Highly polarized emission in spin resolved photoelectron spectroscopy of α-Fe(001)/GaAs(001)

J.G. Tobin(a), S.-W. Yu(a), S.A. Mortona, (b, c), G.D. Waddill(b), J.D.W. Thompson(d), J.R. Neal(d), M. Spangenberg(d) and T.H. Shen(d)

a Lawrence Livermore National Laboratory, Livermore, CA, USA

b Missouri University of Science and Technology, Rolla, MO, USA

c Lawrence Berkeley National Laboratory, Berkeley, CA, USA

d Joule Physics Laboratory, University of Salford, UK

Abstract

Highly spin-polarized sources of electrons, integrated into device design, remain of great interest to the spintronic and magneto-electronic device community. Here, the growth of Fe upon GaAs(001) has been studied with photoelectron spectroscopy (PES), including Spin Resolved PES. Despite evidence of atomic level disorder such as intermixing, an over-layer with the spectroscopic signature of α-Fe(001), with a bcc real space ordering, is obtained. The results will be discussed in light of the possibility of using such films as a spin-polarized source in device applications.

1. Introduction

For many years, the technological possibilities of spintronic or magneto-electronic devices [1], particularly when coupled with potentially pure spin sources such as half-metallic ferro-magnets, [2] have engendered great interest. Despite the limitations encountered in such potential sources [3], there is still ample reason to pursue such concepts. This is because, in part, even with sources that operate below 100% polarization, technologically important devices should emerge [1]. However, the challenges of device integration remain significant even for cases with lowered expectations, because often the physical realities of intermixing, disorder and alloying can creep into the attempts to fabricate structures based upon ideal conceptual designs. Issues related to surface and interface properties plague integration of half-metallics into device materials and consequently less exotic materials may be the preferred route to spin injection [3]. Within this context, ferromagnetic metal to semiconductor interfaces are potentially critically important for the future applications of spintronic devices. One possibility for a room temperature spin injector is Fe/GaAs. Fe/GaAs displays a wide range of magnetic behavior depending upon factors such as substrate preparation, termination, thickness and

temperature. Previously, our earlier work indicated that the Fe/GaAs also grows in a complex island domain structure: this subject will be discussed in more detail below [4], [5], [6] and [7]. Here, we report a surprising and potentially important result: the manifestation of order coming out of chemical chaos. Despite experimental proof of intermixing, alloying, floating surface layers and the seeming triumph of entropy, the Fe overlayers grown on GaAs exhibit very strong spin polarization, with the signature of a bcc α-Fe (001) single crystal [8]. The spectral features include the observation of strong spin polarizations in the spin resolved photoelectron spectroscopy (SRPES, Fig. 1) and photon energy dependencies in photoelectron spectroscopy (Fig. 2). These results are observed despite the overwhelming evidence of chemical mixing from the core-level photoelectron spectroscopy of the Ga 3d and As 3d levels (Fig. 3 and Refs. [4] and [6]). The generation of such highly polarized emission from Fe layers, grown directly on GaAs(001), may open the door to significant technological applications.

