

Experimental determination of Boltzmann's constant Ab initio properties of gaseous helium

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Abstract

Recent *ab initio* calculations of the interaction potential of helium, and the effects of the new potentials on the calculated properties of helium gas (density and acoustic virials, viscosity, and thermal conductivity) are reviewed. **To cite this article:** *J.B. Mehl, C. R. Physique 10 (2009).*

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Résumé

Propriétés de l'hélium en phase gazeux : Calculs ab initio. L'article présente une revue de nouveaux calculs du potentiel d'interaction dans l'hélium gazeux, et des résultats obtenus avec ces nouveaux potentiels pour certaines caractéristiques thermophysiques de l'hélium (développement du viriel, viscosité, conductivité thermique...). **Pour citer cet article:** *J.B. Mehl, C. R. Physique 10 (2009).*

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Keywords: Helium; Density virial; Theoretical interatomic potential; Thermal conductivity; Viscosity

Mots-clés: Hélium ; Développement du viriel ; Potentiel d'interaction interatomique ; Calcul ab initio ; Conductivité thermique ; Viscosité

1. Introduction

Primary acoustic thermometry and acoustic methods for determination of the Boltzmann constant require accurate values of the acoustic virial coefficient $\beta_a(T)$ and thermal conductivity λ (see [1] and references therein). Other primary-thermometry methods [2] require values of the first and second density virials $B(T)$ and $C(T)$. In 1995 Aziz et al. [3] noted that some of these properties can be calculated more accurately than they can be measured, using values of the helium interatomic potential ϕ_{th} determined using *ab initio* theoretical methods. In 2000 Hurly and Moldover [4] fit an empirical potential $\phi_{00}(r)$ to the existing theoretical values of ϕ_{th} and calculated an extensive set of properties of ^4He , ^3He , and ^4He – ^3He mixtures. Following improvements in values of ϕ_{th} , Hurly and Mehl [5] proposed a new potential ϕ_{07} and calculate the virials and transport coefficients of ^4He . Since that work, new values of ϕ_{th} were reported by Patkowski et al. [6], Jeziorska et al. [7], and Hellmann et al. [8], and Bich et al. [9] reported new calculations of $B(T)$, $\lambda(T)$, and the viscosity $\eta(T)$ for ^4He and ^3He and $C(T)$ for ^4He .

Computation of thermophysical properties is a two-step process. The first step is numerical integration to determine the radial term in the Schrödinger equation and the phase shifts $\delta_\ell(E)$ as functions of energy E and angular momentum

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index ℓ . This step requires an analytical expression for the interaction potential, Jeziorska et al. [7] proposed the form

$$\phi_{\text{JCPJS}}(r) = \left(A + Br + Cr^2 + \frac{C'}{r} \right) e^{-\alpha r} + (A' + B'r + Dr^2) e^{-\beta r} - \sum_{n=6,8,10-16} \frac{C_n}{r^n} \mathcal{D}_{n+1}(br) \quad (1)$$

where $A, B, C, D, \alpha, \beta$, and b are fit parameters and A', B', C' follow from an exact asymptotic limit relating the energies of beryllium and helium atoms. Literature values of very high accuracy have been used for the van der Waals constants C_6, C_8 , and C_{10} [10] and C_{11} – C_{16} [11]. The van der Waals terms in the potential are multiplied by a damping term

$$\mathcal{D}_n(br) = 1 - e^{-br} \sum_{m=0}^n (br)^m / m! \quad (2)$$

of the form proposed by Tang and Toennies [12]. The forms of the potential used in Refs. [4,5] and [9] did not include the odd C_n coefficients, multiplied C_n/r^n by $\mathcal{D}_n(br)$ instead of $\mathcal{D}_{n+1}(br)$, and used simpler, less physical forms for the repulsive part of the potential with fewer parameters (6 in [4,5], 9 in [9]). Because of the more general forms for the repulsive terms in $\phi_{\text{JCPJS}}(r)$ and $\phi_{\text{HBV}}(r)$, and the more accurate values of ϕ_{th} to which these potentials were fit to, thermophysical properties calculated with these potentials are expected to be more accurate than those calculated with ϕ_{07} . The binding energy of the single bound state of the ^4He dimer was found to be 1.7 mK; this value was used in computing the virials.

