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### **Charge-Transfer Plasmon Polaritons at Graphene/ $\alpha$ -RuCl<sub>3</sub> Interfaces<sup>1</sup>**

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The fundamental opto-electronic properties of two-dimensional (2D) materials can be tailored based on their nanoscale charge environment. While electrostatic doping offers a means of wholesale tuning of 2D charge densities, the minimum size of charge features is limited by fields fringing through relatively thick gate insulators. Conversely, charge transfer at the interface of two atomically-thin layers with different work functions should not be subject to such limitations. Specifically, the large work function of  $\alpha$ -RuCl<sub>3</sub> (6.1 eV) makes it an ideal 2D electron acceptor. In our study, we exploit this behavior to generate charge-transfer plasmon polaritons (CPPs) in graphene/ $\alpha$ -RuCl<sub>3</sub> heterostructures. Using infrared near-field optical microscopy we measure the CPP dispersion, yielding a quantitative measure of the graphene Fermi energy ( $\sim 0.6$  eV) and thus the charge exchanged between  $\alpha$ -RuCl<sub>3</sub> and graphene ( $\sim 2.7 \times 10^{13}$  cm<sup>-2</sup>). Concurrently, we observe dispersive edge modes and internal “circular” CPPs which reveal sharp (<50 nm) changes in the graphene optical conductivity that correspond to nanoscale modulations in the graphene doping level. Further analysis of the CPP losses implies the presence of emergent optical conductivity in the doped interfacial layer of  $\alpha$ -RuCl<sub>3</sub> and suggests that it no longer possesses a Mott insulating ground state. Our results demonstrate that using high work function materials such as  $\alpha$ -RuCl<sub>3</sub> in Van der Waals heterostructures presents new opportunities for controlling the local charge carrier density of graphene and other 2D materials on nanometer length scales in excess of what can be achieved with an external gate.

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