

**Zhu *et al.* Reply:** We provide a reply to the Comment [1] by Lee and Yeom on results from our Letter [2].

Their first issue is that they thought our metallic state spectral weight was marginal with a V shape and it was not clear whether the system was indeed metallic. In our study [2], we defined the state when the Mott gap is closed as the “metallic state.” We presented the evolution of the spectral weight during the K doping to demonstrate the process of metallization, so whether or not the 0.32 ML is marginal does not affect our analysis. We showed the gap evolution with the K coverage in Fig. 2(c), including two coverages above 0.32 ML where the gap is also closed. The V-shaped feature (which in fact it is not a gap because of the finite density of state at the bottom of the V shape) does not influence our definition of the metallic state.

The Comment authors were also concerned about the inhomogeneity of the K-doped TaS<sub>2</sub> surface, the surface defects, and the subatomic variation in the scanning tunneling spectroscopy (STS) spectra, according to their own paper [3]. What we need to clarify is that the STS spectra in Fig. 2 of our Letter [2] are individual data taken at the center of the David stars without any adsorbed K atoms or defects, in order to exclude the influence of surface defects and the sub-unit-cell spectral variation. More importantly, as can be seen in our Letter [2], what we discovered is that the size of the Mott gap is almost unchanged, which is distinctly different from the defect- or misfit-induced metallization where the gap size is drastically reduced. When we claimed that the K-doped surface becomes metallic, we have already checked the STS spectra at many points of the surface. In fact, the metallic state is very uniform, which is ascribed to the uniformly distributed K atoms on the TaS<sub>2</sub> surface.

Their second issue is that they argued our calculation does not provide any sign of the metallic state, and also reinterpreted our theoretical result by moving the position of the Fermi level. The Comment authors may not have fully understood the purpose of our calculations. First, as stated in our Letter [2], the calculation is only used to explain the origin of the unconventional additional in-gap excitations near the top of the lower Hubbard band (LHB), which is in contrast to a conventional electron-doped Mott insulator. In our calculations, the coverage of K atoms cannot be varied continuously because high coverage will break the rotation symmetry of the clusters we used, so we only present the results for one K atom per 13 David stars. Thus, the system for such small doping is still an insulator. Second, the Comment authors moved the calculated Fermi level in their analysis to make an alignment with our experimental data at 0.32 ML, as shown in Fig. 1 of the Comment [1]. We emphasize that the way they analyzed our data is not correct because the Fermi level is only self-consistently determined by the electron concentration in the calculation and cannot be arbitrarily moved. In addition, the

K concentration of 1/13 in our calculation as described above does not correspond to the 0.32 ML for the experimental data.

The metallization of the Mott insulator by filling up the gap with additional excitations has been clearly demonstrated by our experimental data. From Fig. 2(a) of our Letter [2], one can see that the additional excitation starts to appear near the LHB at  $\sim 0.05$  ML, and its spectral weight increases and extends gradually to a larger region of the gap with doping. While in this process, the Mott gap defined as the difference between the upper Hubbard band (UHB) and LHB is basically unchanged. It is a typical demonstration of the filling up of the gap. Because the total spectral weight should remain constant, the increased spectral weight of the in-gap excitations is transferred mainly from the UHB as shown by the experiment, and leads to a decrease of the spectral weight of UHB. At  $\sim 0.32$  ML, the additional excitations eventually extend to the whole gap region, and the spectral weight of UHB nearly vanishes. The Mottness is thus collapsed as the Comment authors pointed out, and the transition from the Mott insulator to a metallic state occurs. The phenomenon that the additional excitation appears near the LHB is in contrast to the conventional picture of doping a Mott insulator by electrons. So, we carried out the theoretical calculation to give an explanation in our Letter [2]. As shown in Fig. 4 of Ref. [2], our theoretical result indeed clarifies that the additional excitation appears near the LHB and the spectral weight UHB decreases accordingly.

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