New calculations of the properties of helium based on Eq. (1) are used as a baseline for comparisons within this paper. These properties are listed in Table 2. The generally small differences between results obtained with other potentials and with Eq. (1) are presented graphically and discussed below.

The values of ϕ_{th} used for determining the fit parameters were determined in the Born–Oppenheimer (BO) approximation. Jeziorska et al. [7] provide an uncertainty estimate $\sigma_{\text{JCPJS}}(r)$ so that the input values fall in the range $\phi_{\text{JCPJS}}(r) \pm \sigma_{\text{JCPJS}}(r)$. They note that these limits are smaller than some post-BO corrections, the most important of which are the diagonal BO correction (DBOC) [13–15], relativistic effects [16], and QED effects [17]. New calculations of these effects, cited as Ref. [33] of [7], are in progress. A full assessment of the accuracy of computed helium properties must await completion of this work. However, some estimates can be made. Hurly and Mehl [5] included the DBOC values of Komasa et al. [15] in ϕ_{07} , and also reported results without the DBOC. Hellmann et al. [8] used their own DBOC calculations in ϕ_{HBV} .

Formally, nuclear masses are used in the BO approximation for computation of interatomic effects. Handy and Lee [13] and Kutzelnigg [14] have argued in favor of using atomic masses in the next order approximation. As the accuracy of the potential increases, this issue eventually dominates the uncertainty in the calculation of helium properties. It is shown below that the uncertainty of the room-temperature viscosity of ^4He is smaller than the difference between values computed with nuclear and atomic masses. On the other hand, the use of atomic, rather than nuclear masses decreases the room-temperature B by $0.0001 \text{ cm}^3 \text{ mol}^{-1}$. This is an order of magnitude less than the uncertainty B calculated with ϕ_{JCPJS} .

In the new calculations of helium properties reported here, as well as those reported in Refs. [5] and [9], relativistic retardation corrections $f_n(r)$ [18] were applied to the van der Waals terms. These functions approach unity for small r and are proportional to $1/r$ for large r , so that $C_n f_n/r^n$ is proportional to $1/r^{n+1}$ in the far field.

Fig. 1 shows computed values of the density virials. Values computed with ϕ_{07} and the values of Bich et al. [9] lie uniformly above values computed with the reference values ϕ_{JCPJS} . The recent measured values of $B(T)$ for ^4He in the range 220–330 K by McLinden and Losch-Will [19] were compared with calculated values in Fig. 9 of Ref. [5] and in Fig. 2 of Ref. [9]. The measurements have an average uncertainty of $0.04 \text{ cm}^3 \text{ mol}^{-1}$; the average differences between the measured and calculated values are significantly smaller than this, as shown in Table 1. Virials interpolated from the table in Ref. [9] agree with measurements better than the values calculated with ϕ_{07} . The latter calculations were done with both atomic and nuclear masses, and with and without the DBOC. It is thus possible to assess the effects of these post-BO effects on the virials. The use of atomic rather than nuclear masses increases the average by $0.0001 \text{ cm}^3 \text{ mol}^{-1}$, equal to the uncertainty of the values calculated with ϕ_{JCPJS} . Using a version of ϕ_{07} without the DBOC reduced the average by $0.0016 \text{ cm}^3 \text{ mol}^{-1}$. In summary, the experimental uncertainties must be reduced by a factor of 5 or more before they can distinguish among the various theoretical alternatives. The effects of the DBOC

