Nanoparticles as a possible moderator for an ultracold neutron source

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Ultracold and very cold neutrons (UCN and VCN) interact strongly with nanoparticles due to the similarity of their wavelengths and nanoparticles sizes. We analyze the hypothesis that this interaction can provide efficient cooling of neutrons by ultracold nanoparticles at certain experimental conditions, thus increasing the density of UCN by many orders of magnitude. The present analytical and numerical description of the problem is limited to the model of independent nanoparticles at zero temperature. Constraints of application of this model are discussed.

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I. INTRODUCTION

A series of experiments performed by a number of research groups have brought light to the phenomenon of the quasi-elastic scattering of UCN at surfaces displaying surprisingly small energy changes in the order of 10^{-7} eV [1, 2, 3, 4, 5, 6, 7]. A detailed study of this process [8, 9, 10, 11, 12, 13] has allowed us to conclude that, for solid surfaces at least, this is due to the inelastic coherent scattering of UCN on nanoparticles or nanostructures weakly attached to the surface in a state of permanent thermal motion. This conclusion trigged the idea considered in this article of neutron cooling at ultracold nanoparticles. Complete control of the corresponding UCN losses from storage bottles and/or a significant increase in UCN density are of utmost importance for neutron-based research in fundamental physics. This research includes the measurement of the neutron lifetime [14, 15, 16, 17, 18], the search for the non-zero neutron electric dipole moment [19, 20], the study of the gravitationally bound quantum states of neutrons [21, 22, 23], and the search for the non-zero neutron electric charge [24].

In any experiment with trapped neutrons, nanoparticle temperature is equal to the trap temperature T in the typical range of 10-10^3 K, while UCN energy corresponds to UCN temperature T $\approx$1 mK. Ultracold neutrons therefore preferentially increase their energy in collisions with such "warm" nanoparticles. The probability of such inelastic UCN scattering on the surface is small, since the surface density of such weakly attached nanoparticles is typically small. However, the problem of neutron heating due to neutron-nanoparticle interaction can in principle be reversed: the interaction of warm neutrons with ultracold nanoparticles at a temperature of $\approx$1 mK can cool down the neutrons [8, 25, 26]. If the density of weakly bound nanoparticles is high (these nanoparticles not only cover the surface but also fill the volume) and if, as the neutrons cool, the probability of their absorption and β-decay is low, the neutron density will increase. This process can, for the first time, allow equilibrium cooling of neutrons down to the UCN temperature. One should note that in this case the moderator temperature should be as low as the UCN temperature in contrast to traditional methods to produce UCN.

In order to produce UCN, nuclear fission in nuclear reactors is used, releasing neutrons of energy $\approx 10^7$ eV. The energy of neutrons in pulsed sources based on proton accelerators is commensurate with that in reactors. However, the cooling of neutrons by a factor of about 10^8! is achieved by just a few dozen collisions with nuclei in reactor moderators (hydrogen, deuterium). The energy transfer is very efficient since the mass of the moderator nuclei is equal to (or approximates) the neutron mass and the neutron losses during the cooling process are low due to small number of collisions needed to slow down neutrons. However, no further cooling occurs: the lower the neutron energy, the larger the neutron wavelength. When the wavelength becomes commensurate with the distance between the nuclei of the moderator, the neutrons do not "see" individual nuclei any longer – they are just affected by the average optical potential of the medium. The neutron energy becomes lower than the bounding energy of the atoms in the medium. A further cooling of the neutrons due to their interaction with collective degrees of freedom (such as phonons) is less efficient than the moderation of the neutrons due to their collisions with nuclei. However, this does allow the cooling of the neutrons to the energy range of cold neutrons (about 10^{-3} eV). That is insufficient however to cool the main portion of the neutrons to the UCN energy region [27, 28, 29, 30, 31, 32]. The idea of neutron cooling on ultracold nanoparticles consists in reproducing the principle of neutron cooling in reactor moderators using multiple collisions. However,

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there is a difference in scale: the sizes are greater by a factor of $\approx 10^2$; this increases the energy range of application of this mechanism by a few orders of magnitude. It should be noted that a UCN source of this type is based on the principle of UCN density accumulation, as in a super-thermal source [29], but not on the use of a UCN flux from a source in the flow-through mode. In conventional sources used to select UCN, thermal equilibrium is not achieved. These sources are much hotter than UCN. Only a very small portion of the neutrons is used – the other neutrons are lost. Actually, these are sources of cold or very cold neutrons (VCN), and experimentalists have to select a narrow fraction of a broad energy spectrum. For instance, the most intense flux of UCN available for users is now produced in a liquid-deuterium source placed within the core of the high-flux reactor at the Institut Laue-Langevin (ILL) [28]. It increases the UCN flux by a factor of about 10^2 in relation to that available otherwise in the reactor in the thermal equilibrium spectrum. Only a fraction of the neutron flux of about 10^{-9} is thus actually used. On the other hand, the cooling of neutrons on ultracold nanoparticles could provide for further neutron cooling in a significant energy range, thereby increasing the neutron density.

The new method for producing UCN consists in the equilibrium cooling of VCN – through their many collisions with ultracold nanoparticles made from low-absorption materials (D_2, D_2O, O_2 etc.) – down to the temperature of these nanoparticles of ≈ 1 mK, during the diffusion motion of these neutrons in a macroscopically large body of nanoparticles. The principle of equilibrium cooling allows an increase in the neutron phase-space density, in contrast to the method of selecting a narrow energy range out of a warmer neutron spectrum. The use of nanoparticles provides a sufficiently large cross section for coherent interaction and an inhomogeneity of the moderator density, on a spatial scale of about the neutron wavelength; it also shifts the energy transfer range far below a value of about 10^{-3} eV, the characteristic limit for liquid and solid moderators. Many collisions are needed since the nanoparticles' mass is much larger than the neutron mass; the energy transfer to nanoparticles and nanostructures is only moderately efficient. The need for a large number of collisions limits the choice of materials: only low absorption materials are appropriate. The nanoparticles' temperature must correspond to the minimal energy to which neutrons can still be cooled using this method. The diffusion motion of neutrons in the body of nanoparticles allows us to minimize the thermalization length and, accordingly, to increase the achievable UCN density.

