Mapping Disorder-Order Induced Changes To The Fermi Surface Of Cu_3Au Using A New Toroidal Electron Energy Analyser

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Abstract

A second-generation toroidal spectrometer has been used to investigate the Fermi Surface topology of the binary alloy system Cu_3Au when prepared as an ordered or substitutionally disordered alloy. We demonstrate using two complementary Fermi surface mapping techniques and an empirical FS model for the disordered phase, that structures in the data arise from two different mechanisms. We observe distinct effects on the FS upon ordering, and compare the results with a recent bandstructure calculation.

Key words: Angle Resolved Photoemission, Fermi Surface Mapping, Binary Alloys,Wien2k, DFT

Introduction

The binary alloy Cu_3Au is a classical example of a system that undergoes a 1st order order-disorder phase transition. Below T_c = 663K, the structure is simple cubic (SC), resulting from a regular arrangement where Au occupies the corner sites and Cu the face sites of an fcc lattice; above T_c, the atoms are randomly distributed over the same sites, resulting in an fcc structure [1]. The Fermi Surface (FS) of the disordered phase is expected to be similar to that of Cu, with perturbations mainly to the neck region[2]. The changes to the electronic structure near E_F across the order-disorder transition are expected to be small[3], but due to the introduction of new Brillioun Zone boundaries band back folding will occur. Thus the expected FS of the ordered structure should possess a more intricate topology, presenting an ideal test case for FS measurements using a high resolution electron spectrometer.

Experimental

Fermi Surface measurements of the ordered and disordered phases of Cu_3Au were made by angle-resolved photoemission (ARPES) using synchrotron radiation provided at the storage ring BESSY2, Germany. Measurements were made at beamline TGM-4 using p-polarised light with the sample at an angle of 45˚ to the photon beam. The energy resolution of the experiment was determined by the FWHM of the beamline monochromator as approximately 200meV. Sample cleaning was achieved using Ar^+ bombardment (500eV) and annealing at 773K. The disordered phase was obtained via quenching from 773K to room temperature (RT), while the ordered phase was obtained over a 5 day temperature ramp following a method similar to that used by Morris [4].

To perform high resolution ARPES, we have developed a next generation toroidal electron spectrometer system that simultaneously detects both the photoelectron emission angle and its kinetic energy. All polar emission angles are detected simultaneously in a selected azimuthal plane, with an energy "window" covering ~0.8% of the selected analyser pass energy visible on a CCD/channelplate detection system. Fig. 1 shows an image obtained from Cu(111) illustrating a 0.8 eV energy region about the Fermi energy. The data clearly shows the well-known Cu(111) surface state at $\overline{\Gamma}$ and in adjacent Brillouin zones.

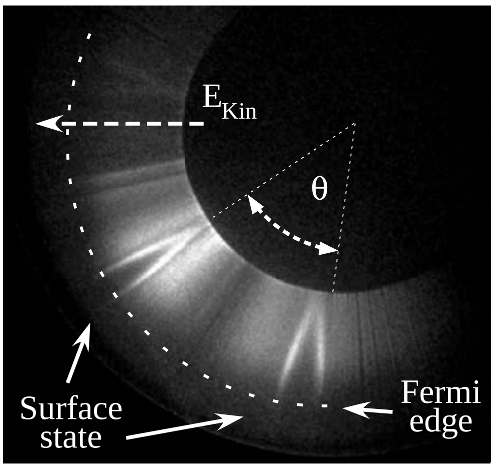

Fig. 1. Toroidal analyser output as imaged by the CCD detection system. Polar angle (θ) direction and kinetic energy (E_{kin}) directions are indicated

Two complementary techniques were used to map the Fermi Surface. The first, defined here as the Azimuthal Scan (AzScan) method, was used to map a wide area of the FS by measuring photoemission intensity at the Fermi energy (E_F) over the