Fe/GaAs has been studied in detail with a variety of techniques. The complicated growth mode of Fe/GaAs(001) has been probed with Reflection High Energy Electron Diffraction (RHEED) and Scanning Tunneling Microscopy (STM) by Moosbuehler et al. [9] and Bensch et al. [10]. Aktas et al. [11] utilized RHEED and Ferromagnetic Resonance (FMR) to investigate magnetic anisotropies and surface reconstructions. The formation of a body-centered-cubic (bcc) Fe-based alloy at the Fe/GaAs(001) interface was probed by Schieffer et al. [12], using photoelectron spectroscopy, Photoelectron Diffraction and RHEED. The interface magnetization profile was determined via the X-ray Magnetic Circular Dichroism in X-ray Absorption Spectroscopy (XMCD-XAS) of marker impurities by Giovanni et al. [13]. Similarly utilizing XMCD-XAS, Claydon et al. [14] reported the observation of a bulk-like spin moment on the Fe atoms at the Fe/GaAs(001) interface. Thomas et al. [15], applying Grazing Incidence X-ray Diffraction to the Fe/GaAs(001) system, have described the interplay of anisotropic strain relaxation and uniaxial interface magnetic anisotropy. In a different approach, Zhu et al. [16] investigated the room temperature spin injection from Fe into GaAs. In general, these studies indicate that the Fe/GaAs system is a complicated interface with substantial island formation and chemical intermixing. Our earlier work, using high-resolution core-level photoelectron spectroscopy (Fig. 3 and Refs. [4], [5] and [6]) has given rise to similar conclusions: that while a bcc structure is growing on the GaAs(001) template, an immense amount of chemical intermixing is occurring. In summary of this previous result, Fig. 3 shows the interface development as determined in our earlier study on interface formation for Fe/GaAs and also in general agreement with most of the other studies of this interface cited herein (Refs. [9], [10], [11], [12], [13], [14] and [15]). At the interface there is significant intermixing of Fe, Ga, and As. We did not determine the precise stoichiometry of this layer, and it likely varies throughout the intermixed region. This region extends for not, vert, similar 7.5–9 Å, with the upper section composed of an Fe–Ga phase formed from the Ga that has segregated to this region. Above that is a relatively bulk-like Fe layer that still does, however, contain some intermixed As. This bulk-like Fe region occurs only for films thicker than not, vert, similar 10 Å. Finally, topmost, the film is covered by an Fe–As phase from As that segregates to the surface. Given this, it is clear that the “mostly” Fe layer is no more than 8–10 Å̊ in extent for our 20 Å̊ films. This amounts to, at most, 4–5 atomic layers, the very edge at which three dimensional effects might begin. What is surprising is that despite all the chemical

chaos, a high-degree of spin polarization is realized in the Fe layer immediately above the intermixed region (> 10 Å Fe deposition) with spin-polarized photoemission in remarkable agreement with that of single crystal Fe(001) (Fig. 3 and Ref. [8]).

2. Experimental details and sample preparation and characterization

The experiments were performed at Beamline 7.0 at the Advanced Light Source of Lawrence Berkeley National Laboratory in Berkeley, CA, USA, [17] using a spin resolving spectrometer described elsewhere [18]. The x-rays were linearly polarized. The Poynting vector of the x-ray, the linear polarization vector of the x-ray and the emission direction of the collected electrons were all in the horizontal yz plane as defined in Fig. 3 [18]. The angle between the incoming x-rays and the outgoing collected electrons was 55°. For the clean GaAs(001) and Fe/GaAs(001), the emission direction (− z in Fig. 3) was normal to the surface and along the [001] direction of the bulk GaAs(001). For the vicinal surface results, a 3 or 6° offset was used, which corresponds to (001) steps with 60 or 30 Å widths, respectively. The magnetization direction (+/−x in Fig. 3) was perpendicular to the horizontal plane and in the plane of the surface [18]. Spin integrated spectra were collected using a multi-channel detector at the exit plane of the hemisphere and spin resolved spectra were collected using the Mott detector, as described in detail in Ref. [18]. The quality of the spin resolved spectra in Fig. 1 is confirmed by a consideration of the polarization and instrumental asymmetry [19], [20] and [21]. The instrumental asymmetry is constant and almost 1.0, for all binding energy values below the Fermi Energy (EF). The polarization is also quite large, ranging from about − 20% below EF and about 40% at EF, with small error bars, as with the instrumental asymmetry [19], [20] and [21].

A synopsis of sample preparation, the earlier coverage study and supporting measurements is as follows. The substrates considered in this study were prepared at the III-V facility at The University of Sheffield and consisted of highly doped n-type GaAs epilayers grown upon singular n + GaAs (001) substrates and capped with amorphous As. The capped substrates were then relocated to the ALS, where clean GaAs (001) surfaces with a range of Ga:As ratios were prepared in situ by thermal desorption (‘decapping’) of the capping layer. Fe was then sequentially deposited by e-beam evaporation at a rate determined by a quartz crystal oscillator to be not, vert, similar 1 Å/min. The same procedure had been employed in earlier in-house experimental work, where clear patterns of low energy electron diffraction (LEED) were observed from both the decapped substrates and the Fe films subsequently deposited, suggesting well ordered surface structures. Between each deposition step of the growth study, the sample was transferred from the growth position to the analysis position for scanning; this process involved both vertical and angular translations and necessitated the use of an additional normalization step if core-level intensities for different Fe thicknesses were to be compared. Energy Distribution Curves (EDCs) for the evolving Fe/GaAs interface were obtained at a chamber pressure better than 3 × 10–10 mbar using a surface sensitive photon energy of 120 eV. Typically, survey scans were taken in the binding energy (BE) range – 70 to 5 eV, thereby incorporating all peaks under

