J. Synchrotron Rad. (1996). 3, 316–317

First observation of inelastic X-ray scattering from condensed ^{4}He using high energy resolution

N. Schell,[a] R. O. Simmons[b] and E. Burkel[c]

[a]*Forschungszentrum Rossendorf, Institut für Ionenstrahlphysik und Materialforschung, PF 510119, D-01314 Dresden, Germany,* [b]*Department of Physics, University of Illinois, Urbana, IL 61801, USA, and* [c]*Institut für Angewandte Physik, Universität Erlangen-Nürnberg, D-91054 Erlangen, Germany*

(*Received 19 May 1995; accepted 2 July 1996*)

Recently, the suitability of inelastic X-ray scattering for the investigation of solid and fluid ^{4}He has been demonstrated. For the test experiments an energy resolution of the order of 10–15 meV was used at the backscattering spectrometer INELAX at the storage ring DORIS of DESY, Hamburg. Lattice excitations were observed for momentum transfers along the c axis of h.c.p. helium crystals which were grown *in situ* at pressures of 54–63 MPa and at temperatures of 4.2–6.4 K. At 10 K above the melting point, energy-loss signals could also be detected from the liquid helium at equivalent momentum transfers.

Keywords: hexagonal close-packed helium-4; liquid helium-4; dynamical structure factor; phonons; inelastic X-ray scattering.

1. Introduction

Condensed helium isotopes ^{4}He and ^{3}He are prototypes of quantum solids and fluids of interacting bosons and fermions, respectively. Because of their simple atomic structure the experimental results can often be explained with first-principle theoretical studies. However, the remarkable quantum nature of the atomic constituents allows solidification only under externally applied pressure, even at low temperatures. The necessary pressure cell and the low-temperature environment restrict the experimental investigations.

Owing to the penetrating power of neutrons through sample containers, experimental data concerning lattice and single-particle dynamics have been available for some time. An overview of these activities, the different theoretical approaches and open questions can be found in the review of Glyde & Svensson (1987) and in the article of Blasdell, Ceperley & Simmons (1993). So far, there is little experimental data on the optical and electronic properties of solid helium. Only recently has the application of new synchrotron radiation techniques allowed this to change (Schell, Simmons, Kaprolat, Schülke & Burkel, 1995).

The relatively new technique of inelastic scattering of X-rays with high energy resolution (Burkel, 1991) is an excellent tool for giving valuable information on the excitation spectrum of condensed helium. For the isotope ^{4}He this will be complementary to inelastic neutron scattering. However, for ^{3}He this information will be unique because this isotope is a strong absorber of neutrons.

2. Experimental set-up

The test experiments on helium were performed at the INELAX (INELAstic X-ray scattering; for details see Burkel, 1991) spectrometer at the storage ring DORIS at DESY in Hamburg. This triple-axis spectrometer uses a scattering geometry close to back-reflection at the monochromator and the analyzer crystals. Grooved and spherically bent silicon crystals allow an energy resolution in the range 10–15 meV to be achieved. With this spectrometer the energy transfer is varied by temperature tuning of the analyzer crystal with respect to the monochromator crystal.

A modified liquid-helium flow cryostat with a high-pressure polycrystalline Be cell was installed at the sample position. The helium crystals ($\sim 15\,mm^3$ volume) were grown within this Be cell by pressurizing it through a steel capillary up to 62.5 MPa and cooling it down to 4.2 K. This pressure was generated by a thermal pump consisting of a pressure vessel which was cycled between 77 and 300 K (Venkataraman, Schell & Simmons, 1996).

At 56 MPa ^{4}He solidifies at 5 K in the h.c.p. modification with its c axis usually normal to the long axis of the cell, *i.e.* normal to the growth direction, owing to the large anisotropy of the thermal conductivity in h.c.p. ^{4}He. The crystal orientation was controlled by the registration of the (002) Bragg reflection. The growth process was reproducible and led to various crystals of similar quality with $\sim 0.25°$ mosaic spread.

3. Results

The main problem of these test experiments lies in the low scattering rate of ^{4}He. In addition, there are always inelastic scattering contributions of the Be pressure cell leading to a significant background signal. The data were taken at several values of the momentum transfer Q by recording up to ten individual scans. The energy was typically ramped from −15 to 30 meV in energy transfer using steps of 1.2 meV and a counting time of 2 min. The scans were normalized to a monitor signal and added up [details can be found in Schell (1994)].

