Collective Excitations in a Metal-Ammonia System as a Function of Electron Density

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Introduction

Alkali metals dissolve in liquid ammonia without chemical reaction, resulting in a free electron and a positively charged alkali metal ion. Solutions of alkali metals in liquid ammonia have been the subject of many experimental and theoretical investigations [1, 2] for over a century.

The properties of metal-ammonia solutions depend on the concentration of the metal. For instance, the solution undergoes a metal-insulator transition between 3 and 8 mol% metal (MPM) [2] and is metallic for concentrations above 8 MPM [3]. The solution saturates at 20 MPM for lithium-ammonia and 17.5 MPM for the sodium-ammonia solutions [4]. At saturation, the Li-NH_3 solution has an electrical conductivity about three times that of Na-NH_3 and about 1% that of copper. Metal- ammonia solutions have a measured macroscopic viscosity about 10 times lower than that of typical liquid metals [5].

Metallic lithium-ammonia and sodium-ammonia systems have values of r_s ≈7.4-12, substantially larger than that of the common metals, which have values of 2–6. By changing the metal concentrations, we can easily reach any desired value of r_s, which makes this system valuable for testing theoretical calculations at low electron density.

Methods and Materials

The experiments were carried out at the high-resolution inelastic x-ray scattering beamline 3-ID-C at the APS. Some of the IXS data are shown in Fig. 1.

Results

We have used the damped harmonic oscillation function (DHO) convoluted with the instrumental resolution function to fit the acoustic modes in our data. One of the fitting parameters $\Omega(Q)$ is shown in Fig. 2. At low Q, a linear dispersion in $\Omega(Q)$ is observed for all concentrations, which is expected for a soundlike mode. The fitting results for the linear Q-dependence for the acoustic modes are shown in Fig. 2 as thin dashed lines.

The slopes of the linear dispersion are comparable to the adiabatic sound velocity measured by ultrasound [6] (see Table 1).

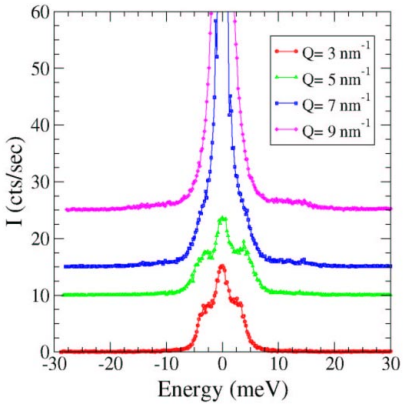

FIG. 1. Some of the raw data for lithium ammonia solution (20 MPM).

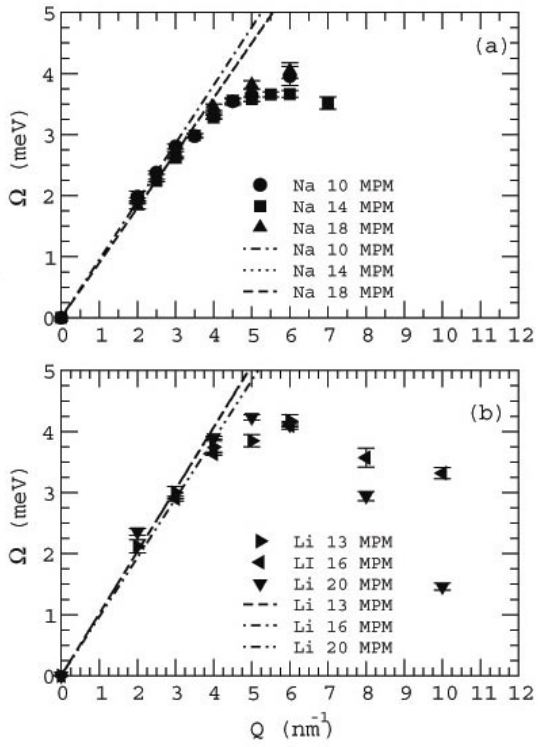

FIG. 2. The dispersion relation for (a) Na-ammonia and (b) Li-ammonia. The dashed lines are linear fits to the low Q dispersions.

TABLE 1. Sound velocities for different metal-ammonia concentrations. The adiabatic sound velocities are taken from Ref. 5.

Metal Ammonia (MPM)	Adiabatic Sound Velocity (m/s)	Sound Velocity from Data (m/s)
Li 13	1450±25	1550±70
Li 16	1450±20	1470±80
Li 20	1460±15	1540±100
Na 10	1400±20	1450±60
Na 14	1340±20	1410±67
Na 18	1340±20	1370±40

The dispersion relations for sodium-ammonia, shown in Fig. 2(a), show similar behavior for the measured concentrations, with the modes dispersing linearly at low Q and reaching a maximum around $Q_0/2$. For lithium-ammonia solutions, the dispersion relations in Fig. 2(b) show linear dispersion at low Q and maximum energy around $Q_0/2$.

Acknowledgments

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