NOTIZEN

Quantum Mechanical Calculation of Collision Integrals for HD

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The magnetic Senftleben-Beenakker effect of the viscosity is mainly determined by two collision integrals of the linearized quantum mechanical Waldmann-Snider collision term, viz. by the relaxation coefficient of the tensor polarization of the molecular rotational angular momenta and by the coefficient which couples the friction pressure tensor and the tensor polarization. Starting from a simple nonspherical potential for HD, the scattering amplitude is evaluated analytically in first order distorted wave Born approximation and the two collision integrals are calculated for room temperature. A fairly good agreement with experimental values is found.

In a magnetic field, the viscosity of a dilute gas of polyatomic molecules becomes a field dependent fourth rank tensor. This phenomenon is known as the "Senftleben-Beenakker effect (SBE)[1,2]. The theory of the SBE starts from the linearized Waldmann-Snider equation[3,4] for the one particle distribution function which is an operator with respect to the degenerate states (magnetic quantum numbers). For the non-equilibrium distribution function of linear molecules it proves sufficient to take into account only the tensor polarization together with the friction pressure tensor. The tensor polarization of the rotational angular momenta of the molecules is proportional to the mean value $\langle \overline{\boldsymbol{J}\boldsymbol{J}} \rangle$ of the irreducible 2nd rank tensor built up from the rotational angular momentum operator $\boldsymbol{J}$. The kinetic theory which is described elsewhere[5,6] then yields the following saturation values of the SBE:

$$(\Delta\eta_3/\eta_1)_{\text{sat}} = 2\,(\Delta\eta_2/\eta_1)_{\text{sat}} = -\,\omega_{\eta\text{T}}^2/\omega_\eta\,\omega_{\text{T}}\,. \tag{1}$$

The viscosity coefficients η_1, η_2, η_3 are those of De Groot-Mazur[7]. The brackets ω_η, ω_{T} of the linearized Waldmann-Snider collision operator are the relaxation coefficients of the friction pressure tensor and the tensor polarization, respectively. The cross coefficient $\omega_{\eta\text{T}}$ describes the coupling of the friction pressure tensor with the tensor polarization. Instead of the ω'..s, equivalent effective cross sections σ.. are also in use, defined by

$$\omega\,.\,. = n_0\,v_0\,\sigma\,.\,.\,. \tag{2}$$

In Eq. (2), n_0 is the equilibrium particle density and $v_0 = (8\,k_{\text{B}}\,T_0/\pi\,m_{12})^{1/2}$ is a mean thermal velocity. For the effective cross sections one obtains[5,8]

$$\sigma_\eta = \tfrac{2}{15}\,[[(\gamma^4 + \gamma'^4 - \gamma^2\,\gamma'^2\,(3\cos^2\chi - 1))\,\sigma]]\,, \tag{3}$$

$$\sigma_{\text{T}} = \tfrac{2}{5}\,\langle J^2(J^2 - \tfrac{3}{4})\rangle_0^{-1}\,[[\sigma_{\text{re}}^{(1)} + \sigma_{\text{re}}^{(2)}]]\,, \tag{4}$$

$$\sigma_{\eta\text{T}} = \frac{2\sqrt{2}}{5}\,\langle J^2(J^2 - \tfrac{3}{4})\rangle_0^{-1/2}\,[[\gamma^2\,\sigma_{\text{ee}}^{(1)} - \gamma'^2\,\sigma_{\text{e}'\text{e}'}^{(1)}]]\,. \tag{5}$$