After the measurements, the solid helium was melted at 20 K and the inelastic scattering of the liquid was registered. In the next step the helium was vented to atmospheric pressure and the Be contribution, including the residual He gas, was measured.

Despite the low photon flux in the scattering experiments the contributions of helium could be detected. This is demonstrated by Fig. 1, which shows the scattering intensities as functions of the energy transfer at momentum transfer $Q = 3.4\,Å^{-1}$, *i.e.* at (0 0 2.8) of h.c.p. ^{4}He at 55.5 MPa and 4.5 K. The intensity data for solid (a) and liquid (b) helium still include the Be signals. The empty Be cell, or in other words the corresponding background, is shown with a dashed line to guide the eye. Because of the low statistics, no difference signals will be shown here. Nevertheless, there are distinct scattering contributions due to the helium which are slightly different for the solid and for the liquid. In Fig. 1(a) a Gaussian curve is drawn using the instrumental resolution of 15 meV with the centre at $\sim$6 meV. Within the obvious error bars this value for a longitudinal phonon is in agreement with the phonon dispersion branch of h.c.p. ^{4}He as determined by neutron scattering after scaling to the appropriate pressure (according to Reese, Sinha, Brun & Tilford, 1971). The increased intensities visible in the range 20–34 meV in Fig. 1 are attributed to phononic contributions from the Be of the pressure cell. This intensity is even more distinct in Fig. 2, showing the inelastic X-ray scattering spectrum of h.c.p. ^{4}He at 62.5 MPa and 6.1 K for the lower momentum transfer $Q = 1.8\,\text{Å}^{-1}$ equivalent to (0 0 1.5) in the same way as in Fig. 1. Apart from the strong Be phonon contribution around 23 meV, there are also more elastic and quasi-elastic scattering intensities from Be close to zero energy transfer. The apparently reduced width of the Be phonon signal in this spectrum is ascribed to a geometrical resolution effect. Despite all these unwanted intensities there is also a distinct scattering signal visible near 6–8 meV due to a phonon in solid helium.

The interpretation of the spectrum of the liquid ^{4}He in Fig. 1(*b*) is not completely understood at present. There could be the indication of a propagating mode in addition to the quasi-elastic contribution within the dynamical structure factor as known from other liquids (Montfrooij, de Graaf & de Schepper, 1992; Sinn, 1991). On the other hand, it is possible that the spectrum consists of an overall broadened peak shifted from zero to ~7 meV. In this case it will have to be explained on the basis of single-particle excitations due to changed effective masses. Such an effective mass has already been found in inelastic neutron scattering experiments (Minkiewicz, Kitchens, Shirane & Osgood, 1973). The statistics of the preliminary inelastic X-ray scattering results available so far do not allow final conclusions. A more detailed study of this

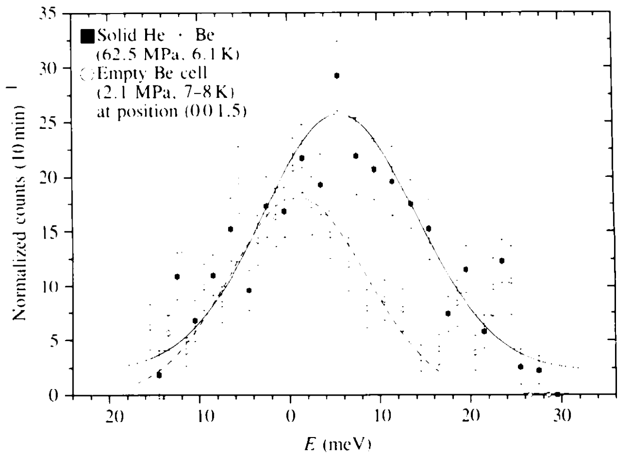

Figure 2
Scattering intensities at $Q = 1.8\,\text{Å}^{-1}$ as functions of the energy transfer for the pressure cell filled with h.c.p. ^{4}He at 62.5 MPa and 6.1 K and for the empty cell. For further details see text.