The cooling itself is provided by the interaction of neutrons with individual degrees of freedom of weakly bound or free nanoparticles, as well as by the excitation of collective degrees of freedom in the body of nanoparticles (e.g. vibrations and rotations), and also by the breaking of inter-particle bonds. Details about gels of nanoparticles can be found in [33, 34, 35]. Even free nanoparticles in the gel have several degrees of freedom: rotation and translation. In this paper, we provide detailed calculations of the cooling of neutrons in a gel of nanoparticles, considering only the collisions on free nanoparticles, and neglecting the rotation. The interaction between the nanoparticles, including the long-range interaction induced by helium, is neglected here. So we deal with an idealized gas of free nanoparticles at 0 K in superfluid helium. Our goal is to determine the behavior of a neutron in such a moderator. In section II, we give a model for the interaction of a neutron with a single nanoparticle in suspension in liquid helium. This model leads to a complete quantum solution for three main quantities, which are, the absorption cross section σ_a, the total scattering cross section σ_s, and at last, the mean relative energy loss per collision $\xi = \frac{\langle \Delta E \rangle}{E}$. This allows us to describe the slowing down of neutrons in a gas of free nanoparticles. In section III, we deal with the ideal situation of the infinite moderator, an infinite medium made with nanoparticles surrounded by helium at 0 K, and where the only loss of neutrons are due to the absorption by a nanoparticle (β-decay is neglected). This naive model provides necessary conditions for an efficient moderation. In section IV, we will present estimations for finite moderators, the characteristic size and the characteristic time of thermalization. We compare efficiency of the moderator with nanoparticles made of different materials, and also for different nanoparticle sizes. We first chose deuterium as the material to illustrate the calculations.

II. FREE NANOPARTICLES MODEL

Let us consider a nanoparticle with radius R, made of hundreds of nuclei, immersed into superfluid helium. A low energy incident neutron only sees the average potential of each nanoparticle. So we will assume the following phenomenological potential for the interaction of a neutron with the considered nanoparticle:

$$V(\mathbf{r}) = \begin{cases} V \equiv V_0 - iV_1 & \text{if } r < R, \\ 0 & \text{if } r > R. \end{cases} \tag{1}$$

We will provide detailed calculations of the collision parameters in the Born approximation using this model. But let us first estimate all the parameters of the potential (1) describing neutron-nanoparticle interaction.

Parameters of neutron-nanoparticle interaction potential

The potential $V_0 - iV_1$ is taken to be the averaged of each nucleus *Fermi* potential in the nanoparticle (neutron-helium interaction potential has to be subtracted):

$$V_0 - iV_1 = \sum_j \rho^{(j)} \int V^{(j)}(\mathbf{r}) d\mathbf{r} - V_{\text{He}} \tag{2}$$

TABLE I: Nuclear data used in these calculations. The experimental data are taken from [38].

Nucleus	b (fm)	σ_a (mbarn)	Mass (m)
^{1}H	-3.74	333	0.999
^{2}H	6.67	0.52	1.997
^{3}He		$5.33{\cdot}10^6$	3.968
^{4}He	3.26	0	3.968
Be	7.79	7.6	8.935
C	6.65	3.5	11.91
O	5.80	0.19	15.86

TABLE II: Nanoparticle characteristics.

Nanoparticle	Density (kg/m^3)	A_0 (nm^{-3})	N_0	V_0 (neV)	V_1 (neV)
D_2	195	488	0.001510	85	$2.2\cdot10^{-6}$
D_2O	1020	2551	0.000292	137	$2.7\cdot10^{-6}$
O_2	1230	3076	0.000242	54	$0.64\cdot10^{-6}$
CO_2	1560	3901	0.000191	85.5	$6.0\cdot10^{-6}$
C (Diamond)	3520	8803	0.000085	290	$45\cdot10^{-6}$
Be	1850	4627	0.000161	235	$68\cdot10^{-6}$

where the sum is done over the different kinds of nuclei in the nanoparticle – for instance, for a heavy water nanoparticle, we must take into account the contributions of both deuterium and oxygen nuclei. $\rho^{(j)}$ is the number of nuclei of type j per volume unit, and $V^{(j)} = V_0^{(j)} - iV_1^{(j)}$ is the interaction potential between a neutron and a j-type nucleus. The real part of this potential can be derived from the coherent scattering length $b^{(j)}$

$$b^{(j)} = \frac{m}{2\pi\hbar^2}\int V_0^{(j)} d\mathbf{r} \tag{3}$$

where m is the neutron mass. The imaginary part of the potential, which describes the possibility of the neutron capture, can be calculated from the absorption cross section via the optical theorem, taking into account the fact that this interaction is not strong and can be treated within the first Born approximation:

$$\frac{2m}{\hbar^2}\int V_1^{(j)} d\mathbf{r} = \sigma_a^{(j)}(k)k. \tag{4}$$

The experimental data are given for thermal neutrons, i.e. for a neutron velocity of 2200 m/s. So we express the result in term of $\sigma_a^{(j)}(k_0)k_0$, where k_0 is the wave vector of a thermal neutron. Eventually:

$$V_0 = 2\pi\frac{\hbar^2}{m}\sum_j \rho^{(j)} b^{(j)}, \tag{5}$$

$$V_1 = \frac{\hbar^2}{2m}\sum_j \rho^{(j)}\sigma_a^{(j)}(k_0)k_0. \tag{6}$$

¿From the nuclear data shown in table I, we can compute all properties of the potential of interaction between neutrons and nanoparticles in superfluid helium. The results are shown in table II, for various materials, taking into account the following points:

- We note A the nanoparticle mass in units of neutron mass. Its variation with the radius is determined by the parameter A_0: $A = A_0R^3$.
- The density of liquid helium is 124.9 kg m^{-3} @ BP=-268.9 C. We calculate the effective potential of the nanoparticles in liquid helium which has a potential $V_{\rm He} = 15.9$ neV.
- We calculate the density of nanoparticles (number of nanoparticles in a unit volume) $N = N_0R^{-3}$, assuming that the total mass of the nanoparticles is 1% of the total mass of the helium. This parameter is only useful for the description of a finite moderator in section IV.

Validity of the model at high energy

At high velocities neutrons can see individual nuclei, and the form of the potential we assumed is not valid. This happens when the wavelength of the neutron is smaller than the inter-atomic distances. So we use the following limit for our model:

$$\lambda_{\rm min} = \frac{2\pi}{k_{\rm max}} = d \tag{7}$$

where d is the mean inter-atomic distances, and $k_{\rm max}$ is the maximum neutron wave vector allowed by our phenomenological model. The table shows this limit for various materials.

Nanoparticle	D_2	D_2O	O_2	CO_2	C	Be
d (nm)	0.32	0.32	0.35	0.36	0.18	0.20
$k_{\rm max}$ (nm^{-1})	19	19	16	17	35	31

As we will see, this constraint is not of actual importance in the range of parameters concerned, corresponding to the optimal conditions of neutron thermalization.

There is another limit at high velocities. The neutron can excite the nanoparticles' internal degrees of freedom, so that the collision can be inelastic. We expect that the probability of phonon excitation is low because the phonon wavelenght should be in this case shorter than the considered nanoparticles' size. Also this process can only increase the efficiency of the neutron cooling.

