Giant Absorption Cross Section of Ultracold Neutrons in Gadolinium

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The transmission of ultracold neutrons through natural Gd and isotopic enriched ^{157}Gd samples has been measured. Absorption cross sections in the order of 100 Mb have been identified. Here one touches a region where fluctuations of the physical parameters influence the result and can cause a basic deviation from the exponential attenuation law and where a purely imaginary interaction potential exists within the sample.

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Gadolinium exhibits the highest absorption cross section of stable isotopes due to resonances near the threshold energy. The absorption cross section at thermal energies for natural gadolinium is reported as σ_a (v = 2200 m/s) = 49 000 b and for ^{157}Gd as σ_a (v = 2200 m/s) = 254 000 b [1]. For cold and ultracold neutrons $1/v$ behavior is predicted, which brings the absorption cross section up to σ_a (v = 10 m/s) = 10.7(3) Mb for natural Gd and to σ_a (v = 10 m/s) = 55.9(1.5) Mb for ^{157}Gd.

The absorption cross section of very slow neutrons of Cd was measured by Palmgren [2] and that of Au, Al, and Cu by Steyerl [3] and Steyerl and Vonach [4]. In all cases the $1/v$ behavior has been verified, and it was observed that inhomogeneous substances cause additional contributions to the total cross section [5]. The largest measured cross section was 30 kb. The inelastic and incoherent scattering contribution has been examined by Binder [6]. A $1/v$ behavior has been predicted, which was found experimentally as well [7–9].

The absorption cross section should be independent of the dynamical state of the absorbing nuclei and is expected to be strictly varying as $\sigma_a \propto 1/v$, where v denotes the group velocity within the material where the absorbing nucleus is embedded [10,11]. v follows from the index of refraction

$$v = nv_0 = \sqrt{v_0^2 - v_c^2}, \qquad \text{with } v_c^2 = \frac{4\pi\hbar^2 N b_c}{m^2}, \tag{1}$$

where v_c denotes the critical velocity, N the particle density, and b_c the coherent scattering length.

On the other hand, Gurevich and Nemirovsky [10] gave for the absorption cross section a saturation value of σ_a of about 77 Mb for ^{157}Gd. This is predicted for pure gadolinium where the interaction potential inside a metallic sample becomes purely imaginary [see Eq. (2)].

The absorption, incoherent, and inelastic processes cause an imaginary part in the interaction potential [12]

$$V(r) = V_r(r) - iV_i(r), \tag{2}$$

where

$$V_r = \frac{2\pi\hbar^2}{m}\sqrt{\left(\sum_i N_i b_{ci}\right)^2 - \sum_i \left(\frac{\sigma_{ri}}{2\lambda}\right)^2}$$

$$\text{and} \quad V_i = \frac{\hbar v}{2}\sum_i N_i \sigma_{ri}$$

($\sigma_r = \sigma_a + \sigma_{\text{incoh}} + \sigma_{\text{inel}}$, at v = 2200 m/s). The imaginary part in most cases is orders of magnitude smaller than the real part ($V_i \ll |V_r|$), except for pure Gd and its compounds. When the thickness D of the barrier is much larger than the wavelength of the neutrons, the oscillatory part of the reflectivity becomes damped due to the finite wavelength spread and one obtains for $V_i \ll |V_r|$

$$R = \left|\frac{v_0 - v'}{v_0 + v'}\right|^2 \quad \text{and} \quad T = \frac{4k\,\mathrm{Re}v'}{|v_0 + v'|^2}, \tag{3}$$

where v' contains an imaginary part as well:

$$(v')^2 = (v)^2 + i\,\frac{\hbar v}{m}\sum_i N_i \sigma_{ri}. \tag{4}$$

When back and forth reflections from the surfaces of the sample are taken into account, the transmission coefficient becomes [13]

$$\tau = \frac{I}{I_0} = \frac{\alpha T^2}{1 - \alpha^2 R^2}, \tag{5}$$

where $\alpha = \exp[-(\sum_i N_i \sigma_{ri})D]$. Calculations have shown that τ equals α within 1% when $v_0 > 1.5v_c$. Wiggles of the quantum transmission function are smeared out due to the velocity spread of the beam and because $D/D_\lambda \gg 1$, where the λ thickness is given as

$D_\lambda = 2\pi/Nb_c\lambda$. Here we have treated the longitudinal component of the neutron velocity with respect to the sample surface only. The tangential component does not have any influence because it is conserved (inverted) at the surfaces and the velocity dependence of the reaction cross section is just compensated by a larger path length through the sample ($\sigma_r \propto \cos\vartheta/v; D_{\text{eff}} = D/\cos\vartheta$) [13].