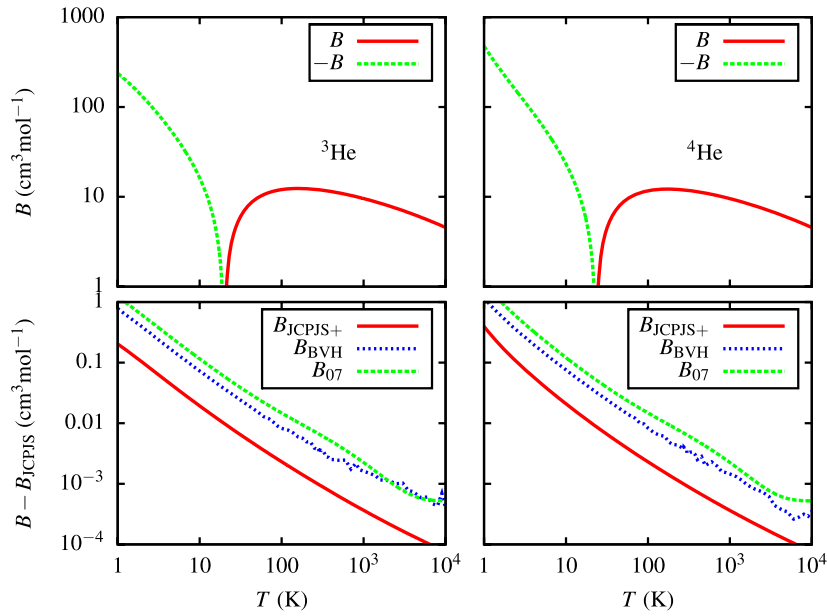


Fig. 1. Density virials of ^4He and ^3He ; the top panels show values computed with ϕ_{JCPJS} . Computations of B with the potentials $\phi_{\text{JCPJS}} \pm \sigma_{\text{JCPJS}}$ are displaced symmetrically from values B_{JCPJS} computed with ϕ_{JCPJS} , that is $B_{\text{JCPJS}+} - B_{\text{JCPJS}} \approx -(B_{\text{JCPJS}-} - B_{\text{JCPJS}})$. The lower panels show this quantity as solid red lines, which represent the uncertainty in B due to the BO-level uncertainty of the potential ϕ_{JCPJS} . The lower panels also show the differences between values of B reported by Bich et al. [9] and values computed with ϕ_{07} . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
Experimental ^4He virials compared with theory.

Potential	Mass	$\langle B_{\text{expt}} - B_{\text{calc}} \rangle$ ($\text{cm}^3 \text{mol}^{-1}$)
JCPJS	n	-0.0014 ± 0.010
JCPJS+	n	-0.0024 ± 0.010
BHV	a	-0.0052 ± 0.010
07	a	-0.0082 ± 0.010
07	n	-0.0083 ± 0.010
07, ndboc	n	-0.0099 ± 0.010

The second column indicates whether nuclear or atomic masses were used in the calculations. The nine measurements B_{meas} of Ref. [19] are in the range $11.7\text{--}12.1 \text{ cm}^3 \text{mol}^{-1}$. The third column is the average difference $B_{\text{meas}}(T) - B_{\text{calc}}(T)$.

and the choice of nuclear or atomic mass are of the same order of magnitude as the uncertainty of calculations with ϕ_{JCPJS} .

The density dependence of the speed of sound is expressed in terms of the acoustic virials. For monatomic gases the second acoustic virial β_a is related to the density virials by

$$\beta_a = 2B + \frac{4}{3}T \frac{dB}{dT} + \frac{4}{15}T^2 \frac{d^2B}{dT^2} \quad (3)$$

