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Thermodynamic Calculation of a Heat of First-Order Phase Transitions

Sobko AA*

Department of Physics and Chemistry of New Materials, AM Prokhorov Academy of Engineering Sciences, 19 Presnensky Val, Moscow 123557, Russia

Abstract

A transition heat is the most important characteristics of first-order phase transitions. Black [1] was first who discovered in 1762 that in the transfer of water to vapor, some quantity of heat is absorbed, which he termed the latent evaporation heat. In spite of more than the two-hundred-year period of the heat transfer concept existence there are no analytical expressions relating the transition heat with other parameters of phase transitions. For example, the fundamental "Physics Encyclopedia", articles devoted to the transition heat, evaporation heat, and so on, comprises no formulae but only tables of experimental data. One can also mention monographs [2-9] which have no relationships except for the conventional definition of the transition heat λ=TΔS. Hence, obtaining the relationships between the transition heat and other parameters of first-order phase transitions will be a substantial contribution into the theory of first-order phase transitions.

Keywords: Thermodynamic; Heat; First-order phase transitions

Experimental Methods

Part I: calculation of the evaporation heat for liquid gases and metals

General expressions for the transition heat of first-order phase transitions: The conventional expression for a transition heat $\lambda=T\Delta S$ has two substantial drawbacks. First, in some phase transitions, for example, in evaporation, not only entropy changes but a system does the work, which can only be supplied by an external source of heat. Second, the transition heat is expressed in terms of the entropy variation ΔS, which cannot be measured experimentally. In the preset work, the transition heat is defined as

$$\lambda=T\Delta S+A$$

Where T is the transition temperature in K^0, ΔS is the change of system entropy, A is the work that the system does. Entropy is calculated from the general definition [3]

$$S=k\ln\frac{V_{ph}}{(2\pi\hbar)^s},$$

Where K is the Boltzmann factor, V_{ph} is the volume of phase space occupied by the system, and S is the number of degrees of freedom. Thus, we obtain:

$$S=k\ln\frac{V_{ph}}{(2\pi\hbar)^s},$$

where $(V_{ph})_o$ and $(V_{ph})_n$ are the volumes of the old and new phases, respectively. The general expression for the heat for a phase transition has the form:

$$\lambda=kT\ln\frac{\left(V_{ph}\right)_n}{\left(V_{ph}\right)_o}+A \qquad (1)$$

The volumes of phase spaces and the expressions for the work are specified for each particular phase transition. In the present work, all calculations are performed for one mole of substance; hence, all extensive values refer to one mole.

Approximate calculation of the phase space volumes for liquid and gaseous states

For liquid and gaseous states, the energy of system has the form:

$$\frac{p_{x1}^2}{2m_1}+\frac{p_{y1}^2}{2m_1}+\frac{p_{1z}^2}{2m_1}+\dots+\frac{p_{zN}^2}{2m_N}+U\left(\vec{r}_1,\dots,\vec{r}_N\right)=E\cdot 1\le i\le 3,1\le n\le N_A\cdot \qquad (2)$$

Since we consider one mole of a single-component substance, all masses are equal and $N=N_A$ is the Avogadro number. The volume of the phase space is:

$$V_{ph}=\int_{6N_A}\!\!\cdot\cdot\int\prod_n^{6N_A}d^3\vec{p}_n d^3\vec{r}_n$$

Equation (2) can be rewritten in the form:

$$\sum_{in}p_{in}^2=2m\left(E-U\left(\vec{r}_1,..,\vec{r}_{N_A}\right)\right),$$

which is the equation of a 3N-dimensional sphere in $\vec{p}$-space; hence, the 3N-dimensional integral over pulses $\vec{p}$ is equal to the volume of this sphere of the radius $\left(2m\left(E-U\left(\vec{r}_1,...,\vec{r}_{N_A}\right)\right)\right)^{\frac{1}{2}}$, and the expression for the volume of phase space takes the form:

$$\iint_{3N}\frac{\pi^{3N/2}}{(3N/2)!}\left(2m\left(E-U\left(\vec{r}_1,...\vec{r}_{N_A}\right)\right)\right)^{3N_A/2}\prod_n^{3N_A}d^3\vec{r}\cdot$$

