Electronic Excitations in Solid and Liquid He-4

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Introduction

There have been few exciton measurements made for helium, all of which are for ^{4}He. The measurements have been of svp (saturated vapor pressure) liquid at 1.2K using EUV reflectance measurements,[1] clusters of sizes up to *N*=5000 using fluorescence measurements,[2],[3] and 13.5 cm^3/mole solid hcp ^{4}He using inelastic x-ray scattering,[4] This experiment was done to take better resolution measurements of helium excitons, to look for dispersion in the hcp solid and density scaling in the liquid.

Methods and Materials

This inelastic x-ray scattering experiment was conducted at ChemMatCARS on the 15-ID beamline. The energy of the monochromator was set to 9.8865 keV, with a measured bandpass of 1.1 eV FWHM. The analyzer used was a spherically bent silicon crystal employing the (555) reflection.

Helium crystals form only under applied pressure, even at the lowest temperature. Our setup (previously described by Venkataraman and Simmons[5]) involves a gas-handling system able to provide 210 MPa of pressure, and a cryostat able to go down to 10K. The helium samples were contained in a cylindrical beryllium sample cell (0.8 mm ID). Empty cell measurements at 25.0K were taken at the same orientation as the data measurements at the end of the run. A preliminary empty-cell measurement with a short count time is shown in Fig. 1, along with a data

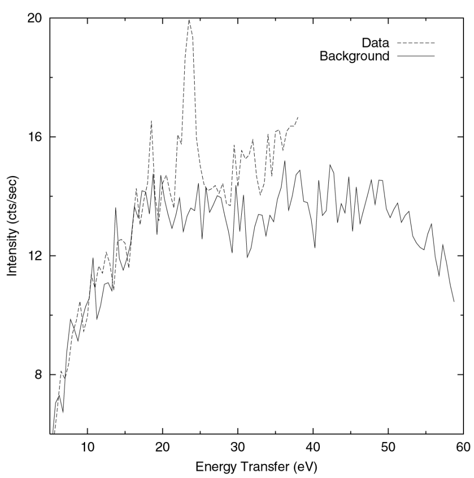

FIG. 1. These are preliminary scans of empty-cell background and data, showing the full range of the background due to beryllium excitations. The data had 30 s/point, while the background had 40 s/pt.

scan, showing the reproducibility of the beryllium excitations at energies lower than the helium exciton.

Three ^{4}He samples were studied: an hcp crystal at 14.0K with a molar volume of 10.72 cm^3/mole and two liquid samples at 25.0 K with molar volumes of 11.22 cm^3/mole (liquid A) and 12.27 cm3/mole (liquid B). The hcp crystal had a c lattice parameter of 4.782 Å, taken from the molar volume found from comparison of its measured melting point to previous melting curve studies.[6],[7] Three measurements were done for the crystal at different points in reciprocal space, near the (002) reflection along the c-axis.

Results

To extract the helium signal, the normalized empty-cell scans were directly subtracted from the normalized data scans without any scaling. Fig. 2 shows the data and empty-cell normalized

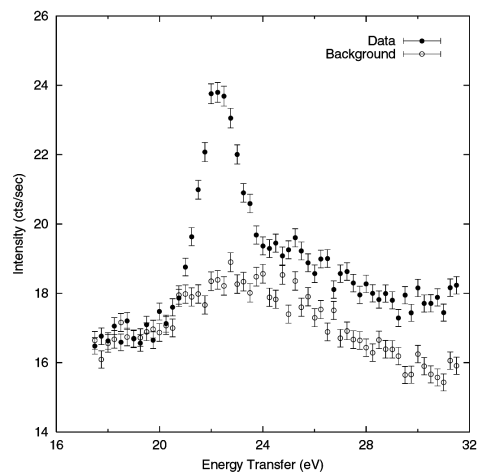

FIG. 2. These are the data and empty-cell normalized scans for 12.27 cm^3/mole liquid. The data scan was taken with 240 s/pt, while the background scan was taken with 300 s/pt.

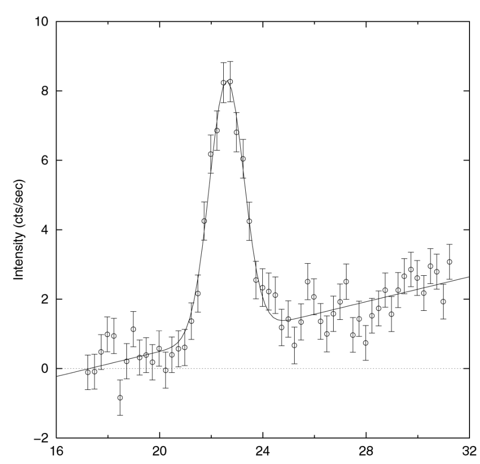

FIG. 3. This is the 30.0° measurement for the hcp crystal. A Gaussian plus a linear function has been fit to the data, giving the exciton energy as the position of the Gaussian.

Table 1.

Sample	**2θ (º)** ± 1.5	***Q*** **(Å^{-1})** ± 0.13	***Q****** (2π/c)	**Exciton** Energy (eV)
crystal	22.0	1.91	1.5	23.24
crystal	30.0	2.59	2.0	22.60
crystal	35.0	3.01	2.3	23.02
liquid A	30.0	2.59		22.60
liquid B	30.0	2.59		22.41

scans for liquid B. The empty cell subtracted data are fitted to a Gaussian plus a linear function. An example of this is in Fig. 3, which is the 30.0º crystal data. The Gaussian position is the energy of the exciton. The summary of results are in Table 1, where Q is the momentum transfer of the photons, and Q^* is the reduced momentum transfer in terms of the c-axis. The uncertainty of 2θ is ± 1.5º, while for Q it is ± 0.13 Å^{-1}.

Discussion

The three crystal measurements can be represented as along the *c*-axis of the third periodic zone at -1.0A, Γ, and 0.6A. The corresponding measured excitons exhibit dispersion with a minimum at the zone center Γ. The data support a calculated value of 31.0 eV for an interband transition across the minimum bandgap which occurs at Γ. The liquid measurements show a shift to higher energy for the exciton as the density is increased.

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