In Eqs. (3), (4), and (5), γ' and γ are the dimensionless relative velocities before and after the collision (unit vectors $\boldsymbol{e}'$ and $\boldsymbol{e}$ of their direction), connected by the energy conservation, and $\chi = \arccos(\boldsymbol{e}\cdot\boldsymbol{e}')$ is the angle of deflection. The bracket symbol $[[\ldots]]$ denotes a weighted averaging over γ and the postcollisional rotational quantum numbers j_1, j_2 as well as an integration over χ and a summation over the precollisional rotational quantum numbers j_1', j_2':

$$[[F]] = 2\,\pi\,Q^{-2} \sum_{j_1 j_2,\, j_1' j_2'} \iint (2\,j_1+1)\,(2\,j_2+1)\;\cdot \exp\{-\gamma^2 - \varepsilon(j_1) - \varepsilon(j_2)\}\,\gamma^3\,F\,\sin\chi\,\mathrm{d}\chi\,\mathrm{d}\gamma\,. \tag{6}$$

In Eq. (6), $F = F(\gamma, \chi \,|\, j_1\,j_2, j_1'\,j_2')$ is any function of γ, χ, and the rotational quantum numbers, $\varepsilon(j) = \hbar^2\,j(j+1)/2\,\Theta\,k_{\text{B}}\,T_0$ is the dimensionless rotational energy and Q is the rotational partition function. The decisive quantities in Eqs. (3), (4), and (5) are the "cross sections" σ, $\sigma_{\text{ee}}^{(1)}$, $\sigma_{\text{e}'\text{e}'}^{(1)}$, $\sigma_{\text{re}}^{(1)}$ and $\sigma_{\text{re}}^{(2)}$. They are defined by

$$\sigma = (2\,j_1+1)^{-1}\,(2\,j_2+1)^{-1}\,\frac{\gamma'}{\gamma}\,\text{tr}_1\,\text{tr}_2\,\{a^{j_1 j_2,\, j_1' j_2'}\,a^{\dagger\, j_1' j_2',\, j_1 j_2}\}\,, \tag{7}$$

$$\sigma_{\text{ee}}^{(1)} = (2\,j_1+1)^{-1}(2\,j_2+1)^{-1}\,\frac{\gamma'}{\gamma}\,\text{tr}_1\,\text{tr}_2\left\{a^{j_1 j_2,\, j_1' j_2'}\,a^{\dagger\, j_1' j_2',\, j_1 j_2}\sqrt{\frac{15}{2}}\,\overline{\boldsymbol{J}^{(1)}\,\boldsymbol{J}^{(1)}} : \overline{\boldsymbol{e}\,\boldsymbol{e}}\right\}, \tag{8}$$

(analogously for $\sigma_{\text{e}'\text{e}'}^{(1)}$)

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[1] H. Senftleben, Phys. Z. **31**, 822, 961 [1930].

[2] J. J. M. Beenakker, G. Scoles, H. F. P. Knaap, and R. M. Jonkman, Phys. Letters **2**, 5 [1962]. — For a review see J. J. M. Beenakker and F. R. McCourt, Ann. Rev. Phys. Chem. **21**, 47 [1970].

[3] L. Waldmann, Z. Naturforsch. **12 a**, 660 [1957]; **13 a**, 609 [1958].

[4] R. F. Snider, J. Chem. Phys. **32**, 1051 [1960].

[5] F. R. McCourt and R. F. Snider, J. Chem. Phys. **47, 4117** [1967].

[6] S. Hess and L. Waldmann, Z. Naturforsch. **26 a**, 1057 [1971].

[7] S. R. De Groot and P. Mazur, Non-Equilibrium Thermodynamics, North-Holland Publ. Co., Amsterdam 1962.

[8] W. E. Köhler, Thesis, Erlangen 1971.

and

$$\sigma_{\mathrm{re}}^{(1)} = \frac{15}{2}\,(2\,j_1+1)^{-1}(2\,j_2+1)^{-1}\,\frac{\gamma'}{\gamma}\,\mathrm{tr}_1\,\mathrm{tr}_2\,\{\overline{\boldsymbol{J}^{(1)}\,\boldsymbol{J}^{(1)}}\,a^{j_1j_2,\,j_1'j_2'} : [a^{\dagger\,j_1'j_2',\,j_1j_2}, \overline{\boldsymbol{J}^{(1)}\,\boldsymbol{J}^{(1)}}]_{-}\}\,. \tag{9}$$