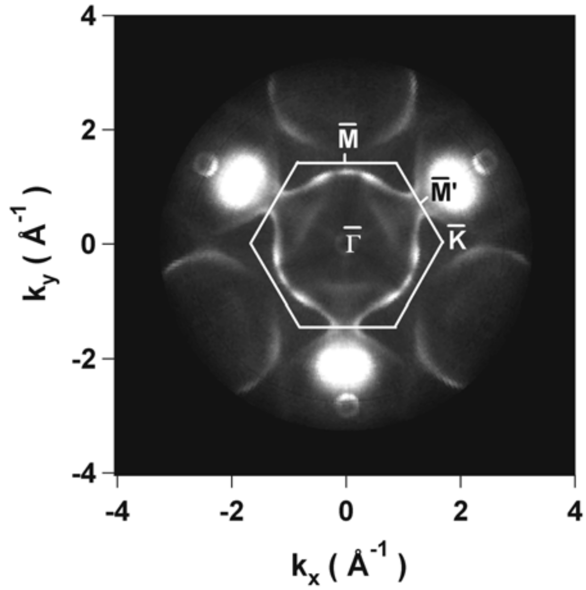

Fig 2. Cu(111) Az Scan. hν = 46eV. k_x, k_y denote surface k-components. Surface directions are shown.

entire emission hemisphere. The resulting I(θ,ϕ) map is converted into a momentum (k) space map resolved in the surface plane. An example taken from Cu(111) using hν = 46eV is shown in Fig 2. This is an established technique that has been used to map the Fermi surface of various materials such as Cu, Ni and Ag[5-7].

To investigate the FS in directions perpendicular to the surface, Angle Resolved Constant Initial State (ARCIS) measurements were made. In this mode, the initial state is chosen as the Fermi energy, and the sample is set at a high symmetry azimuth. The detection kinetic energy is varied synchronously with the photon energy to maintain sampling from E_F. As the photon energy is increased, the photoelectron final state explores the FS in the chosen plane. Using an appropriate model for the final state allows one to determine the absolute k values of observed transitions from the FS. In this study, a free electron final state (FEFS) has been

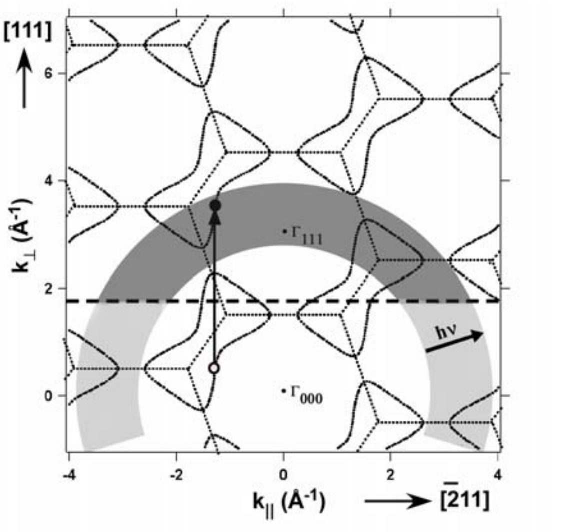

Fig. 3. Phenomenology of the ARCIS experiment. Direct transitions occur whenever the photoelectron final state (shown here as free electron like) intersects the FS. The annulus represents the area sampled by the final state as photon energy is swept over a given range. The dashed line indicates the "PE horizon", or the minimum $k_\perp$ required to overcome the surface potential barrier. The dark grey region thus indicates observable transitions.

employed. Fig. 3 illustrates how this is expected to result in a planar mapping of the FS for Cu(111)[$\bar{2}$11]

AzScans were performed using photon energies ranging from 36 to 64eV, while ARCIS (30-130eV, 0.5eV step size) was employed along the three high symmetry directions indicated in Fig. 2.

