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EXPERIMENTAL DETERMINATION OF HEAVY WATER SCATTERING LAW AS FUNCTION OF TEMPERATURE

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Abstract

The scattering law of the heavy water is determined as a function of temperature. The experimental values corresponding to 295°K, 365°K, 453°K and 553°K are reported.

1. Introduction

The prominent place the water occupies in liquid physics was pointed out by its utilization, both as light and heavy water, as moderator and cooling agent in nuclear power reactors.

In this way there may be explained the continuous and sustained interest the study of these substances has attracted, both theoretically and experimentally, especially after the development of the thermal neutron spectrometry method. By this method it is possible to obtain directly the scattering law, i.e. that physical quantity fully describing the structural and dynamical properties of the condensed systems at microscopic scale.

However, though the heavy water - to which we shall further refer - reaches inside the power reactors temperatures of about 300°C, there are not published up to now any experimental data concerning the scattering law for temperatures higher than 150°C.

The purpose of the present paper is to fill in this failure with the experimental values of the scattering law of heavy water at high temperatures. These results may be used to obtain the effective cross sections for nuclear data libraries as well as to generate the multi-group constants, necessary for

nuclear reactors design.

The choice of this subject was altogether stimulated by the increasing interest shown in literature for the study of effects connected with the dependence upon temperature and pressure of the structure and molecular kinetics in liquid state.

2. Description of the method

The double differential effective cross-section of thermal neutron scattering on the heavy water is related to the scattering law by the relation:

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_b^D}{4\pi}\sqrt{\frac{E}{E_o}}\, e^{-\hbar\omega/2k_BT} S(k,\hbar\omega,T) \qquad (1)$$

where:

E_o - the incident neutron energy

E - the scattered neutron energy

σ_b^D - the bound cross-section of the deuterium atom

K_B the Boltzmann constant

T - the absolute temperature of the sample

$\hbar\omega = E - E_o$ - the energy transfer

$\hbar k$ - the momentum transfer

By using the notations

$$\alpha = \frac{\hbar^2 k^2}{2M_D K_B T} \qquad \beta = \frac{\hbar\omega}{K_B T}$$

where M_D is the deuterium atom mass, one may obtain from (1)

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_b^D}{4\pi}\sqrt{\frac{E}{E_o}}\, e^{-\beta/2} S(\alpha,\beta) \qquad (2)$$

The measurements have been performed at temperatures of 22°, 92°, 180° and 270°C by means of a time-of-flight spectrometer in operation at the horizontal channel no. 1 of the VVR-S reactor at

the Institute of Physics and Nuclear Engineering Bucharest. The performances and the characteristics of the chopper and of the experimental unit are presented in /1/.

For the time-of-flight analysis, a EM96 analyser, employing 256 channels, of 16 μs each for every spectrum, has been used. The neutron are simultaneously detected at four scattering angles.

The heavy water conteiner is made up of vertically positioned steel pipes. Each of the 6 pipes has a diameter of 10 mm and a wall thickness of 0.5 mm. The conteiner is introduced in a thermostat which ensures with the help of an electronic control unit a constant temperature with an accuracy of $\pm$ 1°C. The pressurization system assures an increase of the heavy water pressure up to 100 atm.

Measurements have been performed both with full conteiner as well as with empty conteiner at 8 scattering angles: 20°, 30°, 40°, 50°, 60°, 70°, 80° and 88°.

The calibration measurements have been performed by using a 3 mm thick vanadium plate.

3. Data processing

The calibration measurements performed with the vanadium sample gave standardizing factors as follows:

1. The channel no. N_o corresponding to the cutt-off of the incident neutron spectrum by the Be filter; here the neutron energy is E_c = 5.2428 meV.

2. Knowing N_o and L_1 - the flight path between the chopper center and the sample center - we have determined, by means of a computer programme:

L_2 - means flight path between sample and counter batteries,

E_n - the energies of the inelastic scattered neutrons corresponding to the n-th channel,

K_{nj} - the momentum transfer corresponding to channel n and to scattering angle j.