For $\sigma_{\mathrm{re}}^{(2)}$ the $\boldsymbol{J}^{(1)}$ in the commutator term of Eq. (9) has to be replaced by $\boldsymbol{J}^{(2)}$. In Eqs. (7), (8), and (9) the quantity $a^{j_1j_2,\,j_1'j_2'}$ is the single channel scattering amplitude (matrix with respect to the magnetic quantum numbers) describing a collision process $j_1'+j_2' \to j_1+j_2$ and "tr" denotes the trace over magnetic quantum numbers. The differential cross section σ is equal to that for the time reversed scattering process $j_1+j_2 \to j_1'+j_2'$. The quantities $\sigma_{\mathrm{ee}}^{(1)}$, $\sigma_{\mathrm{e'e'}}^{(1)}$ are called "orientation cross sections for the tensor polarization" since they are a measure for the production of certain components of the tensor polarization in a collision [8, 9]. The quantities $\sigma_{\mathrm{re}}^{(1,2)}$ are called "reorientation cross sections for the tensor polarization". They describe the change of a tensorial alignment in a collision. The orientation and reorientation cross sections vanish for a scattering amplitude due to a pure spherical potential. In this "spherical" case, σ_η reduces to $(8/5\,v_0)\,\Omega^{(2,2)}$. The Chapman integral $\Omega^{(2,2)}$ can be taken from the literature [10]. The terms omitted in σ_η are quadratic in the nonsphericity [8, 9] and can surely be neglected for molecules with small nonsphericity.

In order to obtain the cross sections (8), (9), one has to calculate the magnetic quantum number dependent scattering amplitude from a nonspherical interaction potential. For molecules with small nonsphericity of the interaction this can be done in first order distorted wave Born approximation (DWBA [11]), if the problem of scattering by the spherical part of the potential can be solved exactly. The nonspherical part of the interaction is then treated in first order perturbation theory. A simplified but often used ansatz [12] for the nonspherical potential which takes into account only one leading term of a series (cf. [9]) is given by

$$V(\boldsymbol{u}_1, \boldsymbol{u}_2, \hat{\boldsymbol{x}}_{12}, r) = V_{\mathrm{sph}}(r) + V_{\mathrm{L}}(r)\,[P_{\mathrm{L}}(\boldsymbol{u}_1\cdot\hat{\boldsymbol{x}}_{12}) + P_{\mathrm{L}}(\boldsymbol{u}_2\cdot\hat{\boldsymbol{x}}_{21})]\,. \tag{10}$$

In Eq. (10), $\boldsymbol{u}_1$, $\boldsymbol{u}_2$ are unit vectors in the direction of the molecular axes and $\boldsymbol{x}_{12} = -\boldsymbol{x}_{21} = \hat{\boldsymbol{x}}_{12}\,r$ is the vector connecting the centers of mass of the molecules, and P_{L} is a Legendre polynomial. For homonuclear molecules such as H_2, D_2 one takes the $L=2$ term. For asymmetric molecules such as HD, HT one has $L=1$. The first order DWBA scattering amplitude can then be written as

$$a^{j_1j_2,\,j_1'j_2'} = a_{\mathrm{sph}}\,\delta^{j_1j_1'}\,\delta^{j_2j_2'}\,P^{j_1j_2} + a_{\mathrm{nonsph}}^{j_1j_2,\,j_1'j_2'}\,, \tag{11}$$

where a_{sph} is the part of the scattering amplitude independent of the rotational and magnetic quantum numbers, $P^{j_1j_2}$ is a two-particle projection operator in rotational angular momentum space [9] and $a_{\mathrm{nonsph}}^{j_1j_2,\,j_1'j_2'}$ is the magnetic quantum number dependent ("nonspherical") part of the scattering amplitude. Because of Eq. (10), only one molecule can undergo a transition in a collision. While a_{sph} is determined by the phase shifts corresponding to V_{sph}, the "nonspherical" part of the scattering amplitude is in essence determined by radial integrals involving the radial scattering solutions of the spherical scattering problem and the potential function $V_{\mathrm{L}}(r)$ (cf. [8]).