A transmission experiment of ultracold neutrons from the neutron turbine was performed at the ultracold neutron facility of the ILL research reactor. A sketch of the experimental arrangement is shown in Fig. 1 where the cross section in the range 2.5 to 19 m/s was measured. The energy of neutrons with velocity v_c just equals the height of the optical potential representing the sample. Most of our experiments were carried out with a Gd salt dissolved in D_2O in a quartz cuvette. The critical velocity of the cuvette material (SiO_2) is $v_c = 4.155$ m/s and that of D_2O is $v_c = 5.63$ m/s.

The characteristic spectral distributions and intensities can be found in the literature [14]. The chopper opening time was $\Delta t = 27$ ms, the time-of-flight distance was $L = 0.65$ m, and the detector thickness was $\Delta D = 0.5$ mm. The transport of ultracold neutrons through guide tubes gets a diffuse component when the effect of surface roughness is included. Several diffusion models have been developed to describe this effect (e.g., [9]). In our case where a forward collimated beam enters the time-of-flight tube ($\overline{\cos\vartheta_0} \sim 0.9$) the probability for diffuse scattering is below 5%, the mean number of wall collisions is below 4, and the transmission probability is about 93%. A small slowing-down effect in the time-of-flight system may change the time scale for the slower neutron part of the spectrum by 1%–2% [15]. Figure 2 shows various transmission spectra used for the determination of the transmission function of the samples (Fig. 3). The signal-to-background ratio for the channel counting 5 m/s neutrons was still above 100. The related signal is about 200 counts/channel. Most of the measurements have been made with Gd dissolved in heavy water. The solution had a thickness of 1 mm and was within a quartz cuvette (SiO_2) with a wall thickness of 1.25 mm. Thus, the real part of the interaction potential had a shape as shown in Fig. 1.

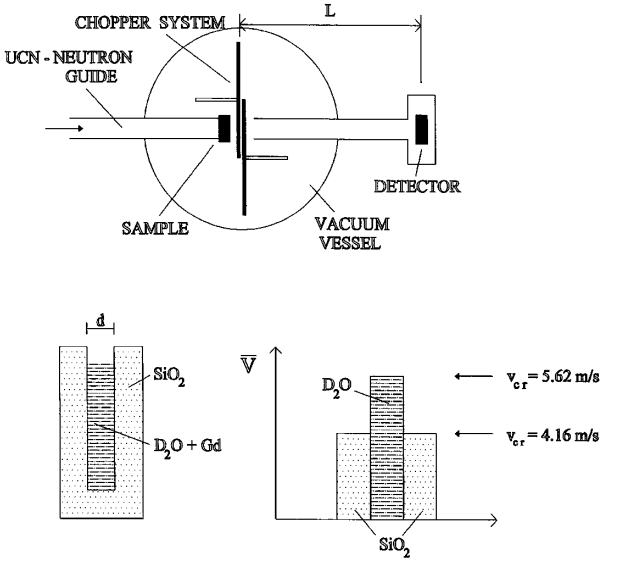

FIG. 1. Experimental setup (top), and the geometrical form and the optical potential of the sample.

The transmission was measured as a function of the time of flight between the chopper and the detector for pure D_2O for various solutions of Gd and ^{157}Gd. The solutions have been prepared from Gd nitrate and Gd metal dissolved in small amounts of HCl. The concentration of Gd in the solvent lies in the range of 10–80 ppm compared to the number of D_2O molecules [corresponding to $(1.2–12.8) \times 10^{17}$ Gd atoms/cm^3] which has also been controlled by total reflection x-ray fluorescence measurements. Because of the very tiny amount of Gd in the solution nearly all scattering effects are determined by the D_2O, whereas Gd contributes to the absorption only. The measured signal (Fig. 2) is a convolution of the transmission spectrum and the known resolution function of the chopper, which can be taken as a Gaussian with a half-width of 27 ms [16]. The related deconvolution was done using an inverse fast Fourier transform algorithm. In order to avoid edge effects the central part of the data was used for cross section evaluation only. Figure 3 shows typical measured transmission coefficients in comparison with an optimal fit curve according to Eq. (5). The transmission was measured for the solvent alone and for the solvent with Gd. Thus all the scattering and absorption effects arising from the cuvette and the D_2O solvent could be compensated up to a high extent.