Scattering amplitude within Born approximation

Our goal is to calculate the three relevant quantities describing the collision of a neutron with a nanoparticle: the total scattering cross section, the absorption cross

section, and the mean energy loss. Let us note that in this problem the absorption probability is extremely small with respect to the elastic scattering one. Therefore the total interaction cross section is approximately equal to the elastic one.

In the first Born approximation we can easily compute this scattering amplitude in the center-of-mass system (c.m.s.), that is, the amplitude for a neutron with incident wave vector $\mathbf{k}$ to be scattered at final wave vector $\mathbf{k}'$:

$$f(\theta) = -\frac{1}{4\pi}\frac{2m}{\hbar^2}\int e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}}V(\mathbf{r})d\mathbf{r} \tag{8}$$

θ being the angle between $\mathbf{k}$ and $\mathbf{k}'$. Let $\mathbf{q} = \mathbf{k} - \mathbf{k}'$ be the momentum transfer. The collision is elastic in the c.m.s. so that $k = k'$ and

$$q = 2k\sin(\theta/2). \tag{9}$$

Finally, we find

$$f(\theta) = -\frac{2m}{\hbar^2}\ R^3\frac{1}{(qR)}j_1(qR) \tag{10}$$

where $j_1(X)$ is the first spherical Bessel function:

$$j_1(X) = \frac{\sin(X)}{X^2} - \frac{\cos(X)}{X}. \tag{11}$$

¿From the scattering amplitude we can calculate the elastic cross section:

$$\sigma_s = \int |f|^2\, d\Omega = 2\pi\left|\frac{2m}{\hbar^2}V\right|^2\ R^6\frac{1}{(kR)^2}I(kR), \tag{12}$$

where

$$\begin{aligned} I(kR) &= \int_0^{2kR}\frac{1}{x}j_1(x)^2dx \\ &= \frac{1}{4}\left(1 - \frac{1}{(kR)^2} + \frac{\sin(2kR)}{(kR)^3} - \frac{\sin^2(kR)}{(kR)^4}\right) \end{aligned} \tag{13}$$

By using the optical theorem, we can calculate the absorption cross section σ_a at the first order Born approximation:

$$\mathrm{Im}(f(\theta=0)) = \frac{k}{4\pi}\sigma_a \tag{14}$$

As $f(0) = -\frac{1}{3}\frac{2m}{\hbar^2}VR^3$, one obtains:

$$\sigma_a = \frac{4\pi}{3}\ \frac{2m}{\hbar^2}V_1\ R^4\ \frac{1}{kR}. \tag{15}$$

Calculated elastic and absorption cross sections are presented in Fig. 1 as a function of neutron velocity. The calculations are performed for different nanoparticle's radii. For low energies where $kR \ll 1$, one can easily see from this figure as well as from (12) and (15) that $\sigma_s \sim R^6$ and $\sigma_a \sim R^3$.

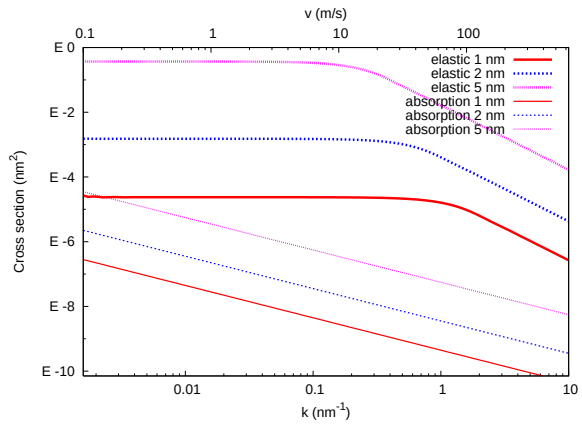

FIG. 1: Elastic and absorption cross sections as a function of neutron velocity, for three values of the deuterium nanoparticles' radii: 1, 2, and 5 nm.

Mean energy loss per collision

We consider a collision between the neutron and a nanoparticle which has a mass A, in units of the neutron mass. Let θ be the scattering angle. As the mass of the nanoparticle is much greater than the mass of the neutron, we know that the energy transfer, in the first order in $1/A$ is given by:

$$\frac{\Delta E}{E} \simeq -4\frac{1}{A}\sin(\theta/2)^2 \tag{16}$$

Since we know the scattering amplitude at the Born approximation, we can calculate the mean relative energy loss:

$$\frac{\langle\Delta E\rangle}{E} = \frac{\int\frac{\Delta E}{E}\frac{d\sigma}{d\Omega}d\Omega}{\int\frac{d\sigma}{d\Omega}d\Omega} = \frac{2\pi}{\sigma_s}\int_0^\pi\frac{\Delta E}{E}(\theta)\,|f(\theta)|^2\sin\theta d\theta \tag{17}$$

In terms of the variable $x = qR = 2kR\sin(\theta/2)$:

$$\frac{\Delta E}{E} = -\frac{1}{A}\frac{x^2}{(kR)^2}\quad\text{and}\quad f = -\frac{2m}{\hbar^2}VR^3\frac{1}{x}j_1(x) \tag{18}$$

Now we can express the mean relative energy loss:

$$\xi = \frac{\langle\Delta E\rangle}{E} = -\frac{1}{A}\frac{1}{(kR)^2}\frac{J(kR)}{I(kR)} \tag{19}$$

with

$$\frac{J(kR)}{I(kR)} = \frac{\int_0^{2kR}xj_1(x)^2dx}{\int_0^{2kR}\frac{1}{x}j_1(x)^2dx}. \tag{20}$$

The first integral in this expression can be expressed in terms of special functions but has to be calculated numerically where the second one was calculated previously in (13).

The relative mean energy loss as a function of neutron velocity is presented in Fig. 2 for different values of nanoparticles' radii. Let us note that, for small energies where $kR \ll 1$, this loss behaves as $\xi \sim R^{-3}$.

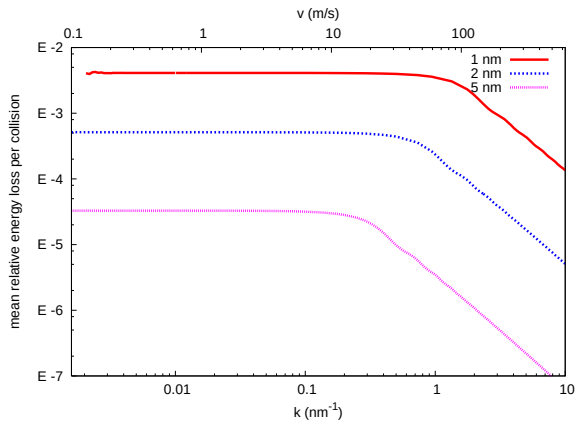

FIG. 2: Relative mean energy loss per collision ξ, as a function of neutron velocity, for three values of the deuterium nanoparticles' radii: 1, 2, and 5 nm.

