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# ARPES Study of X-Point Band Overlaps in $LaB_6$ and $SmB_6$ – Contrast to $SrB_6$ and $EuB_6$

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**Abstract**

In contrast to our recent finding of an X-point band gap in divalent hexaborides, we report here that angle resolved photoemission spectroscopy (ARPES) data shows that the gap is *absent* for trivalent $LaB_6$ and is absent or nearly so for mixed valent $SmB_6$. This finding demonstrates a nontrivial evolution of the band structure from divalent to trivalent hexaborides.

*Key words:* Angle resolved photoemission, Hexaboride, Energy gap

Rare earth hexaborides continue to challenge condensed matter physicists to explain their exotic properties, e.g., the weak high temperature ferromagnetism in lightly doped or nominally stoichiometric divalent hexaborides [1]. An important issue is the existence or the absence of band overlap at the X point of the cubic Brillouin zone appropriate for these materials. Such an overlap is predicted by band calculations and would render divalent hexaborides to be semi-metallic conductors, whereas they would be insulators if there is an X-point gap. Recent angle resolved photoemission spectroscopy (ARPES)

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data reported by our group [2] show a band gap of $\approx 0.8$ eV for the divalent hexaborides $SrB_6$ and $EuB_6$. We have since found the same result for $CaB_6$ and $Ca_{1+\delta}B_6$. We initially [2] interpreted the gap as characteristic only of the surface region probed by ARPES. Strong motivation to re-interpret the gap as characteristic of the bulk was provided by a band calculation [3] which includes a GW self energy and finds for $CaB_6$ an X-point gap of the size we observed. By complementary data from our bulk sensitive x-ray absorption and emission spectroscopy (XAS and XES) [4], we have now shown conclusively that a bulk X-point band gap does indeed exist in these divalent hexaborides. In this paper, we show that this gap is absent in trivalent $LaB_6$ and either absent or nearly so in mixed valent $SmB_6$. This finding demonstrates that the band structure of the hexaborides undergoes a non-trivial change as the valency of the metal atom changes from divalent to trivalent.

Figures 1 and 2 show grey scale intensity images of energy vs. crystal momentum from angle resolved photoemission spectroscopy (ARPES) data obtained at the Ames/Montana beam line, equipped with a 50mm radius VSW spectrometer, of the Synchrotron Radiation Center (SRC) at the University of Wisconsin. The measurements were done on cleaved single crystal surfaces at a temperature of 20 K and in a vacuum of $\approx 4.0\times10^{-11}$ Torr, using 22 eV photons. Shown in panel B of Fig. 1 is a band calculation for $LaB_6$ [5]. The data are presented as a function of the momentum value projected onto the Γ-X line of the Brillouin zone. Strictly speaking, the k paths corresponding to the data are arcs in the Γ-X-M plane, not necessarily close to the Γ-X line. However, extensive data sets show that these data are nonetheless representative of the states along the Γ-X line. We tentatively attribute this to a partial loss of perpendicular k-conservation due to the photo-electron lifetime [6].

The data for $LaB_6$ bear a strong resemblance to the theory of panel B. We show the comparison in panel A. The band $\alpha$ of La 5d character agrees excellently with the data, and the band labeled ($\beta'$) is the theoretical $\beta$ band of B 2p character shifted by 0.38 eV. The theoretical X point band overlap is 0.54 eV, and therefore we estimate the experimental band overlap to be 0.16 eV.

In the $SmB_6$ data of Fig. 2, both the $\alpha$-like band and the $\beta$-like band are visible, the latter more strongly than the former. Since the $\alpha$-like band is very weak in the raw data (A), we enhance its visibility in panel B by dividing the raw data by 11x11-point smoothed data. Relative to $LaB_6$, the Fermi energy lies nearer to the bottom of the $\alpha$ conduction band, as expected from the reduced valency of the Sm ion compared to the La ion. Another difference of the $SmB_6$ data from the $LaB_6$ data is the presence of essentially k-independent Sm 5f emission, faintly visible near $E_F$ in the data of Fig. 2 and clearly observed in data taken with higher photon energy [7]. In Fig. 2, we show as lines experimental band dispersions obtained by following the maxima of the image. The position of the $\alpha$-like band is especially uncertain near the X point, due to the weak emission

strength. Our best estimate is that the two bands barely touch at the X point, but a gap on the order of 0.1 eV is also possible.

Our findings here also agree with XES and XAS data [4] and show that there are non-trivial changes in the band structure as divalent hexaborides evolve towards trivalent hexaborides. Additionally, an anomalous deviation from Vegard's law exists in the $Ca_{1-x}La_xB_6$ series. Rather than the monotonic increase in lattice constant expected from the larger $La^{3+}$ size, the lattice parameter at first shrinks to a minimum at ≈10% La doping before increasing again with higher La-doping [8]. As electrons are forced into the conduction band by the alloying, the purely single particle contribution to the ground state energy is reduced if the material can decrease its conduction band energy by contracting, up to the point where the gap is reduced to zero and overlap begins. It is interesting to speculate that the critical La concentration in this system corresponds to the band crossover. We note a tendency in both $LaB_6$ and $SmB_6$ for the overlap to "stick" near zero as the chemical potential continues to rise in the conduction band.

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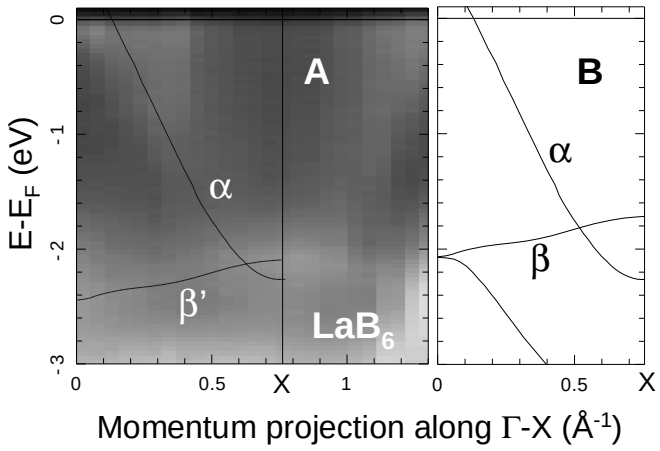


Fig. 1. ARPES data for $LaB_6$ (A) and theoretical band calculation for $LaB_6$ (B) [5]. For details, see text.

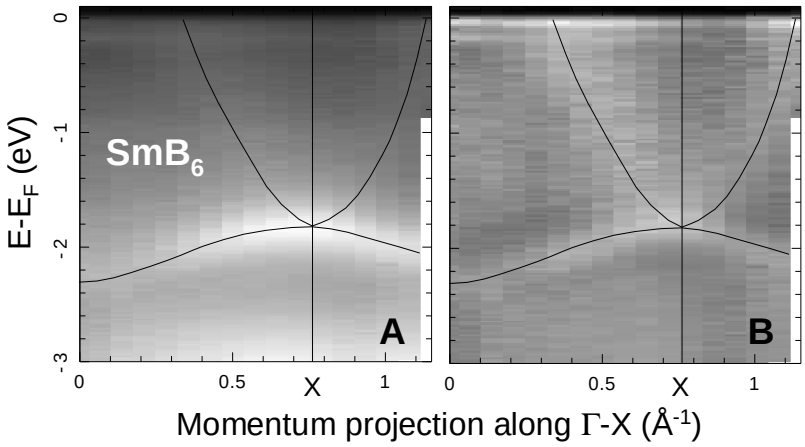


Fig. 2. ARPES data for $SmB_6$. For details, see text.