3. There have been determined the factors f_b necessary for effective calibration of the various counter batteries.

4. The calibration factor γ_j leading to absolute values of the effective cross-section has been determined from:

$$\gamma_j = \frac{\sum_n (N_{nj} - F_j)\, \epsilon_n T_{an} f_o f_b f_M f_s^{-1}}{\frac{\sigma_b^V}{4\pi} e^{-aK_j^2} \rho_V \sqrt{V} \sum_n f_z (E_n^{el})^{3/2} / \Sigma_2} \qquad (3)$$

where:

N_{nj} is the counting rate of scattered neutrons registered in the channel n and at the j scattering angle.

F_j is the continuous background counted at battery j.

ϵ_n is the relative efficiency of the counter batteries.

T_{an} is the correction factor, which takes into account the loss of neutron on the flight-path.

f_o is the monitoring factor.

f_b is the factor specific to every counter battery under the given working conditions

f_M is the multiple scattering correction factor.

f_s is the correction factor due to the atenuation of the neutron beam in the sample.

σ_b^V is the effective cross-section of the vanadium atom.

K_j' is the momentum transfer corresponding to the channel that contains E_o.

a is the mean square displacement of vanadium atoms.

f_z is a factor taking into account the units of measure transfer.

ρ_V is vanadium density.

V is the volume of the sample.

The experimental values of the scattering law of heavy water has been obtained from the following relation:

$$S(K_{nj},\hbar\omega_n) = \frac{(I^p_{nj} - I^c_{nj}) f_b \varepsilon_n T_{an}}{\gamma_j \rho_p V_p p \frac{\sigma^D_b}{4\pi} e^{-\frac{\hbar\omega_n}{2K_B T}} e^{-\frac{\hbar^2 K^2_{nj}}{8MK_B T}} f_z (E^{in}_n)^{\frac{1}{2}} / \bar{L}_z / (E^{el})^{\frac{1}{2}}} \qquad (4)$$

where:

p is a correction factor which takes into consideration the impurities in the sample.

I^p_{nj} and I^c_{nj} are the corrected counting rates corresponding to sample and to empty conteiner as well.

In both cases the multiple scattering corrections factors have been calculated by means of a Monte Carlo programme /2/. This programme was modified so that the resolution of the experimental instrumentation to be taken into account.

4. Experimental results. Conclusions

The experimental values of the scattering law $S(\alpha,\beta)$, as a function of α and for 15 different values of β parameter, are presented in Figs. 1-4, corresponding to the temperatures of 22°, 92°, 180° and 270°C, respectivelly. As compared to the results previously obtained in literature for D_2O at room temperature, our data are included between those reported by Haywood /3/ and Whitemore /4/. However, up to now there do not exist dynamic models for heavy water able to describe the scattering law behaviour within the whole ranges of (k,ω) and of temperatures experimentally accessible.

As compared to the atomic liquids study, where simplified

methods of the Vineyard /5/ or Schofield /6/ types are attempted, the study of molecular liquids is more complex because:

a) The basic component in molecular liquids, the molecule, exhibits in most cases a symmetry much different from the spherical one. This is why the interaction potential will depend not only on the distance between the molecules but also on their relative orientation.

b) The molecules may be composed of atoms having different masses; these have different positions in comparison with the molecular mass center. Thus, what can be observed during a neutron scattering experiment is a sum, generally unresolved, of contributions of various nuclei.

c) In most cases the molecular dynamic cannot be approached classically. This is true even in the case of the translation movement that can be correlated with repositioning movements and with short-life vibrations. Under these circumstances the most available method of theoretical study is the direct simulation of the molecular dynamic on a computer /7/.

The discrepancies between theoretical and experimental results are even more striking in the case of high temperature. The scattering law calculated in the Butler formalism /8/ did not offert a satisfactory correspondence with our experimental results. In this case too, the technique of computer simulation succeeded, till now in explaining some phenomena related to the modification of pressure and temperature /9/; there is expected that development of this technique will lead to the elaboration of some realistic dynamic models for heavy water.