scrutiny in a single sweep. Energy resolutions were estimated by measuring the energy period over which the Fermi Edge of a thin Au film rose from 10%−90% of its maximum value. In this way, a total energy resolution (i.e. encompassing thermal and instrumental broadening, as well as the effects of the finite bandwidth of the beamline's photons) of not, vert, similar 150 meV was determined.

The depositions at the ALS were all at room temperature, and covered a variety of observed reconstructions (Ga-rich) for the singular substrates. It is important to note that the qualitative details of the interface formation have been found to be largely independent of the GaAs reconstruction, and the interfaces for the singular and vicinal substrates are virtually indistinguishable. More detail concerning the coverage study and supporting measurements made “in-house” can be found in Refs. [4] and [6].

The success of the magnetization process was confirmed, independently of the Mott detector, by the performance of X-ray Magnetic Linear Dichroism in PES (XMLD-PES, not shown) [5], [6], [22] and [23]. Here a dichroism of the Fe2p is produced via the reversal of the magnetization. Similar data was collected using the Fe3p as well [5]. The data collection is spin integrated, using the multi-channel detector. The Fe deposition process and its effects upon the underlying GaAs substrate can be followed via the core-level spectra of the Fe 3p, Ga 3d and As 3d levels, as previously discussed [4] and [6]. The strong perturbation of the Ga and As peaks suggests strong intermixing. The net result is that Fe deposition upon GaAs(001) disrupts the substrate order and induces a very chaotic interface due to the substantial intermixing. The question then becomes, is it possible for an ordered structure to emerge from this chaotic situation and can this ordered structure give rise to highly polarized electron emission? The answer is yes, it is, as can be clearly seen in Fig. 1. This will be addressed next.

3. Spin resolved and energy-dependent photoelectron spectroscopy

To begin, consider first the spin integrated PES results in Fig. 2. The spin integrated spectra from the Fe grown on the singular GaAs(001) template exhibit a strong oscillatory behavior in the spectral features as function of photon energy. Interestingly, the phases seems to be reversed, with a maximum (minimum) in the higher binding peak, near − 2 to − 3 eV, corresponding to a minimum (maximum) in the peak near the Fermi Energy. If one looks at the α-Fe(001) Brillioun Zone [8], the reason for these oscillations becomes clear: the maximum at about − 2.5 eV occurs when the emission is from near the Zone Boundary, H, with hν around 145 to 160 eV. Similarly, the maximum in the Fermi Edge peak occurs when the electron momentum is along Δ, away from H, with hν near 100 eV or 190 eV. To make this easier to see, we have added downshifted final states from the extended Brillioun Zone, shown as cross-cutting lines and corresponding to hν = 100, 120, 145, 160 and 190 eV. These downshifted final states are the classical method of traditional experimental band-mapping, manifestations of the underlying energy and momentum conservation that generates “vertical transitions” within the periodic zone scheme [24]. The original work by Kisker et al., using spin resolved PES to investigate α-Fe(001) [8], was

performed at lower photon energies and several examples (hν = 60, 35 and 20 eV) were included to illustrate the relationship between photon energy and electron momentum at normal emission. Using their approach and extending it to higher photon energies, it is possible to get an approximate positioning within the Brillioun Zone (BZ). However, it is the symmetry of the photon energy dependence within the PES plots of Fig. 2 that accurately specifies the positions within the BZ. The minimum of the emission from the Fermi Edge clearly indicates the high symmetry point, H at a photon energy of 145 eV. Using this as a calibration, all of the other photon energy-specific final states are then fixed within the BZ. One way to confirm the nature of the origin of these oscillations is to make them disappear. This can be done, by changing the order: an example of this is shown in the inset in Fig. 2, for the 3° vicinal surface. Here, the oscillations as a function of photon energy have completely disappeared, confirming our analysis above [25].