Measurements have also been performed with pure Gd layers evaporated onto SiO_2 glass. In this case $v \sim v_0$ and the data evaluation can be made by a simple exponential law. Unfortunately, the layer thickness was not very uniform and whether the particle density within the layer matches the bulk value has to be questioned also. Nevertheless, the values are rather compatible with the values measured with the solution.

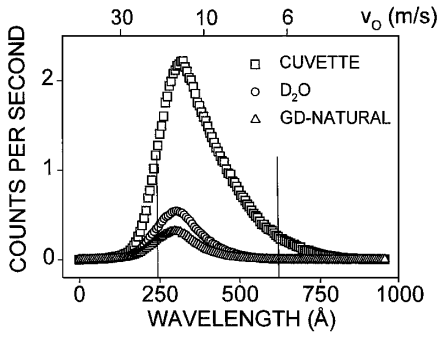

FIG. 2. Measured time-of-flight spectra without and with various samples and an indication for which part the cross section determination has been made.

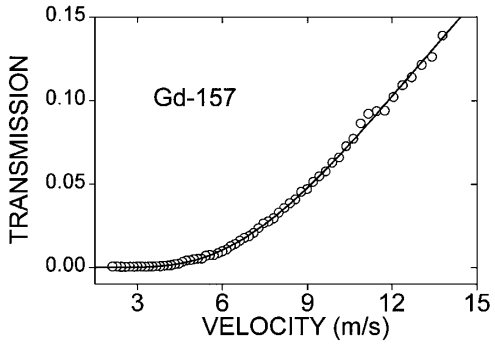

FIG. 3. Measured transmission coefficient of a ^{157}Gd sample and its theoretical fit curves. The abscissa indicates the neutron velocity v inside the sample. (The error bars are generally smaller than the plotted circles and they are marked when they become larger.)

From optimal fit procedures to the velocity-dependent transmission curves an overall $1/v$ cross section can be obtained [Eq. (5)] where the related α values for the solvent and the solvent with the Gd absorber have to be compared. This gives a rather large uncertainty at the higher velocity range where the resolution becomes rather poor $\Delta t/t \sim 0.5$ and at the low velocity range due to low intensity. Corrections according to the enrichment of the ^{157}Gd sample (90.5%) and to the bulk density (95%) of the Gd-layer sample have been taken into account. This procedure yields the values compiled in Table I. For ^{157}Gd and metallic natural Gd these values are 10%–20% below the extrapolated values. The velocity dependence of the cross section has also been evaluated and is shown in Fig. 4 for natural and isotopic pure Gd samples.

Reasonable agreement between measured and expected values according to the $1/v$ law has been achieved. In the lower velocity range ($\sim$3 m/s) a deviation towards lower cross sections appears. Several reasons should be discussed. First, for the metallic Gd layer a smaller cross section can be explained by the saturation effect appearing in the case of a purely imaginary interaction potential. In this case for natural metallic Gd a saturation cross section for $v \to 0$ of 180 Mb is predicted [10]. Second, let us consider the influence of fluctuations of the experimental parameters. When $\alpha = \exp[-\sigma_a N D]$ and $\sigma_a = \sigma_{a,m} v_m / v$ the velocity spread δv around the mean velocity in a certain time channel v_m, the thickness variation δD and the particle density fluctuations δN around their mean values (subscript m) influence the result. For Gaussian distributions of the fluctuations the measured attenuation coefficient becomes

$$\bar{\alpha} = \alpha_m \exp[(\sigma_{a,m} N_m \delta D)^2/2 + (\sigma_{a,m} D_m \delta N)^2/2 + (\sigma_{a,m} N_m D_m)^2 (\delta v / v_m)^2/2], \quad (6)$$

where

$$\alpha_m = \exp[-\sigma_{a,m} N_m D_m].$$

In our case the velocity-dependent part has been taken into account by the deconvolution process. The variation of the thickness can be omitted in the case of the cuvette measurements, but it contributes several percent in the case of the metal layer samples where $\delta D/D_m \sim 10\%$. This has been taken into account for the corrected values cited in Table I. The most interesting influence may arise from the density fluctuation δN. This fluctuation has to be calculated from the number of absorbing nuclei seen by the individual neutron paths through the sample. The related interaction tube $V_{\mathrm{coh}} = \Delta_x \Delta_y D_{\mathrm{eff}} \overline{\cos\vartheta}$ is given by the transverse coherence lengths Δ_x and Δ_y and the effective sample thickness D_{eff} which equals D_m for thin samples ($D_m < 1/N_m \sigma_{a,m}$) and becomes $1/N_m \sigma_{a,m}$ for thick samples as a result of self-shielding. Δ_x and Δ_y are related to the transverse momentum distributions $\Delta \sim (2\delta k)^{-1}$, where δk follows from the critical velocity

TABLE I. Summary of measured data.

