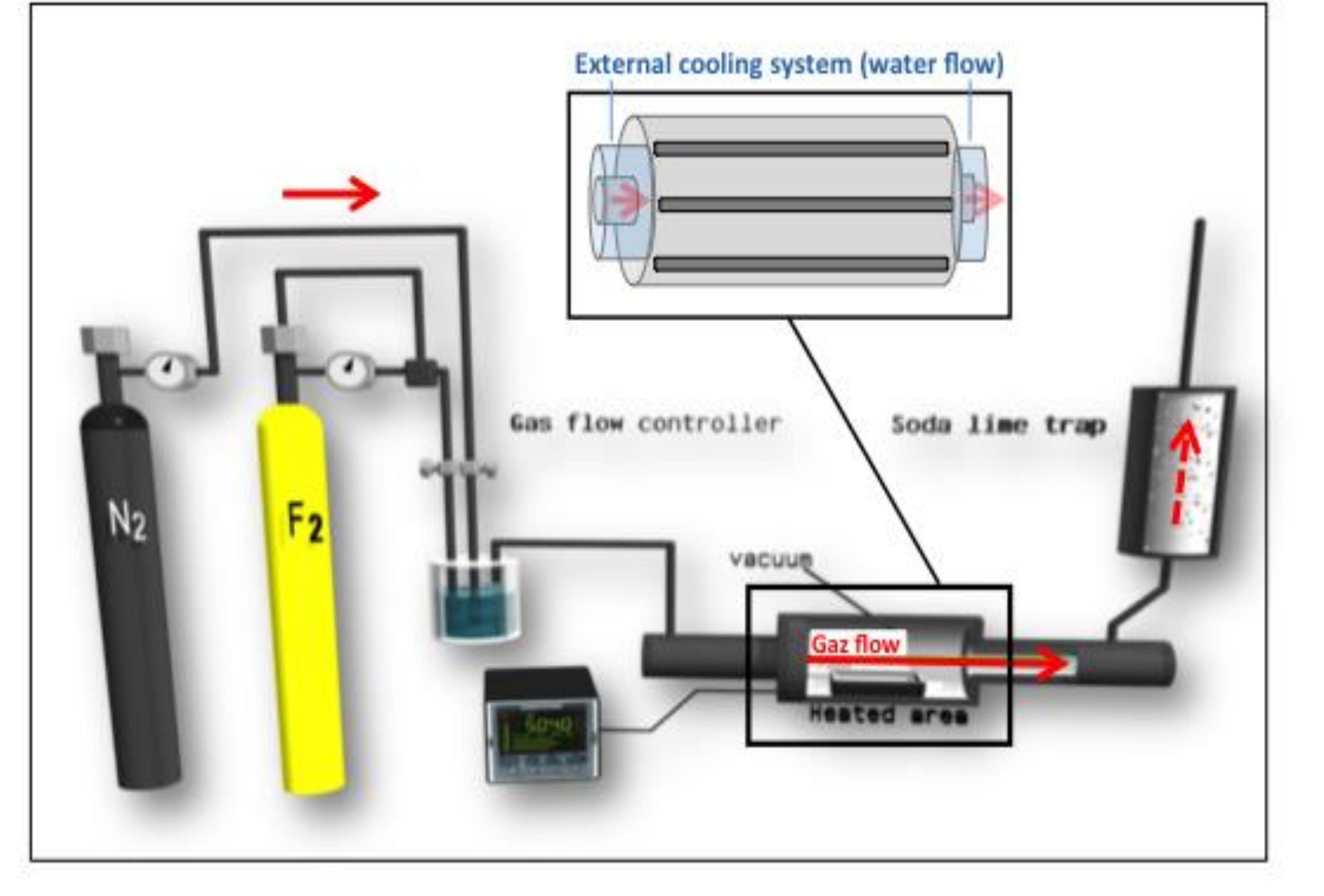
As revealed by neutron activation analysis of raw and fluorinated nano-diamond samples, most disturbing isotopes are ^{64}Cu, ^{24}Na, ^{51}Cr. A chemical treatment reduces their amount by factors of 12, 6 and 2, respectively.

Wet chemical treatment in concentrated hydrochloric acid at the temperature of 140 °C for 18 hours (+ washing, centrifugation, air annealing at the temperature of 600 °C for 5 hours.

The amount of ^{38}Cl increases by an order of magnitude due to the chemical treatment, however, due to a relatively short lifetime of this radioactive isotope, it is not particularly disturbing (but contributes to absorption!).