In the result of integration the dimensionality of the integral has changed from 6 to 3; however, one cannot take the rest integral without further assumptions. For calculating the 3N-dimenstional integral, let us consider the behavior of the function of kinetic energy distribution near a point of a first-order phase transition. Not specifying exactly the distribution function one may assert that it has a bell shape with a maximum that shifts with temperature to right. Near the point of phase transition such an evolution of the distribution function is impossible because the temperature of the system does not change and energy incoming still continues. Hence, the only way for the distribution function to vary is its narrowing and, in the limit, it transfers to the δ-function. In the latter case, the most probable and the average values of the kinetic energy will coincide. It is not a rigorous proof of the distribution function narrowing; however, it suggests a principal

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***Corresponding author:** Sobko AA, Department of Physics and Chemistry of New Materials, AM Prokhorov Academy of Engineering Sciences, 19 Presnensky Val, Moscow 123557, Russia, E-mail: ainrf@mail.ru

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assumption of the present work, which is confirmed by a satisfactory agreement between experimental data and calculation results.

Note: Near the first-order phase transition the most of atoms (molecules, ions) are in the state with the average kinetic energy.

Since (E-U($\mathbf{r}_1$,..$\mathbf{r}_n$)) is the kinetic energy of the system, according to the theory about equal distribution of a kinetic energy over degrees of freedom [4] one can substitute it for the average value $3N_A KT/2=3RT/2$, where $R=KN_A$ is the universal gas constant:

$$V_{ph} = \int \underset{3N}{\cdot\cdot} \int \frac{\pi^{3N/2}}{(3N/2)!} \left(\frac{3}{2} 2mRT \right)^{3N/2} \prod_n d^3 r_n = \frac{\pi^{3N/2}}{(3N/2)!} (3mRT)^{3N/2} V^N$$

Thus, the volume of a phase space for liquid and gas is expressed as

$$V_{phL} = \frac{(3\pi mRT)^{3N/2}}{(3N/2)!} V_L^N \tag{3}$$

$$V_{phG} = \frac{(3\pi mRT)^{3N/2}}{(3N/2)!} V_G^N \tag{4}$$

Where V_L and V_G are the volumes of liquid and gas, respectively.

Calculation of the evaporation heat for liquid gases

By using the general expression (1) for a transition heat one can find the expression for the evaporation heat on the saturation curve. Since the volumes of phase spaces for liquid and gas are known (3), (4), the change of entropy in evaporation has the form:

$$\Delta S = k \ln \left(\frac{V_G}{V_L} \right)^N = kN \ln \left(\frac{V_G}{V_L} \right) = R \ln \left(1 + \frac{\Delta V}{V_L} \right),$$

Where ΔV is the volume jump, V_L is the volume of liquid, V_G is the gas volume.

The work on volume expansion is $A_1=P\Delta V$. In the transit liquid-gas, in addition to the work on volume expansion, also the work against surface tension forces is done $A_2=\sigma FN_1$, where σ is the surface tension coefficient, F is the area of liquid surface, $N_1=V_a/Fd$ is the number of mono-molecular layers, and V_a is the volume occupied by atoms (molecules, ions), *d* is the thickness of a mono-molecular layer (in the present work it is αr, where r is the radius of atom (molecule, ion), α=1.717 is the packing factor).

Thus, we have $A_2=\sigma FN_1=\sigma FV/Fd=\sigma V_\alpha/d$.

The expression for the evaporation heat on the saturation curve has the form

$$\lambda = RT \ln \left(1 + \frac{\Delta V}{V_{Lf}} \right) + P\Delta V + \frac{\sigma V_a}{\alpha r_a}, \tag{5}$$

Where *R* is the universal gas constant; *T, P* are the temperature and pressure on the saturation curve, respectively; ΔV is the jump of volume in the process of evaporation; V_L is the volume of liquid; V_α is the volume occupied by atoms (molecules, ions); r_α is the effective atomic (molecular, ion) radius; and $\alpha=1.817$ is the sphere packing factor. All extensive values refer to one mole of substance.