Sample	$N \times 10^{+17}$ (cm^3)	d (mm)	σ_a (v = 10 m/s) (Mb)	Isotope corr.	From thermal energies extrapolated $1/v$
Gd in D_2O	5.79(2)	1	12.9		
	6.61(4)	1	11.3		
	9.64(2)	1	9.6		
	12.81(3)	1	9.6		
			Average 10.9(8)		10.7(3)
^{157}Gd in D_2O	1.17(5)	1	49.0	53.8	
	2.88(12)	1	43.7	48.0	
	2.88(12)	1	44.0	48.3	
	2.88(12)	1	44.4	48.8	
			Average	49.7(1.6)	55.9(1.5)
	$N \times 10^{+22}$ (cm^3)	d [Å]			
Gd metal layer	2.90(6)	114(11)	9.2(9)		
	2.90(6)	238(11)	8.9(4)		
			Average 8.9(4)		10.7(3)

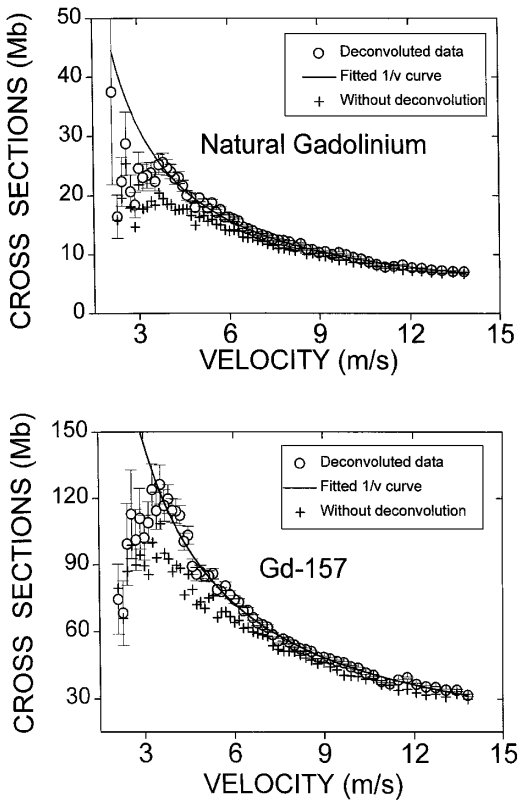

FIG. 4. Velocity dependence of measured ultracold neutron absorption cross section of Gd and ^{157}Gd.

inside the stainless steel guide tube with $v_{\rm cr} = 6.2$ m/s. In our case Δ_x and Δ_y are about 26 Å. The mean transmission cosine is given by the geometry and the index of refraction of the sample and amounts in our case to $\overline{\cos\vartheta} \sim 0.37$. Thus, the mean number of absorbing nuclei seen by the neutron becomes $N_{\rm coh} = N_m V_{\rm coh}$ and its variation $\delta N_{\rm coh} = \sqrt{N_m V_{\rm coh}}$. Because of the low concentration the fluctuation of Gd nuclei inside the coherence tube $\delta N_{\rm coh}/N_{\rm coh} = \delta N/N_m$ reaches values up to 20% and, therefore, a substantial deviation from the standard attenuation law can be expected. One then gets from Eq. (6) an effective absorption cross section

$$\sigma_{a,\rm eff} = \sigma_{a,m}\left(1 - \frac{\sigma_{a,m}\delta N^2 D_m}{2N_m}\right) = \sigma_{a,m}\left(1 - \frac{\sigma_{a,m} D_m}{2\Delta_x \Delta_y D_{\rm eff}}\right). \quad (7)$$

This shows in the case of very high absorption cross sections a marked deviation from the simple exponential attenuation law. Numerically, for ^{157}Gd with a concentration of 10 ppm and $\sigma_{a,m} = 100$ Mb, Eq. (7) predicts a correction to the measured absorption cross section of 6%. Indications of such a deviation of the right order of magnitude can be seen in Fig. 4 at rather low velocities. More precise measurements will be needed to confirm this phenomenon. The influence of the coherence volume on the outcome of scattering experiments has been discussed recently by a similar approach [17]. A $1/v$ cross section indicates an absorption process proportional to the time a neutron spends near an absorbing nucleus. Deviations are expected when fluctuations are taken into account because then the mean number of nuclei with which the individual neutrons are interacting becomes significantly different. This connects these considerations to the debate about general deviations from the exponential decay law which is known as the quantum Zeno effect [18].

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