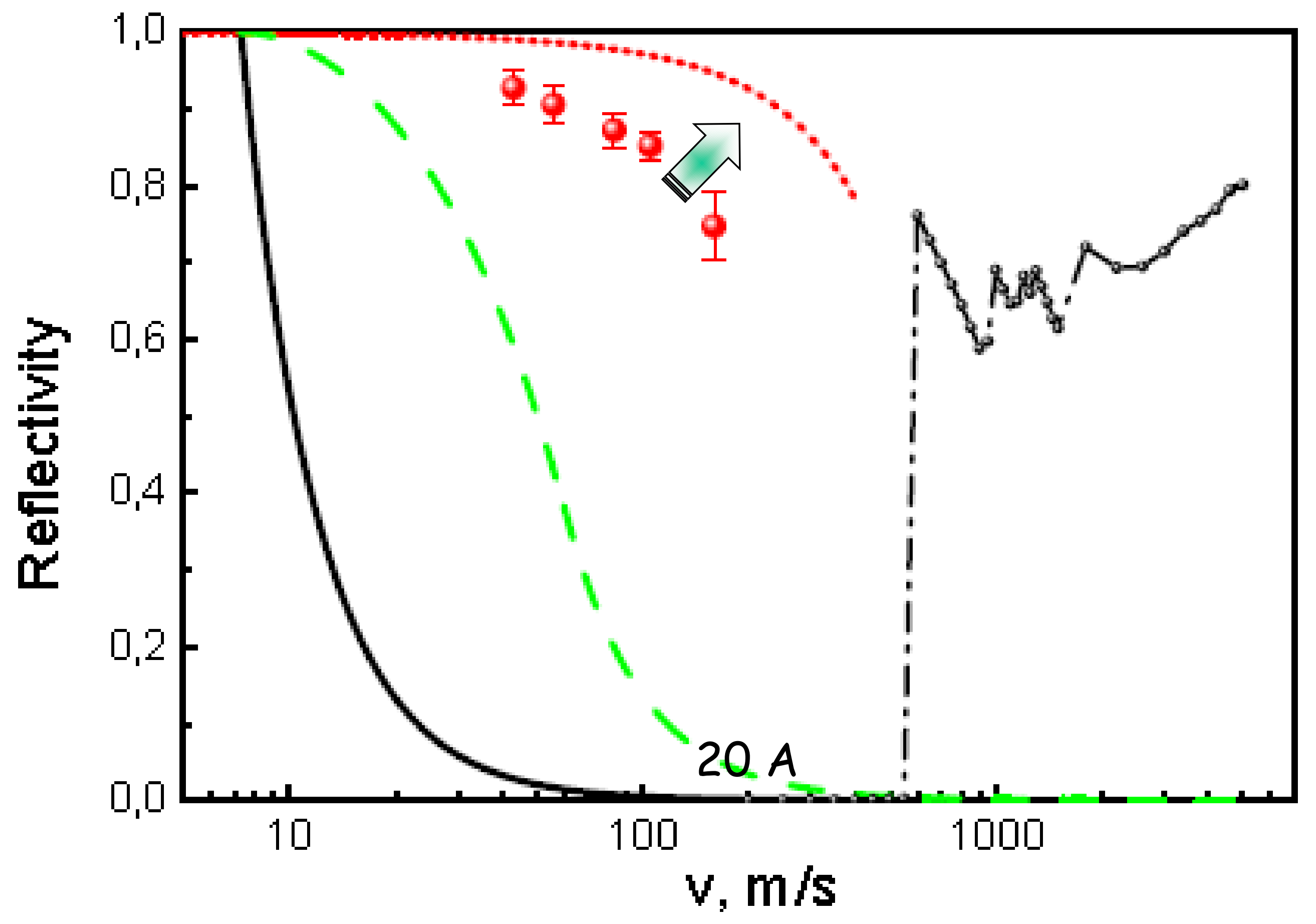
Fluorination procedure

0.5 kg fluorinated nano-diamonds were produced in 2-cycle fluorination of thin (2 mm) layers of raw nano-diamond powder with 99.9 % clean F_2 gas at the pressure of 0.6 bar, at the temperature of 450 °C for 12 hours.

