MULTIPLE ACCELERATION OF NEUTRONS IN AN INVERSELY POPULATED MEDIUM

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The interaction between a rarefied neutron gas and an inversely populated medium consisting of nuclei in an excited isomeric state is considered. The neutron spectrum is approximately described by a Gaussian function with a mean energy E_e which considerably exceeds the isomer excitation energy ϵ_m. The conditions of existence and range of applicability of such a distribution are discussed. It is pointed out that the multiplication factor for systems containing nuclei with a fission threshold can be increased by multiple acceleration of the neutrons by the surrounding isomers.

1. We consider an inversely-populated medium in which all the atoms or nuclei are raised in some manner to an excited state, so that the medium has a negative temperature. Assume that this state can exist for a sufficiently long time. A rarefied gas of extraneous particles, when interacting with such a medium, can either acquire energy through inelastic collisions of the second kind, or else transfer energy to the medium in different deceleration acts. If the fraction of the energy lost by the gas particles in each individual deceleration act is small (for example, in elastic scattering of electrons by atoms and ions, of neutrons by heavy nuclei, etc.), then one can expect from the energy balance that the average energy of the gas particles in the quasiequilibrium state will greatly exceed the excitation energy of the particles of the medium.

By way of example, let us consider the interaction of a rarefied neutron gas with an inversely populated medium. When the neutrons collide with nuclei in an excited isomeric state, we can have besides the pure elastic scattering (n, n) also the inelastic acceleration reaction (n, n″), in which the outgoing neutron carries away the excitation energy ϵ_m of the isomer[1]. Subsequent multiple collisions with isomers can greatly increase the energy of the initially slow neutron.

Let us estimate the average energy E_e to which the neutron is accelerated in a sufficiently large block consisting of isomers only. We assume for simplicity that the leakage and absorption of the neutrons can be neglected, and that the next excited level of the isomer lies much above E_e. The decay of the isomer in the reaction (n, n″) or (n, n) brings the system comprising the isomers and the neutron gas into an equilibrium state. However, if the isomers are long-lived and the neutron density per unit volume n is smaller by many orders of magnitude than the isomer density n_m, then the relaxation time of the entire system is much longer than the time of establishment of the quasistationary distribution of the neutrons.

The energy E_e acquired by the neutron per unit time is approximately equal to $N_m\sigma_g v\epsilon_m$, where σ_g is the cross section of the reaction (n, n″) and v is the neutron velocity. In the case of spherically symmetrical scattering in the c.m.s., the energy loss amounts to

$$N_m(\sigma_g+\sigma_s)v\frac{2E}{A+1},$$

where σ_s is the cross section for pure elastic scattering and A is the ratio of the masses of the nucleus and of the neutron ($A \gg 1$). In the stationary state, both expressions are equal and

$$E_e=\frac{(A+1)\sigma_g}{2(\sigma_g+\sigma_s)}\varepsilon_m. \tag{1}$$

For long-lived isomers, ϵ_m usually amounts to tens and hundreds of keV. Although the reaction (n, n″) has so far not been observed directly, the cross section σ_g can be determined from the inverse reaction (n, n′), the excitation of an isomer level by a neutron, using the detailed-balancing principle. For example, $\sigma_g(E)$ of ^{115m}In (ϵ_m = 0.335 MeV, τ_m = 6.5 h) amounts to several tenths of a barn at E = 0.3 MeV[2]. σ_g is smaller than σ_s because the neutron must overcome the centrifugal barrier. With increasing E, the contribution of the latter decreases and σ_g increases so that E_e can be of the order of several MeV.

2. We now explain the character of the distribution of the neutrons with respect to energy. Since $kT \ll \epsilon_m$, it is possible to assume that the isomer nuclei are at rest in the laboratory frame. For the quasistationary neutron density in phase space, we can write the nonrelativistic kinetic equation

$$dn(E)/dt=N_m\{I_g n(E)+I_s n(E)\}=0, \tag{2}$$

where I_g is the inelastic-collision operator:

$$I_g n(E)=\int d\mathbf{p}'\,\{a^g_{\mathbf{p}',\mathbf{p}}\,\delta(E'+\varepsilon_m-E_q-E)\,n(E')$$
$$-a^g_{\mathbf{p}\mathbf{p}'}\,\delta(E+\varepsilon_m-E_q-E')\,n(E)\},$$
$$E=\frac{p^2}{2m},\quad E_q=\frac{(\mathbf{p}-\mathbf{p}')^2}{2mA},\quad \mathbf{p}=m\mathbf{v}. \tag{3}$$

