High energy neutron scattering and the Bose condensate in He II

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An analysis of the spectrum of deep inelastic scattering of neutrons ($k = 14.1$ Å^{-1}) is carried out on the basis of the regularized Gauss–Newton iteration process. As a mathematical model, a double Gaussian shape is employed, under the assumption that the scattering law can be represented by two Gaussian curves corresponding to the contributions from the condensate and supercondensate parts. From the viewpoint of the statistical criterion a model with a single Gaussian is the more satisfactory at $T = 4.2$°K, indicating that there is no Bose condensate at this temperature. A model with two Gaussians is the best at $T = 1.2$°K, the Bose condensate fraction being $\rho_0/\rho = 0.036 \pm 0.014$.

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1. INTRODUCTION

Although the connection between the superfluidity of He II and the presence of a Bose condensate is not a direct one,[1] the hypothesis was put forth long ago that there is a condensate of atoms with zero momentum below the temperature of the λ transition in liquid He^4.[2,3] This hypothesis is based primarily on the analogy with the nonideal Bose gas;[3] however, to date no sufficiently realistic, exactly solvable model of a nonideal Bose system has been found in which the interaction did not destroy the Bose condensate.[4] The first estimate of the possible fraction of Bose condensate in a system of bosons which interact like solid spheres and which have the observed density of He^4 was proposed by Onsager and Penrose:[5] $\rho_0/\rho = 0.08$. Further estimates, made from theoretical considerations, vary from 0 to 0.55.[4-7]

Hohenberg and Platzman[8] were first to analyze the possibility of experimental observation of the Bose condensate with the help of strongly inelastic neutron scattering, when coherent scattering goes over at high energy and momentum transfers to scattering of neutrons by individual He^4 atoms. In this case, in the opinion of the authors of[8], a "condensate peak" should be observed against the background of a certain broad distribution for the doubly differential cross section. The ideas of Hohenberg and Platzman were developed further by Puff and Tenn,[9] who relied essentially on the exact relations for the dynamic structure factor $S(k, \omega)$ that follow from the rule of sums.[10,11]

Along with the theoretical investigations, experiments have been carried out, beginning with the work of Cowley and Woods[12], to observe and estimate the fraction of Bose condensate in He^4 at temperatures below the λ transition.[13-15] In recent studies, neutron scattering with a high transfer of energy and momentum has been used for this purpose ($k = 14.33$ Å^{-1}).

The results of these studies are given in Table I. It should be noted that in reduction of the experimental data by least squares in[9,13,14], various models were used for the asymptotic behavior of $S(k, \omega)$ at high momentum transfers. Thus, a double Gaussian approximation was used in[9] as the model for $S(k, \omega)$ with a fixed width of the condensate fraction, and in[14] the model that described the shape of $S(k, \omega)$ contained a non-Gaussian increment of the form $\exp\{-[(\omega - \omega_0(k))^4/\sigma^4]\}$. This fact makes it difficult to compare the results obtained.[1)]

The purpose of the present study was to establish the amount of Bose condensate in superfluid helium more precisely within the framework of a model with two Gaussians (see also[15]). The experiment was carried out with improved statistical accuracy. The integrated count in the inelastic-scattering peak amounted to $\sim 2 \times 10^5$ pulses, and the data were reduced by the regularized Gauss-Newton iterative process proposed in[17,18].

2. HIGH-ENERGY NEUTRON SCATTERING IN LIQUID He^4

It is known that the cross section of inelastic coherent scattering of neutrons by a system of atoms of one type at a temperature $\beta^{-1} = k_B T$ has the form

$$\frac{d^2\sigma}{d\Omega\, dE_f} = \frac{M_n^2}{(2\pi)^3\hbar^4}\frac{k_f}{k_i}|\tilde{v}(k)|^2 N \frac{S(k,\omega)}{1-e^{-\beta\omega}}, \tag{1}$$

where M_n is the mass of the neutron, $\hbar k = \hbar(k_i - k_f)$ is the transferred momentum, $\omega = E_i = E_f$ is the transferred energy, and $\tilde{v}(k)$ is the Fourier transform of the interaction potential of the neutron with the helium atom. In the range of energy and momentum transfer considered, one can make use of the pseudopotential approximation, and $e^{-\beta\omega} \ll 1$. Equation (1) can then be rewritten in the form of the cross section per scatterer atom:

$$\frac{1}{N}\frac{d^2\sigma}{d\Omega\, dE_f} = \frac{\sigma_b}{8\pi^2}\left(1 - \frac{\omega}{E_i}\right)^{1/2} S(k,\omega), \tag{2}$$

where $\sigma_b = (1 + M_n/M_{He})^2 4\pi a^2$ and a is the scattering length. The expression for the Van-Hove function $S(k, \omega)$ in terms of the correlation function of the dehsity-density type has the form[19]

$$S(k,\omega) = \frac{1}{\rho}\int d^3r \int_{-\infty}^{+\infty} \frac{dt}{\hbar} e^{-ikr + i\omega t/\hbar}\langle[\rho(r,t), \rho(0,0)]\rangle, \tag{3}$$

where $\rho(r, t) = \Psi^+(r, t)\Psi(r, t)$ is the operator of the number density of particles in the Heisenberg representation, $\rho = \langle\rho(r, t)\rangle$, and $\langle\dots\rangle$ is the thermodynamic average.

For an ideal Bose gas, it is not difficult to obtain

$$S_0(k,\omega) = 2\pi\frac{\rho_0}{\rho}\{\delta[\omega - \omega_0(k)] - \delta[\omega + \omega_0(k)]\} + \left[\frac{\pi\beta}{\omega_0(k)}\right]^{1/2}(\rho\lambda^3)^{-1}\ln\left\{\frac{1-\exp\{-\beta[(\omega+\omega_0)^2/4\omega_0 - \mu]\}}{1-\exp\{-\beta[(\omega-\omega_0)^2/4\omega_0 - \mu]\}}\right\} \tag{4}$$

TABLE I

Literature	Date	k, Å^{-1}	T, K	ρ_0/ρ
Cowley and Woods [12]	1968	5.1	1.1	0.17±0.10
Puff and Tenn [9] (data of [13])	1970	14.33	1.27	0.06±0.03
Harling [13]	1971	14.33	1.27	0.088±0.013
Mook, Sherm and Vilkinson [14]	1972	14.33	1.2	0.024±0.01
Results of present study*	1974	14.1	1.2	0.036±0.014

*In a previous paper of the authors [15], processing was carried out without account of the dependence of the width $\gamma_{1,2}$ on the transferred momentum (see (12)). The result of [15] is $\rho_0/\rho = 0.029 \pm 0.013$.

from (3). Here $\omega_0(k) = \hbar^2k^2/2M_{He}$, $\lambda = (2\pi\hbar^2\beta/M_{He})^{1/2}$, μ is the chemical potential, $\rho_0(\beta, \rho)$ the number density of particles with momentum equal to zero, and $\beta = (k_BT)^{-1}$. The temperature dependence of $\mu(\beta, \rho)$ and $\rho_0(\beta, \rho)$ at fixed density ρ, and also the value of the critical temperature of the Bose condensate for an ideal gas are well known.[2]

Thus the function $S_0(k, \omega)$ is divided into two terms: the first, $S_0^{BC}(k, \omega)$ corresponds to scattering on the Bose condensate and has the shape of δ-like peaks, and the second, $\tilde{S}_0(k, \omega)$, which is connected with scattering by supercondensate Bose particles, is a relatively broad distribution in ω. The latter term in the strongly inelastic scattering region ($\omega \to \infty$, $k^2 \to \infty$, $\hbar^2k^2/2M_{He}\omega$ finite) has the form

$$\tilde{S}_0(k,\omega) \approx \left[\frac{\pi\beta}{\omega_0(k)}\right]^{1/2} \frac{e^{\beta\mu}}{\rho\lambda^3}\left[\exp\left\{-\beta\frac{(\omega-\omega_0(k))^2}{4\omega_0}\right\} - \exp\left\{-\beta\frac{(\omega+\omega_0(k))^2}{4\omega_0}\right\}\right], \tag{5}$$

