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Author(s)	TOYA, Tomiyuki
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NORMAL VIBRATIONS OF POTASSIUM, RUBIDIUM AND CESIUM

By

Tomiyuki TOYA^{*)}

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Frequency *vs.* wave vector relations for K, Rb and Cs metals have been calculated as reported below in the (100), (110) and (111) directions according to the dispersion relation derived by the present author for the calculation of the normal vibrations of Na¹⁾²⁾ and Cu³⁾.

The circular frequency ω of lattice vibration of wave number vector \mathbf{q} propagating along each one of the above principal directions is given by the equation

$$M\omega^2 = Ne^2(C + R + E),$$

where the first term in the parentheses represents the contribution from the COULOMB repulsions between metal ions formulated by EWALD's method in (2.9), (2.10), (2.11) and (3.5) of (I)^{**)}, the second term that from exchange repulsions between ion-cores as given by (3.7) of (I), and the third term that from valence electrons, expressed by (3.1)–(3.4) of (I), as

$$E = E_I + E_{II},$$

where

$$E_I = (4\pi/3),$$

$$E_{II} = \left\{ -4\pi \sum_h \cos^2(\mathbf{e}_q, \mathbf{q} + \mathbf{K}_h) G(t)^2 F(t)^{-1} f(t) \right\},$$

\mathbf{e}_q is the unit vector representing the direction of polarization, $\mathbf{K}_h \equiv (\mathbf{K}_{hx}, \mathbf{K}_{hy}, \mathbf{K}_{hz})$ the reciprocal lattice vector and $t = |\mathbf{q} + \mathbf{K}_h|/2k_F$, k_F being the wave number vector of an electron at the FERMI surface. The functions $G(t)$, $F(t)$ and $f(t)$ are given, with the approximate wave function $\phi(\mathbf{k}) = U_0(\mathbf{r}) \cdot \exp(i\mathbf{k}\mathbf{r})$ of an electron of wave number vector

^{*)} Research Institute for Catalysis, Hokkaido University.

^{**)} Notations are the same as in Ref. 2, which will be referred to as (I) in the text.

\mathbf{k} and of its HARTREE energy $E_0(\mathbf{k})=E_0+(\hbar^2/2m)k^2$, as*)

$$G(t) = \{1 + \gamma(V(r_s) - E_0) (3/\beta\zeta_0) t^2\} g(2k_F r_s t),$$

$$F(t) = \begin{cases} (D_0/D)(2/\beta)t^2 + (1 - Bt^2)f(t) & \text{for } 1 > Bt^2, \\ (D_0/D)(2/\beta)t^2 & \text{for } 1 \leq Bt^2, \end{cases}$$

and

$$f(t) = \frac{1}{2} + \frac{1-t^2}{4t} \log \left| \frac{1+t}{1-t} \right|,$$

where $\gamma = |U_0(r_s)|^2$, $\beta = e^2 k_F / \pi \zeta_0$, $\zeta_0 = (\hbar^2/2m)k_F^3$, $g(x) = 3(\sin x - x \cos x)/x^3$, $V(r_s)$ the HARTREE potential energy at the surface of atomic sphere of radius r_s , D or D_0 the density of states at the FERMI surface with or without the exchange and correlation energies taken into account, and B allows for the effect of the exchange and correlation interactions on the screening field. The values of these constants for the present calculation are the same as used in the calculations of electrical conductivities¹¹⁾.

The results are given in Tables I, II, III, Figs. 1, 2 and 3, in unit of $\omega_0 \equiv (Ne^2/M)^{1/2}$ respectively.

High frequency curves of alkali metals are almost coincident with each other, when plotted in units of ω_0 , indicating that the vibrational spectra may be approximately expressed as $Q(\omega/\omega_0)/\omega_0$ for large ω , where $Q(\omega/\omega_0)$ is the universal function of ω/ω_0 , so that the molar heat capacity is given as a universal function $C(\kappa T/\hbar\omega_0)$ of $\kappa T/\hbar\omega_0$, i. e.,

$$C(\kappa T/\hbar\omega_0) = \kappa \int \frac{(\hbar\omega_0/\kappa T)^2 x^2 Q(x) \exp\{(\hbar\omega_0/\kappa T)x\}}{[\exp\{(\hbar\omega_0/\kappa T)x\} - 1]^2} dx.$$

