Iridium oxides are generating increasing attention [1–13]. In these materials, the entanglement of the spin and orbital degrees of freedom induces a reorganization of the electronic energy levels, described by an effective total angular momentum $J_{\text{eff}}$. This is different from the case of $3d$ and $4d$ oxides, governed by Coulomb interactions and, respectively, Hund’s coupling [14]. The interaction between such composite moments strongly depends on the symmetry of the Ir-O covalent bonds [10,15]. In spite of such differences, iridates built from corner sharing IrO$_6$ octahedra, such as Sr$_2$IrO$_4$, show surprising analogies with the isostructural cuprate compounds, namely, a single band at the Fermi level [2] and a similar magnetic structure [1,4,6]. This motivates a search for connections between iridates and cuprates and, quite naturally, extends the discussion from the cuprates to the iridates [16,17].

Experimental efforts have been so far oriented to the stoichiometric “parent compounds”, and to the role of structural distortions and of dimensionality in determining both the electronic and the magnetic properties. Insight into the former is gained by substituting Sr with, e.g., Ba, which enhances the tetragonal distortion and prevents the rotation of the IrO$_6$ octahedra [16]. The effect of dimensionality can be assessed by comparing the compounds of the series from $n=1$, the two-dimensional (2D) limit, to $n \to \infty$, the three-dimensional (3D) one. Early optical data showed an evolution from a Mott insulator (Sr$_2$IrO$_4$) to a correlated metal (SrIrO$_3$), which was attributed to interlayer coupling and a consequent increase of the bandwidth W [3]. A similar evolution is observed in cuprates where, e.g., La$_2$CuO$_4$ is a charge transfer insulator [17], whereas LaCuO$_3$ is a poor metal [18,19].

In between these two extremes, Sr$_3$Ir$_2$O$_7$ is notable, as a small electronic gap seems to indicate that it lies very close to the critical point marking the metal-to-insulator transition. Its band structure has been mapped for the first time with laser-based angle-resolved photoemission spectroscopy (ARPES) [20], and refined more recently [21,22]. Reference [21] suggests a polaronic ground state with a vanishing quasiparticle pole close to the Fermi level, another possible similarity to the cuprates [23]. Alternatively, Ref. [22] ascribes the reduced gap to the intercell coupling, which would cause a sizable dispersion in the direction perpendicular to the IrO layers and mark a departure from the 2D physics already for $n=2$. Our results suggest a different scenario. We measure a bilayer splitting between bonding and antibonding bands as large as 0.2 eV, which indicates that the interlayer coupling cannot be treated simply as a perturbative term to the electronic structure of the monolayer counterpart. From the intensity modulation of these states with photon energy, we infer the reflection symmetry properties of the corresponding wave functions. We find that the lowest energy state is antisymmetric, opposite to the much studied case of the bilayer cuprates.

Single crystals of Sr$_3$Ir$_2$O$_7$ were grown using the self-flux technique, as described elsewhere [24], and characterized by magnetization measurements, x-ray diffraction, x-ray resonant magnetic scattering, and resonant inelastic scattering. All the ARPES data were measured at beamline 7.0.1 of the Advanced Light Source with a Scienta analyzer, and the momentum and energy resolution were 0.1° and 30 meV at $h\nu = 100$ eV, respectively. The light was linearly polarized in the horizontal plane containing the $k_z$ axis of the sample and the analyzer slits ($\rho$ scattering geometry), and the angular scans were obtained by rotating the sample about $k_x$, in steps of $k_y$. The measurements were performed at $T = 30$ K and no charging effects were observed at this temperature.

Figure 1 shows an overview of the band topology of Sr$_3$Ir$_2$O$_7$. The crystal structure has recently been refined from tetragonal to orthorhombic [25] but, as common practice in these layered iridates, we discuss the data with reference to a square surface Brillouin zone (BZ), as shown in Fig. 1(a). As we will show later, this choice is certainly justified for...
similarities with that of Sr$_2$IrO$_4$, where the states are assigned scales. The planes were small with respect to the other relevant energy.