Fig. 2 shows the differences between the experimental values of β_a measured by Pitre et al. [20] and values computed with ϕ_{JCPJS} . The measurements are in close agreement with the values computed with ϕ_{JCPJS} except for a few points above 200 K. At the lowest temperatures, values computed with ϕ_{07} lie outside the experimental uncertainties. The uncertainties of β_a due to the uncertainties of ϕ_{JCPJS} , shown as green dotted lines, include only the uncertainties in the BO potential. When the work cited as Ref. [33] of [7] is complete, it will be possible to determine the effect of the post-BO effects. Hurly and Mehl [5] computed β_a using versions of ϕ_{07} with and without the DBOC. The use of the version without the DBOC increased β_a (5 K) by $0.18 \text{ cm}^3 \text{mol}^{-1}$. The use of atomic masses instead of nuclear masses

Table 2

Properties of gaseous helium in the limit of zero density calculated with the potential ϕ_{JCPS} in the BO approximation, with nuclear masses.

T (K)	^4He				^3He			
	B ($\text{cm}^3 \text{mol}^{-1}$)	β_a ($\text{cm}^3 \text{mol}^{-1}$)	η ($\mu\text{P s}$)	λ ($\text{mW m}^{-1} \text{K}^{-1}$)	B ($\text{cm}^3 \text{mol}^{-1}$)	β_a ($\text{cm}^3 \text{mol}^{-1}$)	η ($\mu\text{P s}$)	λ ($\text{mW m}^{-1} \text{K}^{-1}$)
1	-476.9	-537.4	0.3285	2.627	-236.9	-299.8	0.5585	5.777
1.2	-371.2	-425.5	0.3399	2.715	-206.0	-252.5	0.6632	6.861
1.4	-303.1	-350.1	0.3577	2.841	-181.4	-216.9	0.7616	7.884
1.6	-255.9	-295.8	0.3838	3.028	-161.4	-189.3	0.8493	8.804
1.8	-221.3	-254.8	0.4177	3.279	-144.9	-167.2	0.9243	9.599
2	-194.8	-222.9	0.4579	3.584	-131.2	-149.2	0.9865	10.270
2.25	-169.30	-191.69	0.5153	4.025	-116.81	-130.83	1.0485	10.949
2.5	-149.45	-167.28	0.5783	4.514	-104.95	-115.87	1.0964	11.484
2.75	-133.55	-147.66	0.6448	5.032	-94.99	-103.43	1.1341	11.908
3	-120.49	-131.57	0.7131	5.564	-86.52	-92.91	1.1648	12.254
3.5	-100.26	-106.73	0.8501	6.631	-72.90	-76.08	1.2150	12.808
4	-85.27	-88.42	0.9823	7.660	-62.45	-63.18	1.2601	13.283
4.5	-73.69	-74.34	1.1068	8.630	-54.18	-52.97	1.3058	13.748
5	-64.46	-63.17	1.2229	9.536	-47.48	-44.69	1.3544	14.234
6	-50.67	-46.54	1.4328	11.179	-37.27	-32.06	1.4607	15.293
7	-40.85	-34.73	1.6199	12.645	-29.87	-22.90	1.5753	16.447
8	-33.49	-25.91	1.7909	13.988	-24.24	-15.96	1.6936	17.650
