Physics of Atomic Nuclei, Vol. 65, No. 3, 2002, pp. 400–408. Translated from Yadernaya Fizika, Vol. 65, No. 3, 2002, pp. 426–434.

NUCLEI
Theory

Interaction of Neutrons with Nanoparticles

V. V. Nesvizhevsky*

Institute Laue-Langevin, Grenoble, France

Received February 13, 2001; in final form, October 5, 2001

Abstract—Two hypothesizes concerning interaction of neutrons with nanoparticles and having applications in the physics of ultracold neutrons (UCN) are considered. In 1997, it was found that, upon reflection from sample surface or spectrometer walls, UCN change their energy by about 10^{-7} eV with a probability of 10^{-7}–10^{-5} per collision. The nature of this phenomenon is not clear at present. Probably, it is due to the inelastic coherent scattering of UCN on nanoparticles or nanostructures weakly attached at surface, in a state of Brownian thermal motion. An analysis of experimental data on the basis of this model allows one to estimate the mass of such nanoparticles and nanostructures at 10^7 a.u. The proposed hypothesis indicates a method for studying the dynamics of nanoparticles and nanostructures and, accordingly, their interactions with the surface or with one another, this method being selective in their sizes. In all experiments with UCN, the trap-wall temperature was much higher than a temperature of about 1 mK, which corresponds to the UCN energy. Therefore, UCN increased their energy. The surface density of weakly attached nanoparticles was low. If, however, the nanoparticles temperature is lower than the neutron temperature and if the nanoparticles density is high, the problem of interaction of neutrons with nanoparticles is inverted. In this case, the neutrons of initial velocity below 10^2 m/s can cool down, under certain conditions, owing to their scattering on ultracold-heavy-water, deuterium, and oxygen nanoparticles to their temperature of about 1 mK, with the result that the UCN density increases by many orders of magnitude. © *2002 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

A long storage of ultracold neutrons (UCN, $V_{\mathrm{UCN}} \sim 5$ m/s, $E_{\mathrm{UCN}} \sim 10^{-7}$ eV) in traps is useful in fundamental-physics experiments. This application of UCN motivates continuing attempts at eliminating their extra losses in the trap wall. Let us recall how the interaction of UCN with surface is described: UCN are reflected from a uniform potential barrier, which is formed upon averaging strong neutron–nucleon interaction, with a critical energy E_{lim} and the corresponding critical velocity V_{lim}. One observes the total reflection if $V_{\mathrm{UCN}} < V_{\mathrm{lim}}$, but UCN can be lost with a small probability because of their absorption by nuclei in the trap walls or because of their upscattering in the trap walls to the energy region around kT, where k is the Boltzmann constant and T is the trap-wall temperature. UCN may penetrate inside if $V_{\mathrm{UCN}} > V_{\mathrm{lim}}$. If the surface is immobile in the laboratory frame, collisions are elastic. Otherwise, UCN may change their energy.

An additional kind of the UCN escape mechanism from gravitational spectrometers [1] due to their scattering on surfaces of beryllium, copper, stainless steel, and liquid fomblin oil was found and investigated in 1997. The phenomenon consists in a small increase in UCN energy owing to their interaction with surface. By analogy with the usual evaporation process, such events will be referred to as the formation of VUCN (Vaporizing UCN). The measured characteristic probability of VUCN generation was $P_{\mathrm{VUCN}} \sim 10^{-7}$–$10^{-5}$ per collision. If the resulting neutron energy is higher than the wall potential barrier, then the neutron can penetrate into the wall material, where it will be absorbed or upscattered; if the wall is sufficiently thin, then it may penetrate through it, as this was measured in [2]. A permanent generation of VUCN presents a spectrum shaping such that all above-barrier neutrons are removed. The new escape channel, described below in detail, is in line with the general hypothesis proposed in [3] to explain anomalous UCN losses [4].

At the liquid-fomblin-oil surface, where the probability P_{VUCN} has the highest value among all measured materials, this process was independently found and studied in [5] and in other publications of this group. Other research groups are expected to publish later their papers, which also confirm the existence of small changes in UCN energy in fomblin-oil-coated traps. It is straightforward to assume that the phenomenon of a small decrease in the UCN energy—it found in [6] at liquid-fomblin-oil surface and in [1] at solid-stainless-steel surface—has the same nature as

*e-mail: nesvizhevsky@ill.fr

1063-7788/02/6503-0400$22.00

the aforementioned phenomenon of a small increase in the UCN energy.

Thus, UCN were expected to be upscattered on surfaces to energies in the region around kT (that is, about 10^{-2} eV at room temperature or at liquid nitrogen temperature). Such upscattering was actually measured in [7, 8] and in other experiments. The total probability of this process is usually 10^{-5}–10^{-4} per collision with the surface. The "tail" of such an energy distribution at energies of about 10^{-7} eV has to be negligible. In all models, the probability of such a scattering process is much lower than 10^{-10}. However the probability of upscattering to this energy region appeared to be surprisingly high ($P_{\mathrm{VUCN}} \sim 10^{-7}$–$10^{-5}$), commensurate with the probability of normal upscattering to the energies around kT. The nature of this phenomenon has to be clarified. Moreover, a simultaneous investigation of VUCN generation and UCN anomalous losses is of interest, since these two phenomena have similar experimental manifestations.