For HD ($L=1$) the following model potential is used:

$$V_{\mathrm{sph}}(r) = V_0\,\Theta(r-a)\,; \quad V_1(r) = V_0\,d\,\delta(r-a)\,, \tag{12}$$

with Θ being the Heaviside step function. The form of the potential (12) follows from the assumption (cf. [12]) that the HD–HD potential is equal to the H_2-H_2 potential if considered as a function of the vector connecting the geometrical centers of the molecules. The H_2-H_2 potential is taken as spherical, in particular as a repulsive step of radius a. The distance between the geometrical center and the center of mass for the HD molecule is called d. By transforming the potential into a function of the vector connecting the centers of mass (which is relevant in scattering problems) and truncating the expansion with linear terms in d, the potential functions (12) are obtained. They allow the first order DWBA scattering amplitude to be calculated analytically. The radial integrals become rather simple for the limit $V_0 \to \infty$ which corresponds to the case of "loaded rigid spheres" the scattering of which has also been studied by CURTISS and coworkers [13] with a different method.

With the use of Racah algebra analytic expressions for the orientation- and reorientation cross sections (8), (9) of the tensor polarization are obtained in the form of a Legendre polynomial series. For the calculation of the effective cross sections (4), (5), the γ-integration and the j_1-summation (only inelastic collisions with $\Delta j_1 = \pm 1$ give contributions) have to be performed numerically. Calculations have been made with $a = 2.72$ Å (taken from the viscosity cross section in the classical rigid sphere model [10]) and a d-value of 0.1233 Å which is determined by the masses of the atoms and the internuclear distance in the HD molecule.

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[9] W. E. KÖHLER, S. HESS, and L. WALDMANN, Z. Naturforsch. **25 a**, 336 [1970].

[10] S. CHAPMAN and T. G. COWLING, The Mathematical Theory of Non-Uniform Gases, Cambridge University Press 1964; L. WALDMANN, in: Handbuch der Physik, ed. S. FLÜGGE, Vol. **12**, Springer-Verlag, Berlin 1958.

[11] L. S. RODBERG and L. M. THALER, Introduction to the Quantum Theory of Scattering, Academic Press, New York 1967.

[12] K. TAKAYANAGI, Suppl. Progr. Theor. Phys. **25**, 1 [1963].

[13] G. GIOUMOUSIS and C. F. CURTISS, J. Math. Phys. **3**, 1059 [1962]. — J. J. MUELLER and C. F. CURTISS, J. Chem. Phys. **46**, 1252 [1967].

The results for σ_T and $\sigma_{\eta T}$ at 293 °K are shown in the following table. They are compared with values taken from experiments of HULSMAN et al. [14] on the transverse SBE of viscosity, and, in the case of σ_T, also with recent measurements on the broadening of the depolarized Rayleigh line [15] by GUPTA [16].

σ_T in Å^2	HULSMAN 1970: GUPTA 1971:	2.3 ± 0.07 2.73 ± 0.14	Calc. 2.58
$\sigma_{\eta T}$ in Å^2	HULSMAN 1970:	$\pm 0.29 \pm 0.015$	Calc. 0.55

The agreement of experimental and calculated values for σ_T is good, but the calculated coefficient $\sigma_{\eta T}$ is too high by a factor 1.9.

The simple potential model could be amended by adding an attractive square well to the repulsive core in the $H_2 - H_2$ potential. The nonspherical HD – HD scattering amplitude could then still be calculated analytically. Hopefully, a better value for $\sigma_{\eta T}$ could be obtained and also the temperature dependence of the effective cross sections could be investigated in the range of 50 – 300 °K and compared with the behaviour found in the experiments.