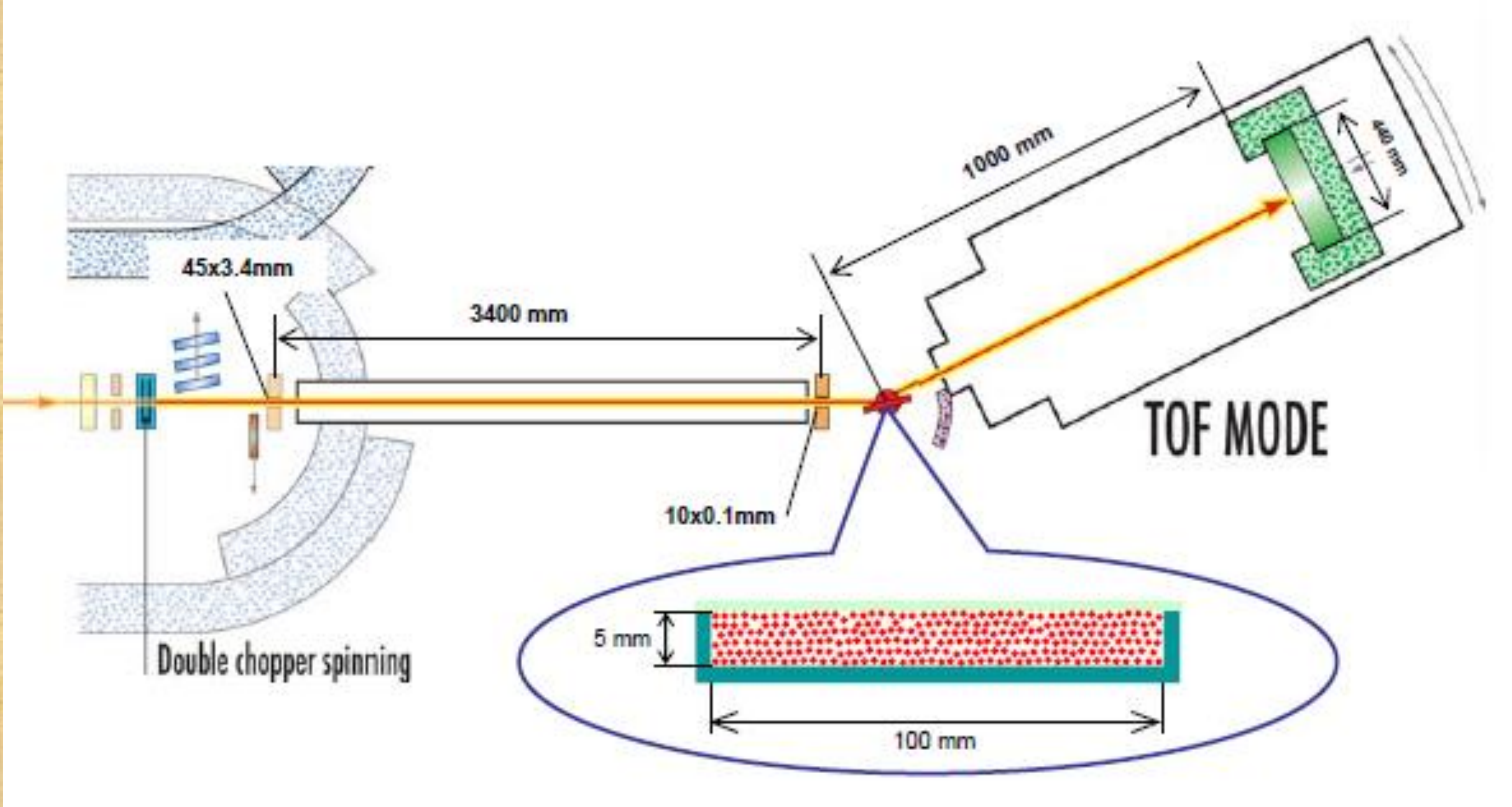


NEUTRONS
FOR SCIENCE

The impact of removal of hydrogen from diamond nanoparticles



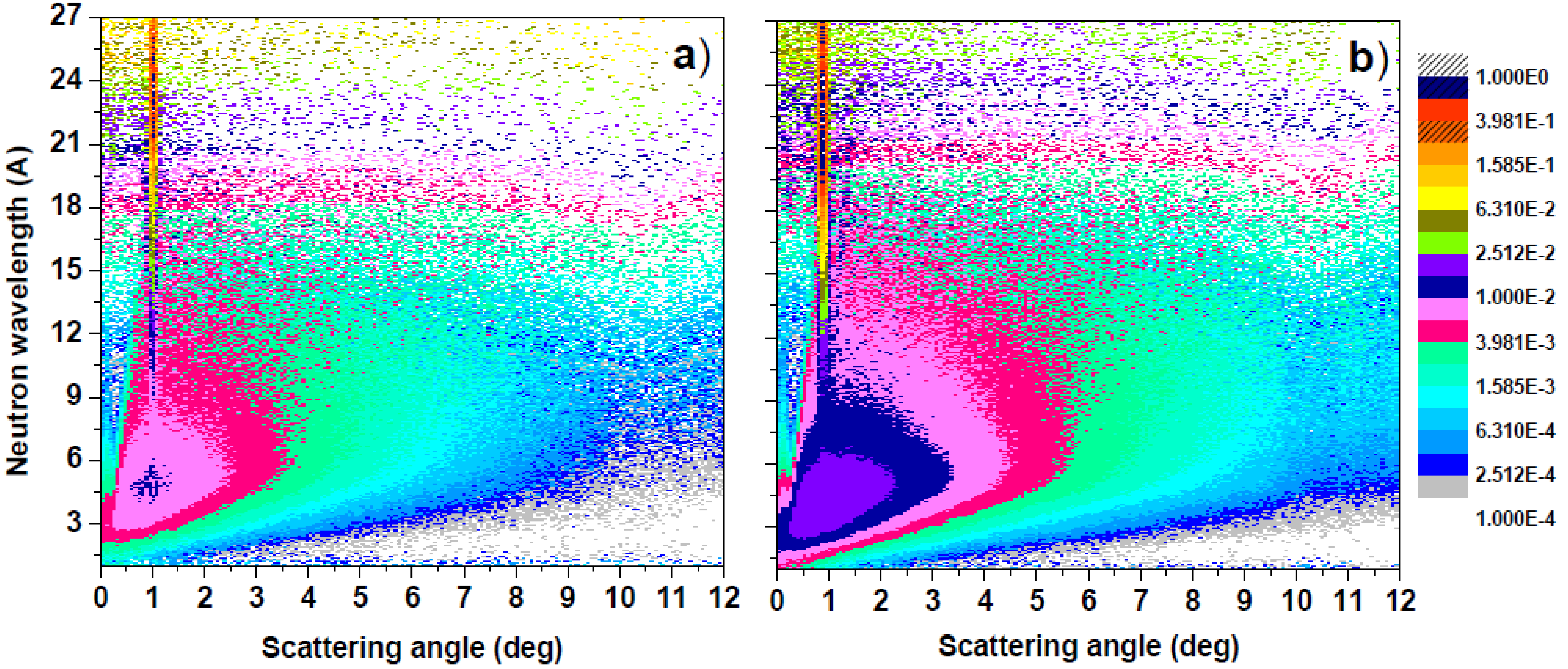
Quasi-specular reflection



Quasi-specular reflection

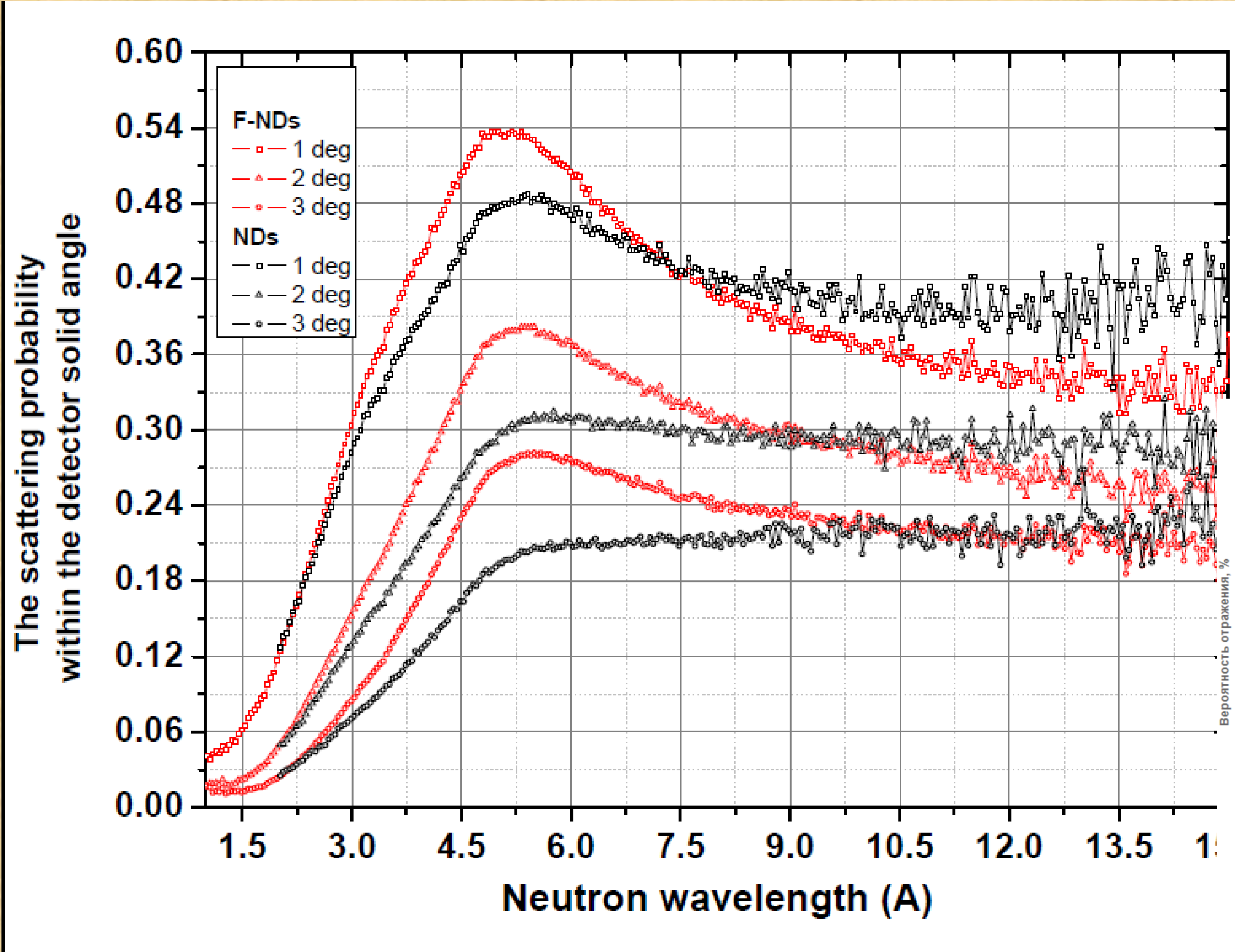
The intensity of neutron scattering (within the angular acceptance of the D17 position-sensitive detector in the vertical direction) as a function of neutron wavelength and scattering angle, for the incidence angle of 1deg.