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References

/1/ *Teutsch H., Mateescu N., Timis P.*, Nukleonik, 4, 4, 165 (1962)

/2/ *Copley J.D.*, Phys. Comm., 7, 209 (1974)

/3/ *Haywood B.C.*, AERE-R-4582 (1964), Harwell

/4/ *Whitemore W.L.*, Nucl. Sci. Eng., 32, 2 (1968)

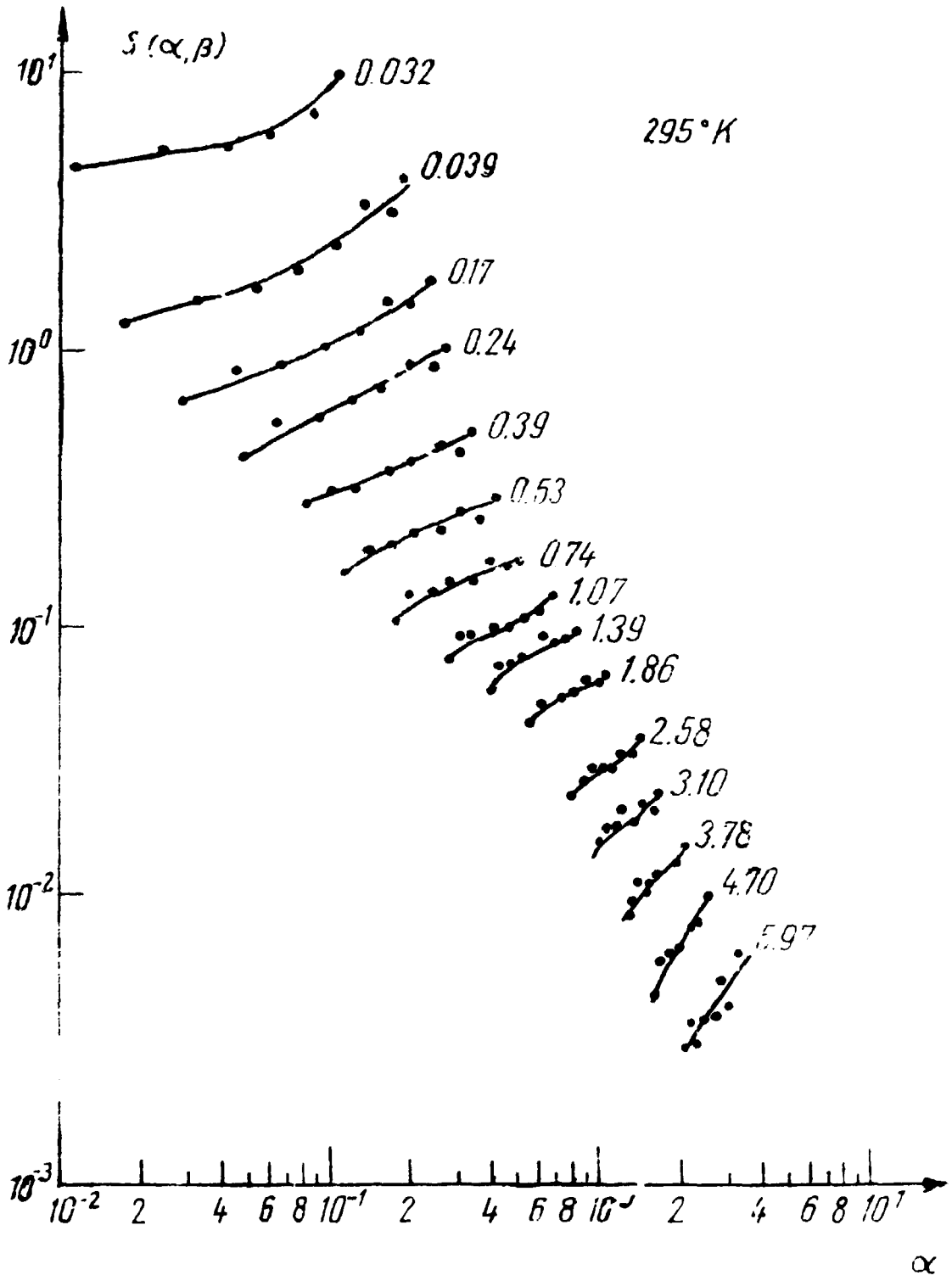
/5/ *Vineyard G.*, Phys. Rev., 110, 999 (1958)

/6/ *Schofield P.*, Phys. Rev. Lett., 4, 239 (1960)

/7/ *Rahman A., Stillinger F.*, Journal of Chem. Phys., 55, 3336 (1971)

/8/ *Buttler D.*, Proc. Phys. Soc. 81, 276 (1963)

/9/ *Stillinger F., A. Rahman*, J. Chem. Phys., 61, 4973 (1974)


