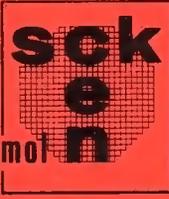


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**NEUTRON INELASTIC SCATTERING STUDY OF  
THE MOLECULAR MOTIONS IN LIQUID SILANE**

**P. VORDERWISCH, S. HAUTECLER**

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Summary. - Several models are compared with experimental neutron scattering cross-sections on liquid SiH<sub>4</sub> at three temperatures. Among these, a very good agreement is only obtained when the translations are described by the Langevin diffusion model while the rotations are treated as free at short times before they are accounted for by the simple rotational diffusion model. The time during which the rotations are free  $\tau_0$  and the rotational diffusion coefficient  $D_r$  are found to increase with temperature. These rotational parameters are not very sensitive to the values of the (not yet measured) self-diffusion coefficient  $D$  used in the analysis. With respect to liquid CH<sub>4</sub>, the rotational motions are much more hindered in liquid SiH<sub>4</sub>.

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Résumé. - On a comparé les sections efficaces de diffusion des neutrons par SiH<sub>4</sub> liquide, mesurées à trois températures, avec les prédictions de divers modèles. On n'obtient un très bon accord que si les mouvements de translation des molécules sont décrits par le modèle de diffusion de Langevin tandis que les mouvements de rotation sont d'abord considérés comme libres pendant un certain temps avant d'être représentés par un modèle de diffusion simple. On observe que le temps  $\tau_0$  durant lequel les rotations sont libres ainsi que le coefficient de diffusion rotationnelle  $D_r$  augmente avec la température. Ces paramètres rotationnels ne sont pas très sensibles aux valeurs du coefficient de diffusion  $D$  (non encore mesuré) utilisées dans l'analyse. Par comparaison avec CH<sub>4</sub> liquide, les mouvements rotationnels sont beaucoup plus entravés dans SiH<sub>4</sub> liquide.

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Samenvatting. - Verschillende modellen worden met de experimentele neutronenverstrooiingsdoorsnede van vloeibaar SiH<sub>4</sub> bij drie temperaturen vergeleken. De beste overeenkomst bekomt men als men de translaties met behulp van het Langevin-diffusie model beschrijft en de rotaties beschouwt als vrije bewegingen die overgaan tot een eenvoudige rotatiediffusie. De gevonden waarden voor de duur  $\tau_0$  van de vrije rotatie en voor de rotatiediffusiekonstante  $D_r$  nemen toe met stijgende temperatuur; ze hangen niet sterk af van de (nog niet gemeten) zelfdiffusiekonstante  $D$  die in de analyse gebruikt werd. De rotationele bewegingen in vloeibaar SiH<sub>4</sub> zijn veel meer gehinderd dan die in CH<sub>4</sub>.

NEUTRON INELASTIC SCATTERING STUDY OF THE MOLECULAR MOTIONS IN LIQUID SILANE

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## ABSTRACT

Several models are compared with experimental neutron scattering cross-sections on liquid  $\text{SiH}_4$  at three temperatures. Among these, a very good agreement is only obtained when the translations are described by the Langevin diffusion model while the rotations are treated as free at short times before they are accounted for by the simple rotational diffusion model. The time during which the rotations are free  $\tau_0$  and the rotational diffusion coefficient  $D_r$  are found to increase with temperature. These rotational parameters are not very sensitive to the values of the (not yet measured) self-diffusion coefficient  $D$  used in the analysis. With respect to liquid  $\text{CH}_4$ , the rotational motions are much more hindered in liquid  $\text{SiH}_4$ .

## 1. INTRODUCTION

The study of the molecular motions appears particularly attractive in the case of tetrahedral molecules  $XY_4$ , the theoretical analysis being favoured by the high structural symmetry. Indeed, the dynamical behaviour of methane molecules in the various phases has been quite extensively investigated by means of a variety of techniques, including the neutron inelastic scattering (NIS).

Silane ( $SiH_4$ ) differs from the isostructural  $CH_4$  molecule in mass, moment of inertia, octupole moment, and exhibits an increased deviation with respect to the spherical shape. When going from methane to silane one changes mainly the orientation-dependent intermolecular forces and the spacing between the (free) rotational levels. It seems thus worthwhile to study how far both systems behave similarly.

Until now silane has not been investigated up to the same extent as methane, so that important information is still lacking. NIS measurements have nevertheless been performed by us some years ago on the gaseous and liquid states [1], and more recently on the various solid phases by VANDERHAEGHEN et al. [2].

As in the case of  $CH_4$ , the neutron data on gaseous  $SiH_4$  were found to be in rather excellent agreement with theoretical predictions based either on an exact quantum mechanical treatment of molecular rotations [3,4] or on the approximate treatment of AGRAWAL and YIP [5] which makes use of classical rotational correlation functions; some slight differences between experimental and computed results could fully be explained by multiple scattering contributions [6].

For solid  $SiH_4$ , the neutron spectra could also be analyzed in terms of molecular motions, at least qualitatively; the scarcity of structural information however prevented a too detailed analysis. In what concerns the nature of the rotational motions, random reorientations about the threefold molecular axes predominate in the plastic phase (above  $T_\lambda = 63.5$  K), the residence times being much too small to allow the existence of well defined periodic motions; however, below  $T_\lambda$  the librations are dominant.