These small changes in the energy of UCN, which occur in their interaction with trap walls, are probably due to the thermal motion of nuclei forming the wall potential. However, the mechanism that is responsible for the transformation of this thermal motion into the spreading of UCN spectrum is not clear. The point is that the interaction of neutrons with nuclei is a quantum process, and the corresponding potential results from the interaction of neutrons with a large number of nuclei even when these nuclei are at rest. When the nuclei move, we have a quantum problem of many interacting bodies, and even methods for solving such a problem are still under discussion.

The reflection of UCN from a flat surface was considered in a series of theoretical studies of Barabanov and Belyaev [9]. The changes in the energy of UCN in their quasielastic reflections from the surface corresponded to values measured in experiments. However the probability of VUCN generation due to the reflection of UCN from a flat surface was estimated to be much lower than that measured in experiments.

In Section 2, we consider the hypothesis that VUCN are generated owing to the inelastic coherent scattering of UCN on nanoparticles or nanostructures weakly attached at the surface in a state of thermal motion. In this case, the problem is much simpler because of the assumption that the interaction of UCN with surface nanoparticles involved in the thermal motion is the dominant mechanism of transformation of the thermal motion of atomic nuclei in walls into small changes in the UCN energy. As a result, the problem becomes equivalent to the well-known quantum-mechanical problem of two bodies—that is, to the problem of a neutron–nanoparticle collision that is elastic in the c.m. frame of the colliding particles. General ideas of additional UCN losses due to small particles at the surface were formulated earlier, for instance, in [10, 11]. In [10], clusters of molecules were considered. In [11], it was indicated that UCN could aid in identifying a new particle with a long-range potential and a weak coupling to surface. In any case, the nanoparticles temperature is obviously equal to the trap temperature T in the range 10^1–10^3 K. The UCN energy corresponds to $T_{\mathrm{UCN}} \sim 1$ mK. Ultracold neutrons increase preferentially 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 small.

However, the mathematical problem of neutron–nanoparticle interaction can in principle be inverted: the interaction of "warm" neutrons with ultracold nanoparticles of temperature about 1 mK can cool down the neutrons. If the density of weakly attached nanoparticles is high (not only do these nanoparticles cover the surface, but they also fill the volume) and if, during cooling of neutrons, the probability of their absorption and β decay is low, then the neutron density increases. This process can allow, for the first time, the equilibrium cooling of neutrons down to the UCN temperature. It is analyzed in Section 3.

In order to produce UCN, one first uses nuclear fission in nuclear reactors, which releases neutrons of energy about 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 just owing to a few tens of their collisions with nuclei in reactor moderators (hydrogen, deuterium). The energy transfer is very efficient, and the neutron losses during the cooling process are low because the mass of moderator nuclei is equal to (or about) the neutron mass. However, a further cooling does not occur: the lower the neutron energy, the larger is the neutron wavelength. When it becomes commensurate with the distances between the nuclei of the moderator, the neutrons do not "see" individual nuclei any longer—they are just affected by average optical potential of the medium. The neutron energy becomes lower than the bounding energy of 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, it allows the cooling of the neutrons to the energy range of cold neutrons (about 10^{-3} eV). But this is insufficient for the cooling of the main portion of the neutrons to the UCN energy region [12–15]. The idea of neutron cooling on ultracold nanoparticles as proposed in Section 3 in this article consists in reproducing

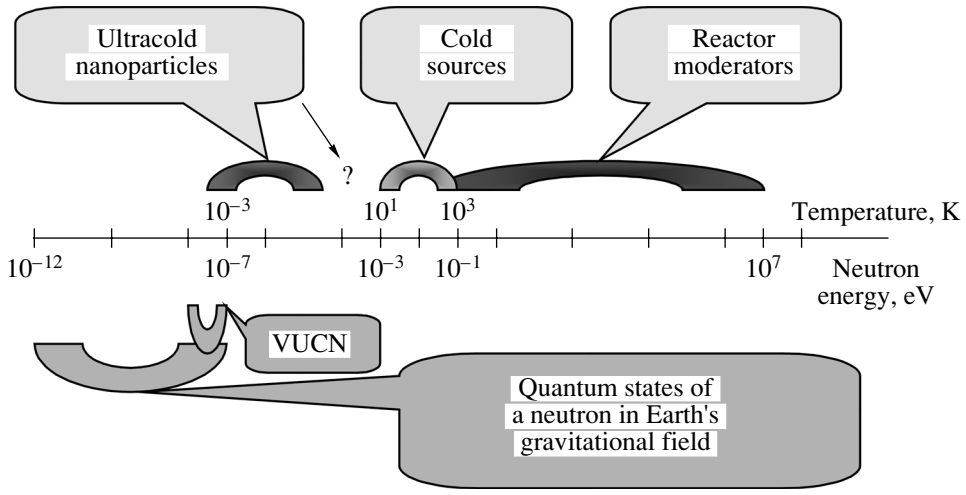

Fig. 1. Neutron energy and temperature ranges that correspond to various moderators, along with a few examples of physical phenomena involving UCN and even slower neutrons.

the principle of neutron cooling in reactor moderators via multiple collisions. However, the scale is different: the sizes are greater by a factor of about 10^2, which increases the energy range of application of this mechanism by a few orders of magnitude. The energy and the temperature scales, which correspond to the mechanisms being considered, are shown in Fig. 1. It should be noted that such a UCN source is based on the principle of UCN density accumulation as in a superthermal source [13] but not on the use of UCN flux from a source in the flow-through mode.