Results: Disordered $Cu_3Au(111)$

Fig. 4 shows three selected AzScan measurements taken from disordered $Cu_3Au(111)$ (hν= 46, 52 and 58eV). The white dots represent the expected direct transition "cut" through the FS, calculated using a FEFS (V_o=14eV) in conjunction with a Fourier Series expansion to dHVA data modified for Cu_3Au[8]. Clearly visible

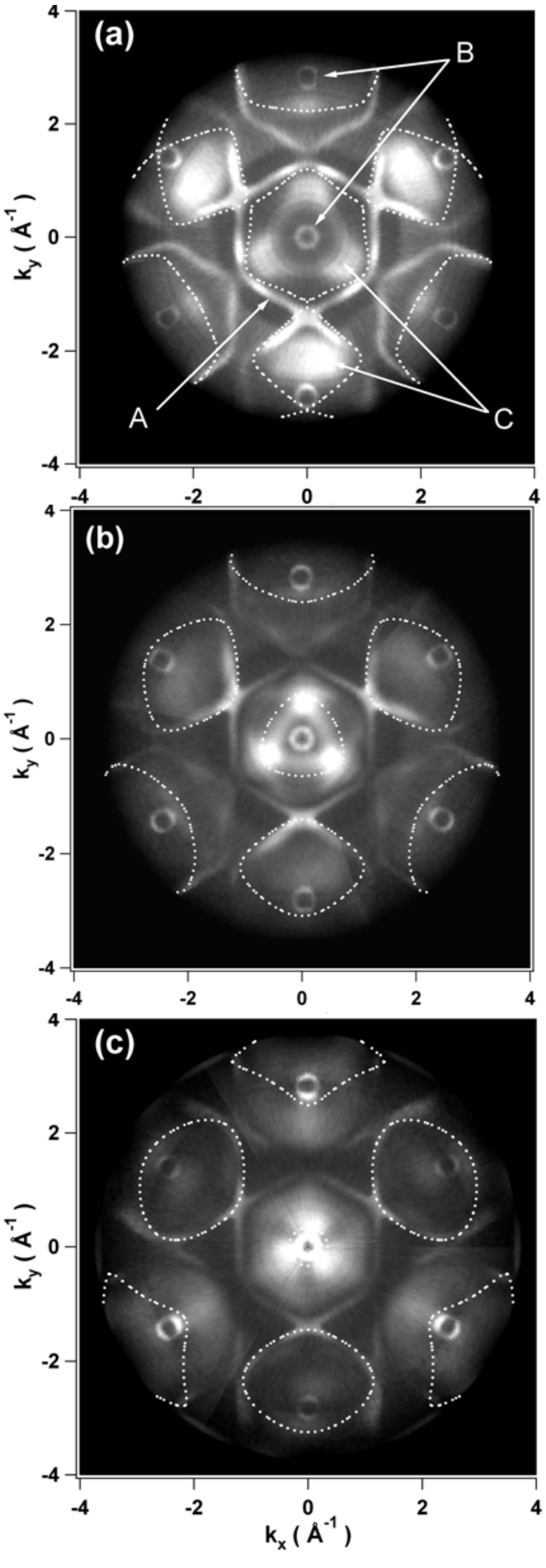

Fig 4. AzScan results for Disordered $Cu_3Au(111)$. (a) hν=46eV (b)hν = 52eV (c) hν = 58eV. White contours are described in the text.

in each scan is what appears to be the relatively sharp contour of a cut through the expected FS (Feature A). This is visible in the central Brillouin zone and its nearest neighbours.The small circles at each $\bar{\Gamma}$ point (Feature B) are due to the $Cu_3Au(111)$ surface state. Also visible are diffuse regions of intensity (Feature C) contained within the sharp contours. These structures alter in position with photon energy, as do the calculated contours, whilst Feature A remains constant in size. This strongly suggests that the diffuse features correspond to the direct transition from the FS, not the sharp contour observed.