S (α, β)
10¹
10⁰
10⁻¹
10⁻²
10⁻³
0.032
0.039
0.17
0.24
0.39
0.53
0.74
1.07
1.39
1.86
2.58
3.10
3.78
4.70
5.97
295° K
10⁻² 2 4 6 8 10⁻¹ 2 4 6 8 10⁰ 2 4 6 8 10¹
α

J Fig. 1

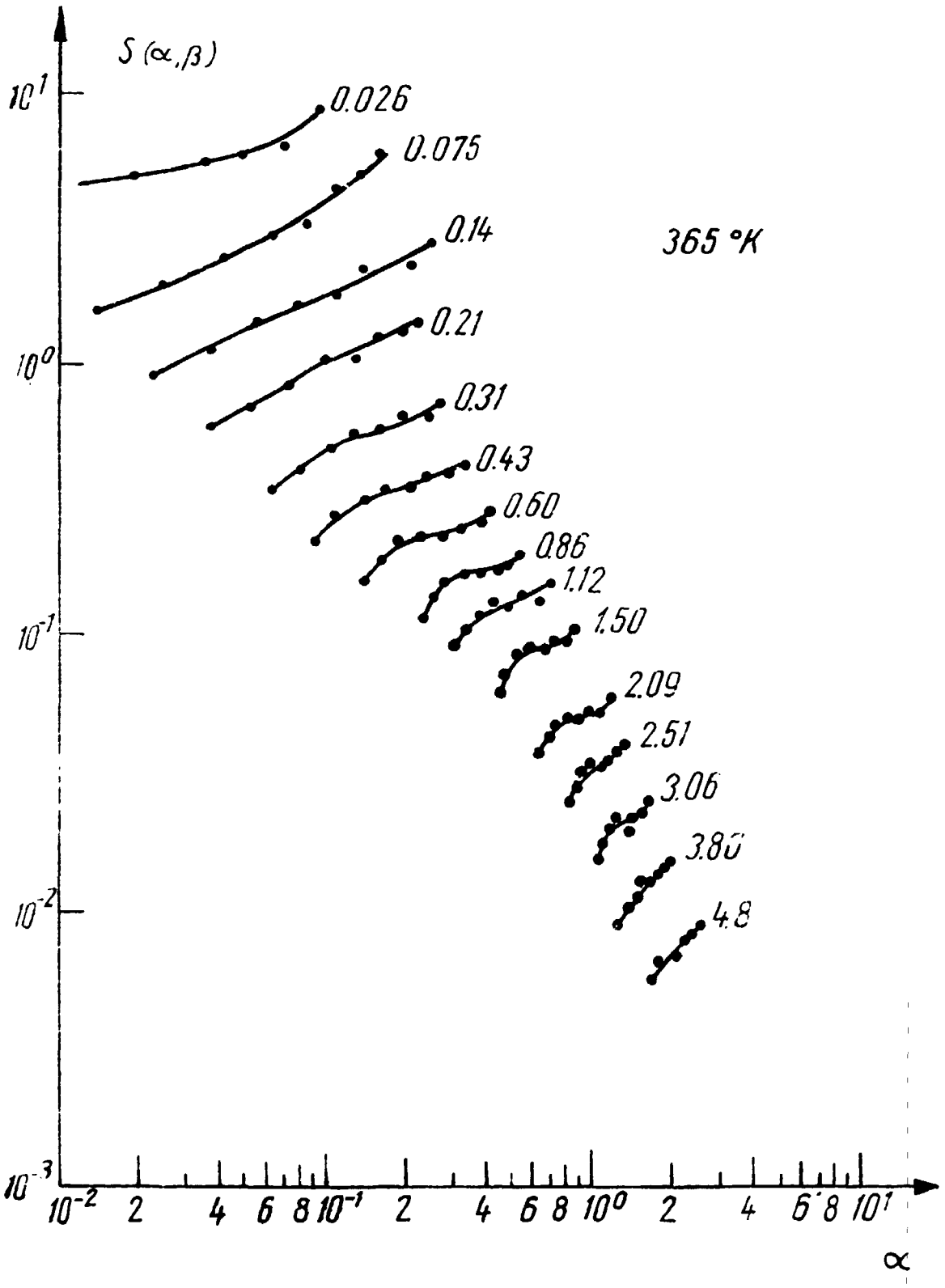

S (α, β)
10^1
10^0
10^-1
10^-2
10^-3
0.026
0.075
0.14
0.21
0.31
0.43
0.60
0.86
1.12
1.50
2.09
2.51
3.06
3.80
4.8
365 °K
10^-2 2 4 6 8 10^-1 2 4 6 8 10^0 2 4 6 8 10^1
α