The intensity in the first eV below the Fermi level is due to the folding potential generated by the structural distortion. The dashed curves outline the dispersion along the same directions for the monolayer compound Sr$_2$IrO$_4$, as reported in Ref. [2]. The relevant dispersion of the J$_{\text{eff}}$ character remains accurate near the M$_1$ and M$_3$ points, respectively. The lattice constant in the direction perpendicular to the Ir-O layers, c, in Fig. 2(a), is $\sim$20.9 Å. For a tetragonal unit cell, the expected $k_z$ periodicity is $4\pi/c \simeq 0.6$ Å$^{-1}$. The range shown in Figs. 2(b) and 2(c) covers almost three BZs, with no detectable dispersion comparable with an 80 meV shift of the leading edge at the M point, reported in Ref. [22].

A possible explanation for these seemingly contradictory results is offered by the analysis of Fig. 3(a), which shows as a color plot the $k_z$ dependence of the ARPES signal at the M point as in Fig. 2(b), but this time without any normalization, except for that by the photon flux. The spectral weight of the bonding and antibonding bands in a bilayer system is expected to have a markedly different, and in fact nearly opposite, dependence on the excitation energy, due to the opposite reflection symmetry of the states with respect to the intermediate plane between the two layers. We use a simple model, accounting only for the phase difference between the wave functions and considering a free electron final state. For a multilayer system of transverse periodicity d, the $k_z$ dependence of the matrix elements essentially follows a sine wave of period $2\pi/d$, dampened with increasing energy [27]. This behavior has been observed for surface states [28], quantum well states [29], and, more recently, for multilayer graphene [30]. The observed intensity modulation of Fig. 3(a) indeed shows good agreement with the

$$\Gamma \overline{M} = 0.78 \text{ Å}^{-1} \quad \text{and} \quad \Gamma \overline{X} = 0.55 \text{ Å}^{-1}.$$
predictions of this simple approach. In Sr$_3$Ir$_2$O$_7$ $d \simeq c/5$, and a sine wave of period $k = 2\pi/(c/5) = 10\pi/c$ (dashed curve) convincingly tracks the transfer of spectral weight between the two branches of the bilayer dispersion.

By choosing the photon energy so that either one or the other of the states are suppressed, the bonding and antibonding bands can almost be mapped separately. Figures 3(c)–3(e) illustrate the dispersion along $\Gamma M \Gamma$ at $k_y = \pi/a$ [dashed lines in panels (f)–(h)], measured for the three photon energies corresponding to the arrows in Fig. 3(a). At the $\Gamma$ point, the three panels show for the $J_{\text{eff}} = 3/2$ states mainly the bonding band (c), both bands with comparable intensity (d), or only the antibonding band (e). The spectrum measured at $\Gamma$ and $h\nu = 96$ eV, shown in panel (b), clearly shows two peaks, separated by 0.18 eV, corresponding to the bonding and antibonding bands. The distinction is less clear for the $J_{\text{eff}} = 1/2$ bands close to $M$, but signatures of the splitting are visible in panels (f)–(h), which present constant energy cuts at $E = -90$ meV, at the top of the valence band. The corresponding intensity patterns are different, as they map in fact different states. Namely, the rectangular contours at the $M$ points are elongated along different directions in (f) and (h), and are nearly square in (g). Notice that the transfer of spectral weight between the two bands could be misinterpreted as an energy dispersion, which may be the origin of the report of transverse dispersion of Ref. [22].

A further analysis of the intensity modulation of Fig. 3(a) reveals an interesting difference with the related case of the bilayer cuprates. The intensity of the bonding and antibonding bands evolves exactly in antiphase with respect to the case of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212) [27,31]. This is the result of a crucial difference in the structure of the two compounds along the $c$ axis. Namely, a bridging oxygen is present within the bilayer between two Ir ions in Sr$_3$Ir$_2$O$_7$, but not in Bi-2212. As a consequence, the wave function of the (bonding) lowest energy state is symmetric in Bi-2212, but antisymmetric in Sr$_3$Ir$_2$O$_7$, as shown in the cartoon of Fig. 3(i).