A further stringent test of this hypothesis would be to determine the spin dependence of these spectral features and compare them to the BZ in Fig. 2 and the previous spin-resolved results for α-Fe(001) [8], [26] and [27]. This has been done and the results confirm the hypothesis, as will be discussed next. The spin resolved spectra for hν = 120, 145 and 160 eV are shown in Fig. 4. As can been seen, there is a strong spin dependence in the spectra that varies with position within the BZ and hν. At H (hν = 145 eV), one can clearly see the spin down bands near − 2 eV binding energy and the spin up bands near − 4 and 0 eV binding energies, corresponding to the H12↓, H12↑, H'25↑ bands, respectively (here, Kisker's nomeclature [8] has been used, following the original work of Wang and Callaway [28]). The agreement is essentially perfect. As one moves away from H (hν = 120 and 160 eV), the spectroscopic clarity becomes diminished, but with the retention of some spectral structure. Near H but along Δ, the spin up spectrum has peaks near − 4 eV and 0 eV that correlate with Δ12↑ and Δ'25↑. The spin down spectrum has a single broad peak near − 1.5 eV, from Δ12↓ and Δ'25↓ bands. The assignment at hν = 120 eV is further bolstered by the strong resemblance to the experimental data from the Kisker Group at hν = 117 eV, in a paper by Jungblut et al. [26]. However, this comparison raises an interesting point about experimental geometries. The Kisker experiment is performed at normal incidence and normal emission, which provides tremendous selectivity [8] and [26]. Our configuration, with p-polarized light, is far less selective. Thus the agreement at hν ≈ 120 eV is due in part to the position within the BZ. Comparisons with Kisker spectra near H are less agreeable, but there is strong agreement between our spectra near H and those collected by Heimann and Neddermeyer [22], who also used a p-polarization configuration. Finally, the vicinal surface spectra, also shown in Fig. 4, are all essentially the same and significantly different from the Fe/GaAs(001) spectra, again confirming the change of the Fe ordering in the vicinal surface depositions.

The highly polarized emission illustrated in [Fig. 1] and [Fig. 4] and the strongly spin-polarized bands of Fig. 2 suggest that this structure, Fe/GaAs(001), could be used as a spin injector. Although there is emission of spin down at hν = 120, 145 and 160 eV, there is a clear domination of spin up, as high as 40% polarization at hν = 145 eV. While true half-metallicity [3] and [28] might provide the desired 100%

polarization throughout the BZ, even a strong partial polarization may be sufficient for device development [1], if materials integration issues could be resolved [4]. Moreover, 100% polarization may be impossible: In Fe3O4, a candidate half-metal, the measured polarization is−40%, with a possible underlying bulk polarization of−65%, both at the Fermi Energy [29].

4. Summary

Using SRPES, it has been demonstrated that despite significant chemical intermixing and disruption of the underlying substrate, an ordered α-Fe(001)-like structure develops in Fe/GaAs(001). Thus, the spin-specific emission characteristics of this interface will be limited primarily to that associated with α-Fe(001) and this material system may be an excellent candidate for a spin injector. Ultimately the utility of the Fe/GaAs interface in spintronics applications depends on the efficiency of spin injection across the interface into the GaAs.

Carrier depolarization, across the FeGaAs and FeGa interfaces into the GaAs, is probably affected by resistance and work-function mismatches, although perhaps it can be mitigated by a smoothing of the gradients in this region, via effects such as chemical mixing. Despite the large resistance mismatch between the metallic Fe and semiconducting GaAs, efficient spin injection for the system was reported by Zhu et al. [16] for a 20 nm thick Fe layer. This Fe thickness is substantially greater than that in our study, and the efficiency of the spin injection was attributed to tunneling across the Schottky barrier formed at the interface. Our study also confirms the existence of bulk-like spin-polarized electronic structure near the interface, and this highly spin-polarized Fe layer is also likely to impact the spin injection efficiency.