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[14] H. HULSMAN, E. VAN WAASDIJK, A. L. BURGMANS, H. F. P. KNAAP, and J. J. M. BEENAKKER, Physica **50**, 33 1970].

[15] S. HESS, Phys. Letters **29 A**, 108 [1969]; Z. Naturforsch. **24 a**, 1675 [1969]; **25 a**, 350 [1970]; Springer Tracts Mod. Phys. **54**, 136 [1970].

[16] B. K. GUPTA, Thesis, University of Toronto 1971.

Nuclear Level Scheme of Au-199 from Double Neutron Capture in Gold

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Several groups have already investigated the level structure of Au-199 by means of the β-decay of Pt-199 [1–3]. Measurements of the gamma- and conversion electron spectrum as well as coincidence and lifetime experiments led to a rather well established level scheme below 1 MeV. The aim of the present investigation was to get further evidence for this scheme through a high resolution study of the Au-198(n,γ)-reaction and to determine very precise level energies.

The measurements were performed with the bent crystal spectrometer at RISØ [4]. The target was a thin gold foil with a weight of 90 mg, which gave a line width of only 1.25 seconds of arc. The energy calibration of the spectrum was performed using the Kα_1-line of gold [5]. About 500 γ-lines have been obesrved. Most of them are due to the Au-197(n,γ)Au-198 reaction [6]. By making several successive runs it was possible to determine lines of increasing intensity, which correspond to double neutron capture. Because of the very high neutron absorption cross section of Au-198 ($\sigma \approx 25\,000$ barn) and the neutron flux 4×10^{13} ns^{-1} cm^{-2} at the target the saturation intensity of those lines is reached already after about 10 days.

The lines of Au-199 are listed in Table 1. Lines, which could not be assigned unambiguously have also been included but they have been marked with "Z". The energies of some Au-199 lines coincide almost exactly with those of Au-198. The intensities of the 746 keV and 786 keV transitions given in the table include those of the Au-198 satellites. The two low-energy lines of 55 and 77 keV found by BÄCKLIN et al. [2] and OKANO et al. [3] are hidden under two strong peaks of Au-198.

The results of the present experiment agree very well with those of the previous studies [1–3] and define the Au-199 levels with an accuracy of about 1×10^{-5} to 5×10^{-5}. The population of the levels in the (n,γ)-reaction gives further evidence for the spin proposals of Bäcklin and Okano. Taking into account the relatively poor resolution of their germanium detector measurements and the difficult intensity determination in the double capture experiment, the overall agreement of the branching ratios is quite good.

Several new transitions were found in the (n,γ)-reaction. For the extension of the present scheme, however, additional information from other experimets is required.

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[1] K. G. PRASAD, R. P. SHARMA, and B. V. THOSAR, Phys. Rev. **149**, 980 [1966].

[2] A. BÄCKLIN, B. FOGELBERG, and S. G. MALMSKOG, Nucl. Phys. **A 103**, 337 [1967].

[3] K. OKANO, Y. KAWASE, S. UCHARA, and T. HAYASHI, Nucl. Phys. **A 136**, 321 [1969].

[4] H. R. KOCH, H. A. BAADER, D. BREITIG, K. MÜHLBAUER, U. GRUBER, B. P. MAIER, and O. W. B. SCHULT, Neutron Capture Gamma-Ray Spectroscopy, Proceedings of the International Symposium, Studsvik, 11.–15. 8. 1969 (IAEA, Vienna 1969) 65–74.

[5] J. A. BEARDEN, Rev. Mod. Phys. **39**, 78 [1969].

[6] D. BREITIG, Dissertation, Techn. Universität München 1971.