In formula (3), $a_{p,p'}$ is the square of the amplitude of the transition from the state p to the state p′, summed over the final spin states and averaged over the initial ones. In the operator I_s it is necessary to put $\epsilon_m = 0$ and replace $a^g_{p,p'}$ by $a_{p,p'}$. If we take the integral with respect to the angles in the first term of (3), then the remaining integral with respect to the energy lies in the range $E_1(E) \le E' \le E_2(E)$, where

$$E_{1,2}(E)=\frac{E}{(A-1)^2}\left[A\sqrt{1-\frac{(A-1)\varepsilon_m}{AE}}\mp 1\right]^2. \tag{4}$$

The region of integration of (4) is shown in Fig. 1. We note that in the absence of pure elastic scattering ($a^s_{p,p'} = 0$) we have $n(E) = 0$ at $E < E_t = (1/4)A\epsilon_m$.

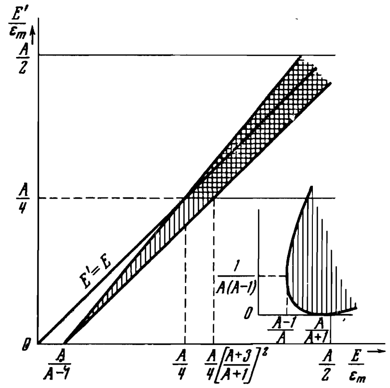

FIG. 1. Region of integration in the inelastic-collision integral (shaded).

Indeed, it follows from the energy conservation law that the recoil energy E_q transferred to the nucleus in backward scattering can compensate for the energy ϵ_m acquired by the neutron, but only starting with a threshold energy E_t. The energy E_t can be determined from the equation $E_t = E_2(E_t)$ (see formula (4) and Fig. 1) or from the condition $E_q = \epsilon_m$ at $p' = -p$ in the lab. When $E < E_t$, the initially slow neutrons will be only accelerated in each collision, until they go beyond the threshold. There will therefore be no neutrons with energy $E < E_t$ in the time-asymptotic quasistationary distribution $n(E)$.

The differential equation for $n(E)$ near E_e can be obtained from (2) by the standard method used to derive the Fokker-Planck equation[3]. We multiply (2) by an arbitrary function $\varphi(E)$ having the required number of derivatives, integrate with respect to dp, and interchange the order of integration with respect to dp and dp′ in the first integral of (3). Expanding then $\varphi(E')$ in a Taylor series about the point E, we obtain

$$N_m \int dp\, n(E) p \sum_{k=1}^{\infty} A_k(E) \frac{1}{k!} \frac{\partial^k}{\partial E^k} \varphi(E) = 0, \tag{5}$$

$$A_k(E) = \frac{1}{p} \int dp' (E'-E)^k \{a^g_{p,p'}\, \delta(E+\varepsilon_m - E_q - E') + a_{p,p'}\, \delta(E - E_q - E')\}. \tag{6}$$

We introduce the density of the collisions per energy interval

$$q(E) = N_m \sigma(E) n(E) 2mE, \tag{7}$$

and also the k-th moment of the energy transferred to the neutron

$$\langle \Delta^k E\rangle = \frac{A_k(E)}{A_0(E)} = \frac{mA_k(E)}{\sigma(E)}, \tag{8}$$

where $\sigma = \sigma_g + \sigma_s$ is the total cross section. We integrate each k-th term of the sum (5) $k-1$ times by parts, setting the expression equal to zero at the integration limits. Then, by virtue of the arbitrariness of the derivative $\varphi'(E)$, which can be taken outside the summation sign, the sum itself must vanish, i.e.,

$$\sum_{k=1} \frac{(-1)^{k-1}}{k!} \frac{\partial^{k-1}}{\partial E^{k-1}} \{\langle \Delta^k E\rangle q(E)\} = 0. \tag{9}$$

Confining ourselves to the first two terms, we obtain an equation for the approximate function $g_0(E)$:

$$\langle \Delta E\rangle q_0(E) - \frac{1}{2} \frac{\partial}{\partial E} \{\langle \Delta^2 E\rangle q_0(E)\} = 0, \tag{10}$$

which in the stationary problem is a consequence of the vanishing of the total neutron current density in energy space. Equation (10) means that the energy transferred by pure diffusion current through the phase-space energy surface compensates for the energy that is acquired (or lost) in the mean by the neutrons as a result of the collisions. The accuracy of such an approximation will be estimated below.