where $\beta\omega_0(k) \gg 1$ and $\beta\,|\,\omega \pm \omega_0(k)| \gg 1$. We now note that for large energy and momentum transfers, formula 5) for the scattering by supercondensate atoms also holds for liquid He^4, since the scattering system in this region of k and ω behaves as an almost free one.2) In this case $\tilde{S}(k, \omega)$ for scattering of high-energy neutrons has a Gaussian shape at high but fixed momentum transfers and is centered on the dispersion curve of free atoms $\pm\omega_0(k)$, while its width is determined by the kinetic energy per atom in the liquid He^4.[8] In order to make these arguments more convincing, we note that the experimental curves for $\tilde{S}(k, \omega)$ in the strongly inelastic region at a fixed k and $T = 4.2°K > T_\lambda$ have the form of Gaussians whose centers lie in the vicinity of the free-atom dispersion curve $\omega_0(k)$. We can also add some considerations based on the rules of sums. The Van Hove function is odd in ω and for large k satisfies the relations[11,15]

$$\frac{1}{2}\int_0^\infty d\omega\,|S(k,\omega)| \approx 2\pi, \quad \frac{1}{2}\int_{-\infty}^{+\infty} d\omega\,\omega S(k,\omega) = 2\pi\omega_0(k),$$
$$\frac{1}{2}\int_{-\infty}^{+\infty} d\omega\,\omega^3 S(k,\omega) \approx 2\pi\omega_0(k)\left[\omega_0^2(k) + 4\frac{\langle E\rangle}{N}\omega_0(k)\right]. \tag{6}$$

If we now use these relations to determine $S(k, \omega)$ in the region of large transfers at $T > T_\lambda$, we find that a Gaussian formula of the form3)

$$\tilde{S}(k,\omega) \approx \left[\frac{\pi}{{}^2/_3\langle E\rangle\omega_0(k)/N}\right]^{1/2}\left\{\exp\left[-\frac{1}{4}\left(\frac{2}{3}\frac{\langle E\rangle}{N}\omega_0(k)\right)^{-1}\times(\omega-\omega_0(k))^2\right] - \exp\left[-\frac{1}{4}\left(\frac{2}{3}\frac{\langle E\rangle}{N}\omega_0(k)\right)^{-1}(\omega+\omega_0(k))^2\right]\right\} \tag{7}$$

satisfies the first two equations of (6) identically and the third with asymptotic accuracy, $\langle E\rangle[\omega_0(k)]^{-1}/N \ll 1$. Thus the rule of sums for large momentum transfers (6) allows us to fix the normalization, width and position of the Gaussian form (7) that is odd in ω, the choice of which for $S(k, \omega)$ in strongly inelastic scattering is dictated by physical considerations. It follows from them that $\tilde{S}(k, \omega)$ for liquid He^4 in this region of k and ω behaves in a fashion similar to $\tilde{S}_0(k, \omega)$ for an ideal Bose gas, and $(3/2)\beta^{-1} = \langle E\rangle_0/N$ (see (5)) is replaced by the real kinetic energy of the atom in liquid He^4, $\langle E\rangle/N$ (cf. (7) and (5)). Analysis of the experimental data for $T = 4.2°K > T_\lambda$ and constant k thus allows us to establish the average kinetic energy of the atoms of the liquid He^4, which is equal to $\langle E\rangle/N$, and to compare it with the corresponding theoretical calculations.

We now consider the features of high-energy neutron scattering in the case of the presence of a Bose condensate in liquid He^4 at $T < T_\lambda$. The corresponding contribution to the Van Hove function for the ideal system $S_0^{BC}(k, \omega)$ has the form of a δ function with amplitudes $\sim\rho_0/\rho$ and with carriers on the dispersion curve of free particles $\omega_0(k) = \hbar^2k^2/2M_{He}$ (see (4)). For He II, the δ functions of $S_0^{BC}(k, \omega)$ should smear out, inasmuch as the lifetime τ of the atom in the Bose condensate is finite because of the interaction: $\tau^{-1} \sim \hbar[\rho\sigma(k)]^2/M_{He}$,[8] where $\sigma(k)$ is the He^4-He^4 scattering cross section, which falls off slowly with increasing4) k, and ρ is the density. Consequently, we can assume that $S(k, \omega)$ has the form

$$S(k,\omega) = S^{BC}(k,\omega) + \tilde{S}(k,\omega)$$
$$= \frac{\rho_0}{\rho}\left\{\frac{4\pi}{\gamma_1(k)}\right\}^{1/2}[\exp\{-\gamma_1^{-1}(\omega-\omega_0)^2\} - \exp\{-\gamma_1^{-1}(\omega+\omega_0)^2\}]$$
$$+\left(1-\frac{\rho_0}{\rho}\right)\left\{\frac{4\pi}{\gamma_2(k)}\right\}^{1/2}[\exp\{-\gamma_2^{-1}(\omega-\omega_0)^2\} - \exp\{-\gamma_2^{-1}(\omega+\omega_0)^2\}]. \tag{8}$$

