Photoelectron Interference Pattern (PEIP): A Two-particle Bragg-reflection Demonstration

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Number of Shifts:	21	
Samples Measured:	Si(9 9 11):In(4×1) Si(1 1 1):In(4×1) Cu_2O (111) Nd-LSCO	

INTRODUCTION

As far as we are aware, the coherence properties of synchrotron radiation from a third generation synchrotron light source have not previously been used to investigate the possibility of photoelectron interference effects in contrast to photoelectron diffraction effects. An interesting question in this context is whether the coherence of the exiting light is enough to create coherent photoelectron emission from different parts of a sample or if the initial state also needs to have spatial coherence. In the latter case one might try to determine to which extent this coherence exists in different systems and how long the coherence length needs to be in order to observe an interference pattern. The proposed experiment was aimed at determining whether photoelectron interference could be observed and under which conditions. If successful the plan was to use the effect to investigate magnetic systems and systems that show signs of phase separation. Materials that are of strong current interest and show signs of phase separation on the nano-scale are the cuprates and nickelates [1-6]. A further possibility would have been to study magnetic ordering by looking at interference patterns in the dichroic on resonance emission.

EXPERIMENTAL METHOD

Our primary goal was to use the angle resolved photoelectron spectroscopy setup of BL25SU to measure to angular and intensity distribution of valence band emission from an ordered sample. Our analysis of data from the recent test-experiments performed at BL25SU (Proposal No.: 2006A1001, Application No.: 1262) showed promising intensity modulations in the angular patterns from a ZnO(0001) sample. Any conclusions about interference effects in the emission were, however, hindered by possible photoelectron diffraction effects. In order to be able to separate any interference effect from photoelectron diffraction we decided to use an overlayer system with a symmetry different from that of the bulk. The In on Si(111) system, both nominal and off cut, were chosen as to provide a system of relative simplicity in preparation and with a substantial valence band emission intensity separable from the bulk Si contribution. Using an off cut system (9 9 11), it was further hoped that the surface steps would provide a natural condition for In 1D wire formation. The In (4x1) reconstruction on Si(111) as well as any 1D wires was thought to provide an ideal testing ground for interference effects well separable from any photoelectron diffraction effects coming from the Si substrate. The Si substrate was cleaned by rapid annealing to 1000^{o}C with the use of specially designed tungsten and tantalum sample holders. The surface quality was then checked by LEED and when a sufficiently high quality pattern was observed, In was deposited on the surface and annealed to provide an ordered overlayer structure. Once an overlayer structure of sufficient quality was obtained angle resolved valence band spectra were collected over a large horizontal and vertical angular range in order to construct the angularly distributed interference pattern.

PEIP RESULTS

With the use of the specially designed sample holders the annealing of preoxidized Si(111) and Si(9 9 11) samples could be carried out rapidly and at low enough pressure. As a result very nice (7x7) reconstructions on both surfaces were obtained as shown by **Fig.1(a)** where a LEED pattern of the clean silicon surface is shown. On top of the clean surface, 1/3 of a monolayer of In was deposited followed by annealing and resulting in a (4x1) reconstruction as shown by the LEED pattern of **Fig.1(b)**. On the other hand, no sign of wire formation was observed on the stepped surface despite continuous efforts.

Once a well ordered (4x1) pattern was obtained, a series of angle resolved photoelectron valence band spectra were then acquired from the samples (see **Fig.1(c)** for an example) at hν = 551 eV. As seen from **Fig.1(c)**, there is clear valence band dispersion in the upper part related to the Si bulk bands. The intensity in the high binding energy part of the valence band is, on the other hand, dominated by emission from the In 4d levels. By integrating over different parts of the VB it is then possible to construct 2D intensity maps corresponding to either the Si part (Ebin=0..3 eV) of the VB or the In-4d part (Ebin=4…5 eV). The results are shown below in **Fig.2(a-d)**. As can be clearly seen from the figures, integration over the Si related part of the valence band results in a clear pattern which can be related to photoelectron diffraction effects in the bulk. Looking at the In-related intensity maps does, however, not display any clear pattern despite the ordered nature of the overlayer.