The characteristic temperature θ is now given by the equation,

$$C(\kappa T/\hbar\omega_0) = 3RD(\theta/T), \quad [D(\theta/T): \text{DEBYE function}]$$

$$= 3RD\left(\frac{\kappa\theta/\hbar\omega_0}{\kappa T/\hbar\omega_0}\right),$$

which implies that $\kappa\theta/\hbar\omega_0$ is approximately a constant for alkali metals, or θ is proportional to $\omega_0 = (Ne^2/M)^{1/2}$, as shown below;

*) The previous discussion in (I) was based on the equation $F(t) = (D_0/D)(2/\beta)t^2 + (1 - Bt^2)f(t)$ for $1 \geq Bt^2$, which is not rigorous, since $\{(D/D_0)(\beta/2)t^2 F(t)\}^{-1}$ may exceed unity at large t , whereas physically it provides the factor which reduces the perturbation of ionic field due to lattice vibration by the redistribution of valence electrons and in consequence should be less than unity. However, the two expressions do not differ appreciably except for T_1 -branches of (110) of Rb and Cs.

	Na	K	Ru	Cs
$\omega_0 \times 10^{-13}$ (c. p. s.)	1.13	0.702	0.429	0.307
θ °K (Ref. 5)	159	99	55~65	35~45
$\pi\theta/\hbar\omega_0$	1.84	1.84	1.68~1.98	1.50~1.92

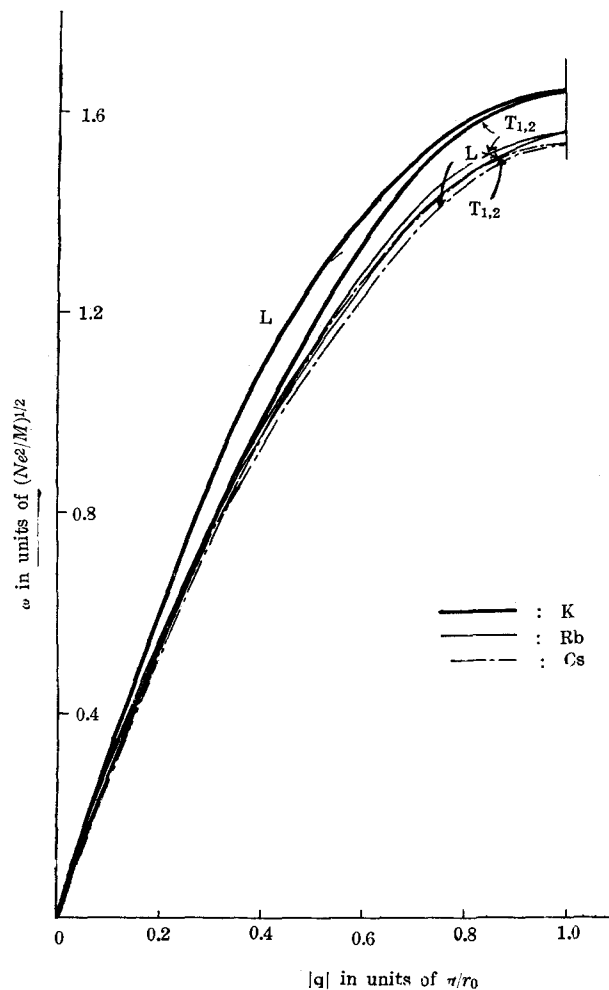


Fig. 1. Circular frequencies ω versus wave number vector $\mathbf{q} = \pi/r_0(\bar{q}, 0, 0)$.

At low temperatures, however, where only low frequency branches are important in determining θ , it is expected that θ of Rb or Cs is lowered appreciably with decrease of temperature rather than in the

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case of Na or K, because of the signurarity $d\omega/dq=0$ at $|q|\approx 0.5(\pi/r_0)$ of the T_1 -branche of the (110) modes, which gives rise to a heap of the vibration distribution at the appropriate low frequencies.

