Phase pure fullerene based superconducting thin films: in-situ growth and ultra-high vacuum Raman characterization.

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Thin films of organic superconductors (SCs) are an attractive platform for studying the effects of doping, layer thickness and substrate on the superconducting (SC) transition temperature and the SC mechanism. The fullerene-based family of A_3C_{60} (A is an alkali metal) SCs is of particular interest because of their high critical temperature (Rb_3C_{60} has Tc=30.9 K) and weak van-der-Waals interaction between the molecular film and the substrate. It has been shown that thin films of C60 grow epitaxially on topological insulators [1] and K_3C_{60} has been synthesized on a semiconductor surface [2]. Amongst the alkali metal intercalated fullerenes, only the A_3C_{60} phase is superconducting. However, it can co-exist with other phases such as the insulating A_6C_{60} phase and can form non-stoichiometric phases. Thus, achieving phase purity is an important open problem in the growth of thin films of organic SCs which needs to be solved e.g. for achieving proximity induced SC. Moreover, A_3C_{60} is air-sensitive and hence it must be studied in an ultra-high vacuum (UHV) environment. Raman is amongst the most direct methods that reveal the phase but the air sensitivity of A_xC_{60} precludes the application of standard Raman that is carried out in air or in high vacuum.
Here, we apply the UHV Raman technique to achieve growth of phase pure SC Rb_3C_{60} films that we perform in a home built optical chamber down to T=4.2K. We perform the full sample preparation (sputtering, annealing, and Rb_3C_{60} growth) and characterization (electron diffraction and UHV-Raman) in-situ [3]. It is found that even ~2-3 monolayers of Rb_3C_{60} have a sufficiently strong Raman response that can easily be measured inside UHV within several minutes. The stoichiometry of the thin film can be accurately controlled in-situ by tracking its Raman response. That is, the main Raman peak of C_{60} is at $1460cm^{-1}$, while in the case of Rb_3C_{60}, the peak moves to $1450cm^{-1}$.Finally, for Rb_6C_{60}, the peak shifts down further to $1435cm^{-1}$, as shown in Figure 1. An extension of the experimental setup it is also discussed to perform four-point transport measurements in tandem with UHV Raman [4].

References

[1] Rogge, S., Durkut, M., & Klapwijk, T. M. (2003). Physical Review B, 67(3), 033410.
[2] Latzke, D. W., Ojeda-Aristizabal, C., Griffin, S. M., Denlinger, J. D., Neaton, J. B., Zettl, A., & Lanzara, A. (2019). Physical Review B, 99(4), 045425.
[3] Nano Lett. 2018, 18, 9, 6045–6056
[4] Nature Communications volume 12, Article number: 2542 (2021)

Figures

Figure 1: Raman spectra of Pristine C_{60} (green), Rb_3C_{60} (orange) and Rb_6C_{60} (blue) with green 532nm laser.