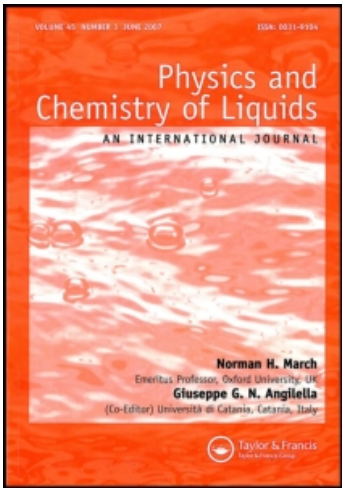

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Sound Velocity in Liquid Binary Alloys as Calculated via the Percus-Yevick Liquid Phonon Dispersion Relation

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In this note, we utilize the recently calculated long wavelength limit of the "liquid virtual crystal" model of a binary alloy liquid structure factor at the melting temperature, $S\overset{\cdot}{T}_M{}^{AB}(0)$, to estimate the long wavelength sound velocity for a group of simple, nearly equi-valent and equi-volume binary liquid alloys. We compare the calculated sound velocities to the available experimental sound velocities in these liquid binary alloys and find reasonable agreement.

Omini[1], utilizing the Percus-Yevick[2] collective coordinate theory of liquids, has calculated the melting entropy of a series of simple metals. Percus and Yevick[2] derived a 3N collective coordinate theory of simple hard sphere liquids in which the potential energy can be written as that of an assembly of 3N harmonic oscillators (liquid phonons)

$$V^{P.Y.} = 1/2 \sum_{\{\underline{k}\}} \nu_{\underline{k}} \, (q_{\underline{k}} \, q_{-\underline{k}} - N) \qquad (1)$$

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where the set $\{\underline{k}\}$ are chosen to optimize the problem, and the $\nu_{\underline{k}}$ are coefficients to be determined. This "liquid phonon" representation was performed in terms of 3N collective boson coordinates

$$q_{\underline{k}}^{\text{P.Y.}} = \sum_i e^{i\underline{k}\cdot\underline{x}_i} \tag{2}$$

These liquid phonons have a frequency spectrum (dispersion relation)

$$\omega_{\underline{k}}^{\text{P.Y.}^2} = k^2 k_B T / m (1 + \nu_{\underline{k}} / k_B T) \tag{3}$$

with a maximum occurring, for simple metals, at about half the cut-off frequency and about half the cut-off wavevector (about one third the cut-off wavevector in Pb)

$$Q = (18\pi^2 \rho / m)^{1/3} \tag{4}$$

and

$$(\omega_{\underline{k}}^{\text{P.Y.}^2})_{\text{MAX}} = (18\pi^2 \rho / m)^{2/3} \; T / 2m (1 + \nu_{\underline{k}} / k_B T) \tag{5}$$

respectively, where m is the liquid particle's mass and ρ is the liquid density. Using the Ascarelli-Harrison-Paskin[3] relation between the long wavelength limit of the liquid structure factor for liquid metals, and extending it to liquid alloys

$$S^{AB}(0) = k_B T / (k_B T + \nu_{\underline{k}}^{AB}) \tag{6}$$

we can rewrite the Percus-Yevick[2] alloy liquid phonon dispersion relation at the melting temperature as

$$(\omega_{\underline{k}}^{\text{P.Y.}^2})_{AB} = k^2 k_B T_M^{AB} / \tilde{m}_{AB} S_{T_M}^{AB}(0) \tag{7}$$

where the alloy "particle" effective mass is defined by

$$\tilde{m}_{AB}^{-1} = m_A^{-1} + m_B^{-1} \tag{8}$$

Thus, the dispersion relation maximum becomes, at the melting temperature

$$(\omega_{\underline{k}}^{\text{P.Y.}^2}(T_M^{AB}))_{\text{MAX}}^{AB} = \\ = (18\pi^2 \rho^{AB}(T_M) / \tilde{m}_{AB})^{2/3} k^2 k_B T_M^{AB} / \tilde{m}_{AB} S_{T_M}^{AB}(0) \tag{9}$$