The comparison of the presently available information on methane and silane in their solid state discloses that both systems behave quite differently [2]. Such a conclusion was already drawn from our first neutron study [1] on liquid  $SiH_4$ : In the case of liquid  $CH_4$  AGRAWAL and YIP [7] were

able to fit the neutron incoherent scattering cross-section reasonably well combining the DESAI and YIP translational model [8] (in which a molecule vibrates in a parabolic potential decaying exponentially) with the Langevin rotational diffusion model [9] (which describes free rotations interrupted after a certain time by small-step angular displacements). In addition AGRAWAL [10] and RAO et al. [11] have shown that the agreement between experimental and theoretical cross-sections is significantly improved when multiple scattering corrections are included. In the case of liquid  $\text{SiH}_4$  we did not succeed to fit our neutron spectra using the model described above. In our analysis the parameters were derived from the values found by Agrawal and Yip for  $\text{CH}_4$  by applying the law of corresponding states, as suggested by VINEYARD [12] and SEARS [13].

Our failure led several authors to reanalyze our data on liquid  $\text{SiH}_4$  in terms of different models. CVIKL [14] using the simple translational diffusion and the extended rotational diffusion models could obtain only a moderate agreement with the experimental results. A much better agreement was reached by THAPER and DASANNACHARYA [15] describing the translational motions within the Langevin diffusion model, and treating the rotations as free during a certain time after which they switch into a simple diffusive behaviour; however, the parameter  $\tau_0$  giving the time during which the rotations are free appears to us to have rather surprisingly low values. In that work multiple scattering corrections were included, but only a small part of our data were used.

For the self-diffusion coefficient  $D$ , involved in all these three models, no direct experimental determination exists in the case of silane. Recently VAN LOEF [16] has studied the temperature and density dependence of the self-diffusion coefficient in quasi-spherical molecular liquids; for  $\text{SiH}_4$  a value of  $D = 3.5 \times 10^{-5} \text{ cm}^2\text{s}^{-1}$  at  $T = 137 \text{ K}$  has been predicted from the measured viscosity  $\eta$ . This result is consistent neither with the value  $2.04 \times 10^{-5} \text{ cm}^2\text{s}^{-1}$  derived by us applying the law of corresponding states to the self-diffusion coefficient of liquid  $\text{CH}_4$ , nor with the value  $4.7 \times 10^{-5} \text{ cm}^2\text{s}^{-1}$  obtained by Thaper and Dasannacharya from the experimental quasielastic width of the neutron spectra at the two smaller scattering angles. Cvikl, however, has obtained from his analysis that value of  $D = 3.5 \times 10^{-5} \text{ cm}^2\text{s}^{-1}$ .

In this paper we present a re-analysis of our whole set of data (six scattering angles, three temperatures) following different models for the translational and rotational motions. Our aim was more particularly to check

whether the best-fitting D parameters agree with the Van Loef predictions, and to what extent a modification of their values would affect the results of Thaper and Dasannacharya.

## 2. THEORETICAL MODELS

The incoherent scattering cross-section per proton is given by

$$\frac{d^2\sigma_{\text{inc}}}{d\Omega d\varepsilon} = \frac{k}{k_0} (a_{\text{H}}^{\text{inc}})^2 S_{\text{inc}}(Q, \omega) \quad , \quad (1)$$

where

$$\vec{Q} \equiv \vec{k}_0 - \vec{k} \quad , \quad (2a)$$

$$\varepsilon \equiv h\omega = E_0 - E = \frac{\hbar^2}{2m} (k_0^2 - k^2) \quad . \quad (2b)$$

$\vec{k}_0$  ( $\vec{k}$ ) represents the incident (scattered) neutron wave vector,  $E_0$  ( $E$ ) the incident (scattered) neutron energy,  $a_{\text{H}}^{\text{inc}}$  the proton bound incoherent scattering length,  $\vec{Q}$  the scattering vector,  $\varepsilon$  the energy transfer, and  $m$  the neutron mass. The other notations used in this paper include : the temperature  $T$ , the molecular mass  $M$ , the distance  $b$  from the proton to the molecular center of mass, the moment of inertia  $I$ , and the macroscopic self-diffusion constant  $D$ .

Following AGRAWAL and YIP [5] the incoherent scattering law is approximated by

$$S_{\text{inc}}(Q, \omega) = \frac{1}{2\pi\hbar} \exp\left[-Q^2\gamma\right] \exp\left[\frac{\hbar\omega}{2k_{\text{B}}T}\right] \exp\left[-\frac{\hbar^2Q^2}{8Mk_{\text{B}}T} G(T)\right] \\ \times \int_{-\infty}^{\infty} dt e^{-i\omega t} e^{-\frac{Q^2}{2} W(t)} \quad . \quad (3)$$

The first exponential is a Debye-Waller factor taking into account the inner vibrations of the molecule (in [1] we have computed  $\gamma$  to be \*  $8.6 \times 10^{-19} \text{ cm}^2$  for  $\text{SiH}_4$ ), the second exponential is the detailed-balance factor compensating for the use of classical correlation functions, while the exponential involving  $G(T)$  is a generalized recoil factor.  $G(T)$  and the width function  $W(t)$

\* In [1] the values of the vibrational parameters  $\gamma_{\text{XX}}$  and  $\gamma_{\text{YY}}$  were given in a wrong order.

can be related to a frequency distribution  $f(\omega)$  :

$$G(T) = 2 \int_0^{\infty} d\omega f(\omega) \frac{\cosh \omega^* - 1}{\omega^* \sinh \omega^*} , \quad (4a)$$

$$W(t) = \frac{\pi}{M} \int_0^{\infty} d\omega f(\omega) \frac{1 - \cos \omega t}{\omega \sinh \omega^*} , \quad (4b)$$

$$\omega^* = \hbar\omega / 2k_B T . \quad (4c)$$

The frequency distribution  $f(\omega)$ , and thus also  $G(T)$  and  $W(t)$  have contributions from molecular translations and rotations. According to Agrawal and Yip the rotational width function  $W_R(t)$  involves only the first order rotational correlation function  $F_1(t)$

$$W_R(t) = \frac{2b^2}{3} \left[ 1 - F_1(t) \right] . \quad (5)$$

Among the many models, developed in order to describe the translational and rotational motions, we have taken into consideration in the analysis of our NIS data :

- for the translations

- (1) the Langevin diffusion model,
- (2) the oscillation-diffusion interpolation model of Desai and Yip.