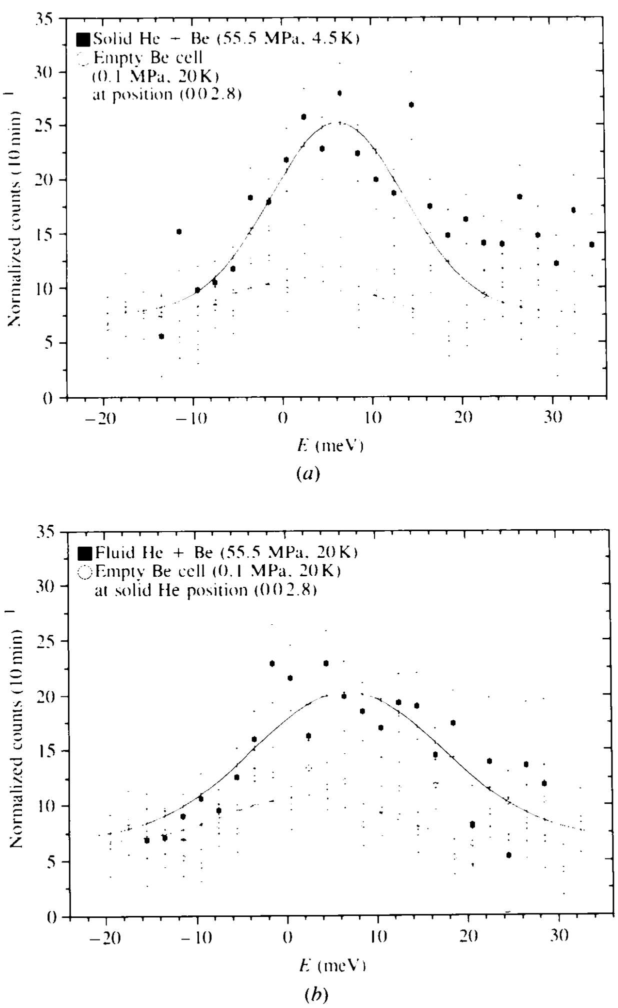

Figure 1
Scattering intensities at $Q = 3.4\,\text{Å}^{-1}$ as functions of the energy transfer for the pressure cell filled with (*a*) h.c.p. ^{4}He at 55.5 MPa and 4.5 K, and (*b*) liquid ^{4}He at 55.5 MPa and 20 K. The intensities for the empty cell are also shown.

phenomenon is necessary and is already underway at a resolution-enhanced successor spectrometer at a high brilliance source at the ESRF.

4. Summary

The first observations presented here already demonstrate the capability of inelastic scattering spectroscopy with high energy resolution to provide information on the lattice dynamics of solid helium and on the dynamical structure factor of liquid helium.

We would like to thank M. Schwoerer-Böhning and H. Sinn (University of Munich, Germany) for their help during beamtime at HASYLAB, and C. T. Venkataraman (University of Illinois, Urbana, USA) for fruitful discussions. This work was funded by the German Federal Ministry of Research and Technology under Contract 03 PE1 LMU 2 and supported by the US Department of Energy, BES-Materials Science Grant DE-FG02-91ER45439 and the Alexander-von-Humboldt Foundation.

References

Blasdell, R. C., Ceperley, D. M. & Simmons, R. O. (1993). *Z. Naturforsch. Teil A*, **48**, 433–437.

Burkel, E. (1991). *Springer Tracts of Modern Physics*, Vol. 125, edited by G. Höhler. Berlin: Springer.

Glyde, H. R. & Svensson, E. C. (1987). *Methods in Experimental Physics*, pp. 303–403. New York: Academic Press.

Minkiewicz, V. J., Kitchens, T. A., Shirane, G. & Osgood, E. B. (1973). *Phys. Rev. A*, **8**, 1513–1528.

Montfrooij, W., de Graaf, L. A. & de Schepper, I. M. (1992). *Phys. Rev. B*.**45**, 3111–3114.

Reese, R. A., Sinha, S. K., Brun, T. O. & Tilford, C. R. (1971). *Phys. Rev. A*, **3**, 1688–1698.

Schell, N. (1994). PhD thesis, Ludwig-Maximilians University, Munich, Germany.

Schell, N., Simmons, R. O., Kaprolat, A., Schülke, W. & Burkel, E. (1995). *Phys. Rev. Lett.* **74**, 2535–2538.

Sinn, H. (1991). Diploma thesis, Ludwig-Maximilians University, Munich, Germany.

Venkataraman, C. T., Schell, N. & Simmons, R. O. (1996). In preparation.