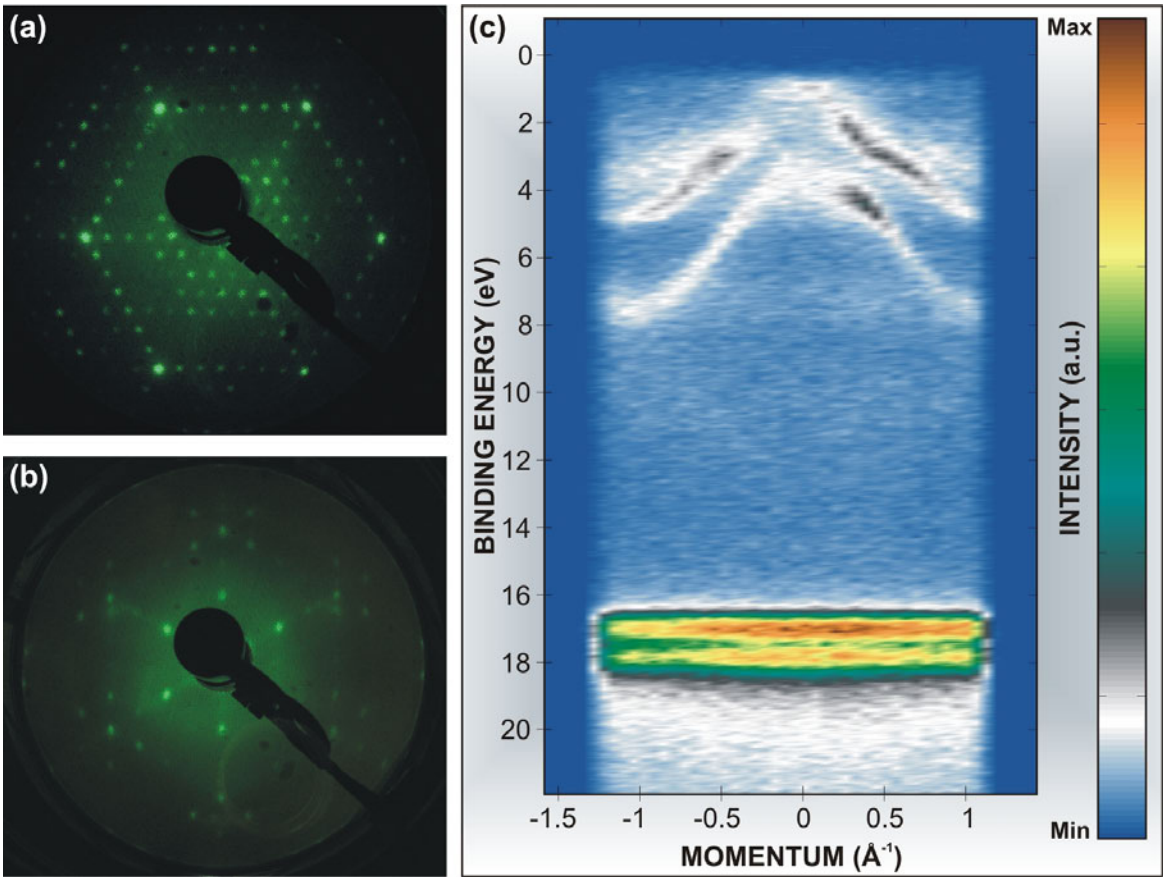