In conventional sources used to select UCN, thermal equilibrium is not achieved. Sources are much hotter than UCN. Only a very small portion of the neutrons is used—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 is now produced in a liquid-deuterium source placed within the core of the high-flux reactor at the Institute Laue–Langevin (ILL) [12]. It increases the UCN flux by a factor of about 10^2 in relation to that available anyway in a reactor in the thermal equilibrium spectrum. Only a fraction of the neutron flux of about 10^{-9} is actually used then. On the other hand, the cooling of neutrons on ultracold nanoparticles could provide a further neutron cooling in a significant energy range, thereby increasing the neutron density available for experiments.

2. ARE VUCN PRODUCED ON SURFACE NANOPARTICLES?

2.1. Description of the Model

Let us assume that inelastic coherent UCN scattering on nanoparticles or nanostructures that are weakly attached at the surface and which are permanently in a state of random thermal motion is the dominant mechanism through which the energy of UCN undergoes small changes in their interaction with the surface. In contrast to the general quantum-mechanical problem of UCN reflection from many moving nuclei, this model allows a radical simplification, which reduces the general problem to that of a UCN collision with a moving nanoparticle, this collision being elastic in the c.m. frame of the two particles. In contrast to the general problem, the proposed model does not obviously describe any features of UCN interaction with the surface other than the aforementioned mechanism of small changes in the UCN energy.

Within the proposed model, no assumptions about the size distribution for such objects at the surface is needed for explaining experimental results. The nature of the interaction itself provides a selection of the particle size d that is important in experiments and which corresponds to the neutron wavelength $\lambda\!\!\!^{-}_n$, which is related to the neutron velocity V_n as

$$\lambda\!\!\!^{-}_n[\mathrm{nm}] = \frac{63}{V_n[\mathrm{m/s}]} \text{ or } \lambda\!\!\!^{-}_{\mathrm{UCN}}[\mathrm{nm}] = \frac{63}{V_{\mathrm{UCN}}[\mathrm{m/s}]}. \quad (1)$$

Thus, we conclude that, if, on one hand, $d \gg \lambda\!\!\!^{-}_{\mathrm{UCN}}$, then such particles are too slow; therefore, the change in UCN velocity, ΔV_{UCN}, is too low and can hardly be measured. If, on the other hand, $d \ll \lambda\!\!\!^{-}_{\mathrm{UCN}}$, then such particles are too small and therefore the probability of their interaction with neutrons P_{VUCN} is too low, since neutrons diffract around such nanoparticles. Only if

$$d_0 \approx \lambda\!\!\!^{-}_{\mathrm{UCN}}, \quad (2)$$

then corresponding VUCN can easily be measured, both owing to their relatively high energy and owing to the high probability of such a process.

Moreover, we do not need any hypothesis about special features of the interaction of nanoparticles with the surface. The probability of inelastic UCN scattering on strongly bound particles (or merely on a flat surface) is too small because of the smallness of their vibration amplitudes. In a rather general case, VUCN are produced on rigid small objects weakly attached to the surface. Nanoparticles can move along surface and (or) oscillate about equilibrium points with a large amplitude. Weak coupling of nanoparticles to the surface is natural because, usually, only a few atoms in nanoparticles interact simultaneously with the surface: the interatomic interaction is small at distances longer than about one angstrom, and any actual surface is rough on a nanometer scale.

Upon formulating the model, we will now justify its statements and estimate its parameters.

2.2. Justification of the Model and Estimation of Its Parameters

(i) Let us show that the thermal motion of nanoparticles or surface nanozones of diameter d equal to the UCN wavelength $\lambda\!\!\!^{-}_{\mathrm{UCN}}$ at a characteristic velocity of $V_{\mathrm{UCN}} \approx 3$ m/s, $d \approx \lambda\!\!\!^{-}_{\mathrm{UCN}} \approx 20$ nm, just corresponds to a change of $\Delta V_{\mathrm{UCN}} \sim 1$ m/s in the UCN velocity, a value that was measured in [1, 5, 6].

In thermodynamic equilibrium at a temperature T, the mean energy of about $kT/2$ is associated with each degree of freedom of a particle. For a nanoparticle of mass M, this allows one to estimate its mean velocity component $\overline{V_M}$ parallel to the momentum-transfer direction:

$$\overline{V_M} \approx \sqrt{kT/M}. \tag{3}$$

The mass of a spherical particle of density ρ is

$$M \approx \frac{\rho d^3}{2}. \tag{4}$$

The change in UCN energy is equal to the doubled nanoparticle velocity:

$$\Delta V_{\mathrm{UCN}} = 2V_M. \tag{5}$$

Thus, the mean change in the UCN velocity is

$$\overline{\Delta V_{UCN}} \approx 2\sqrt{\frac{2kT}{\rho d^3}}; \tag{6}$$

that is, it is just about 1 m/s.