Interpretation of the results

As expected, the absorption cross section follow a $1/v$ law, and is proportional to the nanoparticle's mass A_0R^3. At low velocities, the elastic cross section and the energy loss are constant up to some critical momentum $k_c \approx 1/R$ beyond which the collision becomes anisotropic. The total elastic cross section and the energy loss therefore decrease rapidly.

As we have mentioned previously, the probability of collision is strongly dependent on the size of the nanoparticle, as R^6: due to the coherent scattering of a neutron at nuclei in the nanoparticle, the scattering cross section is proportional of the square of the number of nuclei in the nanoparticle. From the condition $k_c \approx 1/R$, the critical momentum is higher for smaller nanoparticles.

The efficiency of the moderation results from competition between the absorption process and the elastic scattering. The probability of the first process is proportional to the absorption cross section σ_a multiplied by the number of collisions needed to slow down neutrons (this number is inversely proportional to the averaged energy losses ξ). The probability of elastic scattering is proportional to the elastic cross section σ_s. As we have emphasized, all these three values σ_a, ξ, σ_s are strongly R dependent but their combination $\sigma_a/\xi\sigma_s$ which defines the moderator efficiency is not. This combination depends only on the product kR and has its minimum for $kR \sim 1$ (as it can be seen from Fig. 1 and Fig. 2). Therefore it is natural to expect the best moderator properties for neutron velocities close to the critical one $k_c \sim 1/R$. This ratio decreases for both higher and lower velocities. We will see that a more detailed analysis in section III will confirm this general statement.

Validity of the first Born approximation

Sufficient condition for the validity of the first Born approximation is:

$$\frac{2m}{\hbar^2}V_0R^2 \ll 1 \tag{21}$$

The lower the radius, the better the approximation. To see where this condition is verified, let us calculate the maximum radius R_{Born} to have an accuracy better than 10%: $\frac{2m}{\hbar^2}V_0R^2_{\text{Born}} = 0.1$.

Nanoparticle	D_2	D_2O	O_2	CO_2	C	Be
R_{Born} (nm)	4.5	3.7	5.4	4.5	2.6	2.9

Actually, the approximation works much better, because our potential is repulsive [39]. A more accurate calculation, with partial waves given in Appendix A shows that the accuracy of the Born approximation is sufficient enough for our needs.

III. MODEL OF THE INFINITE MODERATOR

The role of any moderator is to increase the density of neutrons in the phase space. In this section, we will determine the influence of the nanoparticles' properties – their material size – on the evolution of neutron density. In order to achieve that, we have to identify the criteria for cooling efficiency.

The most general problem that we perceive is the following. We have a moderator medium made of free nanoparticles in suspension in superfluid helium. There is a neutron source, at a certain location, with a certain velocity distribution. What is the neutron density at every point in the moderator medium and what is the distribution of velocities? The quantity describing the state of the system in this problem is a seven-variable function, the density of the neutrons in the phase space $n(\mathbf{r}, \mathbf{k}, t)$. In general case, this density evolves according to the transport equation, or Boltzmann equation, and all the variables can be coupled.

Solution of this equation in general case can be quite complicated task so we will propose a first necessary criterium of cooling efficiency within the model of infinite moderator. In this model, we eliminate the space variable in the density; we assume that the sources are uniformly distributed, and that the medium is homogeneous and isotropic. The quantity which describes the system is only a function of the energy (and also of time, but it is only the stationary regime that interests us). We can analytically compute the energy spectrum of the neutrons in this moderator, using the three quantities σ_s, σ_a, $\xi = \frac{\langle \Delta E \rangle}{E}$ describing the elementary processes.

The energy spectrum in the infinite moderator

To compute the neutron energy spectrum in the moderator $n(E,t)$, we have to use the equation for the conservation of the number of neutrons:

$$\frac{\partial n}{\partial t} - \frac{\partial q}{\partial E} + \text{Absorption} - \text{Source} = 0 \tag{22}$$

where $q(E,t)$ is the cooling current, i.e. the number of neutrons scattered from an energy greater than E to an energy lower than E, in the unit of time. The absorption term can be related to the macroscopic absorption cross section $\Sigma_a = N\sigma_a$ via the flux variable $\phi(E,t) = vn(E,t) = \sqrt{\frac{2E}{m}}n(E,t)$, so that the conservation equation, using the flux variable, can be expressed as:

$$\sqrt{\frac{m}{2E}}\frac{\partial \phi}{\partial t} - \frac{\partial q}{\partial E} + \Sigma_a \phi = \text{Source} \tag{23}$$

Now we have to specify the cooling current under the assumption that $\xi \ll 1$ which is clear to be a very good approximation for neutron scattering on nanoparticles (see Fig. 2).

Under this assumption the cooling current has simple and natural form [40]:

$$q(E) = E\xi(E)\Sigma_s(E)\phi(E). \tag{24}$$

Let us consider stationary regime. The conservation equation is completely expressed in terms of our three microscopic quantities, and so the time independent equation is given by

$$-\frac{\mathrm{d}}{\mathrm{d}E}\left[\xi\Sigma_s E\phi\right] + \Sigma_a\phi = 0 \tag{25}$$

We have not expressed the source term, because if the source is punctual (a given flux Φ_0 at a given energy E_0) then we can solve the equation without any source in the domain $0 < E < E_0$ and the source is a boundary condition: $\phi(E_0) = \phi_0$. So now, as we have a usual first order linear differential equation, we can solve it analytically with the previous boundary condition:

$$\phi(E) = \phi_0 \frac{E_0\xi(E_0)\Sigma_s(E_0)}{E\xi(E)\Sigma_s(E)} \exp\left(-\int_E^{E_0} \frac{\Sigma_a}{\xi\Sigma_s}\frac{d\epsilon}{\epsilon}\right) \tag{26}$$

We can notice that, if there is no absorption, and if both ξ and Σ_s are energy independent, we find the usual behavior, the so called the $1/E$ flux law.

¿From the flux, it is useful to derive the density $n(k)$ of neutrons in the phase space:

$$n(k) = n(k_0)\left(\frac{k_0}{k}\right)^2 \frac{(\xi\Sigma_s)(k_0)}{(\xi\Sigma_s)(k)} \exp\left(-2\int_k^{k_0} \frac{\Sigma_a}{\xi\Sigma_s}\frac{d\kappa}{\kappa}\right) \tag{27}$$

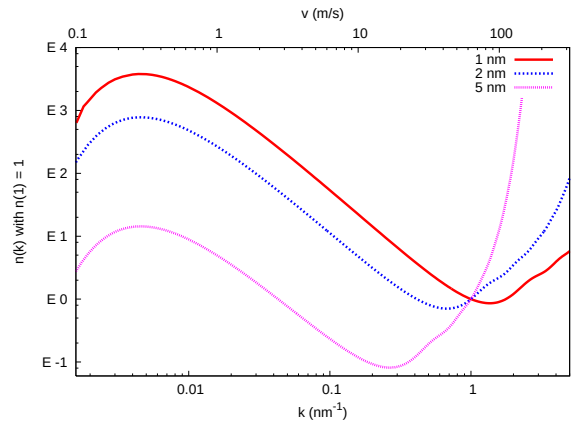

FIG. 3: Phase space density in stationary regime as a function of velocity, for three radii of deuterium nanoparticles. The source is assumed to be monochromatic, with initial energy equal/greater than the maximum energy presented in the graph.