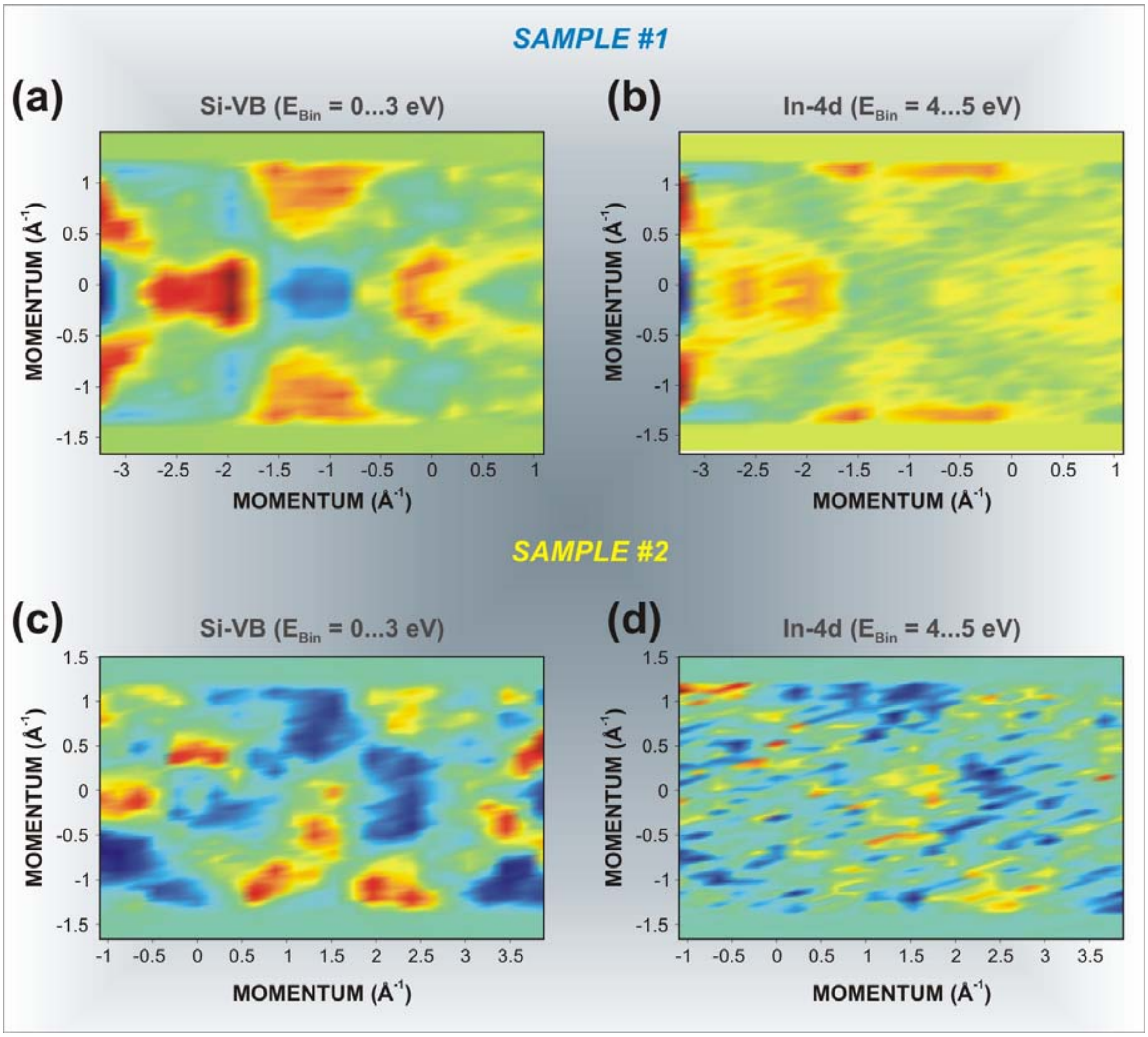