This is a temperature dependent Percus-Yevick[2] cut off in wavevector and frequency, which we choose to evaluate at the melting temperature. We

utilize Siegel's[4] adaption of the Omini[2] and Percus-Yevick[2] theories to liquid binary alloys, the liquid virtual crystal model. Then, the sound velocity at the melting temperature is

$$v_s^{AB}(T_M) = (k_B\ T_M^{AB} / \tilde{m}_{AB}\ S_{T_M}^{AB}(0))^{1/2} \qquad (10)$$

Siegel[4] calculated $S_{T_M}^{AB}(0)$ values for the following liquid binary alloys: *Li*Na, *K*-Rb, *Rb*-Cs, *Al*-Zn, *Zn*-Ga and *Al*-Ga, which were chosen because they were the most nearly equi-valent and equi-volume combinations of the simple metals the Omini[1] theory was performed upon. Since they are equi-valent and equi-volume, $S^{AB}(0) = S^A(0) = S^B(0)$.

In Table I we estimate the long wavelength sound velocities in these alloys at their melting temperatures as a function of solute concentration, and compare them to available experimental data. In Figure 1 we plot the sound velocities, $v_s^{AB}(T_M)$, versus percent solute concentration. We note that in all alloys considered, the sound velocity at long wavelengths drops upon alloying, rising again to some value at 100% pure solute. The calculated sound velocities compared with the experimental agree rather well. The drop

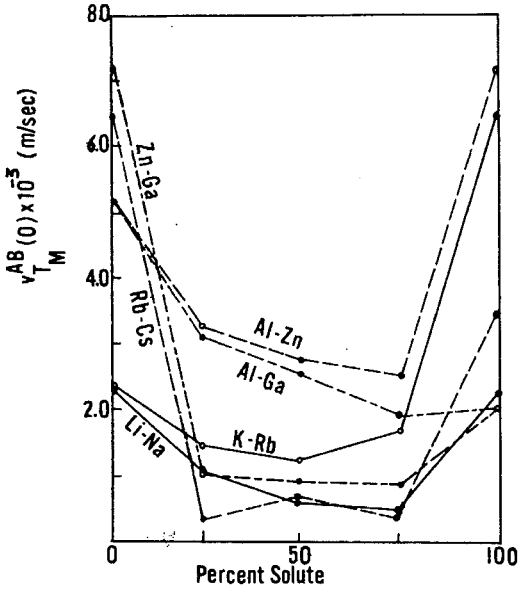

FIGURE 1 Sound velocities at the melting temperature for a variety of simple metal alloys as a function of solute concentration. All of these alloys are equi-valent and equi-volume, and so are virtual liquid crystal like, with one liquid structure factor.

TABLE I

Theoretically predicted sound velocities in the long wavelength limit at the melting temperature from Percus-Yevick liquid alloy structure factors and experimental sound velocities