The expression for the evaporation heat λ comprises the volume occupied by atoms V_α and the volume occupied by liquid V_L. The question arises which volume one should employ – the geometrical (experimental) volume or the free volume $V_{Lf}=V_{\mathrm{Lf}}–N_AV_0$, where V_0 is the volume occupied by atom (molecule, ion). Here, the evaporation heat is calculated by using as the geometrical liquid volume V_L, so and the free volume of liquid V_{Lf}.

Experimental data on the saturation curve are taken from Ref. [10], the radii of atoms and ions are taken from Ref. [11,12]. The radii of two-atomic molecules are taken as half the distance between nuclei centers [13] plus the Van der Waals radii [12]. The results are given in Table 1 and Figure 1.

TK^0 is the evaporation temperature, P*10^{-5} [Pa] is the pressure, V_L*10^5 [m^3/mole] is the molar volume of liquid, ΔV*10^5 [m^3/mole] is the jump of volume in evaporation, σ*10^3 [N/m] is the surface tension coefficient, r*10^{10} [m] is the radius of atom (molecule, ion), λ_{ex} [J/mole] is the experimental value of molar evaporation heat, λ_{T1} [J/mole] is the molar evaporation heat calculated by using the geometrical volume, λ_{T2} [J/mole] is the molar evaporation heat by using the free volume, δ_1 and δ_2 [%] are inaccuracies of λ_{T1} and λ_{T2}, respectively [14].

The calculated values of evaporation heat in Table 1 are only presented for the evaporation lines, for which the experimental values of surface tension coefficient have been found. For hydrogen, data on a surface tension coefficient along the entire evaporation curve are known; the corresponding experimental and calculated values of evaporation heat are plotted in Figure 1. A small difference between experimental and calculated values at low temperatures is related with the fact that the calculation of a phase volume should make allowance for quantum effects.

From Table 1 and Figure 1 one can see that the calculations of the evaporation heat performed by the obtained formula well agree (within several percent) with experimental results. Hence, the assumption, that the most of atoms (molecules, ions) near a point of a first-order phase transition are in the state with the average kinetic energy, is valid. Also valid is the assumption that in a calculation of the evaporation heat one should take into account the work done by the system. The employment of the free volume in calculations also gives a better agreement between experimental and calculated results. Thus, one can assert that the molar evaporation heat λ on the saturation curve is described by the expression:

$$\lambda = RT \ln \left(1 + \frac{\Delta V}{V_{Lf}} \right) + P\Delta V + \frac{\sigma V_a}{\alpha r_a}, \tag{6}$$

where all the values have been defined above.

Specific features of calculating the evaporation heat for liquid metals

As one can see from the results presented in Table 1, calculations of evaporation heat should be performed with the "free volume". For

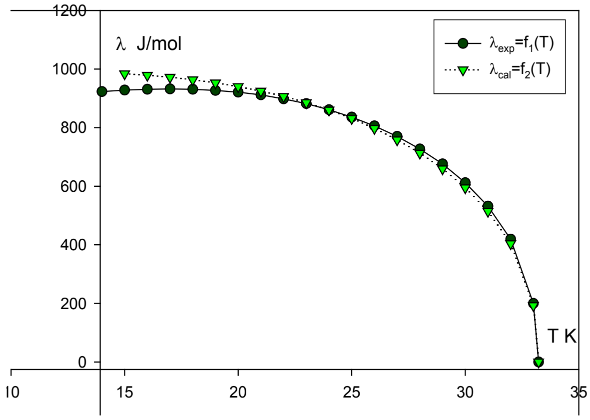

Figure 1: Experimental and calculated evaporation heat versus temperature.

determining the "free volume" of liquid metals one should know the radii of ions. Handbooks [11,12] comprise two metal ion radii: radii for ions M^{+1}, M^{+2} and so on, and for metal. Since it is not clear which radius should be used in the calculations of the free volume, the latter was calculated by using as the metal radius so and the ion radius. The results with the employment of the ion radius are given in Table 2, and the results based on metal radius are presented in Table 3.