- for the rotations

- (1) the Langevin rotational diffusion model,
- (2) the free rotation switching into simple rotational diffusion model.

The explicit formulae are given in the Appendix.

The translational and rotational frequency distributions which are needed to evaluate  $G(T)$  can be approximately derived from the high-temperature form of Eq (4b) which gives

$$f(\omega) = \frac{M}{\pi k_B T} \int_0^{\infty} dt \cos \omega t \left[ \frac{d^2 W(t)}{dt^2} \right] . \quad (6)$$

### 3. DISCUSSIONS

Our results on liquid  $\text{SiH}_4$  at 98, 123, and 137 K are shown in fig. 1. The experimental points are corrected for background and energy dependence of the detector efficiency; the cross-section scale is found by area-normalization to model predictions. For further experimental details we refer to [1].

#### 3.1. Model I

This is the model used by Thaper and Dasannacharya [15] which combines the translational model (1) with the rotational model (2). Three parameters enter in the model :  $D$ ,  $\tau_o$ , and  $D_r$  (rotational diffusion coefficient). The rotational parameters are more conveniently introduced as reduced variables :

$$\tau_o^* = (k_B T/I)^{1/2} \tau_o \quad , \quad (7a)$$

$$D_r^* = (I/k_B T)^{1/2} D_r \quad . \quad (7b)$$

The values of the translational diffusion coefficient  $D$  as derived from fig. 1 of [16] are given in the fourth column of table I (for the lowest-temperature value an extrapolation was required). Using these  $D$ -values and varying the rotational parameters  $D_r^*$  and  $\tau_o^*$ , the best agreement we could reach with the experimental data were obtained for the values given in the second and third columns of table I. In figs. 1a, 1b, and 1c the solid line represents the total scattering cross-section as computed (for each temperature  $T$  and scattering angle  $\varphi$ ) from the parameters from table I, after convolution with the instrument resolution. The dashed curves represent the multiple scattering contributions as computed from the Monte Carlo COPLEY program [17].

As seen on figs. 1a, 1b, and 1c the agreement between the model predictions and the experiment is satisfactory but not perfect. Changing  $D_r^*$  and  $\tau_o^*$  it was not possible to improve the agreement at small  $\varphi$  without deteriorating the agreement at high  $\varphi$  (and vice versa). The quality of this agreement is of the same order as the one obtained by Thaper and Dasannacharya [15] who fitted neutron spectra at only two scattering angles ( $45.3^\circ$  and  $90.3^\circ$ ) and two temperatures (137 K and 98 K). As values of the rotational parameters they

obtained  $\tau_o^* = 0.3 \pm 0.1$ ,  $D_r^* = 0.5 \pm 0.16$  (at 137 K), and  $\tau_o^* = 0.2$ ,  $D_r^* = 0.167$  (at 98 K); within the uncertainties the results of our fits are consistent with these values.

It may seem somewhat surprising that independent fits using different D-values give nearly the same values for the best-fitting rotational parameters. But in fact the results at large  $\nu$  appear rather insensitive to D. We have moreover verified that changing the D-values in both directions could not improve significantly the agreement in the whole  $\nu$ -range.

In fig. 2 the solid lines represent the rotational correlation functions  $F_1(t^*)$  obtained from our fits; the dashed curve shows the free rotation behaviour. It is interesting to notice that in liquid  $\text{SiH}_4$  the time during which "the rotations are free" is small, even at the highest temperature, in comparison with similar values in liquid  $\text{CH}_4$  ( $\tau_o^* = 1$ , GORDON [18]) and in liquid  $\text{NH}_3$  ( $\tau_o^* = 0.55$ , THAPER et al. [19]).

### 3.2. Model II

Here we combine the translational model (1) with the rotational model (1) which involves the parameters D and  $\xi$ , respectively. Again the rotational parameter is more conveniently written as a reduced variable

$$\xi^* = \xi (Ik_B T)^{-1/2} \quad (8)$$

At each temperature an estimate for  $\xi^*$  has been obtained forcing the Langevin rotational correlation function to reproduce at best the corresponding function deduced from Model I, and more particularly its long-time behaviour which is important to predict correctly the scattering cross-section in the quasielastic region. Such Langevin  $F_1(t^*)$  are shown as dotted curves in fig. 2. With these  $\xi^*$  and the Van Loef D values we have calculated the corresponding total scattering cross-sections. Some results are given on the left side of fig. 3 for  $T = 137$  K and three scattering angles  $\nu$ .

The agreement between the predictions and the experimental data is not bad, though nevertheless slightly less satisfactory than for Model I (fig. 1a), mostly in the inelastic region. This deficiency of Model II, also present at the other temperatures, is to be attributed to an overestimation of the Langevin  $F_1(t^*)$  in the short-time region (fig. 2).