Fig.1: **(a)** *LEED-pattern from the clean Si(9 9 11)-(7×7) reconstructed surface,* **(b)** *LEED-pattern from the Si(9 9 11)***:***In(4×1) surface,* **(c)** *ARPES valence-band spectrum from the surface acquired at* $h\nu = 551$ *eV*

The lack of discernible patterns in the Indium emission obviously leads to the conclusion that no interference effects are present. Several explanations for this are possible. First of all, the degree of coherence in the X-rays is limited by the nature of the source and might be too low to observe any effect. Secondly, it is quite possible that the nature of the excited photohole is such that no interference effect occurs. This might be due to localization effects, short coherence length etc. In view of these discouraging results a decision to interrupt the measurements on the In-Si system was taken and instead the remaining beam time was used to perform more conventional ARPES measurements on the two oxide systems Nd-LSCO and Cu_2O. Results from these measurements are briefly described below.



<u>Fig.2</u>: **(a)** *Intensity-map when integrating over the Si-VB (E_{Bin} = 0…3 eV) for Sample #1*
(b) *Intensity-map when integrating over the In-4d (E_{Bin} = 4…5 eV) for Sample #1*
(c) *Intensity-map when integrating over the Si-VB (E_{Bin} = 0…3 eV) for Sample #2*
(d) *Intensity-map when integrating over the In-4d (E_{Bin} = 4…5 eV) for Sample #2*

ANGLE-RESOLVED SPECTRA FROM $Cu_2O(111)$

Cu_2O is a semiconductor with a highly symmetrical simple-cubic crystal structure. It has gained considerable interest in theoretical as well as experimental works during several decades, and many efforts have been made to understand its electronic structure. However, until now there have been very few *angle-resolved* photoemission studies of this compound, as far as we know. During the course of our experiment we have acquired angle-resolved photoemission spectra from a Cu_2O single crystal. The crystal was cleaned by a very gentle sputter-/anneal-cycle and the quality as well as the surface orientation was checked by LEED (**Fig.3**). The surface was oriented along the (111)-direction and ARPES spectra were acquired at two different photon energies hν = 619 eV and hν = 891 and a temperature of 100 K (**Fig.4(a-c)**)

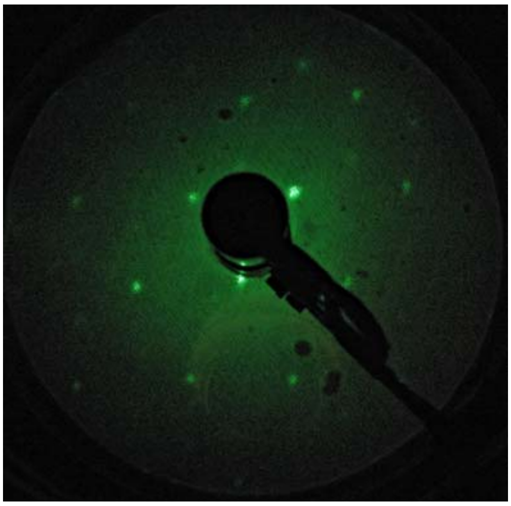


Fig.3: *LEED-pattern from the clean $Cu_2O(111)$ surface*

Since Cu_2O is a fully three-dimensional compound, one has to resort to the assumption of a free-electron-like final state in order to extract the k-resolved band structure from angle-resolved photoemission spectra acquired at *low* photon energies (hν < ~100 eV). This approach was used in a recent paper presenting ARPES data from Cu_2O [11]. However, at the high photon energies used here (619 and 891 eV) this assumption is no longer necessary. By choosing the photon energy appropriately, one is able to precisely select the location in three-dimensional reciprocal space where the band dispersion will be probed. In this study the photon energy was chosen so as to reach two different instances of the Γ-point. The Cu_2O sample was oriented to enable measurement of the dispersion along the R– Γ–R direction in reciprocal space.

Fig.4 presents colormap plots of background-subtracted ARPES intensity as a function of binding energy and electron momentum, for two different photon energies. For comparison, results from LDA band structure calculations on Cu_2O from the WIEN2k software package [12] are displayed. The ARPES results agree reasonably well with the theoretical predictions over almost the entire energy-momentum region.