A necessary condition for efficient cooling

The density of neutrons in the velocity space, for the stationary regime in the infinite moderator, is presented in Fig. 3.

To interpret this figure, we have to assume that there is a monochromatic neutron source at velocity k_0, and we extract the neutrons at the velocity $k_1 < k_0$. Then the relative increase of density in the phase space is given by $n(k_1)/n(k_0)$. As the goal is to increase the density of neutrons in the phase space, we see that the curve gives us a necessary condition for the efficiency of the cooling. Indeed, the cooling is efficient only if $n(k_1) > n(k_0)$, that is in the decreasing part of the curve. This differential necessary condition is then given by $\frac{\mathrm{d}\ln n}{\mathrm{d}k}(k_0) < 0$, that we can put into the form:

$$\frac{\Sigma_a}{\xi\Sigma_s} < 1 + \frac{1}{2}\frac{\mathrm{d}\ln(\xi\Sigma_s)}{\mathrm{d}\ln k} \tag{28}$$

The cooling domain is the velocity domain in which this condition is satisfied. Figure 4 shows that the cooling is not efficient both at low velocities and at high velocities.

At low velocities, the cooling is not efficient because the absorption is important compared to the diffusion cross section and the energy loss, and at high velocities (basically at velocities higher than the critical velocity), the cooling is not efficient because both the scattering cross section and the energy loss fall down. Figure 4 shows the cooling domain as a function of the radius of the nanoparticles, for several materials. These curves leads to three remarks. For certain materials, such as diamond, and for certain radius of nanoparticles, there is no cooling domain at all. The cooling domain is bigger for smaller nanoparticles, because for smaller nanoparticles, the critical velocity is higher. The domain for deuterium nanoparticles include the domain for all the other

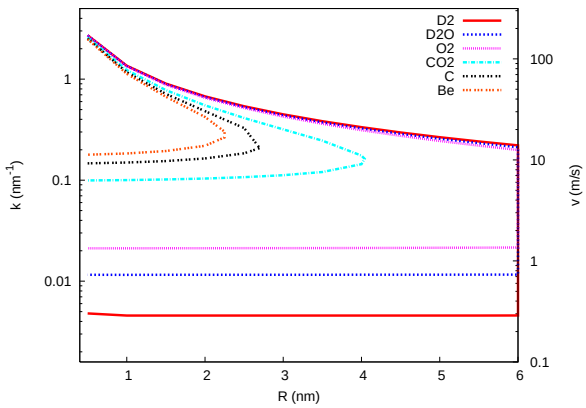

FIG. 4: Efficiency domain of the moderation as a function of nanoparticle's radii, for different materials. Interference effects are not taken into account.

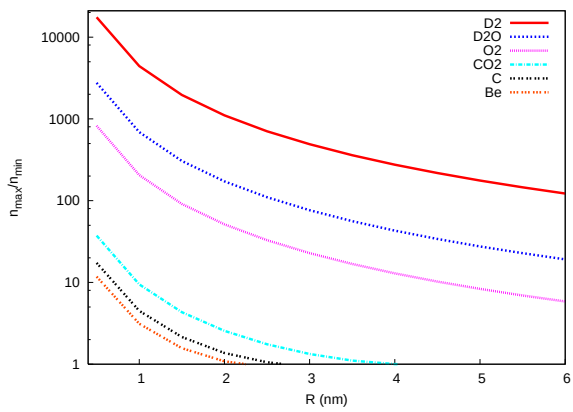

FIG. 5: Maximum compression of phase space density possible, as a function of the nanoparticles' radius, and for different materials. Interference effects are not taken into account.

materials.

We can now put a limit for the total increase of density in the phase space achievable for the infinite moderator and for a monochromatic source. Indeed, in Fig. 3 we see that the best we can do is to have a source with k_0 at the minimum of the curve, and to extract neutrons with k_1 at the maximum of the curve. The maximum gain is given by $n(k_1)/n(k_0)$. Figure 5 shows this limit, as a function of the radius of the nanoparticles, and for different materials. We see that this maximum is bigger for smaller nanoparticles, because the cooling domain is higher. The best material for this criterium is deuterium.

IV. MORE REALISTIC MODERATORS

The consideration of the infinite moderator gives us a necessary condition for the efficiency of the cooling. The results presented do not depend on the density of nanoparticles in the moderator. When it comes to realistic moderators, we have to take two characteristics of the moderation into account: the size of the moderator, and the thermalization time. These two quantities depend on the density of nanoparticles, and for the calculations we assumed that in a given volume, the nanoparticles' total mass represents 1 % of the total mass of helium. That means that for a gel of smaller nanoparticles, the distance between the nanoparticles is assumed to be smaller.

Let us pursue the idea to estimating the size of the moderator and the thermalization time. Suppose a monochromatic neutron source at velocity k_0 inside the efficient domain, and that we want to increase the density in phase space by the factor e. According to Fig. 3, we have to extract neutrons in the moderator with an energy $k_0 - \Delta k$, where Δk corresponds to an increase of factor e for the function $n(k)$:

$$\Delta k = \left[\frac{\mathrm{d}\ln n(k)}{\mathrm{d}k}(k_0)\right]^{-1}. \tag{29}$$

We know that this decrease of velocity by Δk corresponds to a certain number of collisions ΔN:

$$\Delta N = \frac{1}{\xi}\frac{\Delta k}{k_0} \tag{30}$$

During ΔN collisions, neutron assume a Brownian-like trajectory, and we define $L(k_0)$ as the square-mean-root of the distance travelled. We also define the thermalization time $\tau(k_0)$ as the time it takes for the neutron to collide ΔN times.

Estimation of the moderator's size

The square mean distance travelled by a neutron Δr^2 after ΔN collisions is given by [37]:

$$\Delta r^2 = \frac{4}{A}\frac{1}{\xi\Sigma_s^2}\Delta N \tag{31}$$

This expression was obtained under the assumption that the diffusion cross section Σ_s and the mean relative energy loss ξ are energy-independent, and that the scattering is nearly isotropic.

