Spectroscopic wavelength shifts characterizing the phase transition of helium adsorbed on fullerene cations

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Synopsis Using high-precision spectroscopy and elaborate MD simulations, we have studied the absorption lines of the He$_n$C$_{60}$ complex (n ≤ 100) formed via electron ionization of helium nanodroplets doped with C$_{60}$. The observed variation of wavelengths with the number of He atoms provides clear evidence of the phase transition of helium adsorbed on C$_{60}^+$ cations.

The mysterious character of the diffuse interstellar bands (DIBs) had puzzled scientists until very recently, when four DIBs were unequivocally assigned to C$_{60}^+$ by laboratory spectroscopy [1]. Inspired by this seminal work, we have conducted a combined spectroscopic and molecular dynamics (MD) simulation study on the He$_n$C$_{60}^+$ complex with n exceeding 100 via electron ionization of helium nanodroplets doped with C$_{60}$. The addition of more He atoms gives rise to a pronounced blueshift, due to the displacement of He atoms from pentagonal faces into a fullererene adlayer, as revealed by MD simulations [3]. In this second adlayer, He atoms are mobile behaving like liquid. The blueshift continues to be observed up to n = 60, where the adlayer consists of 20 immobile He atoms above hexagons and 40 mobile delocalized ones. Beyond n = 80, the absorption lines do not change, implying the onset of superfluidity.

The observed phase transitions are supported by elaborate MD simulations including quantum effects [3]. By combining MD results with a model that describes the van der Waals interaction of the ground and first excited states of C$_{60}^+$ with the surrounding He atoms, the wavelength shifts have been evaluated, showing an impressive similarity to the experiments.

Figure 1. Center positions for the absorption spectra of He$_n$C$_{60}^+$ around 958 nm (blue circles, left y-axis) and 963.5 nm (red triangles, right y-axis), as a function of n. Simulated wavelengths are plotted as black squares.

References

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