9	-27.77	-19.08	1.9505	15.240	-19.82	-10.54	1.8124	18.868
10	-23.20	-13.64	2.1014	16.424	-16.26	-6.18	1.9299	20.078
11	-19.47	-9.22	2.2454	17.554	-13.32	-2.62	2.0452	21.269
12	-16.36	-5.55	2.3837	18.639	-10.86	0.35	2.1579	22.436
14	-11.488	0.16	2.6464	20.698	-6.971	5.01	2.3753	24.690
16	-7.847	4.39	2.8939	22.638	-4.039	8.47	2.5826	26.842
18	-5.029	7.626	3.1292	24.481	-1.753	11.139	2.7810	28.903
20	-2.787	10.174	3.3543	26.245	0.076	13.242	2.9717	30.884
22	-0.964	12.220	3.5708	27.941	1.570	14.937	3.1556	32.795
23	-0.176	13.097	3.6762	28.767	2.218	15.663	3.2453	33.727
24	0.544	13.893	3.7798	29.579	2.812	16.323	3.3337	34.645
25	1.204	14.618	3.8818	30.378	3.356	16.924	3.4207	35.549
26	1.810	15.280	3.9823	31.165	3.857	17.474	3.5064	36.440
28	2.886	16.443	4.1790	32.705	4.748	18.438	3.6745	38.186
30	3.811	17.429	4.3705	34.204	5.515	19.255	3.8384	39.888
35	5.629	19.320	4.8297	37.800	7.028	20.820	4.2321	43.978
40	6.956	20.648	5.2656	41.213	8.135	21.912	4.6067	47.869
45	7.957	21.607	5.6826	44.477	8.972	22.694	4.9654	51.594
50	8.733	22.314	6.0836	47.615	9.620	23.264	5.3108	55.181
60	9.838	23.240	6.8465	53.584	10.540	23.993	5.9688	62.014
70	10.566	23.766	7.5674	59.225	11.1430	24.385	6.5915	68.478
80	11.065	24.059	8.2550	64.603	11.5515	24.580	7.1858	74.647
90	11.416	24.205	8.9151	69.767	11.8337	24.654	7.7568	80.573
100	11.665	24.258	9.5522	74.748	12.0304	24.650	8.3080	86.294
120	11.970	24.195	10.7692	84.265	12.2598	24.505	9.3618	97.230
140	12.120	24.010	11.9248	93.300	12.3575	24.265	10.3630	107.617
160	12.180	23.767	13.0313	101.949	12.3804	23.982	11.3218	117.565
180	12.1863	23.497	14.0971	110.280	12.3586	23.682	12.2458	127.150
200	12.1587	23.217	15.1286	118.343	12.3093	23.379	13.1402	136.427
225	12.0947	22.866	16.3770	128.098	12.2243	23.005	14.2228	147.656
250	12.0111	22.5222	17.5862	137.548	12.1243	22.6435	15.2716	158.534
273.15	11.9234	22.2137	18.6759	146.062	12.0245	22.3220	16.2169	168.336
275	11.9161	22.1896	18.7618	146.734	12.0163	22.2969	16.2914	169.109
298.15	11.8225	21.8933	19.8242	155.034	11.9129	21.9900	17.2130	178.666
300	11.8149	21.8701	19.9081	155.690	11.9046	21.9661	17.2858	179.421
325	11.7108	21.5644	21.0283	164.442	11.7917	21.6510	18.2577	189.498
350	11.6057	21.2723	22.1253	173.012	11.6793	21.3510	19.2096	199.367
375	11.5010	20.9933	23.2015	181.419	11.5684	21.0653	20.1433	209.049