¿From equations 29 and 30, we can estimate the characteristic length $L(k)$ of the moderator, corresponding to an increase of factor e of the phase space density:

$$\begin{aligned}\frac{1}{L(k)^2} &= \frac{\mathrm{d}\ln(n)}{\mathrm{d}r^2} = \frac{A}{4}(\xi\Sigma_s)^2\frac{\mathrm{d}\ln(n)}{\mathrm{d}\ln(k)} \\ &= \frac{A}{2}(\xi\Sigma_s)^2\left[1+\frac{1}{2}\frac{\mathrm{d}\ln(\xi\Sigma_s)}{\mathrm{d}\ln(k)}-\frac{\Sigma_a}{\xi\Sigma_s}\right]\end{aligned} \tag{32}$$

Note that we find our necessary condition 28, in the sense that the condition is satisfied if and only if the size L is real.

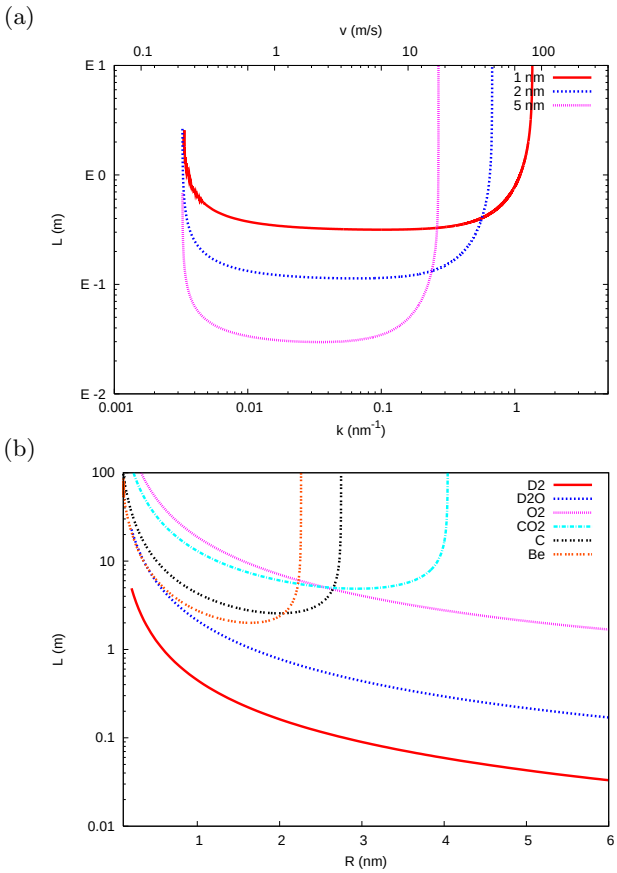

FIG. 6: Moderator size needed to increase the density in phase space by a factor e. Interference effects are not taken into account. (a) Moderator size as a function of velocity, for three radii of deuterium nanoparticles. (b) The minimum moderator size as a function of the nanoparticles radius, for different materials.

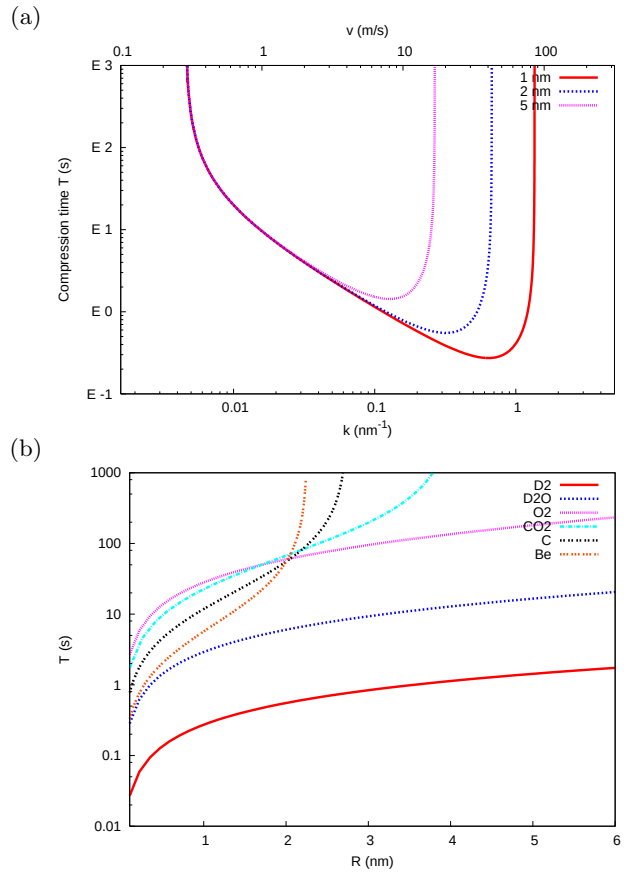

FIG. 7: Thermalization time needed to increase the density in phase space by a factor e. Interference effects are not taken into account. (a) Thermalization time as a function of velocity, for three radii of deuterium nanoparticles. (b) The minimum thermalization time as a function of the nanoparticles radius, for different materials.

Estimation of the thermalization time

Now we must estimate of the thermalization time. We know that the mean time between two collisions is given by:

$$\tau_{\text{coll}} = \frac{m}{\hbar}\frac{1}{k\Sigma_s}. \tag{33}$$

The time needed for a neutron to collide ΔN times is therefore simply $\tau_{\text{coll}}\Delta N$. From equations 29 and 30, we can estimate the thermalization time $\tau(k)$, corresponding to an increase by a factor e of the phase space density:

$$\begin{aligned}\frac{1}{\tau(k)} &= \frac{\hbar}{2m}k^2\xi\Sigma_s\frac{\mathrm{d}\ln(n)}{\mathrm{d}k} \\ &= \frac{\hbar k}{2m}\xi\Sigma_s\left[1+\frac{1}{2}\frac{\mathrm{d}\ln(\xi\Sigma_s)}{\mathrm{d}\ln(k)}-\frac{\Sigma_a}{\xi\Sigma_s}\right]\end{aligned} \tag{34}$$

Again, we find our necessary condition 28, in the sense that the condition is satisfied if and only if the characteristic time τ is positive.

Comments on the results

The results are shown in Fig. 6 and Fig. 7. Let us first comment on the figures for the deuterium example. We see first that outside of the efficiency domain of the cooling, both the thermalization time and the moderator size diverge. We also remark that the moderator size is nearly constant over the efficiency domain, although the thermalization time presents a clear minimum near the critical velocity. But the main difference between the two conditions is the dependence on the radius of the nanoparticles. For small nanoparticles, we need fewer collisions to cool down the neutrons, and we can see in Fig. 7 that the smaller the nanoparticles, the smaller the thermalization time. On the other hand, for small nanoparticles, the mean free path is high, and we see in Fig. 6 that the smaller the nanoparticles, the bigger the moderator size. We cdannot therefore optimize the radius of the nanoparticle in a general way; we must look at more practical considerations.