### 3.3. Model III

As in our first analysis of the silane data [1] we can also combine the oscillation-diffusion interpolation description of the translational motions with the Langevin rotational diffusion model. Computations require here values for three parameters : the rotational friction constant  $\xi$ , the characteristic translational frequency  $\omega_0$ , and the relaxation time  $\tau_1$ . In order to obtain good predictions in the quasielastic region, the  $\xi^*$ -values were again taken from fig. 2. Hoping to enhance the scattering cross-section in the inelastic region we have chosen  $\omega_0 = 2 \times 10^{13} \text{ s}^{-1}$  ( $\hbar\omega_0 = 13.2 \text{ meV}$ ). The relaxation time can then be derived using the relation

$$\tau_1 = \frac{k_B T}{M\omega_0^2 D} ; \quad (9)$$

with the Van Loef values of the self-diffusion coefficient we finally get  $\tau_1 = 0.25 \times 10^{-13} \text{ s}$  at 137 K. A selection of results are presented at the right side of fig. 3 for  $T = 137 \text{ K}$ . The agreement with the experimental results is not bad in the quasielastic region, but it is found that this model predicts a too strong  $Q$ -dependence of the inelastic cross-section.

### 4. CONCLUSIONS

Both Models II and III, which make use of the Langevin diffusion description of the rotations, disclose deficiencies in the inelastic region of the scattering cross-sections. However, with respect to our first attempt to fit the liquid silane neutron data [1], predictions based on Model III have been considerably improved; this result is due partly to a better choice of the rotational friction constant ( $\xi^* = 4.0$  at 137 K instead of 0.5), and partly to the fact that the multiple scattering contribution is taken into account.

Model I, already used by Thaper and Dasannacharya [15], gives definitely the best overall agreement to our experimental data as a result of the better behaviour of the rotational correlation functions at short times; the persisting slight differences could probably be removed by a more complex description of the translational motions. From our fits we have derived, at three temperatures, the times  $\tau_0$  during which the rotations are free as well as the rotational

diffusion coefficients  $D_r$ ; as expected, both are found to increase with the temperature. For liquid silane, where the scattering cross-section in the quasielastic region is strongly influenced by the long tail of the rotational correlation function, it is found that the values of the rotational parameters which give a best fit are not very sensitive to the chosen value of the self-diffusion coefficient. On the other hand, the present data do not allow to extract accurate  $D$ -values, and hence to check the Van Loef predictions. Finally, with respect to liquid methane, it appears that the rotational motions are much more hindered in liquid  $\text{SiH}_4$ .

## APPENDIX - WIDTH FUNCTION AND FREQUENCY DISTRIBUTION

## Translational motions

## (1) Langevin diffusion model

$$W_T(t) = 2D \left\{ t - \tau \left[ 1 - \exp(-t/\tau) \right] \right\},$$

$$\tau = MD/k_B T,$$

$$f_T(\omega) = \frac{2}{\pi} \frac{1/\tau}{(1/\tau)^2 + \omega^2}.$$

## (2) Oscillation-diffusion interpolation model

$$W_T(t) = \frac{2k_B T}{M} \left\{ \frac{\omega_o^2 - 1/\tau_1^2}{\omega_o^4} + \frac{t}{\omega_o^2 \tau_1} + \frac{\exp(-t/2\tau_1)}{\omega_o^4} \right. \\ \left. \times \left[ \left( \frac{1}{8\tau_1^3 \Omega} - \frac{3\Omega}{2\tau_1} \right) \sin \Omega t - \left( \omega_o^2 - \frac{1}{\tau_1^2} \right) \cos \Omega t \right] \right\},$$

$$\Omega^2 = \omega_o^2 - \frac{1}{4\tau_1^2},$$

$$f_T(\omega) = \frac{2}{\pi} \frac{\omega_o^2/\tau_1}{(\omega^2 - \omega_o^2) + (\omega/\tau_1)^2}.$$

The given expressions for  $f_T(\omega)$  have the correct normalization to unity [7].

## Rotational motions

$$W_R(t) = \frac{2b^2}{3} \left[ 1 - F_1(t) \right].$$

## (1) Langevin rotational diffusion model

$$F_1(t) = \exp \left\{ \frac{2 I k_B T}{\xi^2} \left[ 1 - \frac{\xi t}{I} - \exp \left( - \frac{\xi t}{I} \right) \right] \right\},$$

$$\xi = k_B T / D_r.$$

(2) Free rotation switching into simple rotational diffusion model

$$F_1(t) = \begin{cases} \frac{1}{3} + \frac{2}{3} \left(1 - \frac{k_B T t^2}{I}\right) \exp\left(-\frac{k_B T t^2}{2I}\right) & \text{for } t \leq \tau_0 \\ F_1(\tau_0) \exp\left[-D_r (t - \tau_0)\right] & \text{for } t \geq \tau_0 \end{cases}$$

The first expression is valid for a spherical top molecule.

$f_R(\omega)$  can be derived using Eq (6); it can be checked that in both cases this function is correctly normalized to  $M/4\pi$  [7].

## REFERENCES

- [1] S. HAUTECLER and P. VORDERWISCH, Neutron Inelastic Scattering (IAEA, Vienna, 1972) 489.
- [2] J. VANDERHAEGHEN, W. WEGENER, S. HAUTECLER and L. VAN GERVEN, Physica B 95 (1978) 163.
- [3] A. RAHMAN, J. Nucl. Energy A 13 (1961) 128.
- [4] G.W. GRIFFING, Phys. Rev. 124 (1961) 1489; Phys. Rev. 127 (1962) 1179.
- [5] A.K. AGRAWAL and S. YIP, Phys. Rev. 171 (1968) 263.
- [6] P. VORDERWISCH, unpublished work.
- [7] A.K. AGRAWAL and S. YIP, Nucl. Sci, Engng 37 (1969) 368.
- [8] R.C. DESAI and S. YIP, Phys. Rev. 166 (1968) 129.
- [9] W.A. STEELE, J. Chem. Phys. 38 (1963) 2404, 2411.
- [10] A.K. AGRAWAL, Phys. Rev. A4 (1971) 1560.
- [11] K.R. RAO, B.A. DASANNACHARYA, and S. YIP, J. Phys. C. Solid State Phys. 4 (1971) 2725.
- [12] G.H. VINEYARD, Phys. Rev. 119 (1960) 1150.
- [13] V.F. SEARS, Can. J. Phys. 44 (1966) 867.
- [14] B. CVIKL, Phil. Mag. 28 (1973) 1353.
- [15] C.L. THAPER and B.A. DASANNACHARYA, Pramana 6 (1976) 383.
- [16] J.J. VAN LOEF, Physica B 95 (1978) 34.