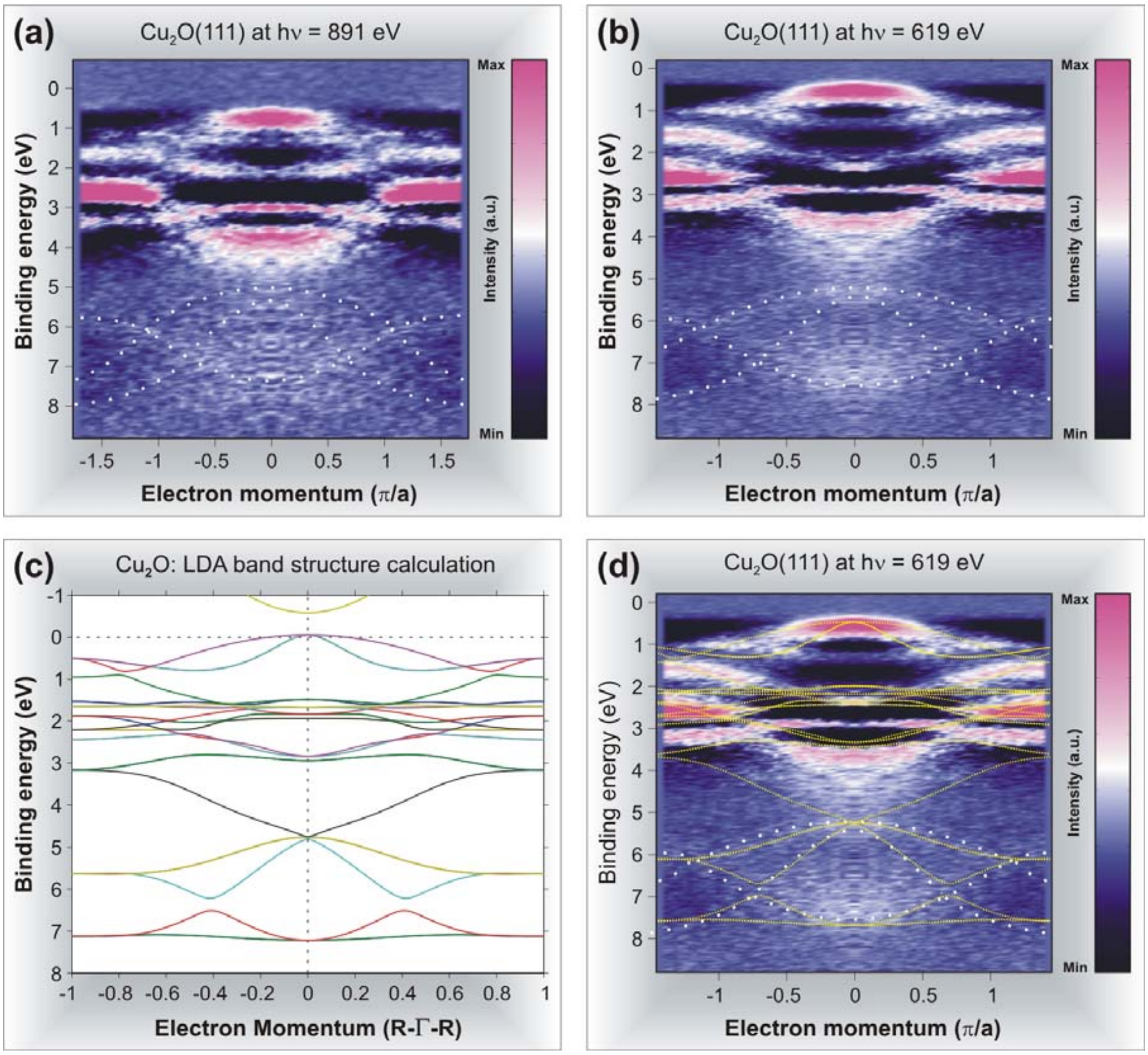


Fig.4: **(a)** *ARPES-data* (R-Γ-R) *from Cu_2O acquired at hν = 891 eV, T = 100 K*
(b) *ARPES-data* (R-Γ-R) *from Cu_2O acquired at hν = 619 eV, T = 100 K*
(c) *Cu_2O LDA band structure calculations*
(d) *LDA band structure calculations compared to ARPES data (hν = 619 eV)*