We can use both the size of the moderator and the

thermalization time as criteria to compare materials. We can see that using both criteria, deuterium is better than any other material by almost one order of magnitude. We also see that the absolute values are very competitive. Indeed, to increase the density in phase space by factor e, the characteristic size is a few tens of centimeters and the thermalization time is less than one second, for a typical radius of 2 nm.

Practical proposal

To achieve a compression in the phase space by many orders of magnitude, we can associate the nanoparticle's moderator to a compression in real space. To explain this idea in more details, Fig. 8 may be of help. We start with a gas of neutrons uniformly located in the initial volume V_0 with a velocity distribution with a mean value v_0 and a width Δv. We decrease the volume by a factor V/V_0. According to the Liouville theorem, the width of the velocity distribution will increase as $\Delta v \longrightarrow (\frac{V_0}{V})^{1/3}\Delta v$, so that the density in the velocity space decreases by the same factor (in absence of absorption this relation would be precise, it is approximately valid in the domain of efficient cooling of neutrons defined above). This is the case when there is no moderator, but if we put a nanoparticle's moderator in the volume (not in all the volume, but in a little box in a corner), the moderation will compensate the increase of density in the velocity space. So finally the maximum density in the phase space achievable is the product of the initial density, the volume compression V_0/V, and the maximal compression due to the moderator shown in Fig. 5. As we can in principle reach as much volume compression as we wish, there is no theoretical limitation of the maximum compression in phase space. With this method, it is not necessary to use all the cooling domain shown in Fig. 4 (two orders of magnitude of velocity in a favorable case); we can concentrate the process around the optimal velocity. This optimal velocity should be chosen according to three practical considerations:

1. We are limited by the size of the moderator, which should not exceed a few tens of centimeters for practical purposes.

2. The thermalization time is of principle importance. If the volume decreases by a factor e, we have to wait the time T shown in Fig. 7.

3. The optimal velocity should correspond to a temperature greater than that achievable with a dilution cryostat, which is about 10 mK. The corresponding wave vector is about 0.2 nm^{-1}.

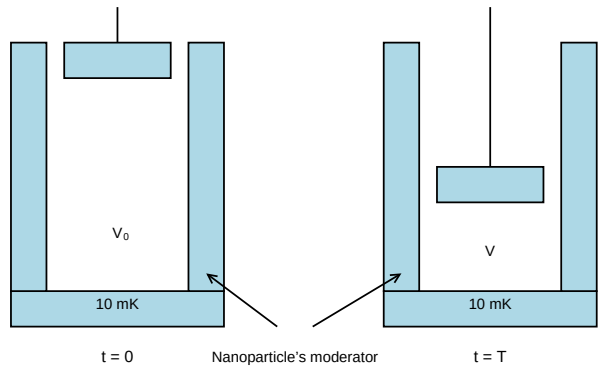

FIG. 8: Practical proposal to increase the neutron density in phase space, combining a compression in the real space with a piston as well as a compression in the velocity space with the nanoparticle's moderator. This picture emphasize the importance of the thermalization time.

V. EXTENSIONS OF THE MODEL

For this special case of moderator nanoparticle gels there are several limitations to the validity of our model. One obvious limitation is that chains of nanoparticles are not taken into account; in this paper we neglect such chains and consider only the cooling of neutrons caused by the collisions whith free nanoparticles. We have already mentionned the limitation at high energies in section II, a limitation is inherent to our assumptions about the interaction of a neutron and a single nanoparticle. But there is a limitation also at low velocities, which is specific to the gel.

Limitation at low energy

At low velocities, we cannot neglect the interferences between the waves diffracted by several nanoparticles. This happens when the wavelength of the neutron is bigger than the distance between the nanoparticles. We see that this limitation does not come from our assumptions about the interaction of a neutron with a single nanoparticle. This limitation is specific to the practical medium that we plan to use, that is, the gel of nanoparticles. Let D be the distance between the nanoparticles in the gel; we can compute D, assuming that the total mass of nanoparticles is 1% of the total mass of helium: $D = R/N_0^{1/3}$. In the following table we give D for nanoparticles with radius 1 nm. We also give the minimal wave vector for the validity of the model $k_{\min}$, defined as follow: $\lambda_{\max} = \frac{2\pi}{k_{\min}} = D$.

Nanoparticle	D_2	D_2O	O_2	CO_2	C	Be
$D_{1\ \mathrm{nm}}$ (nm)	8.7	15.1	16.0	17.4	22.7	18.4
$k_{\min}$ (nm^{-1})	0.7	0.4	0.4	0.4	0.3	0.4

We can see that this is a serious limitation, but not that dramatic. Actually, for the practical purposes, the

lowest energy we are interested in is that corresponding to the lowest temperature achievable with a dilution cryostat, which is about 10 mK. The corresponding wave vector is about 0.2 nm^{-1}. So we only need to know , for practical purposes, the first correction of σ_s, σ_a and ξ due to the interference of neutron waves on the neighboring nanoparticles. And this can be done, because the first correction can be estimated considering only two nanoparticles. This approach is valid until the neutron wavelength covers three or more nanoparticles, that means that it is valid until $k_{\min}/2$.

Let $\mathbf{D}$ be the vector distance between two nanoparticles. At the first order of Born approximation – neglecting multiple diffusions – the scattering amplitude for a neutron colliding on this system of two nanoparticles is given by:

$$f(\mathbf{D},\mathbf{q}) = \left(1 + \mathrm{e}^{i\mathbf{q}\cdot\mathbf{D}}\right) f(\mathbf{q}) \tag{35}$$

where $\mathbf{q}$ is the momentum transfer and $f(\mathbf{q})$ is the scattering amplitude for the collision on a single nanoparticle. ¿From that result, we can conclude that the amplitude for the forward scattering – $\mathbf{q} = 0$ – is simply twice the amplitude calculated for a single nanoparticle. We can conclude that the absorption cross section is not affected by the interferences. Now, to calculate the effects of interferences on the scattering cross section and the energy loss, the physical relevant quantity is the average differential scattering cross section, averaging on all possible directions for $\mathbf{D}$:

$$\begin{aligned}\left\langle \frac{\mathrm{d}\sigma_s}{\mathrm{d}\Omega}(\mathbf{q}) \right\rangle &= \int |f(\mathbf{D},\mathbf{q})|^2 \frac{d\mathbf{D}/D}{4\pi} \\ &= \int \left|1 + \mathrm{e}^{i\mathbf{q}\cdot\mathbf{D}}\right|^2 \frac{d\mathbf{D}/D}{4\pi} |f(\mathbf{q})|^2 \\ &= 2\left(1 + \frac{\sin(qD)}{qD}\right) |f(\mathbf{q})|^2 \end{aligned} \tag{36}$$

Is is now possible to calculate σ_s and ξ using this new effective differential cross section. Figure 9 shows our plot of the relevant quantity, $\xi\sigma_s$, we can call the energy loss cross section, and the one calculated with the estimated correction. One can see that the first order correction is not important.

