Interaction of very cold neutrons with a polystyrene-polybutadiene-polystyrene block copolymer

A. V. Antonov, V. I. Gerasimov,[1)] A. I. Isakov, S. P. Kuznetsov, I. V. Meshkov, A. D. Perekrestenko, and S. G. Tarasov[2)]
P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow

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The total interaction cross sections have been studied as a function of the wavelength of very cold neutrons [$\Sigma_t(\lambda)$] in a very-cold-neutron spectrometer [A. V. Antonov *et al.*, Kratk. Soobshch. Fiz. **10**, 10 (1977)] with a polystyrene-polybutadiene-polystyrene block copolymer at room temperature and at 99 K.

Block copolymers, liquid crystals, and several biological objects may possess an ordered supermolecular structure[2–4] with a period corresponding to the wavelength of very cold neutrons (2 nm $<\lambda<$ 80 nm). Despite this very interesting property, the literature reveals no experimental results of any sort on the interaction of these materials with very cold neutrons.

In the present letter we report a study of the dependence of the total macroscopic cross sections $\Sigma_t(\lambda)$ for the interaction of very cold neutrons with an DST-30 polystyrene-polybutadiene-polystyrene block copolymer with a molecular mass $M\sim 8\times 10^4$ and with a linked-styrene content of 28.3% by weight. The dependence $\Sigma_t(\lambda)$ is measured over the wavelength interval $4<\lambda<91$ nm on a very-cold-neutron spectrometer with a resolution $\delta\lambda/\lambda\cong 8\%$ by passing a neutron beam through a sample either at room temperature ($T=293$ K) or cooled by a cryostat[1] to $T=99$ K. The samples are films 90 μm thick synthesized by slow evaporation of a benzene solution of the polymer from the surface of mercury, followed by drying. According to Ref. 5, such films contain crystallites with an ordered supermolecular structure corresponding to a hexagonal packing of long polystyrene cylinders with a maximum interplanar distance $d_{\max}=22$ nm in a polybutadiene matrix. It follows from an analysis of the possible interactions of very cold neutrons with such objects that the total cross sections are determined by the following processes: nuclear capture of neutrons and the inelastic scattering of neutrons due to the dynamics of the system under study, with cross sections [$\Sigma_c(\lambda)$ and $\Sigma_{is}(\lambda)$, respectively] which are described by a $\sim\lambda$ law; the elastic incoherent scattering of neutrons by a bound nucleus (primarily, hydrogen), with a cross section (Σ_{ei}) which is independent of λ; the elastic coherent scattering of neutrons by the ordered supermolecular structure, which satisfies the Bragg condition $n\lambda=2d\sin\theta$ (n is the order of the reflection, λ is the neutron wavelength, d is the interplanar distance, and θ is the glancing angle); and, finally, the elastic incoherent scattering of neutrons by disordered regions of density fluctuations (inhomogeneities), with a size $\sim\lambda$. The cross sections for the last two processes [$\Sigma_{ec}(\lambda)$, $\Sigma_{ei}^{f}(\lambda)$, respectively[6]] have a more complicated λ dependence than do $\Sigma_c(\lambda)$ and $\Sigma_{is}(\lambda)$. The total cross sections can thus be written as the sum

$$\Sigma_t(\lambda)=\Sigma_c(\lambda)+\Sigma_{is}(\lambda)+\Sigma_{ei}+\Sigma_{ec}(\lambda)+\Sigma_{ei}^{f}(\lambda).$$

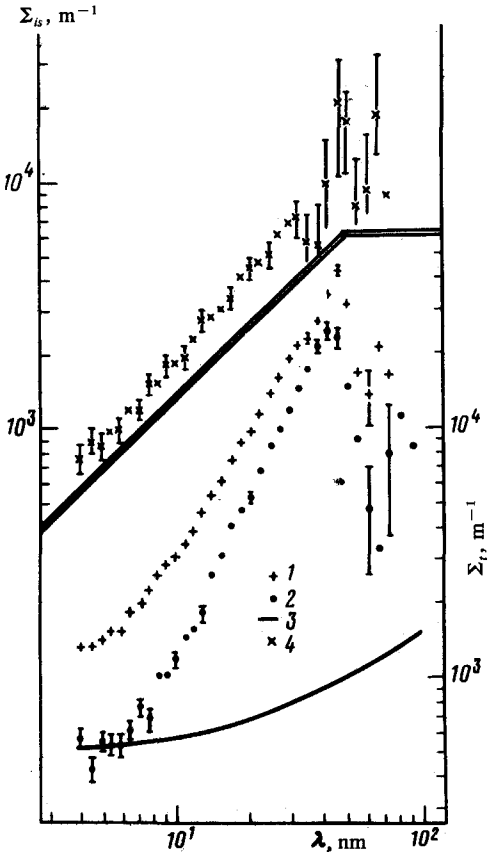

FIG. 1. Wavelength dependence of the cross sections for the interaction of very cold neutrons with a DST-30 block copolymer. 1—$\Sigma_t(\lambda)$ corresponding to a sample at $T = 293$ K; 2—$\Sigma_t(\lambda)$ at $T = 99$ K; 3—$\Sigma_c(\lambda) + \Sigma_{ei}$; 4—$\Sigma_{is}(\lambda)$.