[17] J.R.D. COPLEY, *Computer Phys. Commun.* 7 (1974) 289; 9 (1975) 59, 64.

[18] R.G. GORDON, *J. Chem. Phys.* 43 (1965) 1307.

[19] C.L. THAPER, B.A. DASANNACHARYA, and P.S. GOYAL, *Pramana* 2 (1974) 148.

TABLE I

Values of the reduced rotational diffusion coefficient  $D_r^*$  and of the reduced time  $\tau_o^*$  during which the rotations are free in liquid  $\text{SiH}_4$  deduced from an analysis of the neutron scattering cross-sections, using for the self-diffusion coefficient  $D$  the values of the 4th column. Other sets of  $D$ -values used in similar analyses by different authors are also given in columns 5-7.

T(K)	$D_r^*$	$\tau_o^*$	$D(10^{-5} \text{ cm}^2 \text{ s}^{-1})$			
			this work and [16]	[1]	[14]	[15]
98	0.2	0.25	1.1	0.35	1.55	1.5
123	0.35	0.35	2.54	1.24		
137	0.45	0.45	3.5	2.04	3.5	4.7

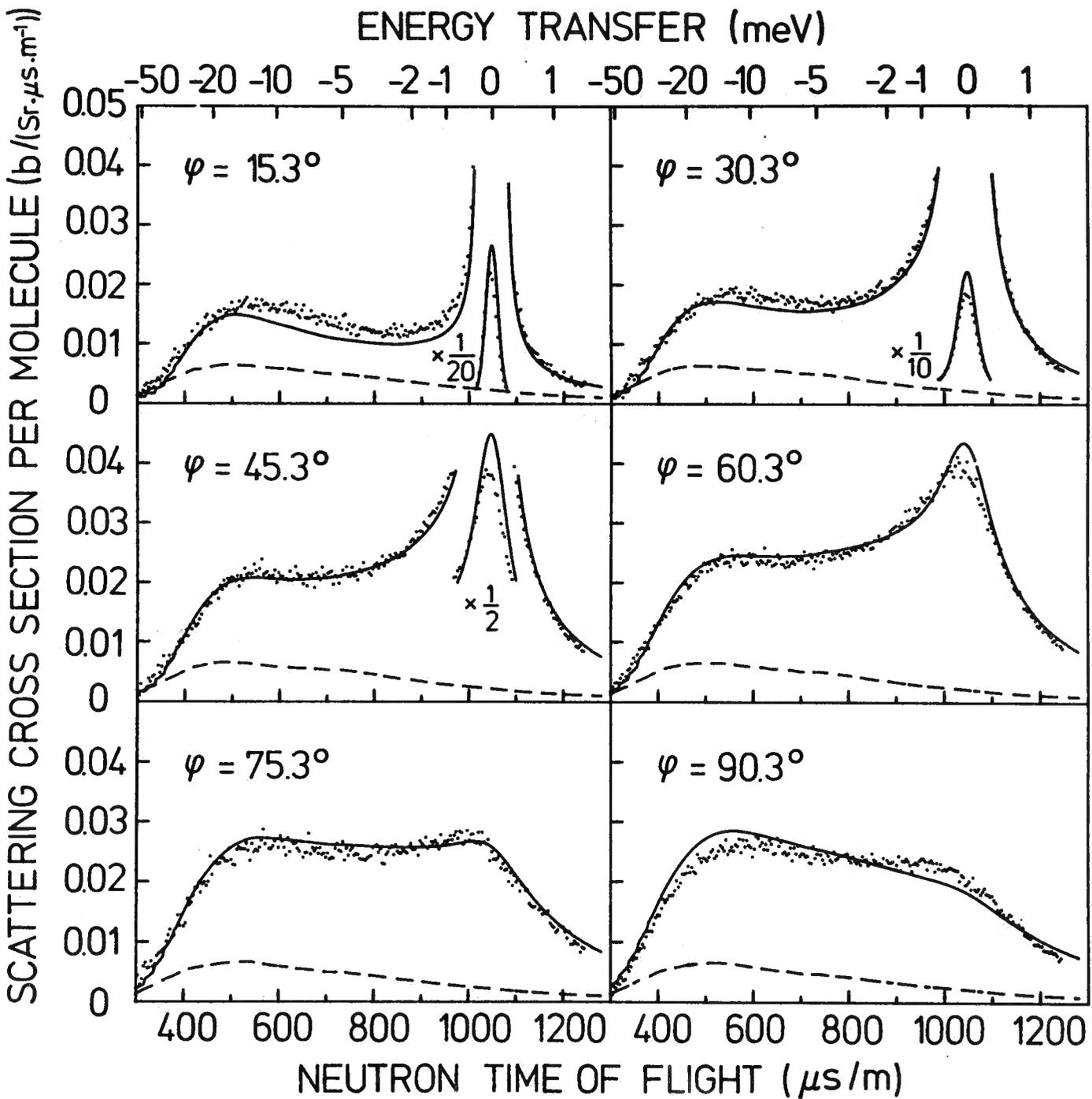


Fig. 1a. Time-of-flight scattering cross-sections per molecule for liquid SiH<sub>4</sub> at 137 K. The solid lines are the total cross-sections computed from Model I using the parameters given in table I; the dashed curves represent the multiple scattering contributions. The experimental points are area-normalized to the theory at each scattering angle  $\psi$ .