On the left – non-fluorinated nanodiamond; on the right – fluorinated nanodiamond

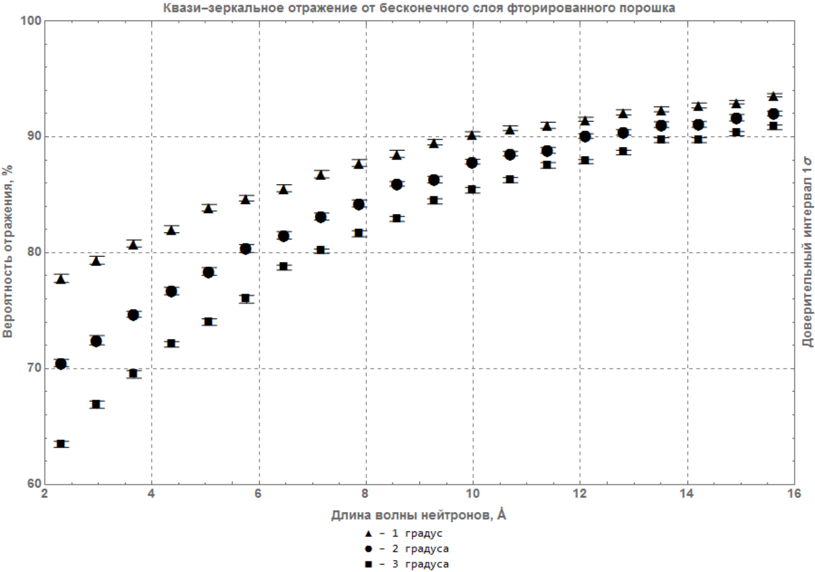


Quasi-specular reflection

The total probability of neutron scattering (within the angular acceptance of the D17 position-sensitive detector) as a function of the neutron wavelength, for the incidence angles of 1deg., 2deg., 3deg.