(ii) How large is the mass of the particles that are responsible for the measured phenomenon? Equations (3), (5), and (6) give the mass of $M \approx kT/\overline{V_M^2} \approx 10^7$ a.u.

(iii) Let us show how the nanoparticle diameter that is of importance in experiments is chosen. If $d \ll \lambda\!\!\!^{-}_{\mathrm{UCN}}$, then the scattering probability is too low and depends strongly on the nanoparticle diameter:

$$P_{\mathrm{VUCN}} \sim \left(\frac{d}{\lambda\!\!\!^{-}_{\mathrm{UCN}}}\right)^6. \tag{7}$$

This is so because neutron scattering on a nanoparticle is a coherent process; therefore, the scattering cross section is proportional to the square of the number of nuclei in the nanoparticle, this number in turn being proportional to the cube of the nanoparticle size.

Equation (6) shows that, if $d \gg \lambda\!\!\!^{-}_{\mathrm{UCN}}$, the change in UCN velocity is too low, so that such VUCN can hardly be measured.

(iv) Let us show that the scattering of UCN on strongly bound nanoparticles corresponds to a low probability P_{VUCN}. The limiting case is that of a piece of material belonging to a flat uniform surface and having a total mass M and a diameter of $d \approx \lambda\!\!\!^{-}_{\mathrm{UCN}}$. In the Debye approximation, the quantum zero-point mean-square displacement of such a particle as a discrete unit in the direction perpendicular to the surface plane is

$$\langle X^2\rangle_\perp^0 = \frac{\hbar}{8\pi^2 M}\int_0^{\nu_D}\frac{g(\nu)}{\nu}d\nu = \frac{3\hbar}{16\pi^2 M\nu_D}, \tag{8}$$

where $g(\nu) = \dfrac{3\nu^2}{\nu_D^3}$ is the phonon density of states and ν_D is the Debye cutoff frequency. Evidently, we have $\sqrt{\langle X_\perp^2\rangle^0} < 10^{-4}$ A. At a finite temperature T, the surface displacement (Debye–Waller factor) is larger, but it is still at least as small as $\sqrt{\langle X_\perp^2\rangle^T} < 10^{-3}$ A. Since $\sqrt{\langle X_\perp^2\rangle^T} \ll \lambda\!\!\!^{-}_{\mathrm{UCN}}$ and since the vibration frequency is much higher than the characteristic frequency of about 10^8 Hz, which is equal to the reciprocal quantum-mechanical time of UCN interaction with the surface, the interaction is of the quantum nature: it is mainly elastic (in the absence of trivial vibrations of the walls), but, with a low probability, inelastic. The probability of such inelastic reflection can be estimated as the square of the ratio of the amplitude of wall vibrations to the neutron wavelength [16, 10]. Evidently, the corresponding value P_{VUCN} is much less than the measured probabilities; therefore, thermal vibrations of strongly bound nanostructures cannot produce the experimentally observed phenomenon.

(v) The opposite case is that of a free nanoparticle (a particle of mass about 10^7 a.u. at room temperature can ascend at an altitude of about 3 cm

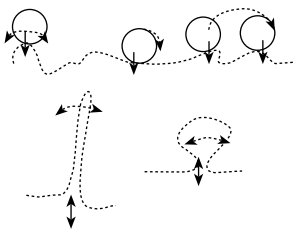

Fig. 2. Interaction of nanoparticles with the surface. The points of attraction of nanoparticles to the surface are indicated by solid arrows. The directions of motion are shown by dotted arrows.

in Earth's gravitational field) or that of weakly attached nanoparticle (it may diffuse along surface or oscillate with a frequency of about 10^8 Hz and with the amplitude commensurate with its size). In this limit, the interaction cross section is nearly equal to the geometric cross section of the nanoparticle if the condition in (2) is satisfied. In all these cases, the probability P_{VUCN} can be estimated as the relative area coated with such "active" nanoparticles. One "active" nanoparticle of size about 20 nm on a square area of size about 20 μm provides the value of P_{VUCN} that was measured in [1]. In such collisions, UCN change the energy of nanoparticles by a negligible value of $\Delta E_{\mathrm{UCN}} \approx 10^{-7}$ eV $\approx \Delta E_M$. Figure 2 illustrates the phenomenon under discussion in an idealized way.

(vi) In an intermediate case of slightly stronger binding ($\nu_{\mathrm{osc}} > 10^8$ Hz and $\sqrt{\langle X^2\rangle_\perp^T} < \lambda\!\!\!^{-}_{\mathrm{UCN}}$), the population of such objects at the surface must be higher than the probability P_{VUCN} of VUCN generation in order to fit measured P_{VUCN} values. It should be noted that the population of such "active" particles seems to have a reasonable value, since the population of any nanostructures, structural irregularities, or nanoparticles of the required size on an actual surface is much higher than that. These were measured by using an atomic-force microscope at the surfaces of all samples that were actually used in the experiments reported in [1].