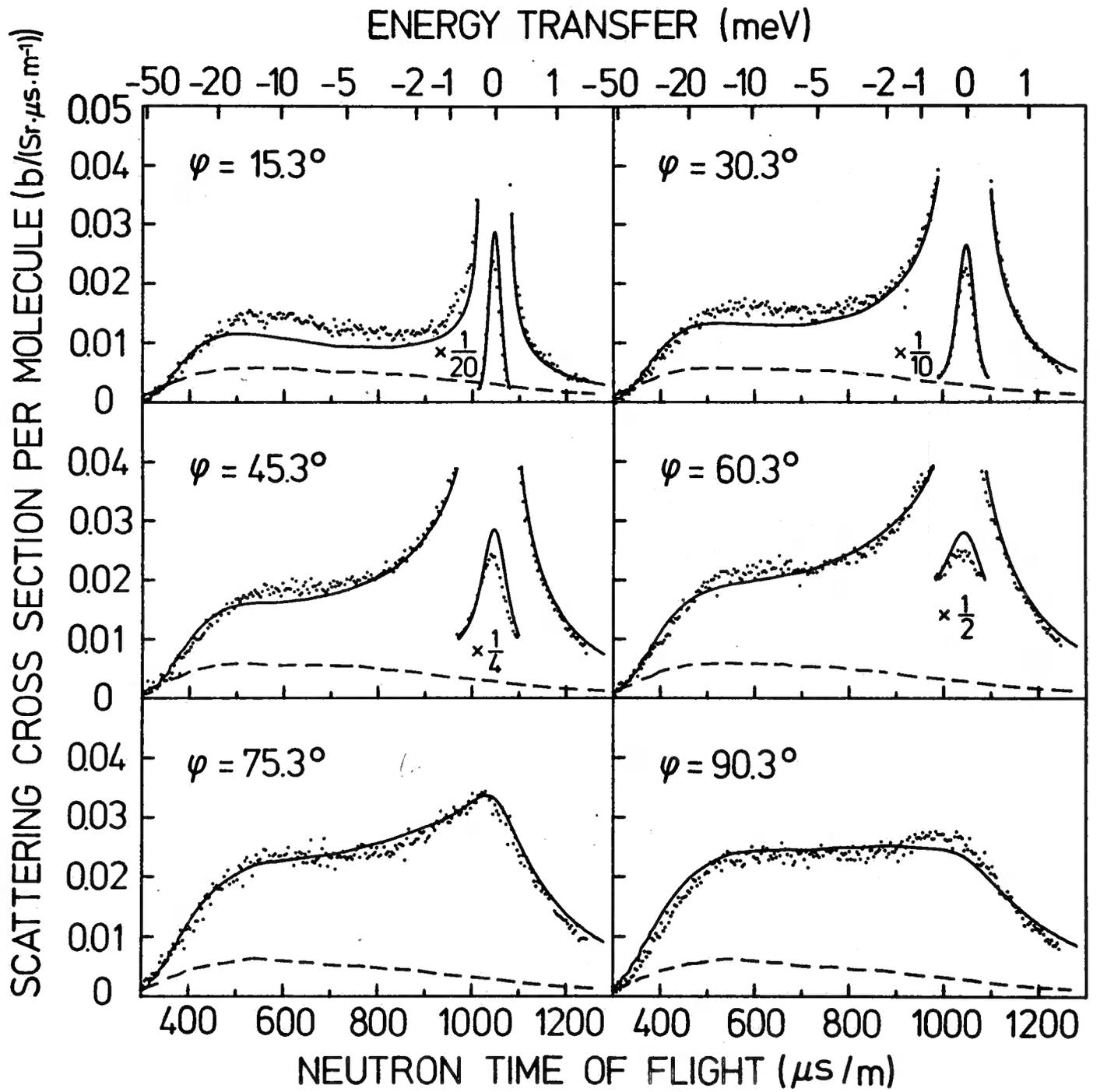


Fig. 1b. Time-of-flight scattering cross-sections per molecule for liquid  $\text{SiH}_4$  at 123 K from Model I (for further details see caption to fig. 1a).