T[K] is the evaporation temperature, $P*10^{-5}$ [Pa] is the pressure, V_L*10^5 [m^3/mole] is the molar volume of liquid, $\Delta V*10^5$ [m^3/mole] is the volume jump in evaporation, $\sigma*10^3$ [J/m^2] is the surface tension coefficient, r_m*10^{10} [m] is the ion radius in Table 2 and the metal radius in Table 3, V_m*10^5 [m^3/mole] is the volume occupied by ions, $V_{Lf}*10^5$ [m^3/mole] is the "free volume" of liquid, λ_{ex} [J/mole] is experimental evaporation heat, λ_T [J/mole] is calculated value of evaporation heat, and δ % is the calculation error λ_T.

From Table 2 one can see that the employment of ion radii in calculations of the free volumes of liquid gives a good agreement with experimental data; however, the presence of free electrons in liquid and screening of ions make one to assume that more realistic are metal radii. Results of calculations with metal radii are given in Table 3. One can see that the calculated values of the evaporation heat in this case are systematically greater than the experimental values. This can be explained by the fact that in evaporation of metals, in addition to endothermic processes, there are also exothermic processes.

Recombination of ions and electrons with the origin of neutral atoms occurs for all metals at the interface liquid-gas (vapor). In this case, the energy is released and completely or partially participates in the evaporation process. In addition, alkali metal atoms in the gaseous state form two-atomic molecules [10] with an energy release. Thus, $\Delta\lambda=\lambda_T-\lambda_{ex}$ is the evaporation heat received by a system due to the exothermic processes considered above. The energy released due to generation of two-atomic molecules is proportional to the fraction of two-atomic molecules in gas, and the energy released in the ion recombination is constant and independent of thermodynamic parameters.

Hence,

$$\Delta\lambda=\beta C+Q_0, \qquad (7)$$

Where C is the part of two-atomic molecules in gas, β is the energy, released in the process of producing 0.5 mole of two-atomic molecules, Q_0 is the heat released in recombination of one mole of metal ions.

Calculation results for the evaporation heat, experimental values of evaporation heat in a wide temperature range on the evaporation curve for all alkali metals and mercury, and data on the part of two-atomic molecules in gas for alkali metals are given in Table 4.

Basing on these data, dependences $\Delta\lambda=f(C)$ for alkali metals were plotted in Figure 2. One can see that the assumption about linear dependence of $\Delta\lambda$ on C is confirmed with a high accuracy. Moreover, one can assert that the energies released in generating two-atomic molecules Na_2, K_2, Rb_2, Cs_2 are similar, and the energy released in generating Li_2 is substantially higher. The values of Q_0 are, respectively, Q_0^{Li}=4510 J/mole, Q_0^{Na}=3510 J/mole, Q_0^{K}=2510 J/mole, Q_0^{Rb}=2030 J/mole, and Q_0^{CS}=1220 J/mole and well correlate with the ionization energy for these metals. This does not mean that the linear dependence will still exist at very high temperatures close to the critical temperature, because λ_{ex} and λ_T tend to zero as the temperature approaches the critical value; hence, $\Delta\lambda$ also tends to zero. The mechanism of λ_{ex} reduction at high temperatures is most probably related with the fact that an exothermic process of dissociation of two-atomic molecules starts. Unfortunately, there are no experimental data for calculating λ_T and no information about the behavior of λ_{ex} and the part of two-atomic molecules in gas at high temperatures close to the critical temperature.

In Figure 3 one can see a dependence of $\Delta\lambda$ on temperature for mercury. A linear extrapolation of $\Delta\lambda=f(T)$ turns $\Delta\lambda$ to zero at a point close to the value of T_c.

Conclusion

We may assert that suggested expression (6), which only comprises measurable parameters, allows one to calculate the evaporation heat of liquid gases with a high accuracy taking into account considerations of Sect. 4, and the evaporation heat of liquid metals.