(vii) Let us analyze the predictions of this model.

As follows from Eq. (6), $\Delta V_{\mathrm{UCN}} \sim T^{1/2}$; therefore, $\Delta E_{\mathrm{UCN}} \sim T$ if $V_M < V_{\mathrm{UCN}}$. These dependences are better satisfied for smaller velocity of the thermal motion of nanoparticles and larger values of the UCN wavelength.

The probability P_{VUCN} depends on the number of active weakly attached nanoparticles. The dependence $P_{\mathrm{VUCN}}(T)$ is smooth if the coupling in the direction of large-amplitude motion (along the surface) is small.

Equation (6) also indicates that $\Delta V_{\mathrm{UCN}} \sim \rho^{-1/2}$; that is, the smaller the nanoparticle density, the larger the change in the UCN velocity. By the way, the density ρ can be calculated by using experimental data or can be obtained by comparing experimental data with calibration measurement involving nanoparticles of known density.

In addition, $\Delta V_{\mathrm{UCN}} \sim d^{-3/2}$; for the optimal ratio of the nanoparticle diameter to the UCN wavelength [see Eq. (2)], this yields $\Delta V_{\mathrm{UCN}} \sim V_{\mathrm{UCN}}^{3/2}$.

Equation (6) estimates the upper boundary of the VUCN spectrum due to the wave properties of neutrons as they tunnel "around" too small nanoparticles. The penetration of neutrons "through" nanoparticles above their potential barrier provides an alternative upper boundary of the VUCN spectrum. Known values of the critical velocity for various materials can provide additional information for verifying the present hypothesis or for identifying the particle material if the validity of the present model would be established. The actual value of $\Delta V_{\max}$ is the smallest value of these two. Of cause, a quantitative analysis of experiments within the present model requires more careful calculations than the above estimations.

2.3. Qualitative Analysis of Experimental Results

This hypothesis assumes an universal reason for VUCN generation on surfaces of different materials; in fact, relevant measurements were performed for the surfaces of stainless steel, fomblin oil, beryllium, and copper [1, 5], with the generation probability being scattered by one to two orders of magnitude.

From [1], it follows that, at solid surfaces, the generation probability $P_{\mathrm{VUCN}}(T)$ at a temperature of 400 K is not considerably higher than that at a temperature of 300 K. New data [12] showed a rather smooth temperature dependence $P_{\mathrm{VUCN}}(T)$ for beryllium and copper samples in the temperature range 100–300 K. On the other hand, the mobility of any nanoobjects at fomblin-oil surfaces (and, accordingly, the P_{VUCN} value) must be much higher than that at solid surfaces. Also, P_{VUCN} must increase strongly at higher temperatures owing to a decrease in the oil viscosity.

The present model is compatible with the measured values for the low energy transfer of $\Delta E_{\mathrm{UCN}} \sim 10^{-7}$ eV.

It assumes higher values of ΔE_{UCN} for greater initial values of E_{UCN}. Such a trend was actually found in [1] for a stainless-steel surface.

The Doppler shift in energy could be both positive and negative. A negative shift was in fact found in [6, 1]. The probability of such a process depends on the phase space available to VUCN. As might have

been expected, measurements showed much a higher probability for heating than for cooling.

It is often stated that, if UCN gain in energy owing to any thermal motion, the energy scale around kT is involved. This is not true in our case since UCN leave traps just after the first event(s) of increase in energy, when no thermal equilibrium of UCN with walls has not yet been achieved. The energy change depends on the nanoparticle velocity, which is small since it is associated with a relatively massive object of energy about kT.

A few factors complicate experiments of this kind. If $V_{\rm UCN} - V_M > V_{\rm lim}$, then $P_{\rm VUCN}$ is low. If $\Delta E_{\rm UCN}$ is too low, such VUCN could hardly be distinguished from UCN because of a finite energy resolution of spectrometers. If $\Delta E_{\rm UCN}$ is too high, then a poor storage time for such VUCN in spectrometers reduces significantly the detection efficiency. If $E_{\rm UCN} + \Delta E_{\rm UCN} > E_{\rm lim}$, the efficiency is negligible. These reasons make difficult quantitative analysis of many previous experiments with UCN. Therefore, a dedicated study of small changes in the UCN energy in a well-optimized spectrometer is required.

3. CAN COOLING ON NANOPARTICLES PRODUCE UCN?

A new method for producing UCN consists in the equilibrium cooling of VCN owing to their many collisions with ultracold nanoparticles made from low-absorbing materials (D_2O, D_2, O_2, etc.) down to the temperature of these nanoparticles of about 1 mK during the diffusion motion of these neutrons in a macroscopically large ensemble of nanoparticles. The principle of equilibrium cooling allows an increase in the neutron phase-space density in contrast to the method of discrimination of 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; they also shift the energy-transfer range far below a value of about 10^{-3} eV, a characteristic limit for liquid and solid moderators. Many collisions are needed since the nanoparticle mass is much higher than the neutron mass; therefore, the energy transfer to nanoparticles and nanostructures is difficult. A large number of collisions constrains the choice of materials: only low-absorbing ones are appropriate. The nanoparticles temperature should correspond to such a minimal neutron energy down to which neutrons can still be cooled by this method. The diffusion motion of neutrons in the ensemble of nanoparticles allows one 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 ensembles of nanoparticles, such as vibrations and rotations, and also by the breaking of internanoparticle bonds.

