Spectroscopic Evidence for the Realization of Two-Dimensional Electride States in Y_2C

Two-dimensional (2D) electrides are a new material concept in which anionic electrons are confined in the interlayer space between positively charged layers. In order to verify that the formation of 2D electride states is realized in actual materials, we carried out angle-resolved photoemission spectroscopy measurements on Y_2C, which is one of the possible candidates for 2D electrides. We clearly observed the existence of "2D electride bands" near the Fermi level, as predicted by *ab initio* calculations. This is direct proof of the realization of 2D electride states in Y_2C.

Electrides are ionic crystals in which electrons serve as anions [1]. The breakthrough discovery of the first room-temperature, air-stable inorganic electride $[Ca_{24}Al_{28}O_{64}]^{4+}\cdot 4e^-$ [2] has opened up a new avenue in the application of electrides, capitalizing on their low work functions, such as their use as electron injection layers in organic light emitting diodes [3] and as catalysts for ammonia synthesis [4].

The electronic properties of electrides are expected to depend critically on the topology and dimensionality of the void space confining the anionic electrons. Thus, electrides have recently attracted considerable attention as a new class of low-dimensional materials. A recent study reported the synthesis of the first possible two-dimensional (2D) electride, Ca_2N [5], and further, the possible existence of 2D electride states in Ca_2N has been supported by *ab initio* calculations [5]. Subsequently, Inoshita *et al.* predicted Y_2C as another possible candidate for 2D electrides [6].

These materials have a unique layered structure, as shown schematically in Fig. 1. In the case of Y_2C, two anionic electrons per Y_2C unit cell ($[Y_2C]^{2+}\cdot 2e^-$) are confined in the interlayer space between positively charged ($[Y_2C]^{2+}$) sheets. The meaning of "2D electrides" is that the orbitals of interlayer electrons overlap each other in plane, but hardly overlap in out-of-plane directions. Thus, the in-plane overlapping of interlayer electrons causes the formation of 2D metallic "electride bands" near the Fermi level (E_F) [5, 6].

The crucial issue in the research of 2D electrides is whether or not the material concept is actually realized. Although there have been various experimental results

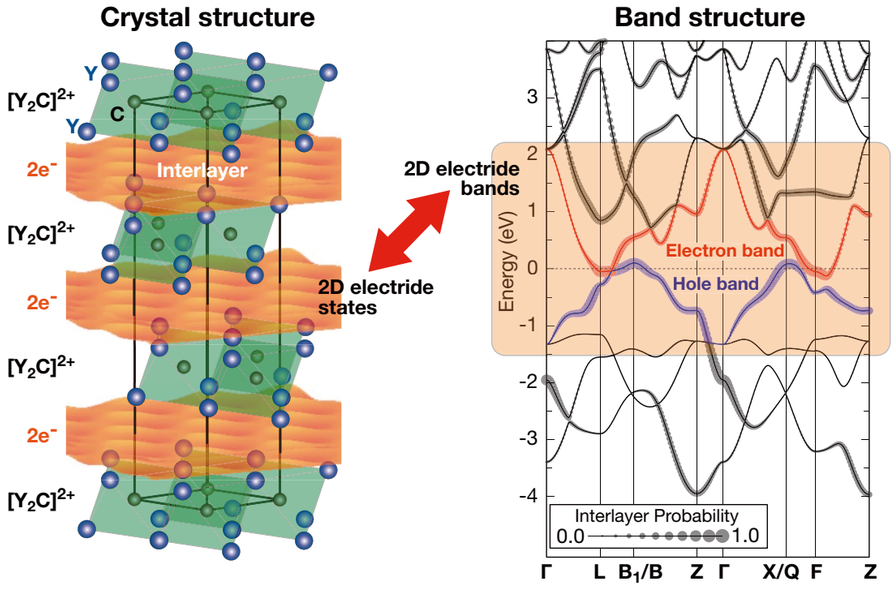

Figure 1: The crystal structure and the theoretical band structure of Y_2C [8].

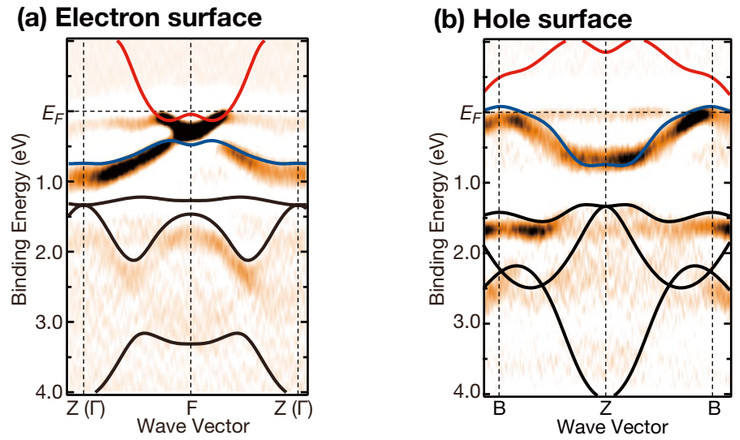

Figure 2: Experimental band structure obtained by plotting the second derivative of the ARPES spectra acquired along the (a) $Z(\Gamma^-)$–F–$Z(\Gamma^-)$ and (b) B–Z–B high symmetry lines. Theoretically predicted band structures are superimposed as solid lines [8].

such as transport measurements [5, 7] and theoretical calculations supporting the realization of 2D electrides, these results can at best be considered as indirect probes of electronic structures. In order to credibly prove that a material is indeed a 2D electride (in which anionic electrons are confined two-dimensionally in the interlayer space), it is necessary to directly probe its electronic structure. As shown in Fig. 1, the existence of mobile anionic electrons in the interlayer space has a one-to-one correspondence with the formation of 2D electride bands near E_F as per band-structure calculations. That is, the direct observation of 2D electride bands near E_F would be direct proof of the realization of 2D electride states. In this study, we directly observed the band structures of Y_2C by using angle-resolved photoemission spectroscopy (ARPES) in order to verify the realization of the 2D electride in Y_2C [8].

Figure 2 shows the band dispersions determined by ARPES along the high symmetry directions, for investigating the electronic structures and their correspondence with the theoretical calculations of Y_2C in detail. It is clear that electride bands exist near E_F, leading to the formation of a small electron and hole pockets around the F and B points, respectively. According to the calculations, the dispersive bands in the energy range from near-E_F to around 1 eV (red and blue lines in Fig. 2) are mainly derived from the anionic electrons in the interlayer space. The overall band structures observed by ARPES are in good agreement with theoretically predicted band structures. Consequently, these results indicate that Y_2C is most evidently a 2D electride, which has electride bands derived from anionic electrons confined in the interlayer space.

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