Integrating (10), we obtain

$$n(E) = \frac{C_1}{\sigma(E) E \langle \Delta^2 E\rangle} \exp\left\{2 \int^E \frac{\langle \Delta t\rangle}{\langle \Delta^2 t\rangle} dt\right\}, \tag{11}$$

where C_1 can be determined from the condition of conservation of the total number of neutrons per unit volume:

$$4\pi \int n(E) p^2 dp = N. \tag{12}$$

The moments $\langle \Delta^k E\rangle$ are more conveniently calculated in the c.m.s. For $A_k^g(E)$ we have

$$A_k^g(E) = m\left(\frac{A}{A+1}\right)^{k+2} \left[1 + \frac{(A+1)\varepsilon_m}{AE}\right]^{1/2} \int d\Omega\, a^g(\mu) \left[\left(\varepsilon_m - \frac{2E}{A+1}\right) + \mu \frac{2E}{A+1}\left(1 + \frac{(A+1)\varepsilon_m}{AE}\right)^{1/2}\right]^k, \tag{13}$$

where μ is the cosine of the scattering angle in the c.m.s. Introducing the notation

$$\langle f(\mu)\rangle_{g,s} = \int a^{g,s}(\mu) f(\mu) d\Omega \Big/ \int a^{g,s}(\mu) d\Omega, \quad \overline{\mu^k} = \frac{\sigma_g \langle \mu^k\rangle_g + \sigma_s \langle \mu^k\rangle_s}{\sigma_g + \sigma_s}, \tag{14}$$

we obtain for $\langle \Delta^k E\rangle$

$$\langle \Delta^k E\rangle = \left[\frac{A}{A+1}\right]^k \left\{\frac{\sigma_g}{\sigma} \left\langle \left[\left(\varepsilon_m - \frac{2E}{A+1}\right) + \mu \frac{2E}{A+1}\left(1 + \frac{(A+1)\varepsilon_m}{AE}\right)^{1/2}\right]^k \right\rangle_g + \frac{\sigma_s}{\sigma}\left[\frac{2E}{A+1}\right]^k \langle (\mu-1)^k\rangle_s \right\}. \tag{15}$$

For $E \gg \epsilon_m$, discarding terms of the order A^{-1}, we get for not too large values of k

$$\langle \Delta^k E\rangle = \frac{\sigma_g}{\sigma} \left\langle \left[\varepsilon_m - \frac{2E}{A}(1-\mu)\right]^k \right\rangle_g + \frac{\sigma_s}{\sigma}\left[\frac{2E}{A}\right]^k \langle (\mu-1)^k\rangle_s. \tag{15a}$$

We see from (15) that when $E \gg \epsilon_m$, with accuracy to $\langle \mu\rangle_g (\epsilon_m/E) A^{-1}$ and $\langle \mu\rangle_g H^{-2}$, we have $\langle \Delta E\rangle = 0$ at the point E_e:

$$E_e = \frac{[A + 1 + \langle \mu\rangle_g] \sigma_g(E_e)}{2[1 - \bar{\mu}(E_e)] \sigma(E_e)} \varepsilon_m. \tag{16}$$

In the case of spherically symmetrical scattering ($\langle \mu\rangle_{gs} = 0$), expression (16) goes over into (1).

Near E_e we can confine ourselves only to the linear term of the expansion of the integrand in (11), and the neutron energy distribution turns out to be Gaussian:

$$n(E) = C_2 \exp\left[-\frac{(E-E_e)^2}{\Gamma_0^2}\right], \quad \Gamma_0^2 = -\frac{\langle \Delta^2 E_e\rangle}{\partial \langle \Delta E\rangle / \partial E|_{E_e}}. \tag{17}$$

If we introduce the probability, normalize the unity, that the neutron has an energy E:

$$w(E) = 4\pi m \sqrt{2mE} N^{-1} n(E),$$

then

$$w(E) = \frac{1}{\sqrt{\pi}\Gamma_0} \exp\left[-\frac{(E-E_e)^2}{\Gamma_0^2}\right], \tag{17a}$$

and the mean-squared deviation from the energy E_e is $\Gamma_0^2/2$.