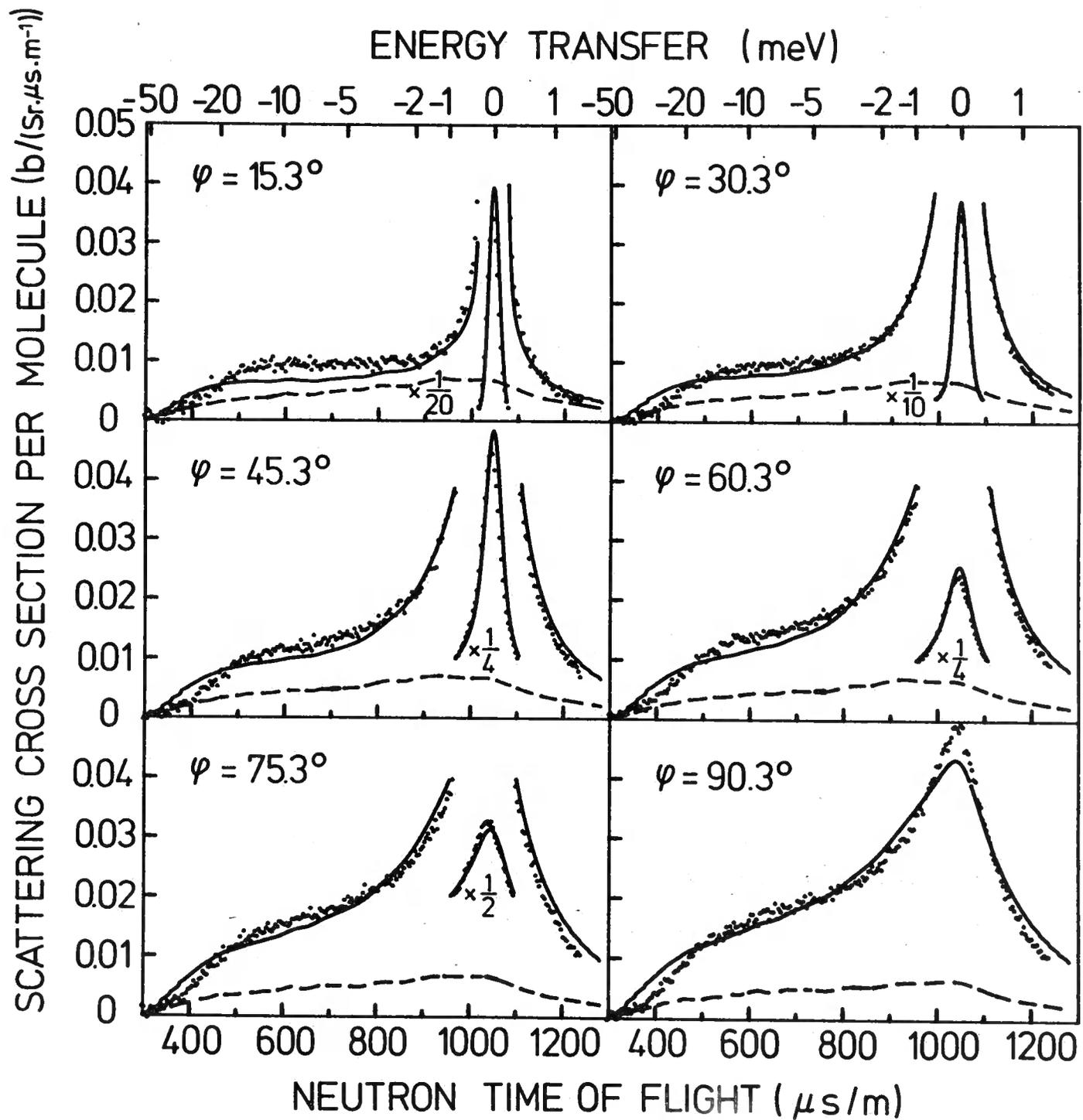


Fig. 1c. Time-of-flight scattering cross-sections per molecule for liquid SiH<sub>4</sub> at 98 K from Model I (for further details see caption to fig. 1a).