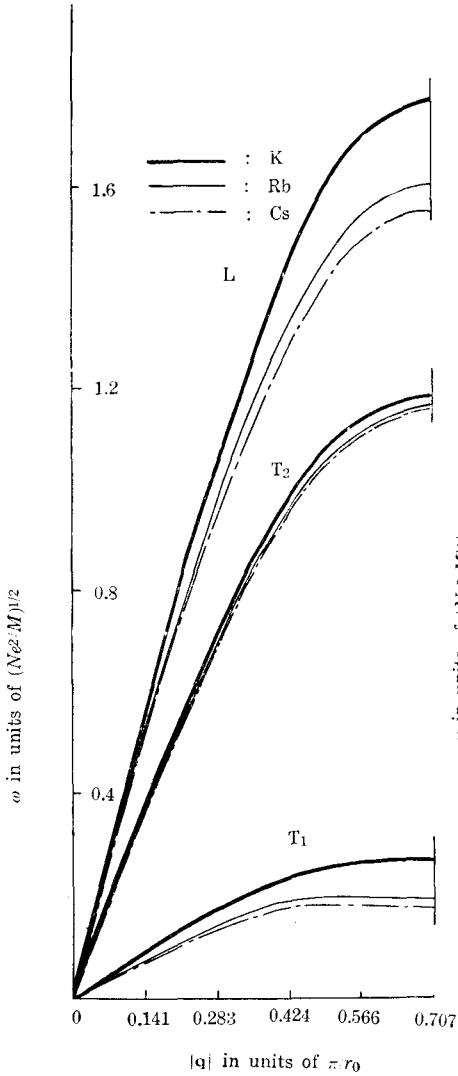


Fig. 2. Circular frequencies ω versus wave number vector $q = \pi/r_0(\bar{q}, \bar{q}, 0)$.

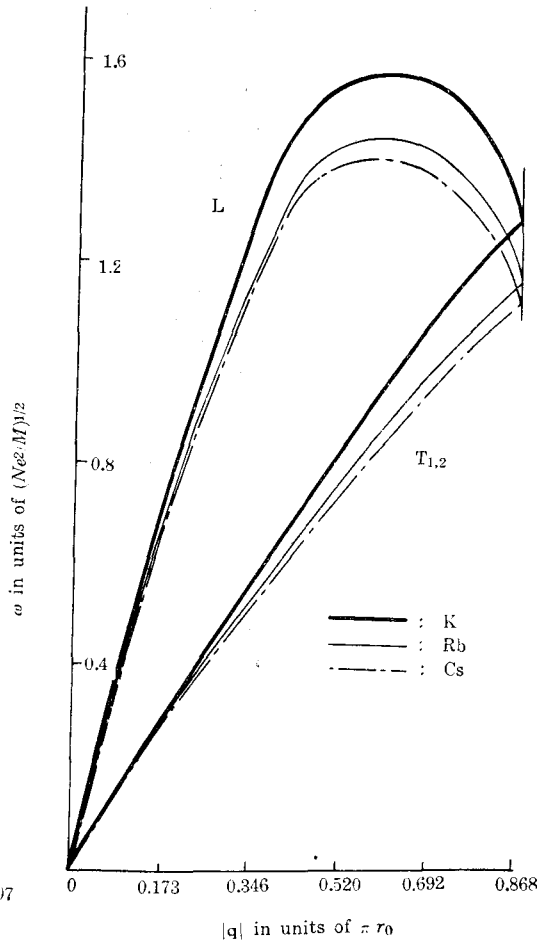


Fig. 3. Circular frequencies ω versus wave number vector $q = \pi/r_0(\bar{q}, \bar{q}, \bar{q})$.

The author would like to express his sincere thanks to Professor J. HORIUTI for his continual encouragement and interests to the present work. He would also like to acknowledge the contributions of Mr. M. TAKAHASHI, who carried out many of the computations in the present work.

TABLE I. Frequencies ω versus wave number vector for potassium. C, R, E_I and E_{II} are given in units of (Ne^2/M) , and ω in units of $(Ne^2/M)^{1/2}$.

(1) $\mathbf{q} = (\pi/r_0)(\bar{q}, 0, 0)$

Longitudinal branch (L), $\mathbf{e}_q = (1, 0, 0)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.2	7.7910	0.0364	4.1888	- 11.5820	- 0.0520	0.381	0.617
0.4	6.0620	0.1210	4.1888	- 8.9884	- 0.2083	1.175	1.084
0.6	3.0510	0.2041	4.1888	- 5.6956	- 0.2732	1.925	1.387
0.8	1.0412	0.2540	4.1888	- 2.7633	- 0.2463	2.474	1.573
1.0	0	0.2680	4.1888	- 0.8970	- 0.8970	2.663	1.632

Transversal branch (T_1 or T_2), $\mathbf{e}_q = (0, 1, 0)$ or $(0, 0, 1)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.2	- 3.8955	0.0264	4.1888	0	- 0.0407	0.279	0.528
0.4	- 3.0311	0.0948	4.1888	0	- 0.2894	0.963	0.981
0.6	- 1.7505	0.1779	4.1888	0	- 0.8491	1.767	1.329
0.8	- 0.5206	0.2440	4.1888	0	- 1.4950	2.417	1.555
1.0	0	0.2680	4.1888	0	- 1.7940	2.663	1.632