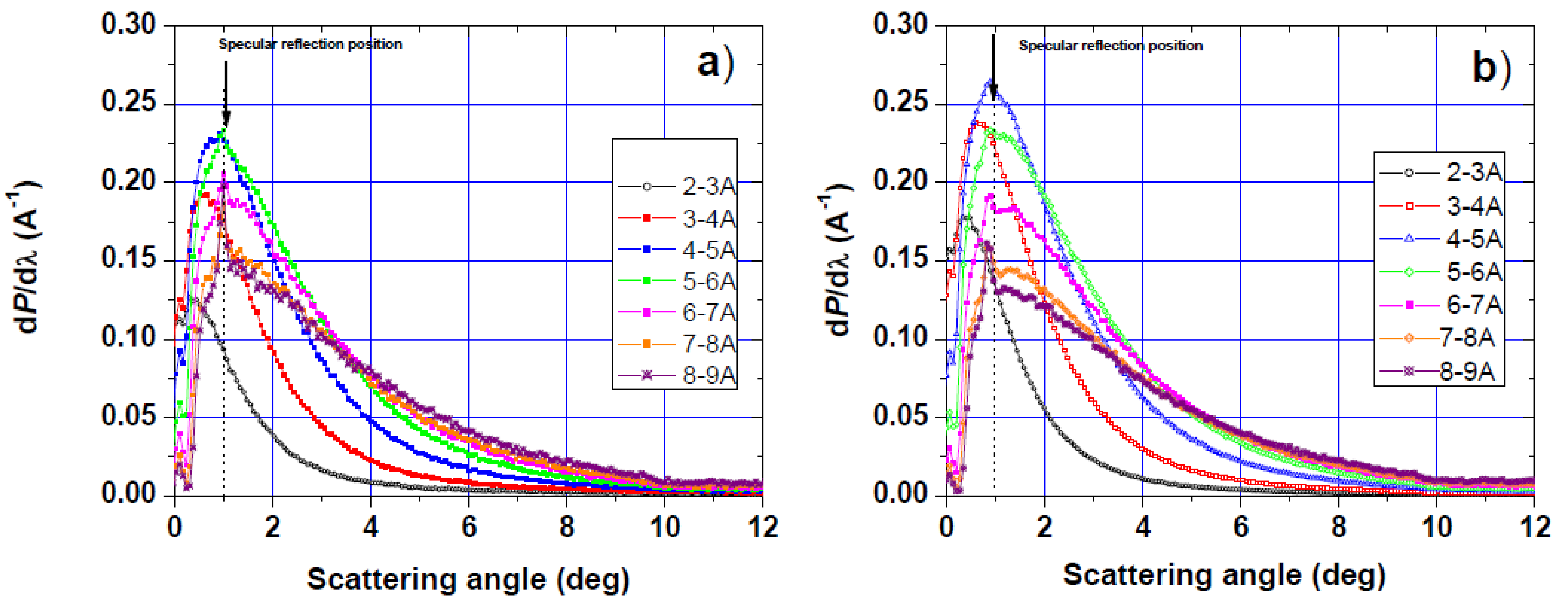


Red color – fluorinated nanodiamond;
Black color – non-fluorinated nanodiamond

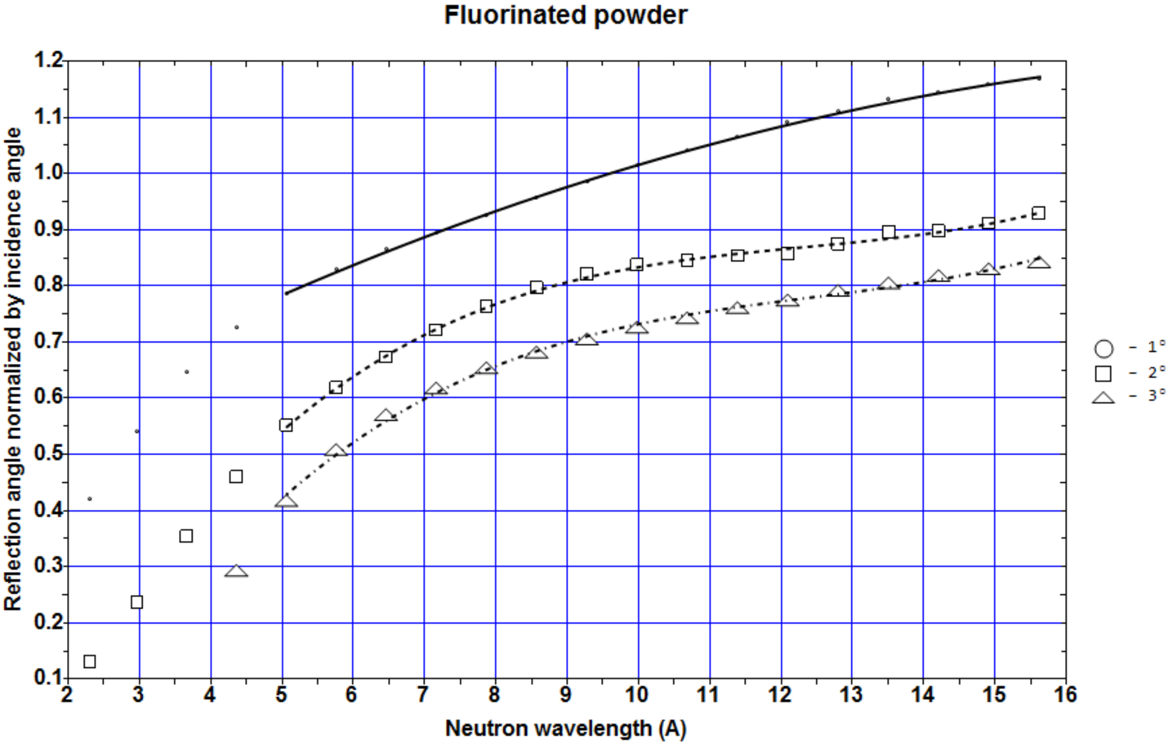


Quasi-specular reflection

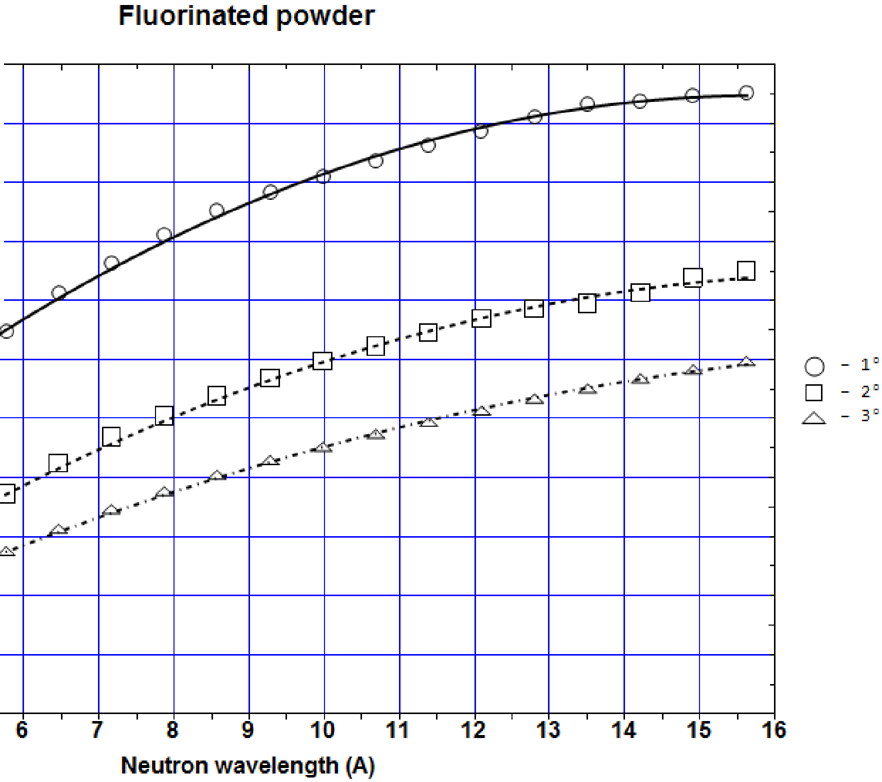
The probability of neutron scattering as a function of the neutron wavelength, for the incidence angle of 1deg.



Quasi-specular reflection



Mean angle of reflection as a function of the neutron wavelength, for incident angles 1deg., 2deg., 3deg.

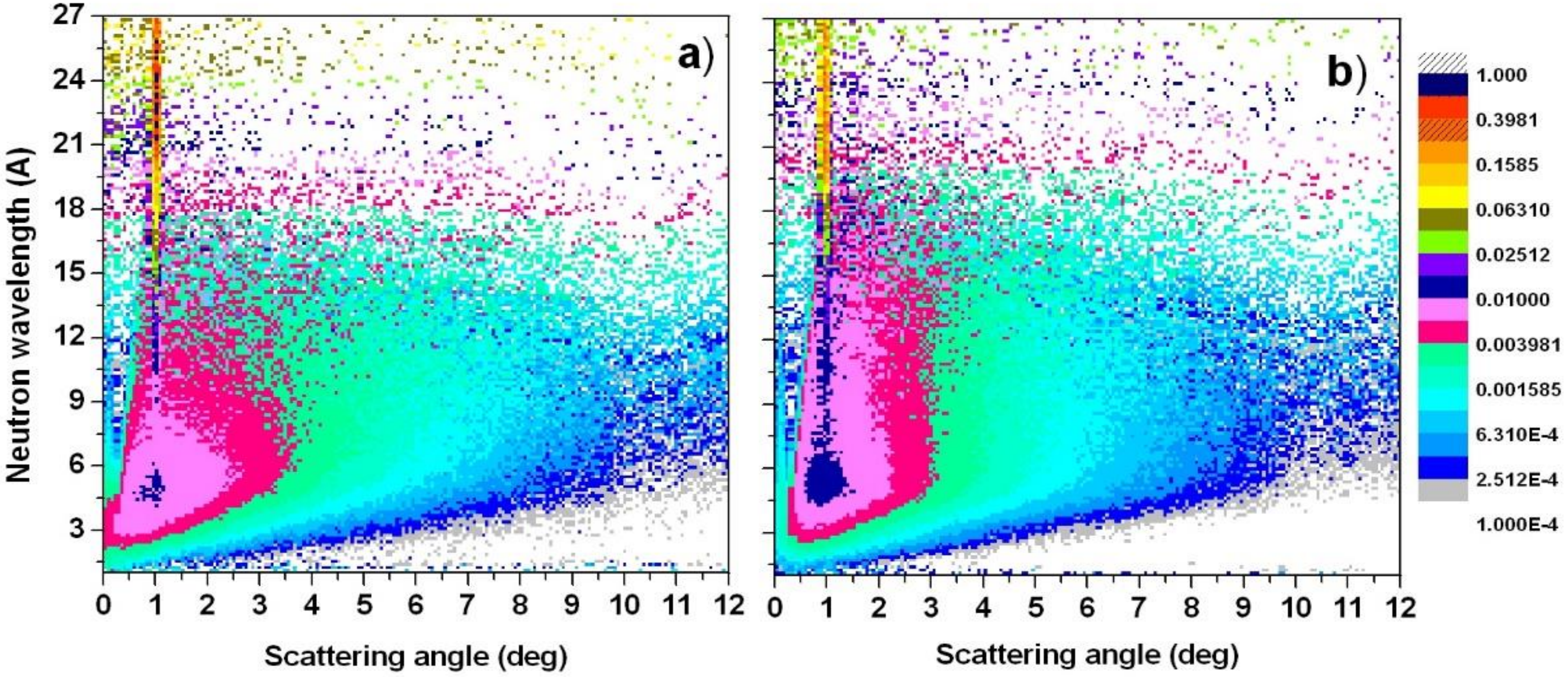


Half-width of the angle of reflection as a function of the neutron wavelength, for incident angles 1deg., 2deg., 3deg.