Fig.2

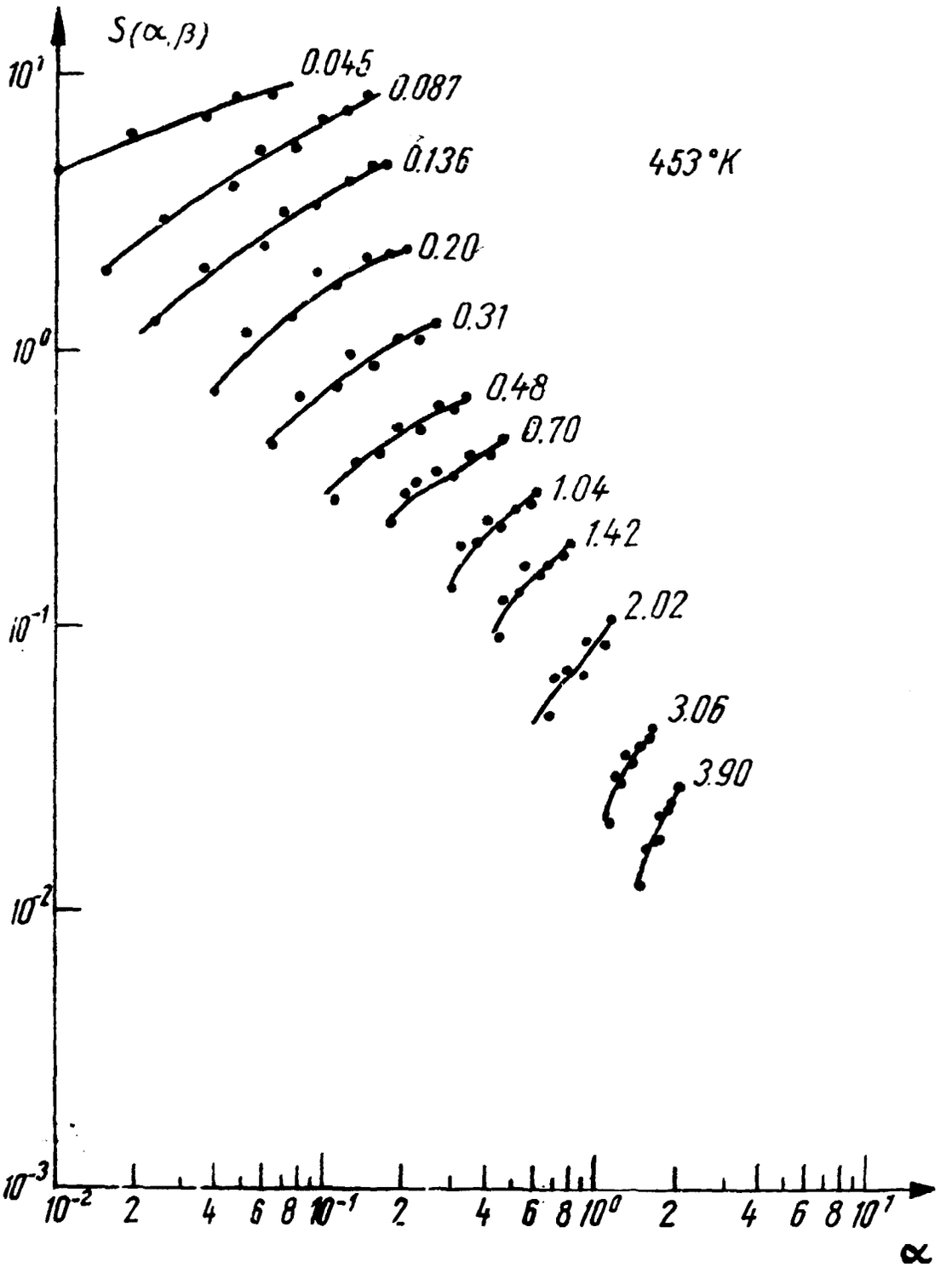

S(α,β)
0.045
0.087
0.136
453 °K
0.20
0.31
0.48
0.70
1.04
1.42
2.02
3.06
3.90
10¹
10⁰
10⁻¹
10⁻²
10⁻³
10⁻² 2 4 6 8 10⁻¹ 2 4 6 8 10⁰ 2 4 6 8 10¹
α

Fig.3.