FERMI-SURFACE MAPPING OF Nd-LSCO

The experiment has been performed on a single crystal of Nd-$La_{1.86}Sr_{0.14}CuO_4$ grown by the TSFZ method. The sample was cleaved using the so-called “post-cleave method” and a nice surface was acquired. As an example of acquired data an ARPES spectrum is shown in **Fig.5(a)** below. One can clearly distinguish three Fermisurface (FS) crossings as indicated by the three dashed yellow lines. By using a photon energy of 500 eV one can cover a large window in k-space by just a single spectra as illustrated by **Fig.5(b)** where we show the slice in momentum space covered by the spectrum in **Fig.5(a)**. Here it is clearly seen how the slice cuts the FS at three different points. This is also clearly visible in the Momentum Distribution Curve (MDC) at the Fermilevel (see **Fig.5(d)**).

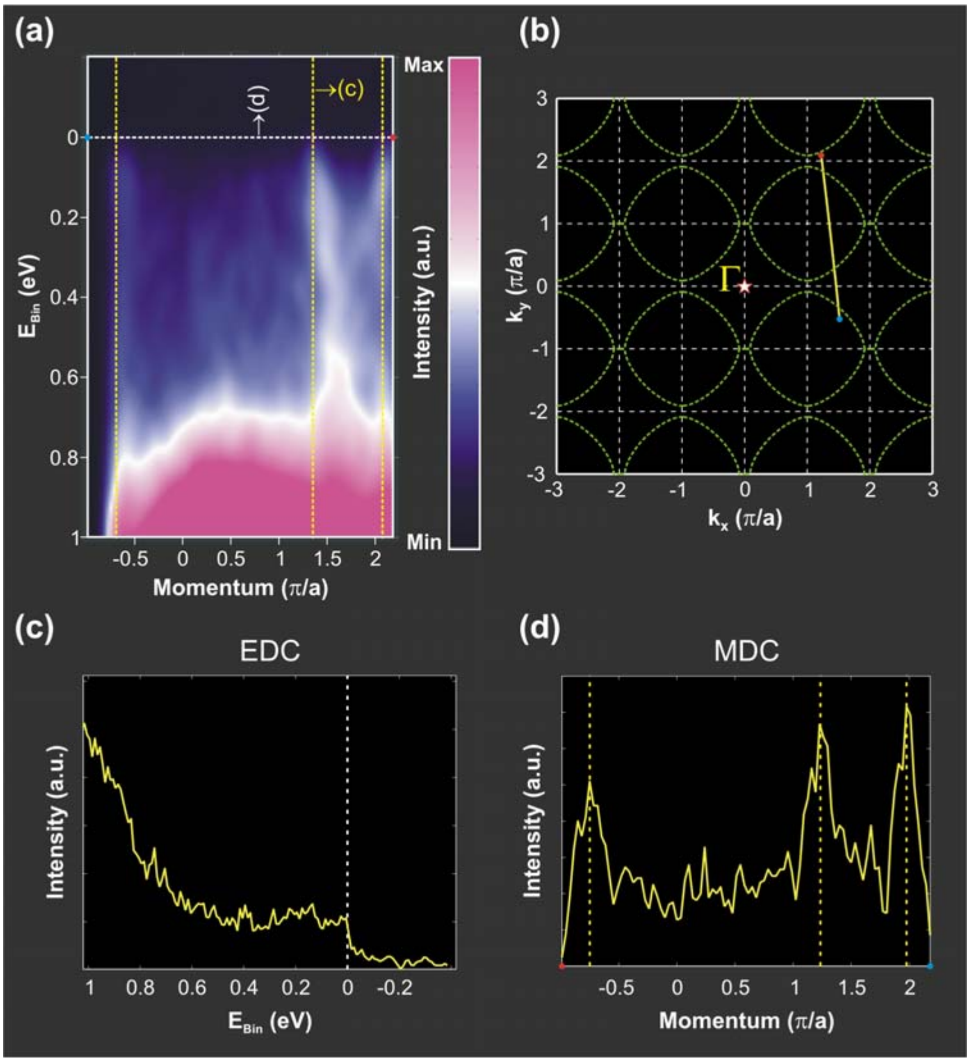


Fig.5: **(a)** *ARPES-data from Nd-LSCO acquired at hν =500 eV, T = 20 K*
(b) *Slice in momentum space covered by the spectrum in (a)*
(c) *EDC taken at one of the maximums of the MDC’s*
(d) *MDC at E_F*