Su-s	T	P	V_L	ΔV	σ	r	λ_{ex}	λ_{T1}	δ_1	λ_{T2}	δ_2
Ne	25	0.51	1.63	394	5.50	1.60	1790	1549	13.5	1757	1.80
Ar	90	1.34	2.90	533	10.53	1.92	6307	5185	17.8	5895	6.53
Kr	150	6.56	3.87	164	10.00	1.98	7886	6356	19.4	7220	8.44
Xe	200	5.22	4.87	284	12.00	2.18	11327	9100	19.7	10362	8.52
H_2	30	8.08	3.67	15.5	0.33	1.44	612	547	10.5	595	2.83
N_2	90	3.60	3.75	182	6.16	2.09	5057	3968	21.5	4668	7.69
O_2	100	2.55	2.95	303	10.70	2.00	6490	5256	19.0	6209	4.32
F_2	95	2.78	2.65	261	10.70	2.06	6775	5024	25.9	6426	5.16
Cl_2	201	0.13	4.26	14570	33.00	2.47	21934	18408	16.1	22110	-0.80
CH_4	105	0.56	3.70	1521	15.80	2.30	8390	7333	12.6	8872	-5.74

T K^0 is the evaporation temperature, $P*10^{-5}$ [Pa] is the pressure, V_L*10^5 [m^3/mole] is the molar volume of liquid, $\Delta V*10^5$ [m^3/mole] is the jump of volume in evaporation, $\sigma*10^3$ [N/m] is the surface tension coefficient, $r*10^{10}$ [m] is the radius of atom (molecule, ion), λ_{ex} [J/mole] is the experimental value of molar evaporation heat, λ_{T1} [J/mole] is the molar evaporation heat calculated by using the geometrical volume, λ_{T2} [J/mole] is the molar evaporation heat by using the free volume, δ_1 and δ_2 [%] are inaccuracies of λ_{T1} and λ_{T2}, respectively.

Table 1: Calculation results of evaporation heat.

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El-s	T	P	V_L	ΔV	σ	r_i	V_i	V_{Lf}	λ_{ex}	λ_T	δ
Li	1600	0,91	1,73	13058	273	0,78	0,12	1,61	134720	133995	0,5
Na	1150	0,96	3,09	8760	120	0,99	0,24	2,84	89590	86899	3,0
K	1000	0,73	5,82	10516	65	1,33	0,59	5,23	75994	72581	4,5
Rb	950	0,92	7,25	7893	50,7	1,49	0,83	6,42	69742	65089	6,7
Cs	800	0,20	8,56	31264	47,5	1,69	1,22	7,34	65126	63799	2,0
Hg	613	0,75	1,57	6820	384	1,12	0,35	1,22	59275	56159	5,3

T[K] is the evaporation temperature, P*10^{-5} [Pa] is the pressure, VL*10^5 [m^3/mole] is the molar volume of liquid, ΔV*10^5 [m^3/mole] is the volume jump in evaporation, σ*10^3 [J/m^2] is the surface tension coefficient, rm*10^{10} [m] is the ion radius in Table 2 and the metal radius in Table 3, Vm*10^5 [m^3/mole] is the volume occupied by ions, VLf*10^5 [m^3/mole] is the "free volume" of liquid, λex [J/mole] is experimental evaporation heat, λT [J/mole] is calculated value of evaporation heat, and δ % is the calculation error λT.

Table 2: Calculation results for the evaporation heat on the saturation curve for metals by using the ion radii.

El-s	T	P	V_L	ΔV	σ	r_m	V_m	V_{Lf}	λ_{ex}	λ_T	δ
Li	1600	0,91	1,73	13058	273	1,57	0,97	0,75	134720	151512	-12,5
Na	1150	0,96	3,09	8760	120	1,89	1,70	1,39	89590	98320	-9,7
K	1000	0,73	5,82	10516	65	2,36	3,31	2,51	75994	82311	-8,3
Rb	950	0,92	7,25	7893	50,7	2,53	4,08	3,17	69742	73767	-5,8
Cs	800	0,20	8,56	31264	47,5	2,74	5,18	3,37	65126	72209	-10,9
Hg	613	0,75	1,57	6820	384	1,60	1,03	0,54	59275	67675	-14,2

T[K] is the evaporation temperature, P*10^{-5} [Pa] is the pressure, V_L*10^5 [m^3/mole] is the molar volume of liquid, ΔV *10^5 [m^3/mole] is the volume jump in evaporation, σ*10^3 [J/m^2] is the surface tension coefficient, r_m*10^{10} [m] is the ion radius in Table 2 and the metal radius in Table 3, V_m*10^5 [m^3/mole] is the volume occupied by ions, V_{Lf}*10^5[m^3/mole] is the "free volume" of liquid, λ_{ex}[J/mole] is experimental evaporation heat, λ_T[J/mole] is calculated value of evaporation heat, and % is the calculation error λ_T.

