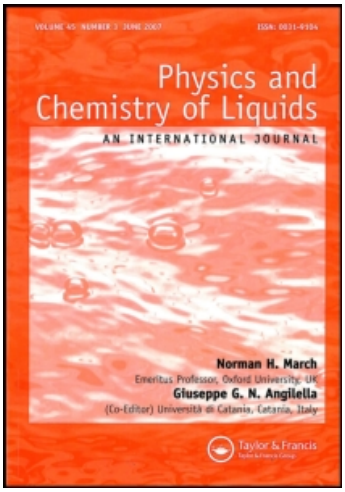

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Are Long-range Dynamical Interactions Between Screened Ions in Liquid Metals Observable in Atomic Transport and Neutron Scattering?

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Letter

Are Long-range Dynamical Interactions Between Screened Ions in Liquid Metals Observable in Atomic Transport and Neutron Scattering?

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Indirect static interactions between two screened ions at separation R in simple liquid metals fall off as $\cos 2k_f R/R^3$ where a sharp Fermi sphere, radius k_f, is assumed. Recently it has been proposed that long-range dynamic interactions exist, of asymptotic form $\cos 2k_f R/R^2$.

Pure isotope measurements on light liquid metals offer a possible means of observing such effects, though order of magnitude estimates of the effects seem presently difficult. However, fairly strong electron-ion interaction, plus large isotopic mass difference, are clearly needed. Li^6 and Li^7 are therefore favourable. The shear viscosities of these isotopes have been measured and seem to support the present ideas at least qualitatively.

One of us (H. S.) has been interested in the electronic contribution to friction in the context of the theory of Kramers for the dynamics of adatoms near metal surfaces.[1] In this framework, Suhl *et al.*[1] recognized that the electronic part of the friction constant was directly related to conduction electron Fermi surface scattering off the adatom. Subsequently, McCaskill and March,[2,3] by examining this scattering off two screened ions at separation R, have pointed out the existence of a long-range dynamical interaction between such ions, having the asymptotic form $\cos 2k_f R/R^2$ in a bulk electron gas. This is to be contrasted with the static energy of interaction between screened

ions in simple metals, which, again with the assumption of a sharp spherical Fermi surface, is known[4] to fall off as $\cos 2k_f R/R^3$.

The purpose of this Letter is to propose a possible test for the existence of such dynamical interactions. It seems clear that one should attempt to compare systems in which the dynamics changes in the simplest possible manner, namely by changing atomic masses only, not force fields. Then one thinks immediately of pure isotopes of liquid metals. To study relative atomic motions, which should reflect the long-range dynamical interactions mediated by conduction electrons, two obvious, and not independent, studies come to mind:

i) To measure the van Hove correlation function $S(k, \omega)$ by inelastic neutron scattering for two different isotopes of the same liquid metal.

ii) To measure shear viscosity η, or sound wave attenuation, related to $\frac{4}{3}\eta + \zeta$, where ζ is the bulk viscosity.

Of course, the availability of suitable isotopes is all important for (i) and therefore, after mentioning briefly the conditions which should be favourable for observing long-range dynamical interactions, the remaining comments will be largely confined to (ii) as possible observational techniques.

Rather obviously, one would need the following conditions met to observe what are inevitably rather small effects of dynamic interactions:

a) As large a ratio of isotopic masses, say M_1/M_2, as possible. This seems to leave one with the light liquid metals Li and Be as much the most favourable. Be seems excluded by its toxicity. As outside possibilities, Na, Mg and Al might merit consideration later.

b) As strong an electron-ion interaction as is consistent with a nearly-free electron situation, in which a sharp Fermi surface is a reasonable assumption. This means an electronic mean free path in the liquid which is large compared with the mean interparticle spacing.

As to (b), Na (and K) would be unfavourable as the metals which are nearest to ideal free electron liquid metals. However, Li remains rather favourable, the electron-ion interaction being quite strong because there are no p-electrons in the core against which to orthogonalize the conduction band wave functions. Thus, the pseudopotential is quite strong and yet the electrical resistivity at the melting point is only 15% higher than that for another, heavier alkali Rb. Clearly then Li is favourable on the grounds of both (a) and (b) above.

Almost 15 years ago, attention was drawn[5] to the challenge to liquid metals theory presented by the measurements of Ban *et al.*[6] on the shear viscosity η of Li^6 and Li^7. This seemed a particularly simple system for

Rowlinson[7] had earlier pointed out that the relation between the viscosity of different isotopes, 1 and 2, could be obtained on essentially dimensional grounds as $\eta_1/\eta_2 = (M_1/M_2)^{1/2}$. Brown and March confirmed this by showing that, with a total potential energy function independent of mass, the van Hove function $S(k, \omega)$ scaled, for a pure isotope of mass M, as

$$S(k, \omega) = M^{1/2} f(k, \omega M^{1/2}, T). \tag{1}$$

This led, using Green–Kubo formulae for transport coefficients, to $\frac{4}{3}\eta + \zeta\alpha M^{1/2}$ and by a slight generalization of the theory, to η proportional to $M^{1/2}$ as obtained by Rowlinson.

Clearly, however, the arguments of Rowlinson, and of Brown and March, no longer hold rigorously in the presence of a dynamical interaction. The measurements of Ban *et al.* of η for Li^6 and Li^7 led to the ratio of the viscosities as more like the ratio of the masses to the power 5/2 at the melting point, reducing to something like a power of 3/2 at about 100°C above the melting point. In view of the arguments set out above, it is tempting to ascribe these effects to the presence of conduction electrons, which mediate a long-range dynamical interaction between screened ions.[2,3]

In summary, it seems of some interest to study further experimentally viscosity, sound wave attenuation, and if there is a feasible system, neutron scattering from isotopes of the same light liquid metal. Departures from established sum rules on $S(k, \omega)$ should occur in the presence of dynamical interactions. We note finally that the assertion above that the conduction electron scattering in Li metal is responsible for the deviations of the measurements of η of Ban *et al.* from classical root mass scaling is supported by the subsequent measurements of Bratby and Harris[8] on Ne isotopes, where, after small corrections for conventional quantal effects, no such difficulties with simple mass scaling remain. Conventional quantal effects above the melting temperature of Li, in contrast, can be safely neglected.

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