During this experiment we have succeeded to map out the complete Fermi surface of this sample. We have collected spectra at points all along the Fermi surface as shown in **Fig.6(a)**. The points represent the FS-crossing attained from the MDC (see e.g. **Fig.5(d)** above). In **Fig.6(b)** the points from **Fig.6(a)** have been folded into the first zone and a nice overlap can be seen. In **Fig.6(c)** a FS intensity map obtained by energy integration of ARPES spectra over a 100 meV window (±50 meV) about the Fermi level. The intensity map reveals a slightly broader intensity distribution than expected but we believe that this has to do with a slight tilt of the sample surface causing the measured k-points not to overlap completely when folded into the first zone. This can though be routinely compensated for by further data analysis and the use of Laue XRD on the measured sample. In the same manner as previously done in [13], the aim for this data is to be compared to data collected at low photon energy.

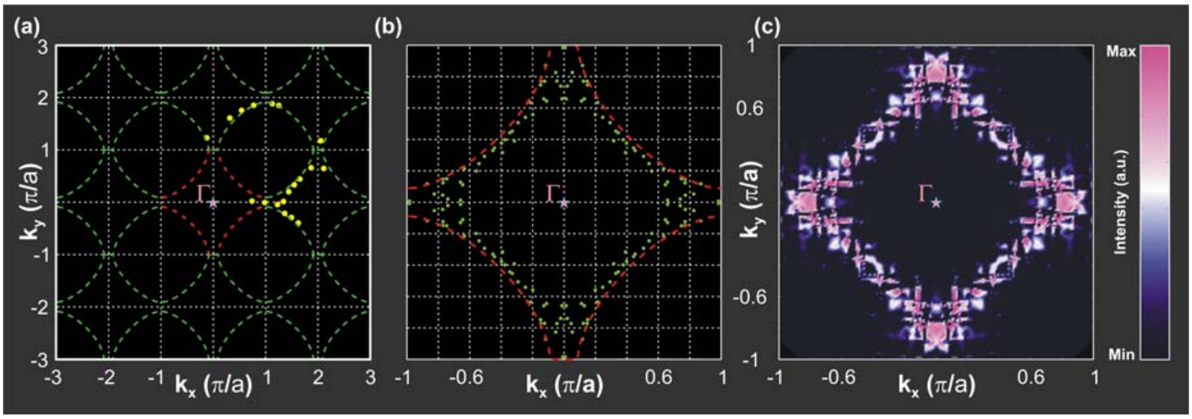


Fig.6: **(a)** *FS-crossing points acquired from MDC's at* E_F
(b) *Points from (a) folded into the first Brillouin-zone*
(c) *FS intensity map obtained by energy integration of ARPES spectra over a 100 meV window (±50 meV) about the Fermi level.*

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