Acknowledgements

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[25] The vicinal GaAs surface is usually considered to help produce epitaxial over-layers of Fe of somewhat better quality [7]. However it is well known that the quality of epitaxial over-layers can be significantly influenced by the actual preparation/growth procedure. The loss of spectral structure in the vicinal samples could arise because of the new periodicity in the surface. Steps with a lateral spacing of 30 Å would induce Brillioun Zone (BZ) nesting. This could affect the vertical as well as the lateral zone: the steps have height and will shift atoms above them. The new zone boundaries would be associated with the 30 Å wide steps, not the 2 Å or so associated with interatomic spacings and lattice parameters. This new BZ would be 1/15 as wide. With our angular acceptance of plus/minus 7° and energy resolution of 0.4 eV in the spin-resolving experiment, we would simply average over the entire BZ, thus removing any oscillatory band structure and instead getting something like a density of states. The averaging effect would be enhanced by the 3 degree or 6 degree vicinal offset: at normal emission, one would be 3° or 6° off. This is exactly what is seen in the inset in Fig. 2 and the second column in Fig. 3. To see the band effects in Fig. 2 and Fig. 3, the BZ needs to be large and the real space zone small. This is true for alpha-Fe(001)-like structure but not the growth with the periodicity of the vicinal steps. The vicinal steps probably provide strain relief at low coverages but add on another level of order that shrinks the BZ.

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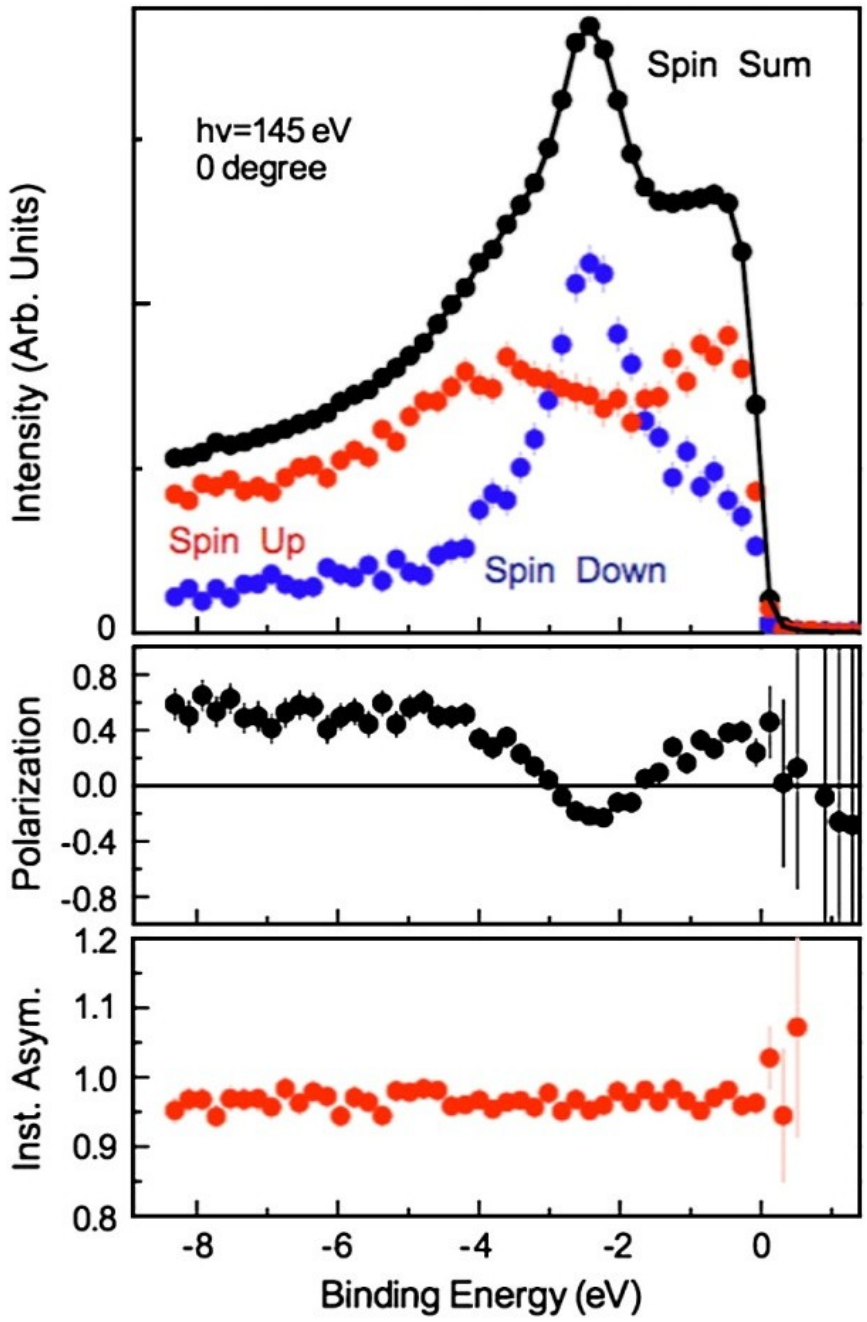


Figure 1.