Quasi-specular reflection

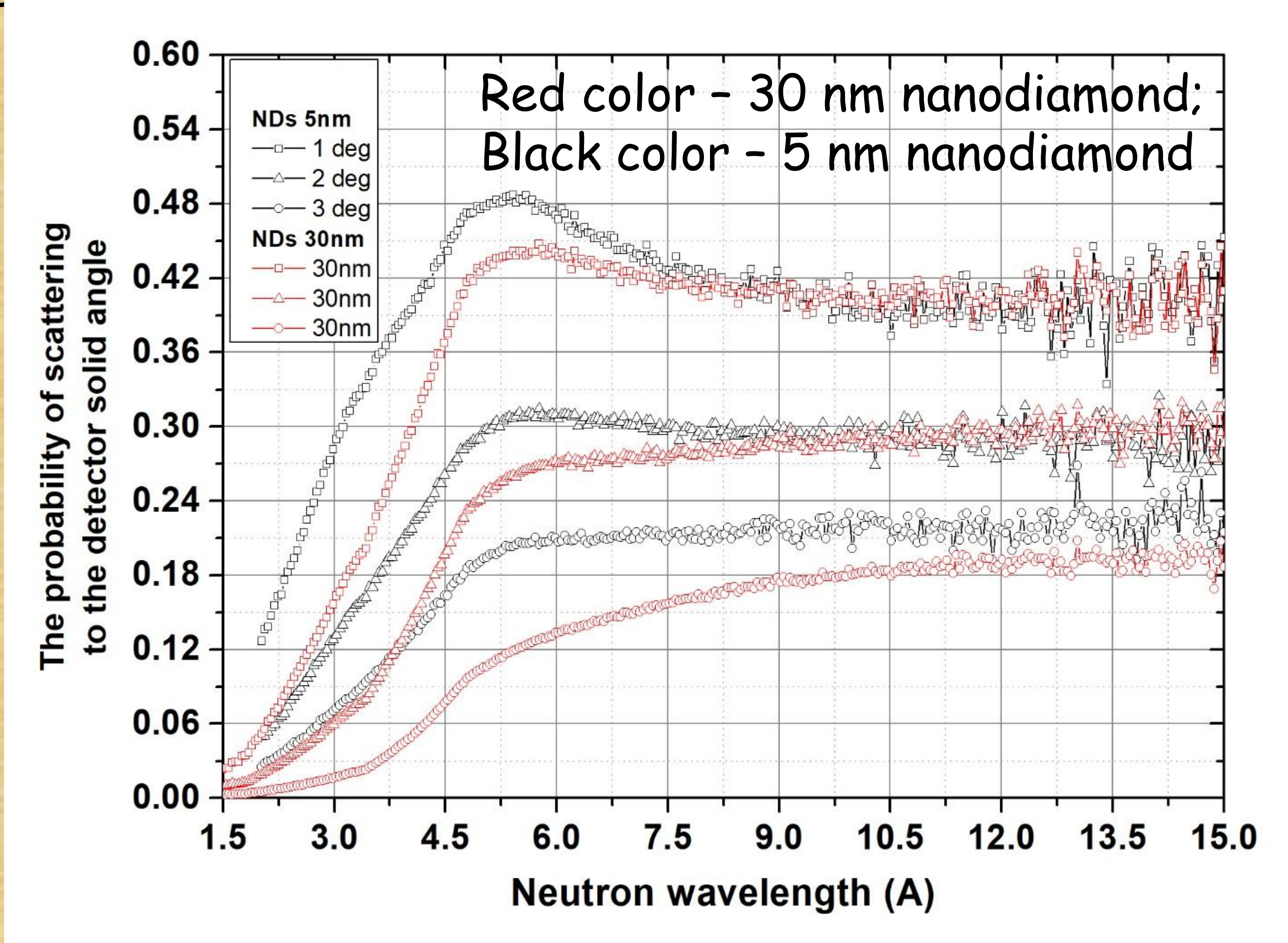
The intensity of neutron scattering (within the angular acceptance of the D17 position-sensitive detector) as a function of neutron wavelength and scattering angle, for the incidence angle of 1deg.

On the left – 5 nm nanodiamond; on the right – 30 nm nanodiamond



Quasi-specular reflection

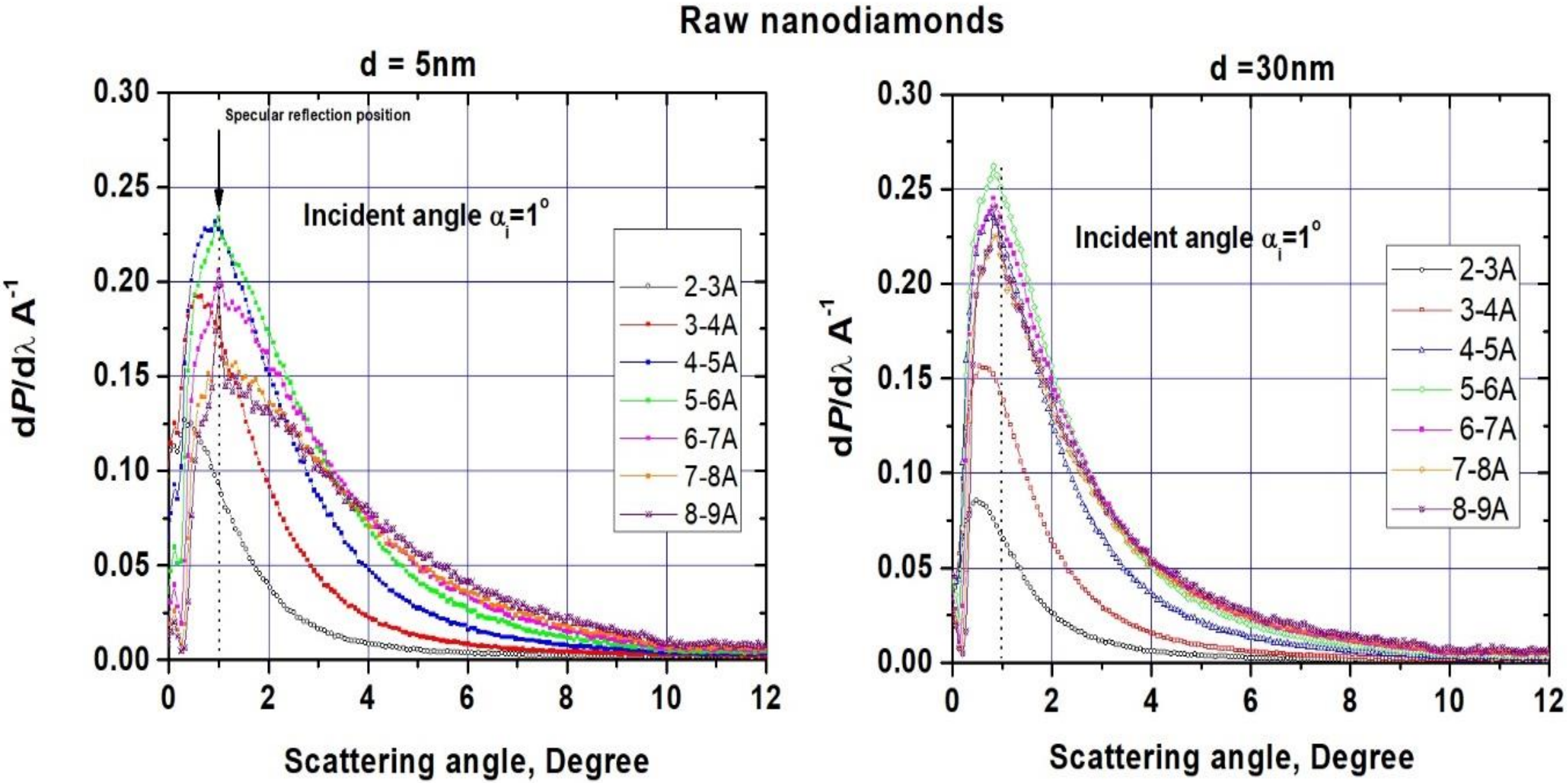
The total probability of neutron scattering (within the angular acceptance of the D17 position-sensitive detector) as a function of the neutron wavelength, for the incidence angles of 1deg., 2deg., 3deg.