In pictorial terms, the oxygen-mediated bond between Ir atoms favors a configuration with an increased $5d$ charge density outside of the bilayer. Since $V_0$ is evaluated by matching the smooth variation of the photoemission intensity with the expected matrix element modulation, it has a considerable uncertainty which we estimate to be $\sim 5$ eV. However, this does not translate into any ambiguity in the assignment of the phase of the wave functions. At these photon energies, an unphysical value of $V_0 > 30$ eV would be needed to translate the data by half a period of the sine wave of Fig. 3(a).

We notice in passing an interesting link between these conclusions and the band structure of SrIrO$_3$, the $n \to \infty$ limit (3D) compound of the series, which remains unavailable. Sr$_3$Ir$_2$O$_7$ provides indeed an $n = 2$ sampling of the band structure of SrIrO$_3$. Given the cubic or nearly cubic structure of SrIrO$_3$ the bands are expected to show a similar, holelike, dispersion along the $c$ axis, as they do in the plane. Therefore, the top of the $J_{\text{eff}} = 3/2$ manifold is expected at the $\Gamma$ point, where the (totally symmetric) wave function has the same phase on adjacent layers, and the bottom at $Z$, where the phase is reversed on each adjacent layer. This is consistent with the previous considerations about the relative energy of the symmetric and antisymmetric states in Sr$_3$Ir$_2$O$_7$.

Figure 4(a) presents the measured band dispersion in the second BZ, where the $J_{\text{eff}} = 3/2$ intensity is higher, for
and, with respect to our previous work on Ba$_2$IrO$_4$ [33], it is known to accurately yield the peak positions [32]. The latter includes additional interlayer hopping terms, which produce a $k$-dependent splitting of all bands. The parameters have been slightly adjusted in order to suppress the $k$ offset between the $J_{\text{eff}} = 1/2$ band maximum and the $M$ point [26], consistent with the experimental data. More details are available in Ref. [34]. The projected weights of each band on a fourfold bilayer basis set [symmetric (S), antisymmetric (AS); $J_{\text{eff}} = 1/2, J_{\text{eff}} = 3/2$] is encoded in the color scale. For simplicity, we do not consider the staggered tilting of the octahedra and neglect electronic correlations, which are essential to capture hybridization and avoided crossings, and therefore to reproduce the observed energy gap. As a consequence, the results, namely, for the $J_{\text{eff}} = 1/2$ manifold, should be considered with caution in the vicinity of the Fermi level. Nevertheless the agreement is excellent further away from $E_F$.

A comparison of the experimental peak positions with the calculated bands in Fig. 4(b) confirms that the bilayer splitting of the $J_{\text{eff}} = 3/2$ states is as large as $\sim 0.2$ eV. As a term of comparison, the separation between bonding and antibonding states is only $\sim 0.09$ eV in Bi-2212 [27,31]. There, the bilayer splitting is directly related to a relevant interlayer hopping term $t_{\perp}$, and the energy splitting $\Delta E$ is simply $2t_{\perp}$, at least in a TB approach [35]. In Sr$_3$Ir$_2$O$_7$ the $t_{2g}$ states are not orbitally ordered, so that the measured ARPES intensity does not exhibit the dramatic matrix element suppression characteristic of the cuprates. On the other hand, the presence of two interacting states and of comparable energy scales makes the link to the perpendicular hopping term less straightforward [26,36]. What is more important, in Sr$_3$Ir$_2$O$_7$ the interlayer hopping is comparable to all other relevant energy scales, i.e., the in-plane hopping terms [26] and the magnetic exchange terms [5]. Therefore, it cannot be treated as a simple perturbative term to the electronic structure of the monolayer counterpart Sr$_2$IrO$_4$.

In summary, a thorough ARPES $k$-space survey of the electronic structure of the bilayer iridate Sr$_3$Ir$_2$O$_7$ reveals a very large splitting of the bonding and antibonding states of mainly $J_{\text{eff}} = 3/2$ character. Moreover, the $k_z$-dependent transfer of spectral weight between these two bands shows that the bonding state has antisymmetric character, opposite to the much studied case of Bi-2212, a typical bilayer cuprate.

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