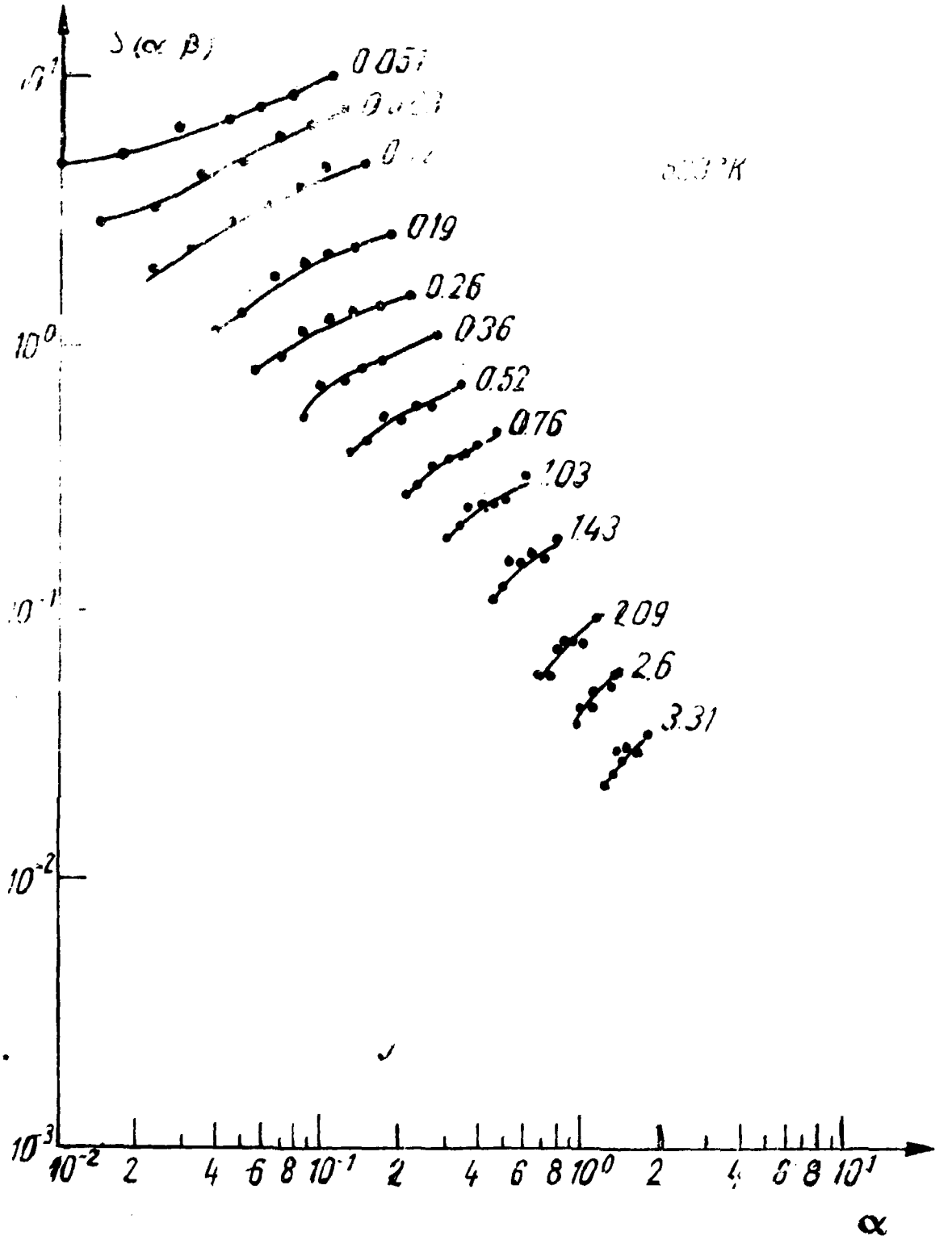

Δ(α, β)
0.019
0.26
0.36
0.52
0.76
1.03
1.43
2.09
2.6
3.31
10^{0}
10^{-2}
10^{-3}
10^{-2} 2 4 6 8 10^{-1} 2 4 6 8 10^{0} 2 4 6 8 10^{1}
α

Fig. 4

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