Review of: "Extreme Ultraviolet Second Harmonic Generation Spectroscopy in a Polar Metal"

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In this work the authors try to understand the microscopic mechanism of the ferroelectricity of the polar metal LiOsO3 by combining the extreme ultraviolet second harmonic generation (XUV-SHG) spectroscopy measurements with the density functional theory (DFT) calculations. Though it has been proposed theoretically that the displacement of Li atoms plays a central role in stabilizing the ferroelectric phase, a direct detect detection of the corresponding variation of the electronic structure were still absent. This work provides clear evidence for broken inversion symmetry involving Li core-level electron excitations. The results are stimulating.

However, I have the following concerns:

1. The authors claim that their results suggest “a lack of OsO6 octahedral rotations”. But my understanding is that their calculation only implies the resultant susceptibility is insensitive to the rotation (Fig. S9.1). It is still possible that this kind of distortion presents in the real material, but because of the half-resonant condition of the experiment, the signal associated with it is weak.
2. In the last paragraph, the authors propose to study the phase transitions and critical fluctuations with the XUV-SHG technique. But for LiOsO3, I guess this would still be very difficult. In Fig.2e, the actual data near 58 eV are quite scattered and it would be really difficult to extract an accurate value of the Li displacement, which is important for the ferroelectric transition.