Evidence for an Instability Near $2k_f$ in $Li(NH_3)_4$*

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Introduction

When an alkali metal is put in liquid ammonia the outermost electron of the alkali metal separates from the ion, resulting in a free electron and an alkali metal ion. At the solubility limit, about 20 mole percent metal (MPM) for lithium $Li(NH_3)_4$, the system is a good liquid metal down to its freezing point, T_F = 89K, but it has a low electronic density compared to typical metals. We describe the electron density by *rs,* which is approximately the ratio of the Coulomb-to-kinetic energies for the electrons. For a saturated solution of $Li(NH_3)_4$, $r_s \cong 7.4$. We carried out high-resolution (~2 meV) IXS measurements at T = 240K of the low-energy excitations of $Li(NH_3)_4$. The system has a Fermi momentum $k_F \cong 0.49$ Å^{-1}.

Methods and Materials

These experiments took place at beamline 3-ID at the Advanced Photon Source. A primary monochromator provided 21.65-keV x-rays, which were further monochromatized by a four-bounce monochromator and focused onto a 150 x 350 mm^2 spot. Scattered x-rays were reflected by a temperature controlled, spherically bent, diced Si (18 6 0) analyzer 6 m from the sample in a near backscattering geometry. The measured energy resolution was 2.4 meV.

Results

Figure 1 shows the results of the fit to the data for the energy of the excitations. At low q the phonons disperse to higher energies, as is expected for a sound-like mode, and the intensity of the mode increases. The mode position and intensity have maxima near q = 0.5 Å^{-1} and then are reduced dramatically (to zero within our resolution) as q goes to $2k_F$. If the observed phonon-like mode has only softened due to its interaction with the electrons, the minimum in the dispersion curve is perhaps a hint that we may be close to an instability of the ground state. But if the mode truly goes to zero energy, than it would imply a new ground state

Figure 2 shows the structure factor for $Li(NH_3)_4$ and pure ammonia measured with a ~1-eV incident beam (therefore integrating over the inelastic cross section). The large first peak occurs within error at the same momentum transfer where our data in Fig. 1 showed an extremely soft collective mode. To explain the greater height of the first peak compared to the second peak (which is the nitrogen-nitrogen distance), we must assume some reasonable degree of long-range spatial ordering of the coupled lithium ion free electron systems.

Discussion

As a first approximation to the Li-NH_3 system we take the two component ion-electron jellium model of Bardeen and others,[1] where the strongly interacting electrons are only coupled by the Coulomb interaction to mobile monovalent ions. The low-lying excitations of the jellium model are given by $\omega^2(\mathbf{q}) = \frac{\Omega_p^2}{\varepsilon_e(\mathbf{q},\omega)}$. Here, Ω_p is the ion's plasma frequency, and ε_ε is the dielectric function for the electron gas. At higher momentum transfer, weak coupling approximations such as the Lindhard dielectric function predict weak singularities in derivatives of the dielectric function at 2 k_F. Such non-analytic behavior can result in so-called "Kohn anomalies," i.e., softening in the phonon spectrum as well as a periodic variation, the so-called "Friedel or Ruderman-Kittel oscillations," in the real space pair potential between Li ions. In our case it is clear that a dramatic reduction in the phonon frequency implies that, in this model, there must be, at least, a strong peak in the dielectric function near 2 k_F.

In the early 60s, Overhauser, using a Hartree-Fock description, argued that the ground state of jellium at any *rs* might include spin and charge density waves (CDW) at even multiples of 2 k_F. Overhauser also proposed that many of the anomalous properties of the alkali metals could be explained by the formation of a CDW. We believe that the structure in $Li(NH_3)_4$ may be driven by the tendency of the electrons to order at 2 k_F.

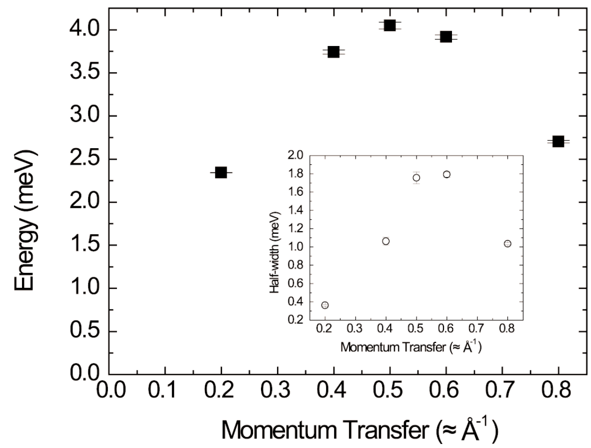

FIG. 1. Fits of the raw data for $Li(NH_3)_4$. The central peak width is shown in the insert.

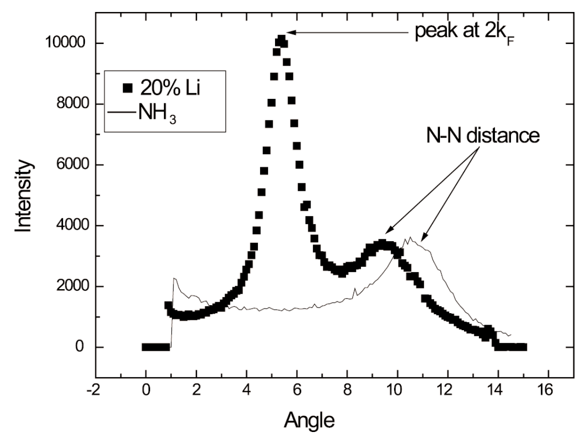

FIG. 2. The structure factor in pure NH_3 and $Li(NH_3)_4$.

Acknowledgments

This work was supported by the DOE, Division of Materials Science grant DE-FG02-99ER45772 and by Sector 3-ID, SRI-CAT, under DOE/BES contract no. W-31-109-ENG-38.

References

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[1] See, for example, J. R. Schrieffer, *Theory of Superconductivity*, (W. A. Benjamin, New York, 1964).

[2] A. W. Overhauser in *Highlights in Condensed Matter Theory*, F. Bassani, F. Fumi, and M. P. Tosi, eds. (North-Holland, Amsterdam, 1985), and references therein.