Data taken in the ARCIS mode along the [$\bar{1}10$] and [$\bar{2}11$] surface directions (Fig. 5) helps to support this hypothesis. In this planar k-space representation, the surface state now appears as two closely spaced vertical lines just within the "neck" of the FS. The increased brightness of the surface state near the neck region has been reported previously for Cu[9]. The dHVA model of the FS (white dots) agrees quite well with the more diffuse contours in the data, even for low photon energies where the FEFS model is expected to fail. Importantly, it can be observed that the intensity from certain locations within Fig. 5 extends vertically well beyond the expected outline of the FS, implying a significant degree of $k_\perp$ broadening. Such features have been circled. Thus the sharp structures seen in Fig. 4 (Feature A) that do not alter in $k_{||}$ location with increasing photon energy can now be unambiguously associated with these dispersionless features. We have also observed this effect for Cu, although to a lesser extent[1]; this may be due to the generally broadened electronic structure of disordered alloy systems.

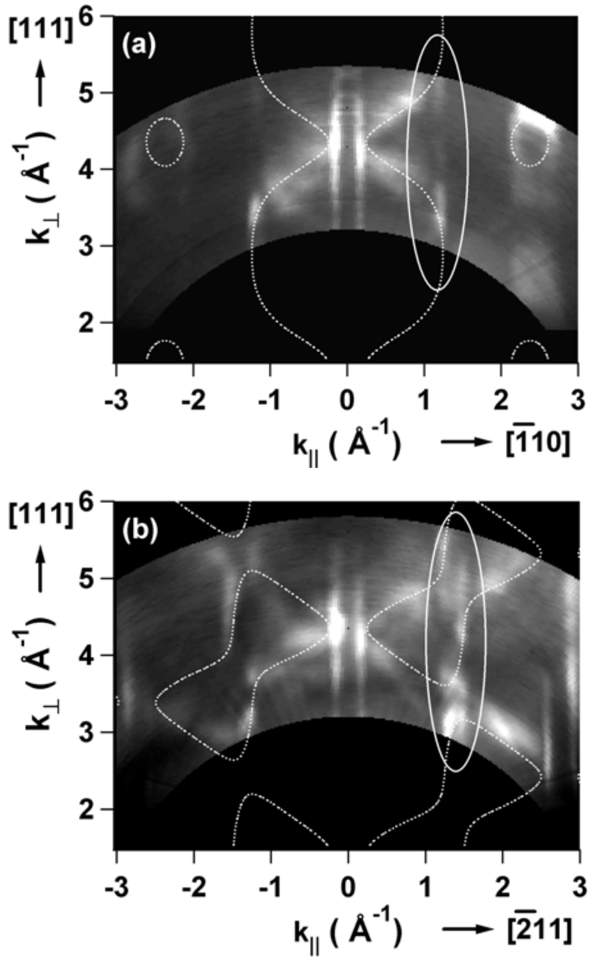

Fig. 5. ARCIS measurements of the disordered - phase FS. (a) $Cu_3Au(111)$- [$\bar{1}10$] (b) $Cu_3Au(111)$-[$\bar{2}11$] Circled features indicate significant areas of $k_\perp$- broadening.

These results are in contrast to a previous AzScan study of Cu(111)[10], which discussed the sharper features in terms of direct transitions. The results also help to explain features previously observed in an AzScan measurement on Ag(111). In that case, similarly diffuse 3-fold structures were observed, however they were assigned to final state diffraction effects, while some of the sharp contours were postulated to be umklapp processes[5].

[1] To be published

In summary, structure in the AzScans of disordered $Cu_3Au(111)$ that is common to all photon energies originates from transitions made possible by distinct $k_\perp$-broadening processes, whereas the more diffuse structures which vary in location with photon energy are the result of anticipated direct transitions from the FS.