for strongly inelastic scattering of high-energy neutrons by He II at $T < T_\lambda$5) (compare with (4)). Here the quantity $\gamma_1(k)$ is determined by the interaction of the Bose condensate atoms in the final state (finite lifetime τ):

$$\gamma_1(k) = \frac{1}{2\ln 2}\frac{\hbar^2[\rho\sigma(k=0)]^2}{M_{He}}\omega_0(k). \tag{9}$$

The second term corresponds to scattering by supercondensate atoms, and we get

$$\gamma_2(k) = \left[\frac{8}{3}\frac{\langle E\rangle}{N} - \frac{\rho_0}{\rho}\frac{\gamma_1(k)}{\omega_0(k)}\right]\left(1-\frac{\rho_0}{\rho}\right)^{-1}\omega_0(k). \tag{10}$$

from the sum rules (6) for $\gamma_2(k)$.

Equation (8) is basic for the subsequent numerical analysis of the experimental data.

3. DESCRIPTION OF EXPERIMENT

The experiment was carried out on an IBR-30 pulsed reactor in an operating regime with linear acceleration, using a DIN-1M spectrometer.[20] A monochromatic beam of neutrons with energy $E_i = 189.4$ MeV was analyzed, after scattering on the sample at an angle of 122.62°, by the time-of-flight method between the sample and the detector.

Values of the transferred momentum that were most favorable for the experiment were chosen in the range $k \sim 13$–$15\,Å^{-1}$. The lower bound is determined by the closeness of the approach to unity of the ratio of the energy transferred in the neutron scattering to the energy of the free helium atom that corresponds to the momentum k: $\omega_0(k) = \hbar^2k^2/2M_{He}$. This ratio is of the order of 0.96 at $k = 14.1\,Å^{-1}$ in our experiment.[21] This interval is limited above by the resolving power of the spectrometer. In this experiment, the width of the resolution function in the region of the "helium" peak was equal to ~9 MeV. We did not strive for the limiting parameters of the resolution function, since the condensate part of the "helium" peak is broadened by the interaction in the final state to the extent that it is not possible to separate it in explicit form. Therefore, attention was concentrated on lowering the statistical error and obtaining the highest possible accuracy in measurement of the shape of the "helium" peak (see the figure). Over the time of the neutron-scattering experiment at 1.2°K, the integrated count in the "helium" peak amounted to $\sim 2 \times 10^5$ pulses.

The experimental spectra of inelastic neutron scattering by liquid helium at temperatures of 1.2 and 4.2°K are shown in the figure. The width of the time-spectrum analyzer channel was 8 μsec. The total time of measurement amounted to 240 hr. The measurements at the different temperatures were not normalized. The energy resolution, indicated by the horizontal line on the figure, was determined with the help of a vanadium sample and converted for the inelastic-scattering region.

The background was measured during evacuation of the helium vapor over the liquid at the bottom of the cryostat. The center of the elastic peak is located in the 172nd channel. The relatively large scatter in the results at the wings of the "helium" peak is explained by the fact that the background was measured over times less than the effect and was not smoothed but was calculated from the channel. The experimental results were corrected for the effectiveness of the detector.

4. MATHEMATICAL MODEL AND ANALYSIS OF EXPERIMENTAL RESULTS

The experimental spectra for the doubly-differential inelastic-scattering cross section were obtained at a