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(2) $\mathbf{q} = (\pi/r_0) (\bar{q}, \bar{q}, 0)$

Longitudinal branch (L), $\mathbf{e}_q = \left(\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0 \right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	8.2164	0.0328	4.1888	- 12.0666	- 0.0439	0.328	0.572
0.2	7.7905	0.1188	4.1888	- 10.6595	- 0.3056	1.132	1.064
0.3	7.2563	0.2250	4.1888	- 8.6044	- 0.9722	2.094	1.447
0.4	6.8177	0.3110	4.1888	- 6.2627	- 2.2031	2.852	1.689
0.5	6.6486	0.3438	4.1888	- 4.0220	- 4.0220	3.137	1.771

Transversal branch (T_1), $\mathbf{e}_t = \left(\frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}, 0 \right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.1696	- 0.0023	4.1888	0	- 0.0086	0.003	0.091
0.2	- 4.1179	- 0.0084	4.1888	0	- 0.0297	0.033	0.181
0.3	- 4.0509	- 0.0158	4.1888	0	- 0.0642	0.058	0.240
0.4	- 3.9941	- 0.0219	4.1888	0	- 0.1020	0.071	0.266
0.5	- 3.9718	- 0.0242	4.1888	0	- 0.1178	0.075	0.274

Transversal branch (T_2), $\mathbf{e}_t = (0, 0, 1)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.0468	0.0123	4.1888	0	- 0.0172	0.137	0.370
0.2	- 3.6726	0.0446	4.1888	0	- 0.0594	0.501	0.708
0.3	- 3.2054	0.0846	4.1888	0	- 0.1284	0.940	0.969
0.4	- 2.8236	0.1169	4.1888	0	- 0.2041	1.278	1.130
0.5	- 2.6768	0.1292	4.1888	0	- 0.2355	1.406	1.186

(3) $\mathbf{q} = (\pi/r_0) (\bar{q}, \bar{q}, \bar{q})$

Longitudinal branch (L), $\mathbf{e}_q = \left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}} \right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	8.1781	0.0539	4.1888	- 11.8229	- 0.0635	0.534	0.731
0.2	7.4328	0.1735	4.1888	- 9.7963	- 0.3595	1.639	1.281
0.3	5.8257	0.2667	4.1888	- 7.0329	- 0.7876	2.461	1.569
0.4	3.2057	0.2651	4.1888	- 4.2335	- 0.9.80	2.448	1.565
0.5	0	0.1780	4.1888	- 2.0129	- 0.6710	1.683	1.297

Transversal branch (T_1 or T_2), $\mathbf{e}_q = \left(\frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}, 0 \right)$ or $\left(\frac{1}{\sqrt{6}}, \frac{1}{\sqrt{6}}, -\frac{2}{\sqrt{6}} \right)$

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.0890	0.0075	4.1888	0	- 0.0220	0.085	0.292
0.2	- 3.7164	0.0307	4.1888	0	- 0.1834	0.320	0.565
0.3	- 2.9128	0.0701	4.1888	0	- 0.6509	0.695	0.834
0.4	- 1.6029	0.1223	4.1888	0	- 1.5231	1.180	1.086
0.5	0	0.1780	4.1888	0	- 2.6838	1.683	1.297

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TABLE II. Frequencies ω versus wave number vector for rubidium.
 C, R, E_I and E_{II} are given in units of (Ne^2/M) , and ω in
units of $(Ne^2/M)^{1/2}$.

(1) $\mathbf{q} = (\pi/r_0)(\bar{q}, 0, 0)$

Longitudinal branch (L), $\mathbf{e}_q = (1, 0, 0)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.2	7.7910	0.0377	4.1888	- 11.6449	- 0.0642	0.308	0.555
0.4	6.0620	0.1265	4.1888	- 9.1846	- 0.2528	0.940	0.969
0.6	3.5010	0.2165	4.1888	- 5.9346	- 0.3203	1.601	1.265
0.8	1.0412	0.2733	4.1888	- 3.0538	- 0.2926	2.157	1.469
1.0	0	0.2912	4.1888	- 1.0302	- 1.0302	2.420	1.556