Table 3: Calculation results for the evaporation heat on the saturation curve for metals by using the metal radii.

El-s	T	P	V_L	ΔV	σ	r_m	V_m	V_{Lf}	λ_{ex}	λ_T	C	Δλ
Li	1400	0,18	1,64	64895	265,5	1,55	0,939	0,701	139563	154081	8,67	14518
	1500	0,43	1,69	29223	251,5	1,55	0,939	0,751	137114	153172	10,55	16058
	1600	0,91	1,73	14622	237,5	1,55	0,939	0,791	134719	152305	12,60	17586
	1700	1,77	1,77	7983	223,5	1,55	0,939	0,831	132394	151553	13,57	19159
	1800	3,19	1,82	4691	209,5	1,55	0,939	0,881	129487	150689	15,05	21202
Na	900	0,05	2,87	145285	145,3	1,89	1,702	1,168	95107	102618	6,37	7511
	1000	0,20	2,95	41885	135,3	1,89	1,702	1,248	92849	102068	8,58	9219
	1100	0,60	3,05	15340	125,3	1,89	1,702	1,348	90628	101145	10,79	10517
	1200	1,50	3,15	6636	115,3	1,89	1,702	1,448	88481	100064	12,89	11583
	1300	3,22	3,25	3360	105,3	1,89	1,702	1,548	86469	99339	14,74	12870
	1400	6,26	3,37	1859	95,3	1,89	1,702	1,668	84434	98268	16,58	13834
	1500	11,01	3,49	1129	85,3	1,89	1,702	1,788	82547	97294	18,12	14747
K	800	0,06	5,43	108807	78,75	2,36	3,313	2,117	79866	85079	4,05	5213
	900	0,24	5,62	30668	72,15	2,36	3,313	2,307	77942	84272	5,98	6330
	1000	0,73	5,82	11355	65,55	2,36	3,313	2,507	75944	83606	7,97	7662
	1100	1,86	6,04	4904	58,95	2,36	3,313	2,727	73899	82454	9,88	8455
	1200	3,91	6,28	2703	52,35	2,36	3,313	2,967	71823	82814	11,61	10991
	1300	7,30	6,54	1475	45,75	2,36	3,313	3,227	69739	80698	13,10	10959
	1400	12,44	6,81	899	39,15	2,36	3,313	3,497	67674	78990	14,31	11316

El-s	T	P	V_L	ΔV	σ	r_m	V_m	V_{Lf}	λ_{ex}	λ_T	C	Δλ
Rb	700	0,03	6,61	183451	65,0	2,53	4,082	2,528	72632	76715	3,44	4083
	800	0,16	6,85	42015	59,35	2,53	4,082	2,768	70718	76303	5,46	5585
	900	0,55	7,11	13667	53,55	2,53	4,082	3,028	68777	75483	7,61	6706
	1000	1,47	7,39	5661	47,75	2,53	4,082	3,308	66862	74680	9,70	7818
	1100	3,30	7,70	2768	41,95	2,53	4,082	3,618	64931	73783	11,62	8852
	1200	6,47	8,03	1535	36,15	2,53	4,082	3,948	62999	72816	13,26	9817
	1300	11,43	8,40	937	30,35	2,53	4,082	4,318	61075	71729	14,62	10655
Cs	700	0,04	8,26	132419	50,3	2,68	4,851	3,409	69366	72071	3,03	2705
	800	0,20	8,56	32798	47,5	2,68	4,851	3,709	67731	71982	4,91	4251
	900	0,66	8,89	11300	42,7	2,68	4,851	4,039	66016	71321	6,95	5305
	1000	1,69	9,24	4843	37,9	2,68	4,851	4,389	64209	70410	8,98	6201
	1100	3,63	9,65	2478	33,1	2,68	4,851	4,799	62361	69605	10,86	7244
	1200	6,79	10,1	1440	28,3	2,68	4,851	5,249	60487	68785	12,05	8298
	1300	11,41	10,7	924	23,5	2,68	4,851	5,849	58600	67779	13,85	9179
Hg	373	0,0004	1,50	8285191	452	1,55	0,939	0,561	60848	70421		9573
	423	0,0038	1,52	931431	439	1,55	0,939	0,581	60518	69241		8723
	473	0,0232	1,53	169895	429	1,55	0,939	0,591	60194	68471		8277
	523	0,0996	1,54	43651	416	1,55	0,939	0,601	59872	67662		7790
	573	0,3302	1,56	14417	402	1,55	0,939	0,621	59546	66801		7255
	623	0,8990	1,57	5748	378	1,55	0,939	0,631	59206	65695		6489