3. It follows from (15a) that

$$\frac{\partial \langle \Delta E\rangle}{\partial E}\Big|_{E_e} = -\frac{2}{A}(1-\bar{\mu}) + \varepsilon_m \frac{\partial}{\partial E}\left[\frac{\sigma_g}{\sigma}\right]_{E_e} + \frac{2E_e}{A} \frac{\partial \bar{\mu}}{\partial E}\Big|_{E_e}. \tag{18}$$

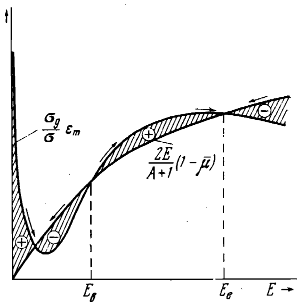

FIG. 2. Average energy gained or lost by the neutron vs E. The region of values of ⟨ΔE⟩ is shaded.

If σ_g/σ increases in such a way that $\partial\langle \Delta E \rangle/\partial E\,|_{E_e} > 0$, then $\Gamma_0^2 < 0$ and the energy $E_e = E_b$ corresponds to the minimum in the distribution of the neutrons. According to formulas (8) and (13), when $E \ll \epsilon_m$ we have $\sigma_g \sim E^{-1/2}$. When $E > \epsilon_m$, the cross section $\sigma_g(E)$ should increase with increasing E, owing to the decreased contribution of the centrifugal barrier ($kR \sim 1$ at neutrons energies on the order of tenths of a MeV). At an energy of several MeV, $\sigma_g(E)$ and $\sigma_s(E)$ vary slowly, whereas $\bar{\mu}(E)$ increases. Figure 2 shows qualitatively one of the possible variants of the behavior of $\epsilon_m\sigma_g/\sigma$ and $2E(1-\bar{\mu})/(A+1)$. The region of the values of $\langle \Delta E \rangle$ is shown shaded. The arrows indicate the direction of the growth of the neutron spectrum. The energy E_b is the limiting energy that divides the double-hump distribution of the neutrons. Neutrons with initial energy near E_b will be accelerated in the main at $E > E_b$ and slow down at $E < E_b$. Thus, a low-energy neutron trap can be produced at $E < E_b$. Depending on the behavior of the cross section of the concrete isomer, the curves in Fig. 2 can have two or one intersections. If the cross sections σ_s and σ_g have broad resonances, the neutron spectrum becomes even more complicated.

When determining the high-energy part of the spectrum near E_e, we can assume that $E_e \gg \epsilon_m$. Since allowance for the derivatives of the cross sections and $\bar{\mu}(E)$ calls for detailed information on the concrete isomer, we calculate Γ_0 for a simple model, in which $a^g(\mu)$ and $a^s(\mu)$ depend little on the energy, so that we can confine ourselves to the first term in (18). According to (17), (18), and (15) we obtain discarding terms of order A^{-1} and above,

$$\Gamma_0^2 = \varepsilon_m^2 \frac{A\sigma_g}{2\sigma(1-\bar{\mu})}\left\{1 - \frac{\sigma_g}{\sigma}\left[2 - \frac{\sigma_g\overline{(1-\mu)^2}}{\sigma(1-\bar{\mu})^2}\right]\right\}. \qquad (19)$$

In the limiting cases we have

$$\Gamma_0^2 = \begin{cases} \varepsilon_m^2 \dfrac{A\sigma_g}{2\sigma(1-\bar{\mu})} = \varepsilon_m E_e, & \sigma_g \ll \sigma; \\ \varepsilon_m^2 \dfrac{A}{2}\dfrac{\overline{\mu^2}-\bar{\mu}^2}{(1-\bar{\mu})^3} = \varepsilon_m E_e \dfrac{\overline{\mu^2}-\bar{\mu}^2}{(1-\bar{\mu})^2}, & \sigma_s \ll \sigma. \end{cases} \qquad (19a)$$