Red color – 30 nm nanodiamond;
Black color – 5 nm nanodiamond

Quasi-specular reflection

The probability of neutron scattering as a function of the neutron wavelength, for the incidence angle of 1deg.



Intermediate conclusions. Prospects. Applications.

- Powder of diamond nanoparticles provided the first efficient reflectors for VCNs and efficient quasi-specular reflection of CNs;

- Such powders can provide a major gain in fluxes and densities of slow neutrons in various configurations;

- Losses of neutrons in raw diamond nano-powders are dominated by hydrogen impurities; the amount of hydrogen has been largely reduced by fluorination;

- Samples of diamond nanoparticles with "close-to-ideal" parameters have been produced and successfully studied. We explore a possibility of larger-scale production in order to provide their real applications in neutron science/technology.

- More powerful sources of slow neutrons (CNs, VCNs, UCNs) !
  - A new generation of general-purpose neutron sources !?
    - More efficient nuclear reactors of the future ?

UCNs versus VCNs

To remind advantages of VCNs:

- Longer observation times (compared to CNs and TNs) that is interesting for particle physics (nñ oscillations, neutron beta-decay etc);
- Larger absorption and fission cross-sections (compared to CNs and TNs) that is interesting for nuclear and particle physics;
- Relaxed geometrical constrains and higher sensitivity (compared to CNs and TNs) that is interesting for neutron reflectometers;
- Broader dynamical range for large-structure diffractometers, time-of-flight spectrometers and reflectometers (compared to CNs and TNs);
- Much higher sensitivity for spin-echo techniques (compared to CNs and TNs);
- Higher fluxes than UCNs etc…

UCNs versus VCNs

However, huge **disadvantages**:

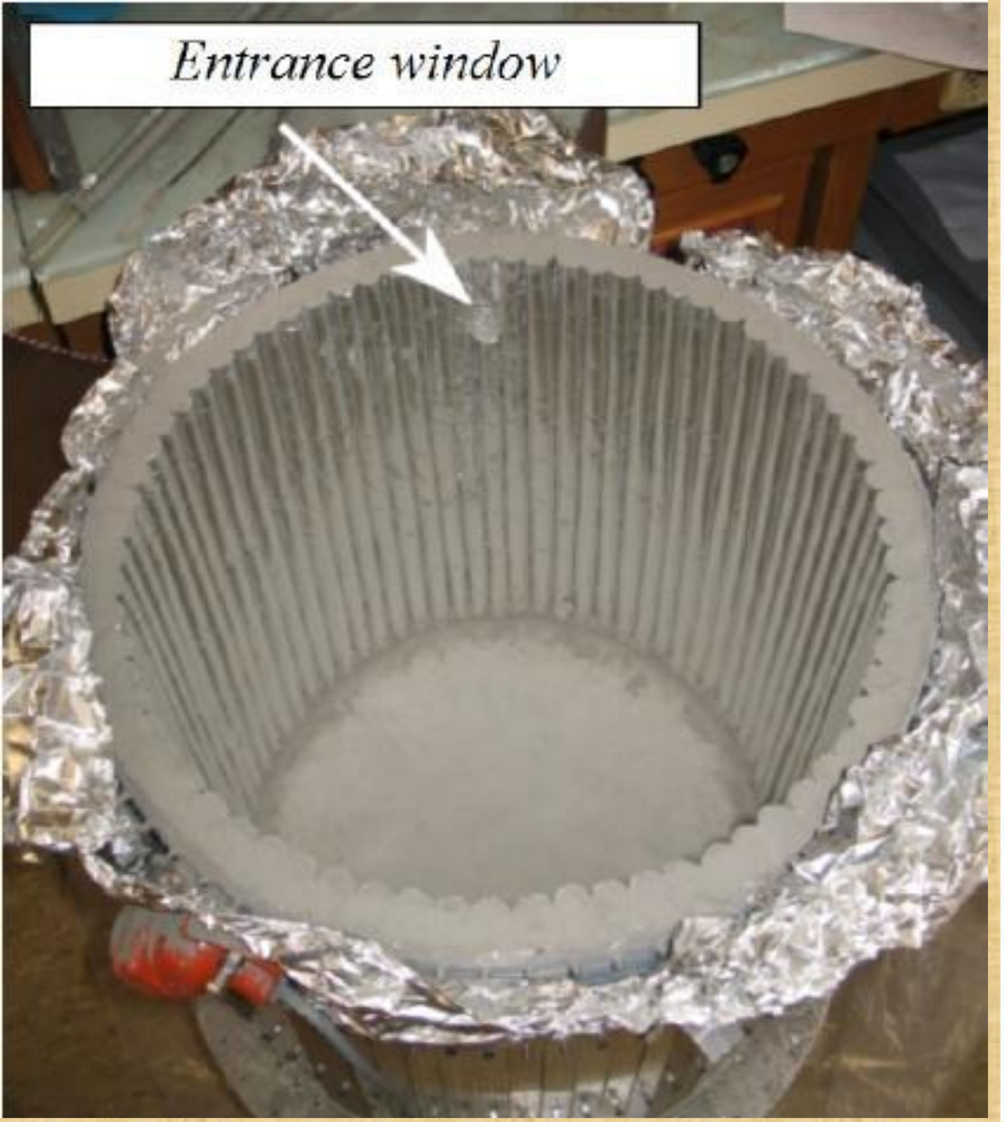
- Relatively low fluxes compared to CNs and TNs (as no efficient VCN reflectors have been available…), and

- Short observation times compared to UCNs (also … as no efficient reflectors have been available).