T [K] is the evaporation temperature, $P*10^{-5}$ [Pa] is the pressure, V_L*10^5 [m³/mole] is the molar volume of liquid phase, $\Delta V*10^5$ [m³/mole] is the volume jump, $\sigma*10^3$ [J/m²] is the surface tension coefficient, r_m*10^{10} [m] is the metal radius for ion, V_m*10^5 [m³/mole] is the volume occupied by ions, $V_{Lf}*10^5$ [m³/mole] is the "free volume" of liquid, λ_{ex} [J/mole] is the experimental value of evaporation heat, λ_T [J/mole] is the calculated value of evaporation heat, C[%] is the concentration of two-atomic molecules in a gas of alkali metals, $\Delta\lambda=\lambda_{ex}-\lambda_T$ [J/mole].

Table 4: Calculation of Δλ.

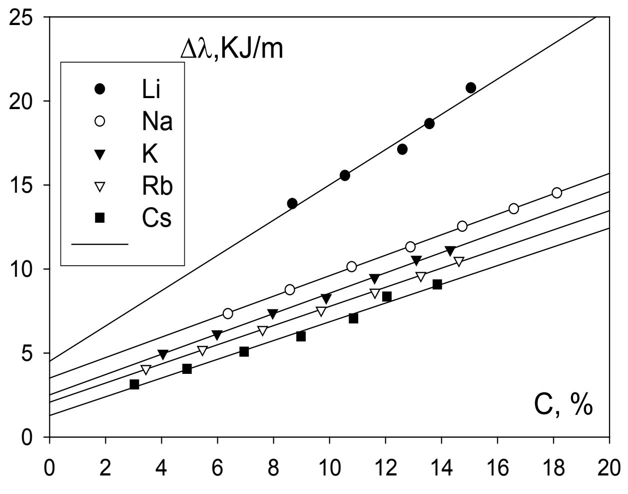

Figure 2: Dependence of $\Delta\lambda=\lambda_{ex}-\lambda_T$ on the concentration of two-atomic molecules in gas for alkali metals.

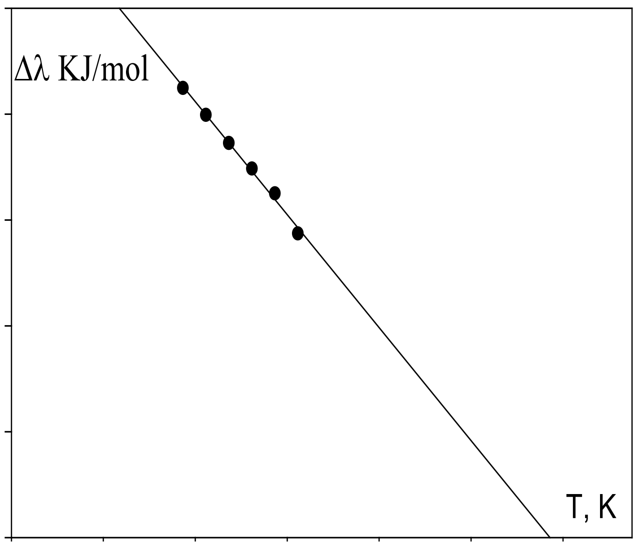

Figure 3: Dependence of $\Delta\lambda=\lambda_{ex}-\lambda_T$ on temperature for mercury.

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