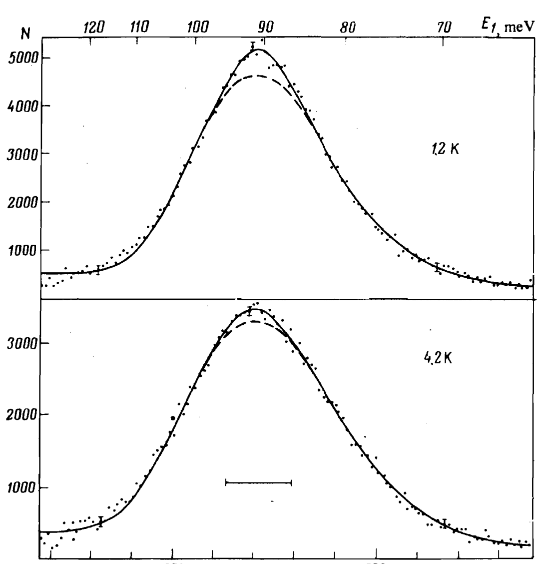

FIG. 1. Experimental spectra of neutrons scattered by liquid helium at temperatures of 1.2 and 4.2°K (E_i = 189.4 MeV, θ = 122.62°, time of measurement at a single temperature ~100 hr). The solid lines indicate the theoretical curves, which consist of two Gaussians with $\rho_0/\rho = 0.036$. The dashed line is the curve referring to the supercondensate fraction; n is the number of the analyzer channel, and N the number of pulses counted in the channel.

fixed angle θ. Therefore, we use the kinematic relation

$$k^2(E_i, \omega, \theta) = \hbar^{-2} \cdot 2M_n\{2E_i - \omega - 2\cos\theta[E_i(E_i-\omega)]^{1/2}\}, \quad (11)$$

for investigation of these data with the help of Eq. (8). This relation expresses the transferred momentum k in terms of the transferred energy $\omega = E_i - E_f$ and the scattering angle θ. For the numerical analysis, we used the following mathematical model for the double differential cross section

$$\frac{1}{N}\left(\frac{d^2\sigma}{d\Omega\, dE_f}\right) = y(t)$$

as a function of the time of flight t:

$$y(t) = \frac{\sigma_b}{4\pi^2}\left(\frac{\rho_0}{\rho}\right)\left(\frac{\pi c^2}{\gamma_1}\right)^{1/2}\frac{t_0}{t^4}\exp\left[-\frac{(2.5t^{-2} - 1.5t_0^{-2} + \omega_0' + 0.53(tt_0)^{-1})^2}{2\gamma_1 c^{-2}}\right]$$
$$+ \frac{\sigma_b}{4\pi^2}\left(1 - \frac{\rho_0}{\rho}\right)\left(\frac{\pi c^2}{\gamma_2}\right)^{1/2}\frac{t_0}{t^4}\exp\left[-\frac{(2.5t^{-2} - 1.5t_0^{-2} + \omega_0'' + 0.53(tt_0)^{-1})^2}{2\gamma_2 c^{-2}}\right]$$
$$+ a + b\left(\frac{t - 413.86}{8} - 218\right), \quad \gamma_{1,2} = \tilde{\gamma}_{1,2}(t_0^{-2} + t^{-2} + 1.06(tt_0)^{-1}). \quad (12)$$

This expression is obtained with the help of the kinematic relation (11) from the two-Gaussian model for $S(k, \omega)$ (8) and Eq. (2) (the exponentials of the form $\exp[-\gamma j^{-1}(\omega + \omega_0)^2]$ were omitted in (8) as unimportant).

The conversion from the energy to time of flight t was perfomed by the formulas

$$E_i = \frac{c}{t_0^2}, \quad E_f = \frac{c}{t^2}, \quad \omega = c\left(\frac{1}{t_0^2} - \frac{1}{t^2}\right), \quad (13)$$

where $c = 603.53 \times 10^6$ meV-μsec^2, and t_0 corresponds to the center of the elastic peak. The connection between t and the time analyzer channel number n has the form $t_n = 413.86 + n \cdot 8$. In expressions (12), the possibile existence of a linear background $a + b((t - 413.86)/8 - 218)$ and a shift in the positions of the desired Gaussians ω_0' and ω_0'' relative to the energy of the free particles $\omega_0(k)$ was also taken into account (see notes 2), 3)).

A numerical analysis of the experimental data[6)] was carried out on the basis (12), reducing to solution of an overdetermined nonlinear set of equations obtained for different values of n, for the unknowns ρ_0/ρ, $\tilde{\gamma}_1$, $\tilde{\gamma}_2$, a, b, ω_0', and ω_0'' (the latter two being set equal to one another). For both temperatures, the number of equations used in analysis of the experimental data was $N_{T1} = N_{T2} = 121$, and the number of unknowns m_i was varied from 5 to 7.

