Probing the Contact and Sliding of Elastomer/Polymer Interfaces\footnote{Financial Support from NSF (DMR-0512156)} BETUL YURDUMAKAN, The University of Akron, KUMAR NANNJUNDIAH, ALI DHINOJWALA — In this study, we have designed a novel approach to couple interface sensitive infrared-visible sum frequency generation (SFG) spectroscopy with adhesion and friction experiments. This provides a direct probe of the interfacial structure in terms of orientation and density of molecules during contact and sliding which is important in understanding the molecular origin of adhesion and friction. Here, we show that the friction forces between poly(dimethyl siloxane) (PDMS) lens and glassy poly( styrene) (PS) are $\approx 4$ times higher than PDMS sliding on surfaces of crystalline alkyl side chain comb polymers. This cannot be explained by the differences in adhesion energy or hysteresis. The in-situ SFG measurements indicate local interdigitation during contact, which is evident from the decrease in the number of oriented phenyl groups at the interface. The local penetration is unexpected at room temperature ($T_R$) that is much below the $T_g$ of PS. For comparison, we have also studied poly(n-butyl methacrylate) and poly(n-propyl methacrylate) having $T_g$ above and below $T_R$, respectively. Both of these polymers show similar adhesion and friction forces as PS. The SFG results indicate that local changes in interfacial structure affect friction, regardless of the bulk $T_g$. These results also show that the adhesion energy and hysteresis are not sufficient to predict friction, which makes the characterization of the molecular structure during contact and sliding essential.