Results: Ordered $Cu_3Au(111)$

Shown in Fig 6 are two selected AzScans taken with hν = 46 and 52eV. Data from the ordered and disordered samples (Fig 4) are very similar at first glance, although significant changes can be discerned, the most distinct being 3-fold "spikes" pointing toward the centre. As described earlier, the SC structure introduces new Brillouin Zone boundaries, which should lead to back folding of bands at the Fermi level. This should introduce additional structures commensurate with the original FS topology. A DFT calculation of the ordered phase bandstructure has been performed using the Wien2k package. We have overlaid the ordered data with the results of a FEFS intersection with this calculation. Due to the 3 fold symmetry, only one third is shown for clarity. The calculated contours indeed bear some similarity to the ordered FS. Significantly, the predicted 3-pointed star pattern pointing towards the centre of each BZ is well reproduced in the calculation. Other features contained in the data (eg: the presence of the diffuse features from the disordered FS) are not so evident in the theory and require further study.

It should be mentioned that the sample used was ordered in UHV for 5 days prior to measurement. This may be compared with the 1-year ordering used by Diemel and Higgins in their dHVA study[11]. It is therefore possible that our sample was not completely ordered. One may speculate

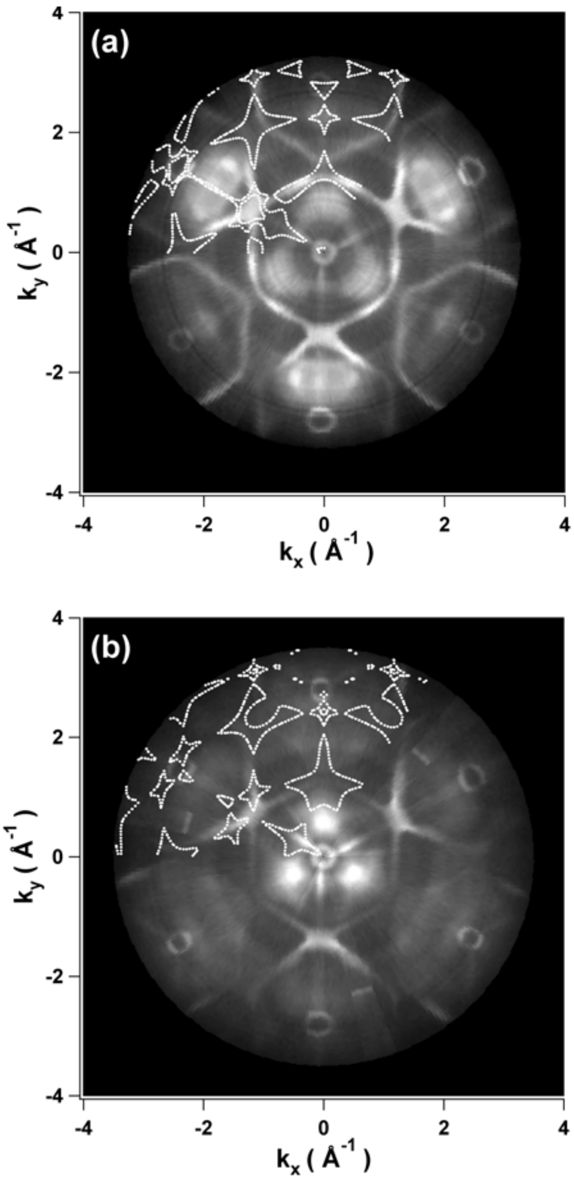

Fig. 6 AzScan results for Ordered $Cu_3Au[111]$. (a)hν=46eV (b) hν = 52eV White contours are described within the text.

that the possible existence of a surface layer ordered differently from the bulk may partially explain why the degree of $k_\perp$ -broadening is greater in Cu_3Au than is the case in Cu. For example, it has been postulated for $Cu_3Au(111)$ that the ordered solid is not simply bulk truncated, but possesses a surface disorder profile[12].

Conclusion

The FS of disordered and ordered Cu_3Au has been investigated using a combination of AzScan and ARCIS scans. Changes to the FS across the disorder-order transition have been observed and are accounted for well by a recent bandstructure calculation of the ordered phase. Structures observed in the disordered FS data have been assigned to large $k_\perp$-broadening effects, which can manifest themselves as somewhat misleading contours in AzScan experiments. With the aid of ARCIS, direct transition-type structures have been identified in the scans which agree well with the expected FS.

Acknowledgements

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