Purity of Deuterium and Helium

In all our calculations, we assumed that the helium was purely ^{4}He, with no absorption, therefore, by the Helium medium. We also assumed that the deuterium in the nanoparticles was pure. Let us estimate the purity we actually need.

First, consider the presence of hydrogen – which is an efficient neutron absorber – inside the deuterium. We can compute the purity needed so that the absorption differs from the ideal case by less than 10 %. If x is the proportion of hydrogen relatively to deuterium, then the

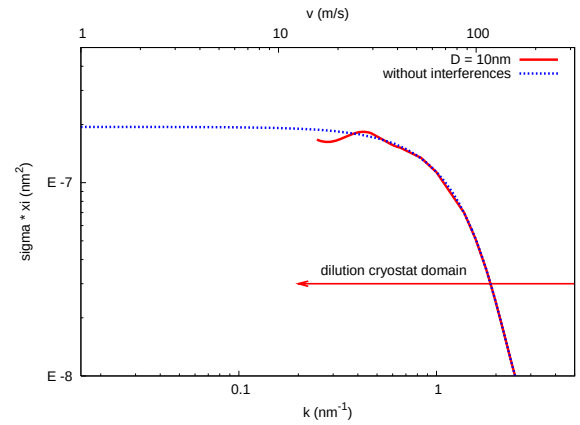

FIG. 9: $\xi\sigma_s$ is plotted as a function of velocity, for a deuterium nanoparticle of radius 1 nm. The dot line is calculated without interference effects, and the continuous line take into account the first correction due to interference effects.

absorption cross section is:

$$\sigma_a(x) = (1-x)\sigma_a(x=0) + x\sigma_a(x=1). \tag{37}$$

Using the table I we can find that the requirement of less than 10 % increase is satisfied if $x = 1.5 \cdot 10^{-4}$. We can easily achieve this purity.

Let us now consider the presence of ^{3}He. We can estimate the lifetime τ_a of a neutron due to the absorption by ^{3}He. If x is the proportion of ^{3}He, σ_a is the absorption cross section for thermal neutrons (with velocity $v_0 = 2200$ m/s), and N_{He} is the density of helium nuclei, then:

$$\tau_a = \frac{1}{x}\frac{1}{N_{He}\sigma_a v_0} \tag{38}$$

If we have a purity $x = 0.5 \cdot 10^{-14}$, the absorption lifetime τ_a is ten times the intrinsic neutron lifetime. The natural abundance of ^3He is $1.4 \cdot 10^{-6}$, and the purity we need is achievable.

VI. CONCLUSIONS

A new concept for producing high UCN density is analyzed within the framework of the free nanoparticles model. This concept is based on neutron cooling using ultracold nanoparticles of deuterium, heavy water, etc. We have shown that increase in the phase space density of neutrons, within the model of free nanoparticles, is possible, given certain parameters of nanoparticles and neutron velocity. Thus, solid deuterium, which is shown to be the best material, provides efficient cooling of neutrons in the range 1 - 10^2 m/s, in an infinite medium of free nanoparticles of radius 1 - 2 nm, sufficiently spatially separated. The characteristic cooling time is much shorter than the corresponding absorption time, or the neutron β-decay lifetime for optimum parameters of neutrons and nanoparticles. The moderator size of, at most,

a few times 10 cm allows in principle the realization of the cooling mechanism presented.

We examine the different constraints on the model, such as scattering at individual nuclei (for too short wavelengths), excitation of internal degrees of freedom in a nanoparticle (such as phonons), extensions of the Born approximation description used (the partial waves expansion), neutron optical effects due to diffraction at several nanoparticles simultaneously, the purity of deuterium or helium. We show that these constraints do not change our main conclusions. We do not consider in this present article such effects as rotation of the nanoparticles, interaction between nanoparticles, in particular excitation of the collective degrees of freedom for nanoparticles in gels or any breaking of the inter-nanoparticles bounds. We do not consider neither the influence of the non-zero temperature of the nanoparticles on the cooling process. These phenomena are expected to be analyzed in further publications.

APPENDIX A: PARTIAL WAVE EXPANSION

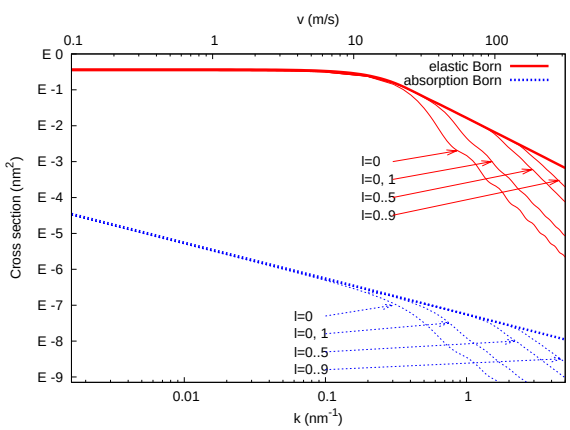

FIG. 10: Elastic and absorption cross-sections as a function of neutron velocity, for a deuterium nanoparticle of radius 5 nm. The two bold lines are calculated using the first Born approximation, and the thin lines are calculated using the partial wave method, till the order 0, 1, 5 and 9.

Since the potential interaction between a neutron and a nanoparticle is approximated by spherical rectangular barrier, it is possible to give exact solution for the scattering amplitude, using the partial wave expansion.

The analytical solution for the scattering amplitude η_l for lth partial wave is known [39] to be equal to

$$\eta_l = \frac{\gamma_l \; h_l^-(kR) - k \; h_l^{-'}(kR))}{k \; h_l^{+'}(kR) - \gamma_l \; h_l^+(kR)} \tag{A1}$$

where $h_l^{\pm}$ are spherical Hankel functions and

$$\gamma_l = K \frac{j_l'(KR)}{j_l(KR)} \tag{A2}$$

where j_l is spherical Bessel function and $K^2 = k^2 - 2m(V_0 + iV_1)/\hbar^2$.

The elastic and absorption cross sections are then given by

$$\sigma_s = \frac{\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \left|1 - \eta_l\right|^2, \tag{A3}$$

$$\sigma_a = \frac{\pi}{k^2} \sum_{l=0}^{\infty} (2l+1)(1 - |\eta_l|^2). \tag{A4}$$

The result is shown in Fig. 10, for a deuterium nanoparticle of radius 5 nm. We first estimated this size to be the limit of application of the Born approximation. The figure shows that the Born approximation is actually precise enough. The width of the bold line in the figure corresponds to a relative error of 5%, and there is no deviation between the born approximation result and the partial wave result at this precision level.

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