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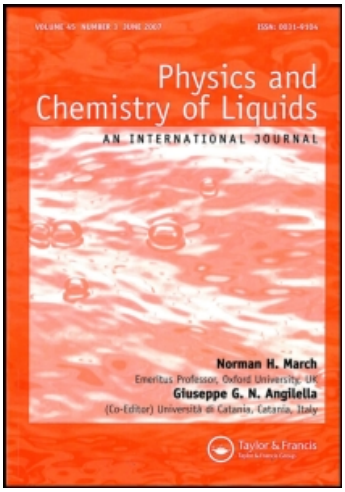

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Surface Thickness of Liquid Metals Related to Surface Energy and Bulk Compressibility: A Universal Relation

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LETTER

Surface Thickness of Liquid Metals Related to Surface Energy and Bulk Compressibility: A Universal Relation

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Recent experimental data for the surface tension σ and isothermal compressibility K of the rare-earths, combined with similar data for other metals known previously, allows us to conclude that the product $(\sigma K)_{Tm}$ near the melting point Tm varies little over the whole range of metals. Since $(\sigma K)_{Tm}$ can be interpreted as the width L of the liquid–vapour interface, the conclusion is that this interface is very sharp for all metals. The rather small variation of L between different groups of metals appears to be related to the different valence.

KEY WORDS: Liquid rare earth metals, surface tension, bulk compressibility, surface thickness.

Previous work[1,2] has shown that the product $(\sigma K)_{Tm}$ of the surface tension σ and the isothermal compressibility K of liquid metals near the melting temperature Tm varies only by a factor between 2 and 3 over the range of liquid metals for which data on both σ and K exists. The range of metals on which this observation is based includes simple and polyvalent nontransition metals, transition metals and noble metals. This observation is interesting because the separate values of σ and K vary by as much as a factor of 30 through this range of liquid metals. The product σK has the units of length. Then the values of the length.

$$L = (\sigma K)_{Tm} \tag{1}$$

range from about 0.2 Å to 0.5 Å. This length was interpreted by Egelstaff and Widom[3] as a fundamental length, characteristic of a substance, proportional to the thickness of the liquid–vapour interface. The proportionality constant depends on the precise definition of this thickness but it is of order unity. The conclusion is that such an interface is very sharp. Brown and March [4] have justified Eq. (1) from a simple density functional theory for a model of liquid metals strictly applicable, however, to the alkali group only.

Very recently, a compilation of the properties of liquid rare-earth metals has been published[5], allowing for a test of the correlation pointed out above for this group of metals.

In Table 1 we give the experimental values of the surface tension and isothermal compressibility for liquid rare earth metals near the melting temperature. The data

Table 1 Surface tension σ (Ref. 5), isothermal compressibility K (Ref. 6; values in brackets from Ref. 5), product σK and product Vn_b of atomic volume V and interstitial electron density n_b.

Metal	σ ($dyn\ cm^{-1}$)	K ($10^{-12}\ dyn^{-1}\ cm^{-2}$)	σK (Å)	Vn_b (e)
La	718	4.29 (4.24)	0.30 (0.30)	3.6
Ce	706	4.64 (5.30)	0.33 (0.37)	3.7
Pr	707	5.23 (4.17)	0.37 (0.30)	3.6
Nd	687	3.41	0.23	3.6
Eu	264	6.46	0.17	2.0
Gd	664, 810	3.07	0.20, 0.25	3.5
Tb	669, 840	3.40	0.23, 0.24	3.5
Dy	648	2.97	0.19	3.5
Ho	650	1.92	0.12	3.4
Er	637	2.28	0.15	3.4
Yb	320	4.48 (11.69)	0.14 (0.37)	1.9
Lu	940	1.88	0.18	3.4

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for the compressibilities have been obtained by two different techniques. Waseda and Ueno[6] obtained K from X-ray diffraction experiments through the relation of fluctuation theory[7]

$$S(0) = \rho K_B T K \tag{2}$$

where $S(0)$ is the long wavelength limit of the structure factor $S(q)$, ρ is the number density of atoms, K_B is the Boltzmann constant and T the temperature. On the other hand, the values in brackets for La, Ce, Pr and Yb have been derived from sound velocity measurements[5,8]. The large discrepancy between the two experimental values for the compressibility of Yb is especially noticeable.

The product $L = (\sigma K)_{Tm}$ varies between 0.12 Å and 0.37 Å for the metals in Table 1, with the average value of 0.23. For comparison we recall that the average value of L is 0.43 for alkali metals, 0.35 for alkaline-earths, 0.19 for other polyvalent nontransition metals and 0.21 for transition metals. We then conclude that a universal property of liquid metals is that the liquid–vapor interface is sharp, with a thickness that varies relatively little between different metal groups or between the different metals within a group. This universality should motivate the investigation of a theoretical description of the phenomenon which, except for the oversimplified case of alkali metals treated by Brown and March[4], is to our knowledge still lacking.

The variation of L within the rare-earth group is difficult to analyze, considering the considerable uncertainty in the compressibilities (compare the two sets of measurements for La, Cr, Pr and Yb). The average value of L, namely $\bar{L}$ (rare-earths) = 0.23, is similar to $\bar{L}$ (transition metals) = 0.21. In Ref. 2 Alonso and Silbert use some empirical relations between σ, the interstitial electron density n_b, K and the atomic volume V, to arrive, starting from Eq. (1) above, at the following expression for L:

$$L = c/Vn_b, \tag{3}$$

where c is a constant and the product Vn_b can be interpreted as an effective valence. Using values of V and n_b from de Boer *et al.*[9] we have arrived at the results given in the last column of Table 1. The effective valence has values consistent with current ideas about rare-earths, that is Eu and Yb are divalent and the rest trivalent. Also these values are similar to those obtained for transition in metals in Ref. 2, which explains the similar values of the surface thickness of liquid transition and rare-earth metals.

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