The experimental results are shown in Fig. 1. In calculating the total cross sections we make corrections for the refraction of the neutron wave in the medium and its multiple reflection from the boundaries of the sample.[7] The resulting $\Sigma_t(\lambda)$ curves typically show a marked deviation from the $\sim\lambda$ law toward larger cross sections for neutrons with $\lambda \gtrsim 7$ nm (at shorter wavelengths, the curves become noticeably flatter because of the substantial contribution from $\Sigma_{ei} = 462$ m^{-1}) and a "Bragg cutoff" for neutrons with a wavelength exceeding $\lambda_{\text{lim}} = 46$ nm (at room temperature) or 42 nm (at a sample temperature $T = 99$ K). This result is evidence that the observed deviation is due in large part to the elastic coherent scattering of neutrons by the ordered supermolecular structure. The shift of the position of the cutoff, which is observed regularly as the sample temperature is lowered, is apparently caused by a compression of the supermolecular structure primarily due to vitrification of the polybutadiene matrix, which occurs at $T_{\text{vit}} = 180$ K. From the cutoff position we can easily estimate $d_{\max} = \lambda_{\text{lim}}/2$, which in our case turns out to be 23 nm at $T = 293$ K or 21 nm at $T = 99$ K. The value $d_{\max}$ (293 K) agrees with the value found for $d_{\max}$ from the small-angle x-ray diffraction patterns of the same samples at room temperature, and it also agrees with a result reported previously.[5] The absence from the $\Sigma_t(\lambda)$ curves of peaks due to

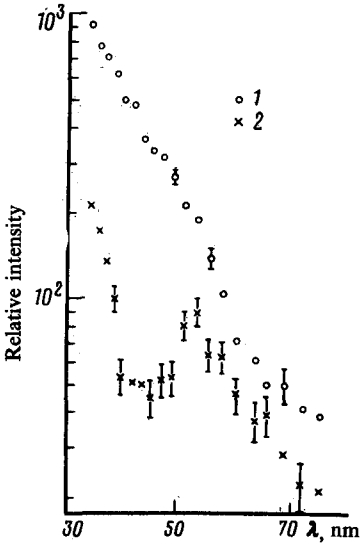

FIG. 2. Neutron spectra. 1—Measured without a sample; 2—after passage through a sample at $T = 99$ K.

higher reflection orders is evidence that the sample is "polycrystalline." The same conclusion follows from the x-ray structural analysis.

A comparison of the neutron spectra (Fig. 2) measured without a sample and after passage through the cooled sample reveals that these DST-30 block copolymer films can serve as a filter for neutrons with $\lambda > 45$ nm. The use of the deuterated analog of DST-30 would make it possible to substantially increase the luminosity of such a filter.

The substantial difference between the total cross sections at the two temperatures, which is described by a $\Sigma_{is}(\lambda) \sim \lambda$ law, as can be seen from Fig. 1, is a measure of the inelastic scattering of neutrons by thermal vibrations of the system under study. The coincidence (within the measurement errors) in the region $\lambda \lesssim 6$ nm of the $\Sigma_t(\lambda)$ curve corresponding to the cooled sample and the theoretical curve of $\Sigma_c(\lambda) + \Sigma_{ei}$ indicates that lowering the temperature to 99 K basically suppresses these vibrations. The observed discrepancy between the curves at $\lambda > \lambda_{\text{lim}}$ can be attributed to an elastic incoherent scattering of neutrons by disordered inhomogeneities (structural defects, pores, foreign inclusions, etc.) over the entire wavelength range of these measurements.

It can be seen that a study of the total cross sections for the interaction of very cold neutrons is a very informative method for studying the structure and dynamics of substances having an ordered supermolecular structure. The "growth" of large-area samples with an ordered supermolecular structure (analogous to single crystals) would make it possible to develop unique devices such as a monochromator for very cold neutrons, which would substantially expand the experimental capabilities for studying the condensed state of matter.

[1)]M. V. Lomonosov State University, Moscow.
[2)]L. Ya. Karpov Physicochemical Scientific-Research Institute.

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