It is important to emphasize that numerical analysis of the exponential dependences is a problem whose solution depends strongly on small distortions of the initial data; therefore to obtain a correct result it is necessary to apply special stable methods of solution.[17] In our case, the analysis was carried out on the basis of the regularized iterative process of Gauss-Newton[17,18] (library program CØMPIL, C-401, Dubna). The resultant nonlinear systems were solved with statistical weights of the form σ_n^{-2} ($n = 1, 2, \ldots, N_T$), where σ_n are the standard deviations of the measured quantities. This makes it possible to use the closeness to unity of the quantity χ_i^2/s_i ($i = 1, 2$; $s_i = N_T - m_i$ is the number of degrees of freedom) as a statistical criterion of the quality of the approximations found[7)] [22]. The results of the numerical analysis of the experimental data for $T_1 = 1.2$°K and $T_2 = 4.2$°K (see[15]) are given in Table II, together with the statistical criteria of the approximation χ_i^2/s_i.[18] The quantities χ_i^2/s_i correspond to the results of approximation of the differential cross section

TABLE II

Parameters	$T_1 = 1.2$ K	$T_2 = 4.2$ K
Model with a single Gaussian		
$\tilde{\gamma}_2$	340.5±15	434.2±8
ω_0''	4.42±0.11	4.02±0.15
χ_1^2	129.2	97.2
χ_1^2/s_1	1.13	0.85
Model with two Gaussians		
$\tilde{\gamma}_2$	366.6±10.6	434.5±15
ω_0''	4.45±0.12	4.02±0.15
$\tilde{\gamma}_1$	41.2±17.1	45.8±43
ω_0'	4.45±0.06	4.02±0.14
χ_2^2	100.3	87.7
χ_2^2/s_2	0.88	0.77
ρ_0/ρ	0.036±0.014	0.017±0.011

Note: The quantities $\tilde{\gamma}_1$, $\tilde{\gamma}_2$, and ω_0', ω_0'' are expressed here in MeV.

by a single Gaussian with the number of unknowns $m_1 = 5$; the χ_2^2/s_2 describe the results of the analysis within the framework of the model with two Gaussians, $m_2 = 7$. Account of the resolution function reduces to convolution of expression (13) with this function. Since the convolution operation does not change the ratio of the areas, which is $\sim\rho_0/\rho$, account of the resolution function is unimportant for determination of the Bose condensate, since it affects only the determination of the width of the experimental peak. The width of the function which describes the Bose condensate contribution turns out in our case to be somewhat greater than the width of the resolution function.

5. CONCLUSION

As a result of this analysis, it has been established that the model with two Gaussians for the doubly-differential cross section of inelastic neutron scattering by He^4 describes the experimental data sufficiently well. It was also established that complication of the model by the addition of new Gaussians does not improve it.

Further, on comparison of the quantities χ_1^2/s_1 and χ_2^2/s_2 for the two temperatures (see Table II), we verify that the model with two Gaussians is better from the viewpoint of the statistical criterion at $T_1 = 1.2°K$ (see Sec. 4), and the model with one at $T = 4.2°K$. As is seen from Table II, we obtain the estimate $(3.6 \pm 1.4)\%$ for the value of the fraction of the Bose condensate ρ_0/ρ. However, the problem of the origin of the non-Gaussian corrections and their effect on the amount of BC determined remains unresolved (see[14]). We hope to turn our attention in the future to study of this problem. In conclusion, we also note that to lower the interaction of the helium atoms in the final state, the Bose condensate must be studied at high energies of the incident neutrons and with improvement of the resolution function.

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1)The experimental data of [14] were subjected to further analysis by Jackson. [16] This paper also gives a short review of the state of the problem of the Bose condensate in HeII.

2)If we leave out of account the interaction of the helium atoms with one another in the final state, an approximation that evidently improves with increasing energy and momentum transfer. [8]

3)Actually, the interaction in the final state leads to a certain shift of the position of the Gaussians (7) from the free-particle dispersion curve (see note2) and Secs. 3 and 4).

4)In the transfer region to be considered below, $\sigma(k)$ differs little from $\sigma(k = 0) \approx 2 \times 10^{-15} cm^2$.

5)See Note2).

6)The experimental data that were reduced can be found in [15].

7)
$$\chi^2 = \sum_{n=1}^{N_T} \frac{[y(t_n) - \tilde{y}(n)]^2}{\sigma_n^2},$$
where $\tilde{y}(n)$ is the experimental count in the n-th channel of the analyzer (see [15]) and $y(t_n)$ is the function (12).

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