Transversal branch (T_1 or T_2), $\mathbf{e}_q = (0, 1, 0)$ or $\mathbf{e}_q = (0, 0, 1)$

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.2	- 3.8955	0.0272	4.1888	0	- 0.0502	0.270	0.520
0.4	- 3.0311	0.0990	4.1888	0	- 0.3511	0.906	0.952
0.6	- 1.7505	0.1890	4.1888	0	- 0.9952	1.632	1.278
0.8	- 0.5206	0.2628	4.1888	0	- 1.7260	2.205	1.485
1.0	0	0.2912	4.1888	0	- 2.0604	2.420	1.556

(2) $\mathbf{q} = (\pi/r_0)(\bar{q}, \bar{q}, 0)$

Longitudinal branch (L), $\mathbf{e}_2 = \left(\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0\right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	8.2164	0.0339	4.1888	- 12.0989	- 0.0542	0.286	0.535
0.2	7.7905	0.1226	4.1888	- 10.7745	- 0.3663	0.961	0.980
0.3	7.2563	0.2322	4.1888	- 8.8166	- 1.1145	1.746	1.321
0.4	6.8177	0.3209	4.1888	- 6.5434	- 2.4362	2.348	1.532
0.5	6.6486	0.3548	4.1888	- 4.3106	- 4.3106	2.571	1.603

Transversal branch (T_1), $\mathbf{e}_2 = \left(\frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}, 0\right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.1696	- 0.0034	4.1888	0	- 0.0107	0.005	0.071
0.2	- 4.1179	- 0.0123	4.1888	0	- 0.0367	0.022	0.148
0.3	- 4.0509	- 0.0233	4.1888	0	- 0.0789	0.036	0.189
0.4	- 3.9941	- 0.0322	4.1888	0	- 0.1250	0.038	0.194
0.5	- 3.9718	- 0.0356	4.1888	0	- 0.1441	0.037	0.193

Transversal branch (T_2), $\mathbf{e}_2 = (0, 0, 1)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.0468	0.0136	4.1888	0	- 0.0215	0.134	0.366
0.2	- 3.6726	0.0491	4.1888	0	- 0.0735	0.492	0.701
0.3	- 3.2054	0.0931	4.1888	0	- 0.1578	0.919	0.958
0.4	- 2.8236	0.1286	4.1888	0	- 0.2500	1.244	1.115
0.5	- 2.6768	0.1422	4.1888	0	- 0.2882	1.366	1.169

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(3) $\mathbf{q} = (\pi/r_0) (\bar{q}, \bar{q}, \bar{q})$

Longitudinal branch (L), $\mathbf{e}_l = \left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}} \right)$.

\bar{b}	C	R	E_l	E_{ll}		Sum	
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	8.1781	0.0562	4.1888	- 11.8701	- 0.0783	0.475	0.689
0.2	7.4328	0.1709	4.1888	- 9.9556	- 0.4353	1.402	1.184
0.3	5.8257	0.2754	4.1888	- 7.2968	- 0.9128	2.080	1.442
0.4	3.2057	0.2701	4.1888	- 4.5246	- 1.1054	2.035	1.426
0.5	0	0.1758	4.1888	- 2.2347	- 0.7443	1.385	1.177

Transversal branch (T_1 or T_2), $\mathbf{e}_t = \left(\frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}, 0 \right)$ or $\left(\frac{1}{\sqrt{6}}, \frac{1}{\sqrt{6}}, -\frac{2}{\sqrt{6}} \right)$

\bar{q}	C	R	E_t	E_{tt}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.0890	0.0068	4.1888	0	- 0.0271	0.080	0.282
0.2	- 3.7164	0.0282	4.1888	0	- 0.2221	0.279	0.528
0.3	- 2.9128	0.0660	4.1888	0	- 0.7544	0.588	0.767
0.4	- 1.6029	0.1179	4.1888	0	- 1.7272	0.977	0.988
0.5	0	0.1758	4.1888	0	- 2.9797	1.385	1.177

TABLE III. Frequencies ω versus wave number vector for cesium.
 C , R , E_I and E_{II} are given in units of (Ne^2/M) , and ω in
units of $(Ne^2/M)^{1/2}$.