4. Let us estimate now the accuracy of Eq. (10) and the region of its applicability. According to perturbation theory, the contribution of each of the discarded terms of (9) to q(E) is

$$q_0(E)\psi_k(E) = -q_0(E)\int^E \frac{dE'}{q_0(E')\langle\Delta^2 E'\rangle}\frac{(-1)^{k-1}}{k!}\frac{\partial^{k-1}}{\partial E'^{k-1}}\{\langle\Delta^k E'\rangle q_0(E')\}, \qquad (20)$$

where $g_0(E)$ is the solution of (10). If $a^g(\mu)$ and $a^s(\mu)$ have a weak dependence of the energy, then it follows from (15a) that

$$\frac{1}{k}\left|\frac{d}{dE}\langle\Delta^k E\rangle\right| \sim \frac{2}{A}|\langle\Delta^{k-1}E\rangle|, \qquad \frac{\varepsilon_m}{kE^2}|\langle\Delta^k E\rangle|.$$

Therefore when $E \gg \epsilon_m$ we can take $\langle \Delta^k E' \rangle$ from outside the differentiation operator in (20) with accuracy of order A^{-1} in comparison with the term ψ_{k-1}.

For the case $\sigma_g \ll \sigma$ we can confine ourselves to the region $\epsilon_m \ll E \ll \frac{1}{2}A\epsilon_m$. When $k \geq 2$, it follows from (15a) that $\langle \Delta^k E \rangle \sim \epsilon_m^k \sigma_g/\sigma$. Near E_e we can replace $q_0(E)$ in (20) by the Gaussian function (17). Using the definition and properties of Hermite polynomials $H_{k-1}(x) = (2k)^{-1} dH_k(x)/dx$,[4], we obtain

$$\psi_k(x) \sim -\frac{1}{k!\,k}\frac{\varepsilon_m^{k-2}}{\Gamma_0^{k-2}} H_k(x), \quad x = \frac{(E-E_e)}{\Gamma_0}. \qquad (21)$$

Thus, the expansion of (9) near E_e is an expansion in the parameter $\epsilon_m/\Gamma_0 = (\epsilon_m/E_e)^{1/2}$. The largest contribution to q(E) is made by $\psi_3(x)$:

$$\psi_3(x) \sim (\varepsilon_m/E_e)^{1/2}[{}^2/_3 x - {}^4/_9 x^3].$$

In order for the main part of the spectrum to be described by a Gaussian distribution, it is necessary that formula (17) be valid up to values $x^2 \sim 1$. The discarded terms are small if $\psi_3(x) \ll 1$, i.e., under the condition

$$0.4(1-\bar{\mu})A^{-1}x^6 \ll \sigma_g/\sigma \ll 1,$$

where x^2 can amount to several units.

For the case $\sigma_s = 0$ and spherically symmetrical scattering in the c.m.s. we have

$$E_e = {}^1/_2(A+1)\varepsilon_m, \quad \Gamma_0^2 = {}^1/_6(A+2)\varepsilon_m^2,$$
$$\langle\Delta^k E_e\rangle \sim \varepsilon_m^k \overline{\mu^k}$$

and the quantity $\langle \Delta_k E_e \rangle$ vanishes for odd values of k. This means that to estimate the accuracy of (10) it is necessary to retain in ψ_3 the linear term of the expansion of $\langle \Delta^3 E \rangle$ in x, and also to make use of ψ_4. By performing the calculations we can show that the sum $(\psi_3 + \psi_4)$ contains terms of order A^{-1}, $A^{-1}x^2$, and $A^{-1}x^4$. If these terms are neglected, then we can retain terms of order $A^{-1/2}x$ and $A^{-1/2}x^3$ in n(E) calculated by formula (11):

$$n(x) = \frac{N}{2(2\pi m)^{1/2}\Gamma_0 E_e^{1/2}}(1-3bx)\exp\left[-x^2 + \frac{4}{3}bx^3\right],$$
$$x = \frac{E-E_e}{\Gamma_0} = \frac{1}{b}\frac{(E-E_e)}{E_e}, \qquad b = \frac{\Gamma_0}{E_e} = \left(\frac{2}{3A}\right)^{1/2}. \qquad (22)$$

The normalization factor in (22) has been calculated accurate to A^{-1}. In Fig. 3, the solution (22) for A = 100 is compared with a numerical computer solution of the exact integro-functional equation $I_g n(E) = 0$ (see formula (3)) under the condition that a^g does not depend on either the energy or the angles. As seen from the figure, the two solutions coincide at the accuracy indicated above. We note incidentally that Eq. (10) and formula (22) could be obtained by expanding the δ-function in (3) in terms of the difference $(\epsilon_m - E_q)$ and retaining only the quadratic terms.