Upon formulating our main ideas, we will now proceed to justify them within the simple model of free nanoparticles and to estimate the model parameters.

3.1. Model of Free Nanoparticles and Estimation of Parameters

(i) Let us estimate the loss of neutrons due to their capture in nuclei during their cooling in a gas of free molecules. At low temperatures, all gases become liquid (helium) or solid. Therefore, a consideration of neutron scattering on free molecules at temperature of about 1 mK is only the first step in analyzing neutron interaction with nanoparticles. From the theory of neutron cooling in reactor moderators, it follows that, for an isotropic angular distribution of scattered neutrons in the c.m. frame, the cooling of neutrons in gases of free atoms (or molecules) with an atomic mass A is efficient if

$$\frac{\sigma_{\rm coh}}{A\sigma_{\rm abs}} > \ln\left(\frac{V_i}{V_f}\right), \qquad (9)$$

where V_i is the initial neutron velocity and V_f is the final neutron velocity. It should be noted that the coherent-scattering cross section $\sigma_{\rm coh}$ is independent of the neutron velocity, while the absorption cross section $\sigma_{\rm abs}$ is proportional to the reciprocal neutron velocity:

$$\sigma_{\rm abs}(V_n) \sim \sigma_{\rm abs}(V_0)V_0/V_n. \qquad (10)$$

This circumstance limits the minimal velocity $V_{\rm min}$ that can be achieved owing to the cooling of neutrons in a free molecular gas even at zero temperature. On the other hand, the losses of neutrons are negligible when the neutron velocity is higher than this "dangerous" limit. The condition in (9) constraints the list of candidates to a very few: deuterium, oxygen, probably carbon, or a combination of these atoms. The table compares different materials for nanomoderators. The cooling of neutrons down to velocities even lower than that presented in the table is not efficient, but, fortunately, such neutrons have already been cooled sufficiently in order to trap them.

(ii) Evidently, a decrease in the thermalization length increases the density of cooled neutrons. This condition requires a significant increase in the neutron-scattering cross section in moderators; this may be achieved by assembling of molecules (or atoms) into nanoparticles. The cross section for the

Estimates of the minimal velocity to which neutrons can cooled down in a gas of free atoms, molecules, or nanoparticles of various materials [$\frac{\sigma_{\rm coh}}{A\sigma_{\rm abs}(V_{\rm min})} = 1$]

Molecule/atom	$V_{\rm min}$, m/s
D, D_2	~ 0.4
D_2O	~ 1.0
O, O_2	~ 2.4
CO_2	~ 10.0
C	~ 16.0
Be	~ 20.0

interaction of neutrons with nanoparticles is proportional to the square of the number of molecules in a nanoparticle, while the absorption cross section is in direct proportion to the number of molecules in a nanoparticle. (If $T \sim 10^{-3}$ K, then absorption is supposed to be the only alternative to the cooling of neutrons. This assumption is surely correct if the low-energy upscattering of UCN [1, 5] is actually responsible for anomalous losses of UCN [4] and if other unknown phenomena are not involved.) With increasing the nanoparticle size, there arise two factors that compensate each other. (A) The number of collisions needed for cooling neutrons is proportional to the nanoparticle mass M. (B) The ratio of the coherent-scattering cross section to the absorption cross section is also proportional to the nanoparticle mass, $\sigma^M_{\rm coh}/\sigma^M_{\rm abs} \sim M$. Therefore, the condition in (9) for the efficiency of cooling is valid until the nanoparticle size becomes so great that scattering proves to be anisotropic; that is, if $d < \lambda\!\!\!^{-}_n$, then

$$\frac{\sigma^M_{\rm coh}}{M\sigma^M_{\rm abs}} \approx \frac{\sigma_{\rm coh}}{A\sigma_{\rm abs}}. \qquad (11)$$

If the neutron velocity is higher or if the nanoparticle size exceeds these limits (or if both these conditions are satisfied), the angular distribution of scattered neutrons is directed forward. This change in the angular distribution of scattered neutrons increases the relative importance of absorption. The condition in (11) is not valid in this case, since the coherent-scattering cross section increases more slowly than in proportion to N^2; also, the energy transfer per collision decreases. Therefore, the velocity range for neutrons that can still be cooled down is restricted from above by some velocity $V_{\rm max}$.

(iii) Thus, the neutron velocity range $V_{\rm min}$–$V_{\rm max}$ in which the cooling at ultracold nanoparticles is efficient is restricted from both sides: the minimal velocity $V_{\rm min}$ is restricted by neutron absorption in the nuclei of a nanoparticle material, while the maximal velocity $V_{\rm max}$ is restricted by a decrease both in the interaction cross section and in the energy transfer. The broader this range of acceptable velocities, the greater the resulting increase in the neutron phase-space density.