Table 2 (continued)

T (K)	^4He				^3He			
	B ($\text{cm}^3 \text{mol}^{-1}$)	β_a ($\text{cm}^3 \text{mol}^{-1}$)	η (μPs)	λ ($\text{mW m}^{-1} \text{K}^{-1}$)	B ($\text{cm}^3 \text{mol}^{-1}$)	β_a ($\text{cm}^3 \text{mol}^{-1}$)	η (μPs)	λ ($\text{mW m}^{-1} \text{K}^{-1}$)
400	11.3975	20.7268	24.2587	189.677	11.4596	20.7930	21.0607	218.559
450	11.1962	20.2282	26.3226	205.798	11.2496	20.2852	22.8517	237.127
500	11.0040	19.7711	28.3278	221.459	11.0506	19.8208	24.5919	255.165
600	10.6487	18.9605	32.1930	251.643	10.6856	18.9998	27.9464	289.933
700	10.3301	18.2615	35.9011	280.597	10.3603	18.2936	31.1647	323.285
800	10.0434	17.6496	39.4833	308.565	10.0689	17.6766	34.2739	355.503
900	9.7842	17.1073	42.9615	335.718	9.8060	17.1304	37.2928	386.783
1000	9.5483	16.6214	46.3518	362.183	9.5673	16.6416	40.2356	417.271
1200	9.1335	15.7823	52.9159	413.416	9.1485	15.7982	45.9331	476.292
1400	8.7788	15.0776	59.2465	462.821	8.7911	15.0905	51.4281	533.208
1600	8.4701	14.4727	65.3908	510.766	8.4805	14.4835	56.7615	588.444
1800	8.1978	13.9447	71.3822	557.513	8.2067	13.9539	61.9621	642.299
2000	7.9548	13.4775	77.2451	603.253	7.9625	13.4856	67.0512	694.994
2500	7.4439	12.5068	91.4532	714.084	7.4496	12.5128	79.3842	822.679
3000	7.0324	11.7352	105.1665	821.037	7.0369	11.7399	91.2878	945.898
3500	6.6901	11.0997	118.5078	925.075	6.6937	11.1035	102.8684	1065.757
4000	6.3984	10.5625	131.5581	1026.832	6.4015	10.5656	114.1965	1182.988
4500	6.1453	10.0994	144.3746	1126.755	6.1479	10.1021	125.3216	1298.107
5000	5.9226	9.6940	156.9991	1225.171	5.9248	9.6963	136.2801	1411.491
6000	5.5456	9.0130	181.7925	1418.431	5.5474	9.0148	157.8015	1634.141
7000	5.2360	8.4579	206.1211	1608.042	5.2374	8.4594	178.9195	1852.587
8000	4.9749	7.9929	230.1049	1794.946	4.9761	7.9941	199.7382	2067.914
9000	4.7502	7.5950	253.8279	1979.800	4.7512	7.5960	220.3305	2280.880
10000	4.5538	7.2490	277.3516	2163.087	4.5547	7.2499	240.7498	2492.040

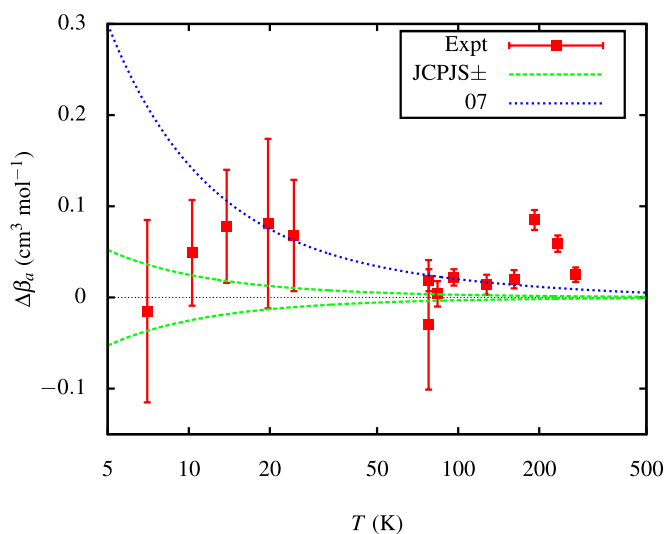


Fig. 2. Comparison of measured values of the acoustic virial coefficient $\beta_{a,\text{expt}}$ [20] and computed values. The measured values and the theoretical values computed using alternative potentials are plotted as the differences $\Delta\beta_a \equiv \beta_a - \beta_{a,\text{JCPJS}}$. For the lines labeled JCPJS \pm (07, HBV), β_a was calculated with $\phi_{\text{JCPJS}} \pm \sigma_{\text{JCPJS}}$ (ϕ_{07} , ϕ_{HBV}). (For interpretation of the references to color, the reader is referred to the web version of this article.)

increased β_a (5 K) by a smaller amount, $0.017 \text{ cm}^3 \text{ mol}^{-1}$. Both of these effects decreased rapidly with increasing T . Improved measurements of β_a , particularly at low T , would provide a sensitive test of the theoretical models.