5. It follows from the foregoing that the process of equilibrium establishment in an inversely populated medium differs from the analogous processes in an ordinary medium, where the energy of the initially fast neutrons decreases continuously as a result of moderation, tending to become equalized with the average energy of the nuclei. To the contrary, in an isomeric

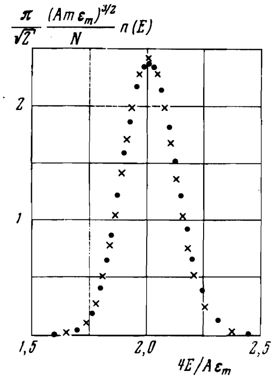

FIG. 3. Stationary distribution of neutron energy at $\sigma_s = 0$ (A = 100); ●—numerical integration of the the integral equation (2); X—solution (22) of the Fokker-Planck equation.

medium the neutrons with initial energies exceeding the average energy of the nuclei will be repeatedly accelerated until a quadistationary Gaussian distribution with average neutron energy $\overline{E} = E_e \gg \epsilon_m$ and a quadratic variance $\overline{(E - \overline{E})^2} = \Gamma_0^2/2$ is established. $\overline{E}$ decreases and the non-equilibrium distribution n(E) begins to deviate gradually from the Gaussian distribution only as the nuclei in the ground state (N_0) accumulate. To describe these processes it is necessary to include in (2) the terms proportional to N_0 and take the inelastic moderation into account. The time τ_0 of transition of the nuclei to the ground state is connected with the lifetime of the nucleus in the isomeric state by the relation

$$\tau_0 = \tau_m (1 + N\sigma_g \bar{v} \tau_m)^{-1}.$$

For long-lived isomers at a low neutron gas density τ_0 can amount to weeks and months. The quasistationary spectrum (17) will exist at times $t \ll \tau_0$. However, t can be increased if the heat is removed from the system at a sufficiently rapid rate and the unexcited nuclei are replaced by isomeric ones.

In the analysis of systems with real isomers, it is necessary to take into account the neutron leakage and absorption, which make it impossible for the neutron to exist for a long time in the system without stationary sources. The quasistationary spectrum n(E) will then be intermediate between the source spectrum S(E) and the Gaussian distribution (17), going over into the latter in the limiting case of a neutron loss that is small in comparison with the (n, n″) reaction.

Another important circumstance that must be taken into account in real systems is the existence of upper excited levels of the isomeric nucleus. The contribution of inelastic moderation to the neutron spectrum can be neglected if the energy ϵ_2 of the second excited level satisfies the inequality $\epsilon_2 - \epsilon_m - E_e \gg \Gamma_0$. In the opposite case, the spectrum begins to decrease already starting with $E \sim \epsilon_2$, and the average neutron energy turns out to be lower than E_e. If the double-humped distribution discussed in Sec. 3 obtains, then the inelastic moderation transfers the neutrons to the low-energy trap, where the capture cross section is usually relatively large, thereby increasing the effective capture.

For most isomers, ϵ_2 amounts to several hundred keV. Near the magic nuclei, the distance between the levels increases and ϵ_2 increases. Thus, for example, for ^{91m}Nb (ϵ_m = 0.1 MeV, τ_m = 92 d) we have ϵ_2 = 1.2 MeV[5].

To calculate the neutron spectra in real systems it is necessary to accumulate detailed information on the dependence of the differential cross sections of the reactions (n, n″) and (n, n) on the angles and energies for concrete isomers. These data will answer the question whether it is feasible in principle to obtain a chain reaction in systems with substances having a low fission threshold, such as ^{234}U, ^{238}U, and ^{240}Pu[6]. The neutrons moderated below the fission threshold could be raised back in these systems above the threshold via multiple acceleration by the surrounding isomers.

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