(iv) Let us show that the range of acceptable neutron velocities in the model of free nanoparticles is broad. An estimate of $V_{\rm min}$ is independent of the nanoparticles size, since the condition in (11) is always valid if $d < \lambda\!\!\!^{-}_n$. But the neutron wavelength is proportional to the reciprocal neutron velocity (1) and, in the low-velocity limit, is just large at the last stage of the cooling. Thus, only the nanoparticle material specifies the value of $V_{\rm min}$, which can be estimated as

$$\frac{\sigma_{\rm coh}}{A\sigma_{\rm abs}(V_{\rm min})} = 1; \qquad (12)$$

that is, it can be as low as about 1 m/s (see table).

On the other hand, the quantum-mechanical problem of neutron interaction with a nanoparticle must be solved in order to estimate $V_{\rm max}$. This is beyond the scope of the present study, but I am going to address the problem in a forthcoming publication. However, $V_{\rm max}$ can be estimated. Let us use the following model: The distance between nanoparticles is three times greater than the nanoparticle diameter; that is, the relative volume of about 1.6% is occupied by nanoparticles. Therefore, neutrons can resolve the variation in the density of nuclei in such a moderator. The macroscopic scattering length in such a moderator is:

$$\Delta l \approx \frac{1}{N^M\sigma^M_{\rm coh}} \approx \frac{(3d)^3A^2m_n^2}{\sigma_{\rm coh}M^2} \approx \frac{108A^2m_n^2}{\sigma_{\rm coh}\rho^2d^3}. \qquad (13)$$

Under the assumption of an isotropic angular distribution of scattered neutrons, the thermalization length L is greater by a factor equal to the square root of the number of scattering events needed for thermalization:

$$L \approx \Delta l\sqrt{\frac{M}{m_n}} \approx \frac{80A^2m_n^{3/2}}{\sigma_{\rm coh}\rho^{3/2}d^{3/2}}. \qquad (14)$$

The thermalization length for heavy-water nanoparticles in this simplified model is

$$L^{\rm D_2O}[\rm cm] \approx \frac{40}{(d[\rm nm])^{3/2}}. \qquad (15)$$

A reasonable moderator size of 10 cm corresponds to the nanoparticle diameter of about 2.5 nm or [see Eq. (2)] to the initial neutron velocity of about 25 m/s. The efficiency of neutron thermalization is still significant for an initial neutron velocity a few times higher than that; therefore, the maximal neutron velocity of interest is about 10^2 m/s.

4. DISCUSSION

(i) In actual nanoparticle moderators at ultralow temperatures, nanoparticles are not free. However, the interaction between them is very weak sometimes. By way of example, we indicate that, if one takes the nanoparticles of needed size and material (D_2O, D_2, O_2, etc.) and drops them into superfluid ^{4}He (not absorbing neutrons and just providing heat transfer), they are immediately coated with a thin layer of solidified helium. This layer screens the nanoparticles from one another and reduces the interaction between neighboring nanoparticles [18]. An important problem to be studied in experiments is that of the conditions under which nanoparticles move rather independently in their collisions with neutrons. Does internanoparticle interaction in such gels leave sufficient freedom for them? Their independent interaction with neutrons is of crucial importance, since, otherwise, the effective mass of nanoparticles increases, with the result that the energy transfer decreases dramatically. On the other hand, additional degrees of freedom (vibrations, rotations, breaking of interparticles bonds) in such gels provide probably an even more efficient cooling of neutrons than collisions, and they should be considered specially.

(ii) In actual moderators, in contrast to the simplified model of free nanoparticles, it is necessary to take into account neutron-optical effects due to finite distances between nanoparticles. As soon as the neutron energy becomes sufficiently low (the neutron wavelength becomes sufficiently large), the neutron wavelength covers simultaneously a few nanoparticles; therefore, the effect of coherent upward scattering of neutrons in the ensemble of nanoparticles takes place. This results in the following: (A) The energy transfer decreases and the cooling process becomes less efficient. (B) The depth of extraction of such neutrons increases, and the moderator becomes more transparent for such low-energy neutrons. In this case, UCN can diffuse out to the moderator surface from its total depth, and this simplifies their extraction.

(iii) The technical feasibility of such a moderator should be carefully studied. The use of ultralow temperatures does not permit placing in the vicinity of the reactor core. It can be installed at the exit of an optimized VCN neutron guide. A detailed experimental and theoretical study of the feasibility of the proposed neutron moderator using ultracold nanoparticles is to be performed in the framework of the project described in [19].

5. CONCLUSION

The proposed hypothesis on the nature of VUCN generation explains small changes in the UCN energy in traps in terms of their coherent scattering on nanoparticles weakly bound to surface or just on nanopieces of a rough surface for which large oscillation amplitudes are allowed. UCN change their energy in such a scattering process because nanoparticles and nanostructures are always in a state of thermal motion. This model is qualitatively compatible with available experimental data. Such weakly attached nanoparticles or nanostructures could be selected in a direct measurement (for instance, with an atomic-force microscope) or in future UCN experiments (for instance, by a selective action on a surface nanostructure with a simultaneous control of VUCN generation). This model provides guidelines for experimental studies but it cannot yet be proven or rejected on the basis of already known results. On the other hand, this hypothesis provides a sensitive method for studying the dynamics of specially introduced nanoparticles or nanostructures and, as a result, their interactions with surface or with one another.