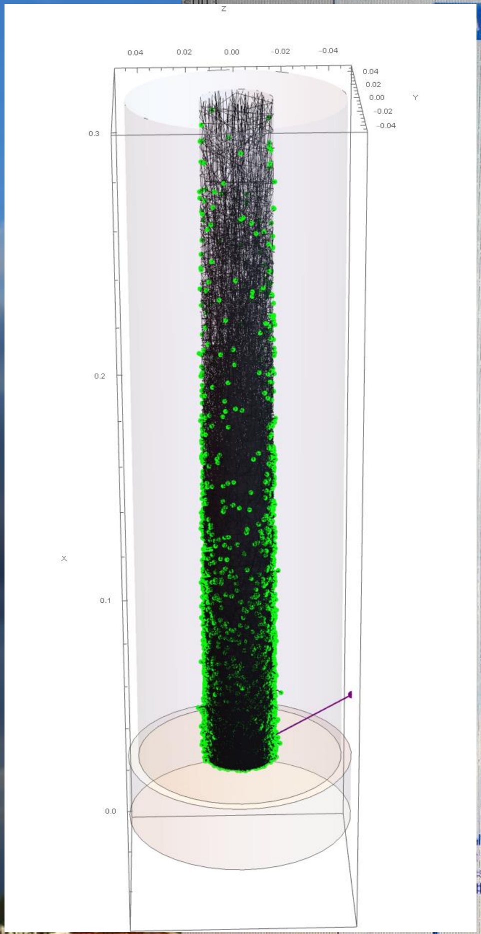
Our method: Keep advantages and reduce disadvantages !

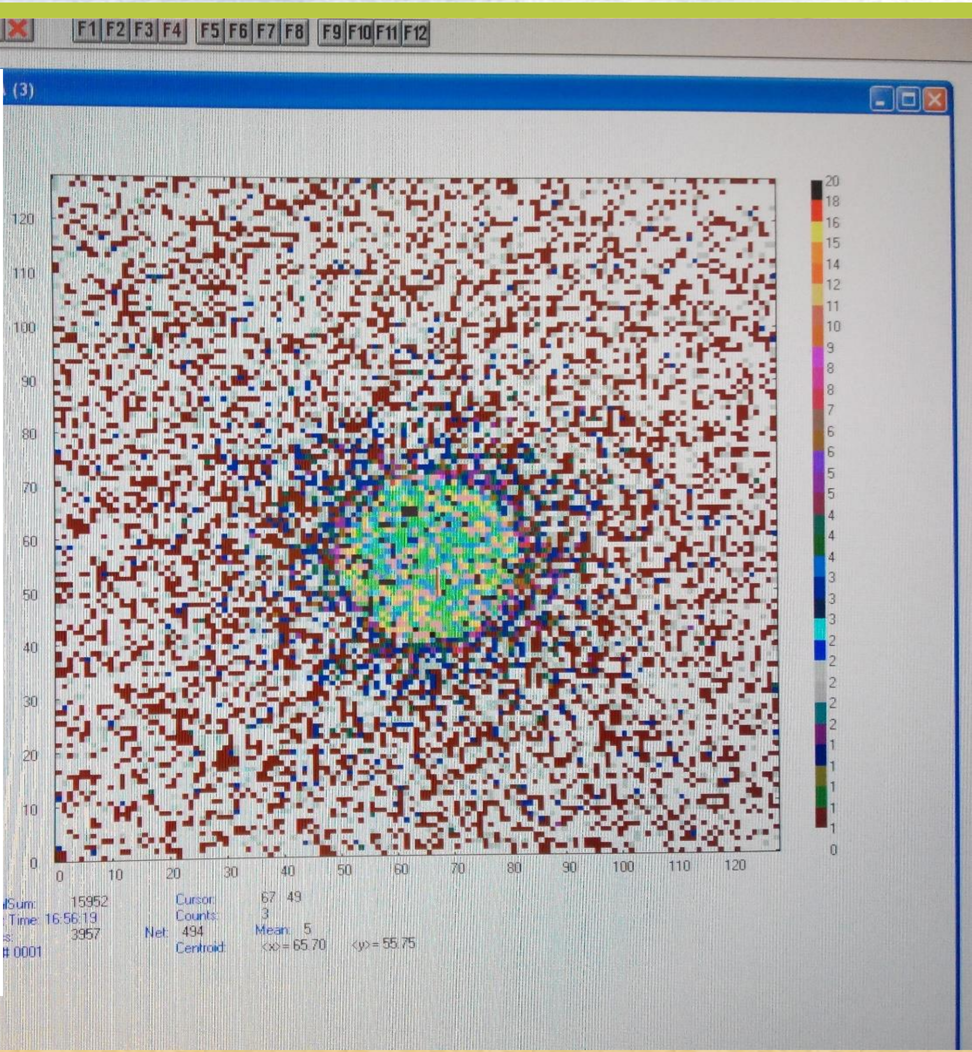
High fluxes and storage in closed traps

The list of possible applications is long


Entrance window

Directional extraction of VCNs





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Mean: 5
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The gain factor is about 20-30 (preliminary, in progress)

References relevant to nano-diamond neutron reflectors:

V.V. N., “*Interaction of neutrons with nanoparticles*", Phys. At. Nucl. 65 (2002) 400; V.A. Artem’ev, “*Estimation of neutron reflection from nanodispersed materials*", At. En. 101 (2006) 901; V.V. N., E.V. Lychagin, A.Yu. Muzychka, A.V. Strelkov, G. Pignol, and K.V. Protasov, “*The reflection of very cold neutrons from diamond powder nanoparticles*", Nucl. Instr. Meth. A 595 (2008) 631; E.V. Lychagin, A.Yu. Muzychka, V.V. N., G. Pignol, K.V. Protasov, and A.V. Strelkov, “*Storage of very cold neutrons in a trap with nano-structured walls*", Phys. Lett. B 679 (2009) 186; V.N. Nesvizhevsky, R. Cubitt, E.V. Lychagin, A.Yu. Muzychka, G.V. Nekhaev, G. Pignol, K.V. Protasov, A.V. Strelkov, “*Application of diamond nanoparticles in low-energy neutron physics*", Mater. 3 (2010) 1768; R. Cubitt, E.V. Lychagin, A.Yu. Muzychka, G.V. Nekhaev, V.V. N., G. Pignol, K.V. Protasov, and A.V. Strelkov, “*Quasi-specular reflection of cold neutrons from nano-dispersed media at above-critical angles*", Nucl. Instr. Meth. A 622 (2010) 182; A.R. Krylov, E.V. Lychagin, A.Yu. Muzychka, V.V. N., G.V. Nekhaev, A.V. Strelkov, and A.S. Ivanov, “*Study of bound hydrogen in powders of diamond nanoparticles*", Crystal. Rep. 56 (2011) 1186; V.V. N., “*Reflectors for VCN and applications of VCN*", Rev. Mexic. Pisic. 57 (2011) 1; V.V. Nesvizhevsky, U. Koester, M. Dubois, N. Batisse, L. Frezet, A. Bosak, L. Gines, O. Williams, “*Fluorinated nanodiamonds as unique neutron reflector*", submitted.