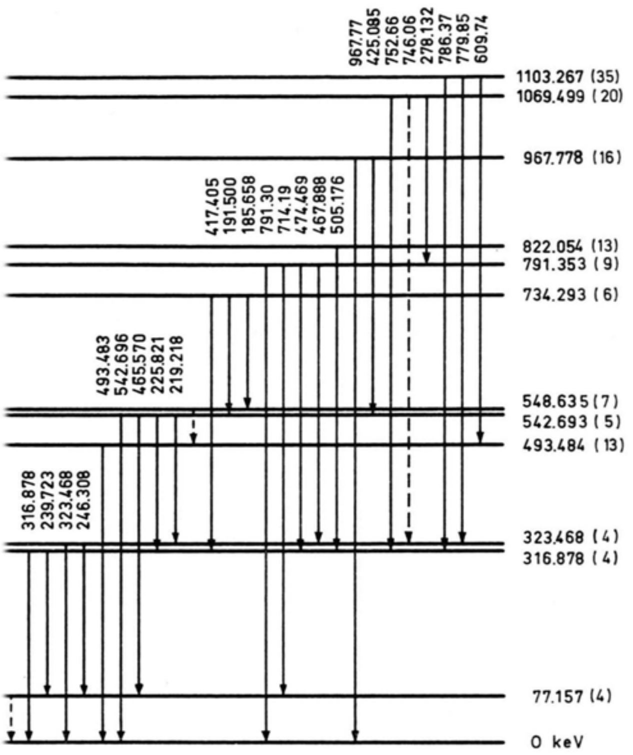

Fig. 1. Level scheme of Au-199.

Table 1. Gamma lines from the reaction Au-198((n,γ). The energy errors dE do not include the calibration error which amounts to 2×10^{-5}.

E_γ relativ		dE/eV	FWHM/keV	I_γ relativ	dI_γ/I_γ (%)
991.97	Z	300	1.2	5.0	30
967.77	*	90	0.59	12.2	20
924.3		350	1.0	3.2	45
901.96		200	0.99	5.5	30
869.5		350	0.92	1.8	50
862.7	?	450	0.90	1.4	80
859.97		250	0.90	1.9	40
819.59		110	0.57	4.4	20
791.30	*	90	0.53	2.6	20
786.37	*	60	0.40	8.5	14
779.85	*	60	0.31	3.7	20
755.04		200	0.70	1.9	35
752.66		70	0.48	3.0	17
746.06	*	40	0.27	5.0	20
735.51		130	0.26	2	35
715.27		150	0.63	1.0	40
714.19	*	45	0.29	3.7	17
706.37		90	0.42	1.3	25
685.28		90	0.40	1.33	20
664.54		50	0.25	2.2	20
660.20	Z, ?	150	0.64	1.1	40
628.09		90	0.49	1.02	30
612.86	Z, C	130	0.46	1.6	35
609.74	*	80	0.31	0.57	25
603.58		200	0.45	0.7	60
600.969		25	0.18	3.2	15
589.847		25	0.17	3.5	14
586.34		80	0.43	0.79	25
562.570		30	0.18	3.1	14
542.696	*	14	0.060	24.2	12
523.783		25	0.13	1.24	20
521.040		30	0.15	1.45	17
505.176	*	12	0.13	8.9	13
493.483	*	13	0.12	41	11
474.469	*	12	0.11	2.26	12
467.888	*	12	0.11	1.90	13
465.570	*	13	0.11	1.53	14
427.166		30	0.20	0.41	25
425.085	*	15	0.11	0.71	20
417.405	*	17	0.10	0.72	20
417.114		25	0.10	0.45	25
323.468	*	6	0.066	3.70	15
318.55	?	45	0.13	0.39	50
316.878	*	5	0.025	40	11
278.132	*	25	0.11	1.04	25
246.308	*	3.5	0.044	30.1	11
239.723	*	4.0	0.042	1.64	12
225.821	*	6	0.036	0.35	17
219.218	*	4.5	0.034	0.91	20
191.5998	*	3.0	0.037	6.1	12
190.863	Z	9	0.049	0.25	30
185.658	*	3.5	0.049	9.9	13
183.324		5	0.048	0.96	17
180.220	Z	6	0.046	0.80	20

Z assignment uncertain   C Complex structure
* line placed in level scheme   ? questionable line

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