In addition, a new concept for increasing the UCN density has been proposed. This concept is based on the cooling of neutrons on ultracold nanoparticles of heavy water, deuterium, or oxygen in superfluid helium. This method can be applied at initial neutron energies in the range from 10^{-8}–10^{-7} eV to 10^{-5}–10^{-4} eV. It differs from traditional methods for UCN production by a high efficiency of employing the initial neutron flux. Detailed theoretical and experimental study of such a cooling process, as well as reliable estimations of UCN density gain and the maximal neutron energy at which such a process is still efficient, is required.

ACKNOWLEDGMENTS

I am grateful to my colleagues for criticism and discussions, especially to H. Börner, V.K. Ignatovich, R.U. Khafizov, E.V. Lychagin, A.V. Muzychka, J.M. Pendlebury, and A.V. Strelkov.

REFERENCES

1. V. V. Nesvizhevsky, A. V. Strelkov, P. Geltenbort, and P. S. Iaydjiev, ILL Annual Report (1997), p. 62; Eur. J. Appl. Phys. **6**, 151 (1999); Yad. Fiz. **62**, 832 (1999) [Phys. At. Nucl. **62**, 776 (1999)]; A. V. Strelkov, V. V. Nesvizhevsky, P. Geltenbort, *et al.,* Nucl. Instrum. Methods Phys. Res. A **440**, 695 (2000).
2. P. Geltenbort, A. G. Kharitonov, V. V. Nesvizhevsky, *et al.,* ILL Preprint 97GE5051 (1997).
3. A. V. Strelkov in *Proceedings of IV School on Neutron Physics, Alushta, Russia, 1990*, p. 325.

4. V. P. Alfimenkov, V. V. Nesvizhevsky, A. P. Serebrov, *et al.*, Pis'ma Zh. Éksp. Teor. Fiz. **55**, 92 (1992) [JETP Lett. **55**, 84 (1992)].
5. L. N. Bondarenko, P. Geltenbort, E. I. Korobkina, *et al.*, Yad. Fiz. **65**(1) (2002) (in press) [Phys. At. Nucl. **65** (2002) (in press)].
6. L. N. Bondarenko, E. I. Korobkina, V. I. Morozov, *et al.,* Pis'ma Zh. Éksp. Teor. Fiz. **68**, 663 (1998) [JETP Lett. **68**, 691 (1998)]; in *Proceedings ISINN-6, JINR, Dubna, Russia, 1998,* p. 101.
7. A. V. Strelkov and M. Hetzelt, Zh. Éksp. Teor. Fiz. **74**, 23 (1978) [Sov. Phys. JETP **47**, 11 (1978)].
8. A. D. Stoica, A. V. Strelkov, and M. Hetzelt, Z. Phys. B **29**, 349 (1978).
9. S. T. Belyaev and A. L. Barabanov, Nucl. Instrum. Methods Phys. Res. A **440**, 704 (2000); A. L. Barabanov and S. T. Belyaev, Yad. Fiz. **62**, 824 (1999) [Phys. At. Nucl. **62**, 769 (1999)]; Eur. Phys. J. B **15**, 59 (2000).
10. V. K. Ignatovich, *The Physics of Ultracold Neutrons* (Clarendon, Oxford, 1990).
11. J. M. Pendlebury, Annu. Rev. Nucl. Part. Sci. **43**, 687 (1993).
12. A. Steyerl *et al.,* Phys. Lett. A **116**, 347 (1986); P. Ageron, P. DeBeaucourt, H. D. Harig, *et al.*, Cryogenics **9**, 42 (1969).
13. R. Golub and J. M. Pendlebury, Phys. Lett. A **62**, 337 (1977).
14. I. S. Altarev, N. V. Borovikova, A. P. Bulkin, *et al.*, Pis'ma Zh. Éksp. Teor. Fiz. **44**, 269 (1986) [JETP Lett. **44**, 344 (1986)].
15. A. P. Serebrov, V. A. Mityukhlyaev, A. A. Zakharov, *et al.*, Pis'ma Zh. Éksp. Teor. Fiz. **59**, 728 (1994) [JETP Lett. **59**, 757 (1994)].
16. L. D. Landau and E. M. Lifshitz, *Course of Theoretical Physics,* Vol. 3: *Quantum Mechanics: Non-Relativistic Theory* (Nauka, Moscow, 1989, 4th ed.; Pergamon, New York, 1977, 3rd ed.).
17. E. V. Lychagin, A. Yu. Muzychka, V. V. Nesvizhevsky, *et al.*, Yad. Fiz. **63**, 609 (2000) [Phys. At. Nucl. **63**, 548 (2000)].
18. L. T. Mezhov-Deglin and A. M. Kokotin, Pis'ma Zh. Éksp. Teor. Fiz. **70**, 744 (1999) [JETP Lett. **70**, 756 (1999)].
19. V. V. Nesvizhevsky, ILL Millenium Program (2000), p. 30.