Liquid alloy system	Liquid binary alloy	T_M^{AB}(°K.)	$\tilde{m}_{AB}^{-1} \times 10^{23}$ (gm.)	$S_{T_M}^{AB}(0)$	$v_s^{AB}(T_M)$ Thy(m/sec)	$v_s^{AB}(T_M)$ Exp. (m/sec)
Li-Na	Li_{100}	479.4	0.05424	.04 3079	2,2 83.	
	$Li_{75}Na_{25}$	670.	0.07061	.01 3702	1,1 16.	
	$Li_{50}Na_{50}$	660.	0.07061	.02 2430	7 58.	
	$Li_{25}Na_{75}$	540.	0.07061	.04 0219	5 12.	
	Na_{100}	397.8	0.01637	.00 6409	2,2 88.	2,531
K-Rb	K_{100}	363.	0.00963	.00 9923	2,2 90.	1,880.
	$K_{75}Rb_{25}$	345.	0.01403	.01 6726	1,4 25.	
	$K_{50}Rb_{50}$	338.	0.01403	.02 0837	1,2 63.	
	$K_{25}Rb_{75}$	335.	0.01403	.02 6790	1,6 96.	
	Rb_{100}	338.9	0.00440	.01 1465	6,5 87.	1,260.
Rb-Cs	Rb_{100}	338.9	0.00440	.01 1465	6,5 87.	1,260.
	$Rb_{75}Cs_{25}$	314.	0.00723	.54 3431	3 32.	
	$Rb_{50}Cs_{50}$	309.	0.00723	.19 4913	5 50.	
	$Rb_{25}Cs_{75}$	318.5	0.00723	.34 1094	4 22.	
	Cs_{100}	328.4	0.00283	.01 3117	3,4 95.	967
Al-Zn	Al_{100}	960.	0.01395	.00 3642	5,2 39.	4673.-4730.
	$Al_{75}Zn_{25}$	880.	0.01971	.00 5862	3,2 43.	
	$Al_{50}Zn_{50}$	820.	0.01971	.00 6971	2,8 70.	
	$Al_{25}Zn_{75}$	750.	0.01971	.00 8418	2,4 98.	
	Zn_{100}	719.5	0.00576	.00 3260	7,2 72.	2850.
Zn-Ga	Zn_{100}	719.5	0.00576	.00 3260	7,2 72.	2850.
	$Zn_{75}Ga_{25}$	615.	0.03433	.02 0479	1,0 99.	
	$Zn_{50}Ga_{50}$	550.	0.03433	.02 3157	9 77.	
	$Zn_{25}Ga_{75}$	450.	0.03433	.02 4715	8 57.	
	Ga_{100}	329.8	0.00540	.02 0421	2,0 33.	2873.
Al-Ga	Al_{100}	960.	0.01395	.00 3542	5,2 39.	4673.-4730.
	$Al_{75}Ga_{25}$	830.	0.01935	.00 6212	3,0 87.	
	$Al_{50}Ga_{50}$	715.	0.01935	.00 8212	2,4 92.	
	$Al_{25}Ga_{75}$	555.	0.01935	.01 0998	1,8 97.	
	Ga_{100}	329.8	0.00540	.02 0421	2,0 33.	2873.

in sound velocity upon alloying in the liquid state is to be expected, as alloying contributes another form of disorder scattering in addition to the positional disorder inherent in the liquid state, atom type disorder scattering.

In conclusion, the use of the liquid virtual crystal model of liquid alloys, with Omini's[1] adaption of the Percus-Yevick[2] theory of melting, allows us to compute reasonable sound velocities from the liquid phonon dispersion relation at the melting temperature. One could extend this to any temperature by recalculating $S^{AB}(0)$ at that temperature, and one could try an extension of this work to non-equi-valent and non-equi-volume binary alloys by calculating the correct $S^{AB}(0)$, $S^{A}(0)$ and $S^{B}(0)$, the three unequal structure factors, and utilizing an analog of their relation to $v_s(T)$ when they are all equal. This would go beyond this liquid virtual crystal approximation, which is only a first, simplest assumption. One should be cautious about the use of the liquid virtual crystal approximation for concentrated liquid alloys, both here and in future calculations, since in solid alloys, the virtual crystal approximation breaks down in the non-dilute case. We utilized this approximation here merely as an illustrative first step in the solution of the hard problem of sound wave scattering in a liquid alloy. The Percus-Yevick dispersion relations of Omini[1] have a maximum at Q/2, not at Q, for most simple liquid metals (except Pb, where the maximum is at Q/3) so that the estimated sound velocities are already in error by a factor of two, due to the non-monatonicity of the liquid phonon frequency spectrum.

References

1. M. Omini, *Phil. Mag.* 26, 2, 287 (1972).
2. J.K. Percus and G.T. Yevick, *Phys. Rev.* **110**, 1 (1958).
3. P. Ascarelli, R.J. Harrison and A. Paskin, *Adv. Phys.* **16**, 717 (1967).
4. E. Siegel, to be published.
5. R.T. Beyer and E.M. Ring, in *Liquid Metals*, Ed. by S. Beer, Marcel Deker Inc., New York (1972).