(1) $\mathbf{q} = (\pi/r_0)(\bar{q}, 0, 0)$

Longitudinal branch (L), $\mathbf{e}_q = (1, 0, 0)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.2	7.7910	0.0355	4.1888	- 11.6595	- 0.0663	0.290	0.538
0.4	6.0620	0.1209	4.1888	- 9.2335	- 0.2635	0.875	0.935
0.6	3.5010	0.2111	4.1888	- 6.0621	- 0.3338	1.505	1.227
0.8	1.0412	0.2719	4.1888	- 3.0950	- 0.3041	2.103	1.450
1.0	0	0.2922	4.1888	- 1.0693	- 1.0693	2.342	1.530

Transversal branch (T_1 or T_2), $\mathbf{e}_q = (0, 1, 0)$ or $(0, 0, 1)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.2	- 3.8955	0.0275	4.1888	0	- 0.0518	0.269	0.519
0.4	- 3.0311	0.0999	4.1888	0	- 0.3660	0.892	0.944
0.6	- 1.7505	0.1901	4.1888	0	- 1.0371	1.591	1.261
0.8	- 0.5206	0.2639	4.1888	0	- 1.7935	2.139	1.462
1.0	0	0.2922	4.1888	0	- 2.1385	2.343	1.531

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(2) $\mathbf{q} = (\pi/r_0) (\bar{q}, \bar{q}, 0)$

Longitudinal branch (L_1), $\mathbf{e}_q = \left(\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0 \right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	8.2164	0.0331	4.1888	- 12.1064	- 0.0559	0.276	0.525
0.2	7.7905	0.1196	4.1888	- 10.8020	- 0.3823	0.915	0.956
0.3	7.2563	0.2266	4.1888	- 8.8699	- 1 1560	1.646	1.283
0.4	6.8177	0.3131	4.1888	- 6.6178	- 2.5039	2.198	1.483
0.5	6.6486	0.3462	4.1888	- 4.3912	- 4.3912	2.401	1.550

Transversal branch (T_1), $\mathbf{e}_q = \left(\frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}, 0 \right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4 1696	- 0.0035	4.1888	0	- 0.0110	0.005	0.069
0.2	- 4.1179	- 0.0012	4.1888	0	- 0.0378	0.020	0.143
0.3	- 4.0509	- 0.0240	4.1888	0	- 0.0816	0.032	0.130
0.4	- 3.9941	- 0.0331	4.1888	0	- 0.1296	0.032	0.179
0.5	- 3.9718	- 0.0366	4.1888	0	- 0.1495	0.031	0.176

Transversal branch (T_2), $\mathbf{e}_q = (0, 0, 1)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.0468	0.0137	4.1888	0	- 0.0220	0.134	0.366
0.2	- 3.6726	0.0495	4.1888	0	- 0.0757	0.490	0.700
0.3	- 3.2054	0.0939	4.1888	0	- 0.1632	0.914	0.956
0.4	- 2.8236	0.1297	4.1888	0	- 0.2591	1.236	1.112
0.5	- 2.6768	0.1434	4.1888	0	- 0.2989	1.357	1.165

(3) $\mathbf{q} = (\pi/r_0) (\bar{q}, \bar{q}', \bar{q})$

Longitudinal branch (L), $e_q = \left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}} \right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	8.1781	0.0554	4.1888	- 11.8813	- 0.0808	0.460	0.678
0.2	7.4328	0.1776	4.1888	- 9.9947	- 0.4543	1.350	1.162
0.3	5.8257	0.2704	4.1888	- 7.3654	- 0.9493	1.970	1.404
0.4	3.2057	0.2638	4.1888	- 4.6053	- 1.1429	1.910	1.382
0.5	0	0.1698	4.1888	- 2.2995	- 0.7665	1.293	1.137

Transversal branch (T_1 or T_2) $e_q = \left(\frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}, 0 \right)$ or $\left(\frac{1}{\sqrt{6}}, \frac{1}{\sqrt{6}}, -\frac{2}{\sqrt{6}} \right)$.

\bar{q}	C	R	E_I	E_{II}		Sum	ω
				$h=0$	$h \neq 0$ (Umklapp)		
0.1	- 4.0890	0.0064	4.1888	0	- 0.0208	0.078	0.280
0.2	- 3.7164	0.0267	4.1888	0	- 0.2318	0.267	0.517
0.3	- 2.9128	0.0628	4.1888	0	- 0.7845	0.554	0.745
0.4	- 1.6029	0.1130	4.1888	0	- 1.7857	0.913	0.956
0.5	0	0.1698	4.1888	0	- 3.0660	1.293	1.137

References

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- 5) See, for example, D. K. C. MACDONALD, *Handbuch der Physik* XIV, (Springer-Verlag) p. 172 (1956).