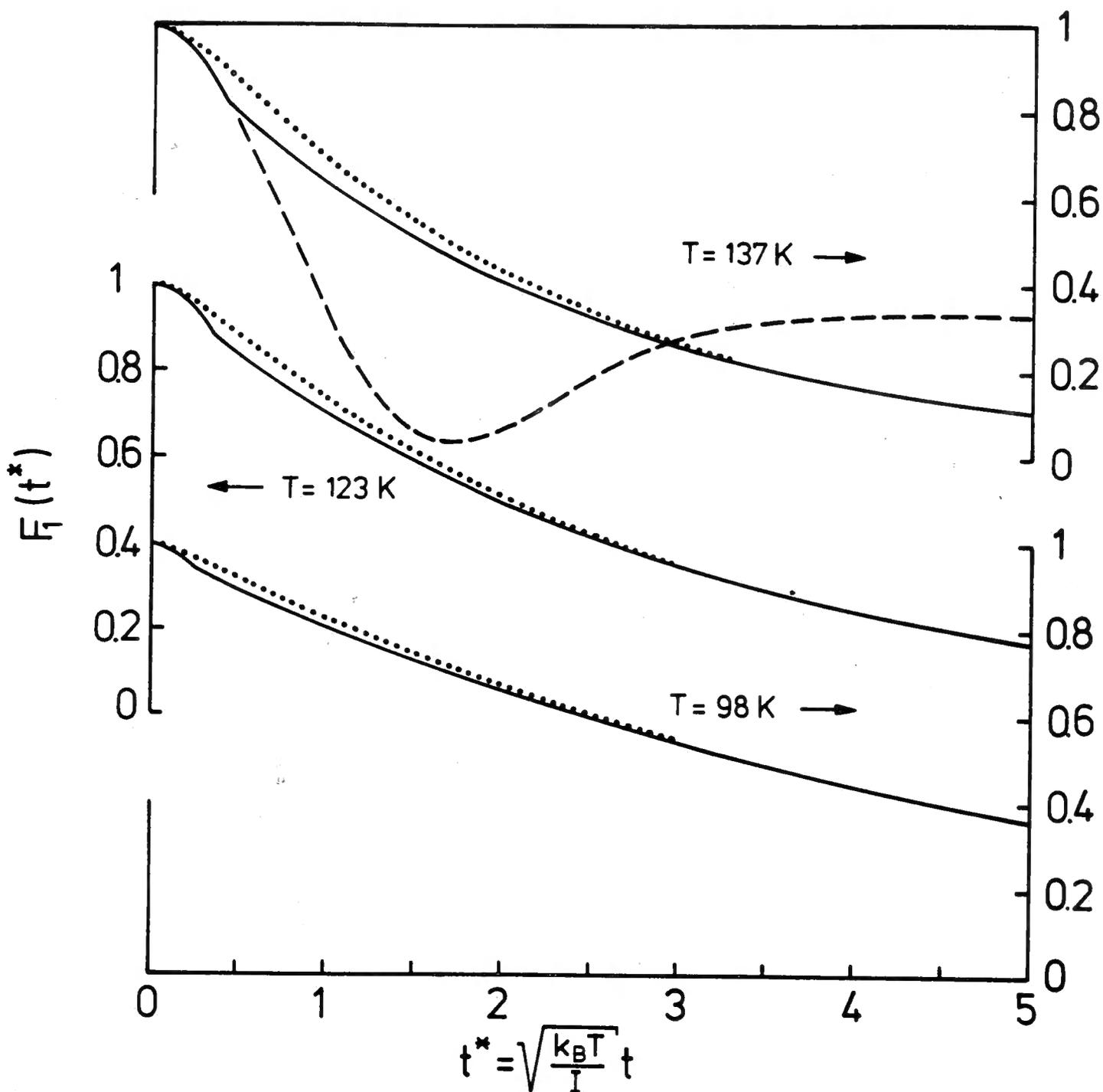


Fig. 2. Rotational correlation functions  $F_1(t^*)$  for liquid  $\text{SiH}_4$  at various temperatures  $T$ . The solid lines are deduced from fits to the neutron scattering data using Model I (figs. 1a, 1b, 1c). The dashed curve shows the free rotation behaviour. The dotted curves are the Langevin rotational correlation functions used in computations with Models II and III ( $\xi^* = 4.0, 5.3$  and  $9.5$  at  $137, 123,$  and  $98$  K, respectively).

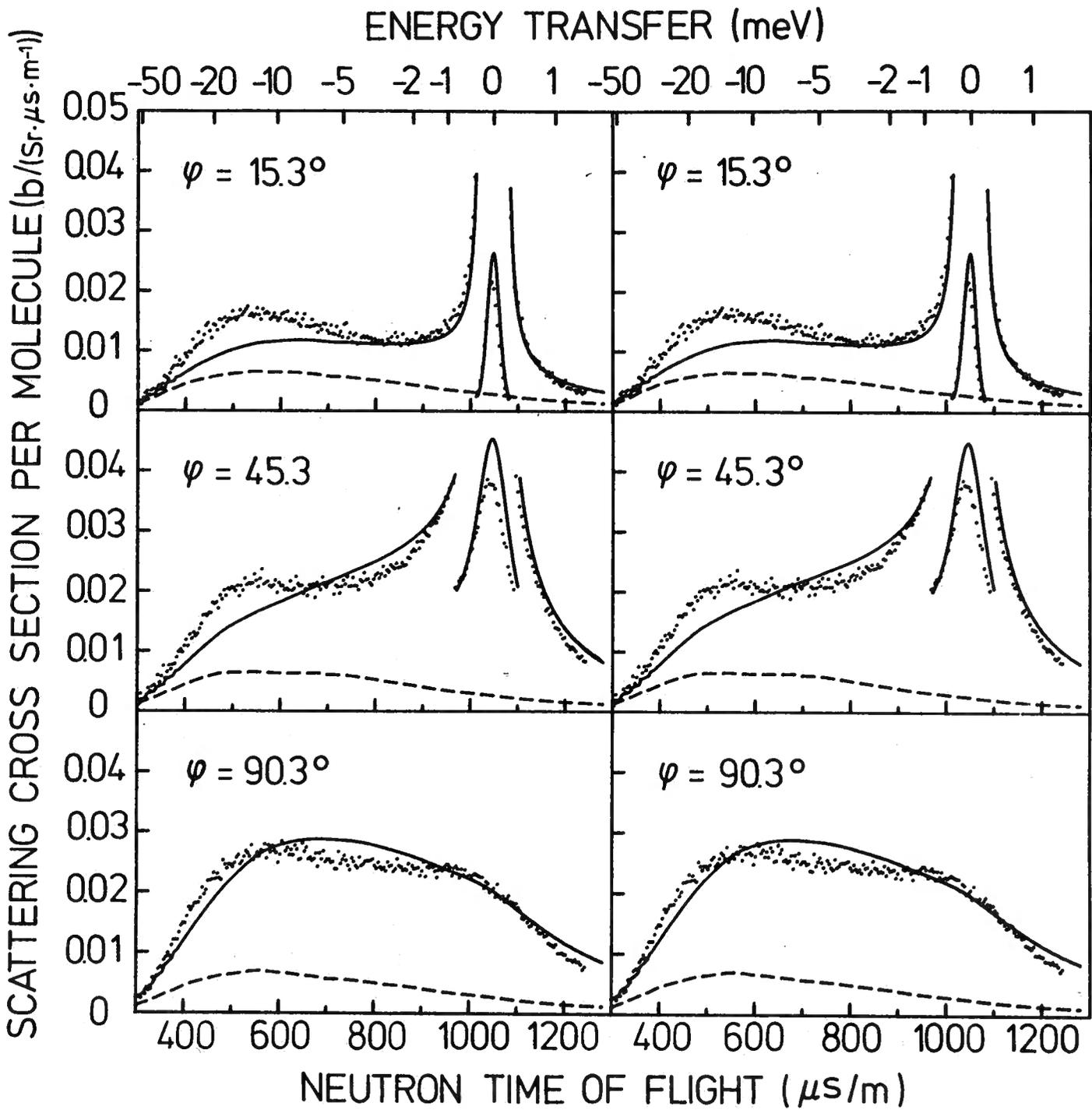


Fig. 3. Time-of-flight scattering cross-sections per molecule for liquid SiH<sub>4</sub> at 137 K. The solid lines are the total cross-sections computed from Model II (left side) and Model III (right side) using the Langevin rotational correlation function shown on fig. 2 and other parameters given in the text; the dashed curves represent the multiple scattering contributions.

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