Electron Diffraction of Superfluid Helium Droplets

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Experimental details

The experimental apparatus combines a superfluid helium droplet source with a gas phase electron diffraction system. The droplet source consisted of a pulsed valve (Digital Technology Trading & Marketing Ltd., E-L-5-8-C-Unmounted Cryogenic Copper Even-Lavie Valve) pressurized to 70 atm, and the lowest achievable source temperature was 8 K. The 90° nozzle had an opening of 100 µm but the gasket had only a 50 µm opening. The resulting droplet pulse had a duration of 50 – 60 µs controlled by the driver from the same manufacturer of the pulsed valve (2009 Model Electronic Driver Unit). Phthalocyanine gallium chloride was enclosed in a cylindrical copper oven with a diameter of 3 cm and heated to 400°C. Two circular apertures of 5 mm in diameter on the wall of the oven were in line with the droplet beam for sample pickup. Under this condition, the vapor pressure of the sample was probably 10⁻⁵ Torr, and most droplets should only pickup one dopant molecule. Doping was confirmed from spectroscopic studies using laser induced fluorescence.¹ The helium droplet beam was divergent with an approximate diameter of 3 mm in the diffraction region, and we estimate that there were probably 10⁵ molecules arriving at the diffraction region per pulse.

The electron beam (Kimball Physics, EGH-6210A) was set at 40 kV, with a flux of 60 µA, a pulse duration of 10 µs, and a beam diameter of 2 mm. The resulting electron density was less than 1‰ electron/nm², far below the recommended damage threshold of 80 electron/nm².² The coherent length of our electron beam was ~ 100 nm (transverse, adjustable) and 1 µm
(longitudinal), and the estimated average distance between the sample molecules was over 3 µm. Under this condition, each molecule diffracted independently of the others without multiple scattering, and the resulting image was a simple addition of the diffraction signals from individual molecules. Diffracted electrons were imaged directly onto a phosphor screen (Kimball Physics, PHOS-UP22GL-CF6 P22) placed 43.2 cm downstream from the diffraction region, and a CCD camera (SBIG Astronomical Instruments, ST-8300M) was used to record the image without further amplification. The un-diffracted main electron beam at the center of the phosphor screen was blocked by a Faraday cup of ¼” in diameter and 2” in height.

The overall apparatus operated at 10 Hz, limited by the pumping capacity of the droplet source chamber. The relative timing among the beams was controlled by delay generators (Stanford Research, DG535). During the experiment, the readout time for a full frame download was 7.5 s, and pictures were typically accumulated for 10 min before being transferred to the computer. To eliminate diffraction of residual gases in the diffraction chamber and any potential optical interference from the alignment laser, a background picture was taken without the droplet beam for another 10 min. The difference between the two images was taken as the diffraction result of the droplet beam. This cycle of ~20 min was repeated for several hours to obtain the final diffraction result. Limited by several spots of blemish on the phosphor screen, only two wedges of 30° each were used to obtain the scattering intensity. Calibration of the diffraction parameters were guided by the diffraction image of evaporated aluminum on a 3 mm grid (Electron Microscopy Sciences, 80044), a standard for calibration of the camera length used in transmission electron microscopes.
Raw Data

Fig. s1 shows the raw image of electron diffraction from PcGaCl doped in superfluid helium droplets. The electron beam had a slight shift in position during the day, so each image had to be centered after a data collection period of ~ 20 min. Consequently, we could not directly add all the images together to get the average. Instead, we added and averaged the obtained radial profiles from each image after centering. Fig. s2 shows the resulting radial profile from an average of 72,000 raw images. This profile has been smoothed and truncated to avoid the effect of the Faraday cup and the edge of the phosphor screen. The dashed line is the calculated contribution from all the atoms of PcGaCl and from 150 helium atoms. The vertical axis of the calculated profile has been adjusted based on the best fit between the theoretical and experiment result. Subtraction of the overall diffraction profile from the contributions of atoms results in the final profile shown in Fig. 3.

![Fig. s1. A raw image of electron diffraction from PcGaCl in helium droplets. The dark profile corresponds to the shadow of the Faraday cup and its support arm.](image1)

![Fig. s2. Average diffraction intensity from images like that of Fig. s1. The dashed line represents contributions from all atoms of PcGaCl and from 150 helium atoms.](image2)
References

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