Fig. 3 shows the differences between the computed viscosities determined with various potentials. The largest fractional differences occur at low T , with poorest agreement for viscosities computed with ϕ_{07} [5]; the results of

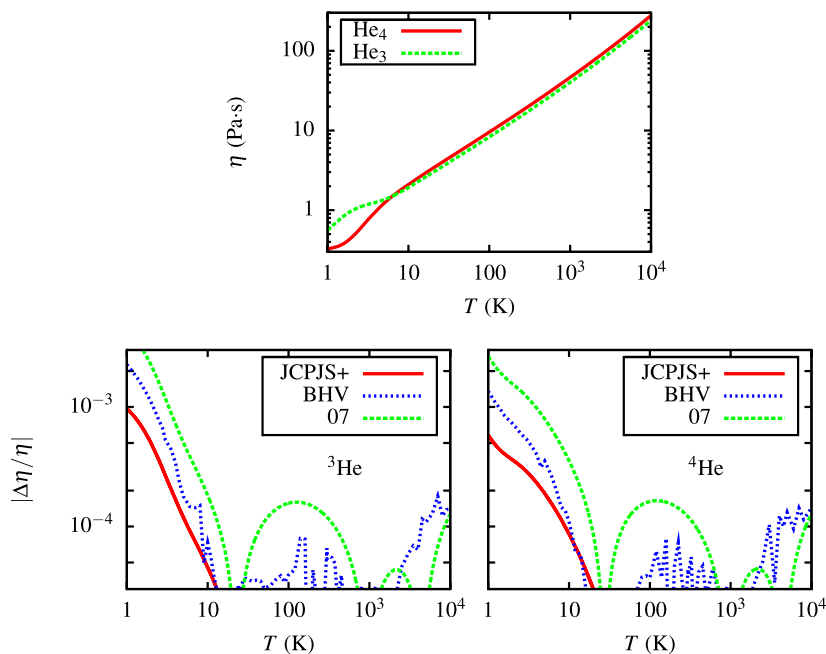


Fig. 3. Calculated viscosity of ^4He and ^3He . The top panel shows values η_{JCPJS} calculated with ϕ_{JCPJS} . The bottom panel shows the absolute value of the fractional differences between values η_x computed with other potentials and η_{JCPJS} . For example, the key JCPJS+, 4 refers to calculations with $\phi_{\text{JCPJS}} + \sigma_{\text{JCPJS}}$ for ^4He .

Bich et al. [9] are in closer agreement at all temperatures. Computed values of the thermal conductivity follow a very similar pattern.

Moldover [1] compares recent measurements and calculations of the viscosity of ^4He at 298.15 K in his Fig. 3. The various calculations of the viscosity near room temperature have small uncertainties and are in close agreement. The viscosity determined with ϕ_{JCPJS} is (19.8269 ± 0.0002) $\mu\text{Pa s}$. Hurly and Mehl [5] obtained (19.824 ± 0.004) $\mu\text{Pa s}$, and Bich et al. obtained 19.8262 $\mu\text{Pa s}$. The theoretical values are thus in close agreement. The measured value obtained in the high-accuracy measurement of Berg et al. [21] at the same temperature is (19.842 ± 0.014) $\mu\text{Pa s}$. This lies just above and just outside the combined uncertainties. The effect of the DBOC can be estimated from Ref. [5]; inclusion of the DBOC lowers η by 0.0015 $\mu\text{Pa s}$. The use of atomic, rather than nuclear masses increases η_{calc} by 0.003 $\mu\text{Pa s}$.

Further theoretical work on the helium interaction potential is in progress, including new calculations of the diagonal Born–Oppenheimer correction [7, Ref. [3]] and relativistic effects [17]. Once that work is complete, it will be of interest to update the calculations presented in this article. It will also be helpful if the theoretical questions about the use of atomic rather than nuclear masses [13,14] are resolved.

Acknowledgements

The author has benefited from long collaboration with M.R. Moldover and J. Hurly; and is grateful to them for useful comments on this paper.

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