SRPES results for the photon energy of 145 eV, for the valence bands of 20 Å Fe/GaAs(001). The energy resolution was approximately 0.4 eV, based upon the 10%–90% width of the Fermi Edge at 0 eV Binding Energy. The angular acceptance was +/−7°. Top panel: spin resolved spectra. Middle panel: polarization. Lower panel: instrumental asymmetry. See text for details.

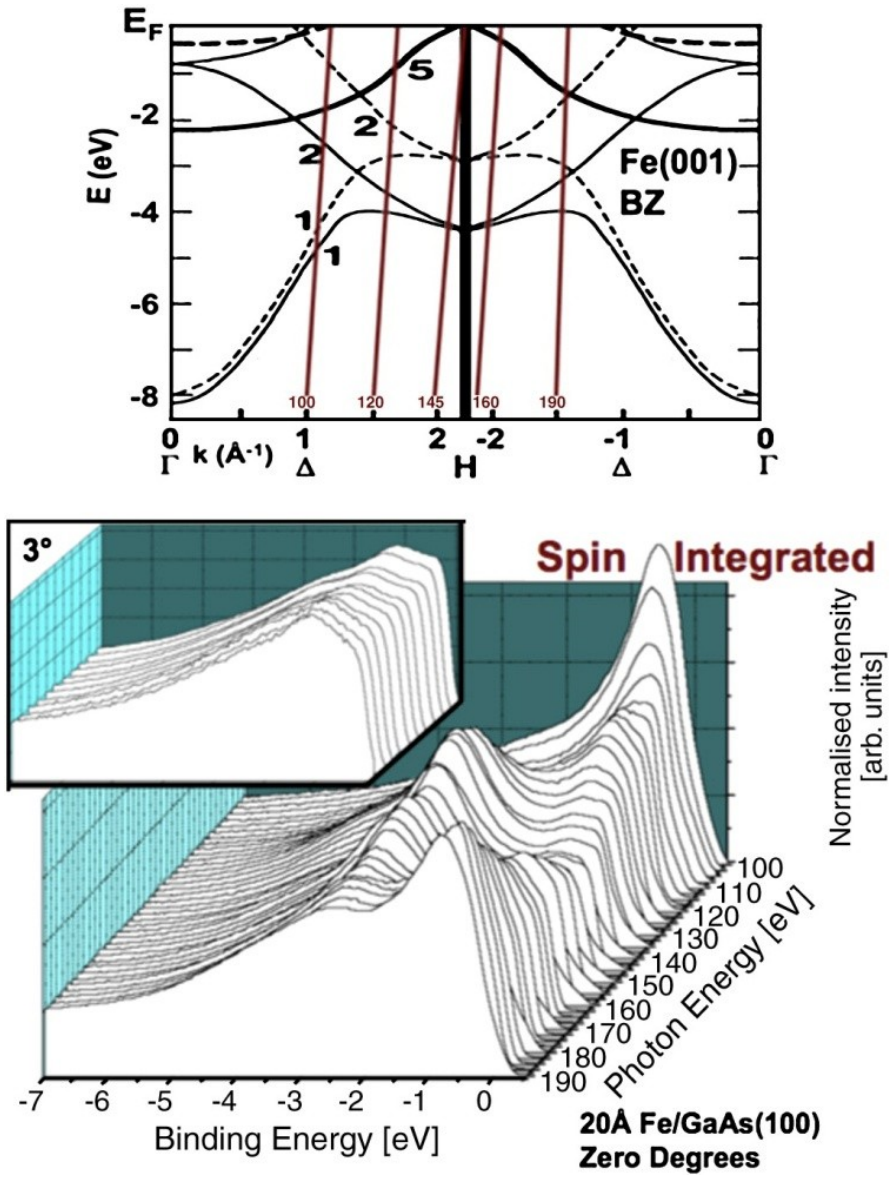
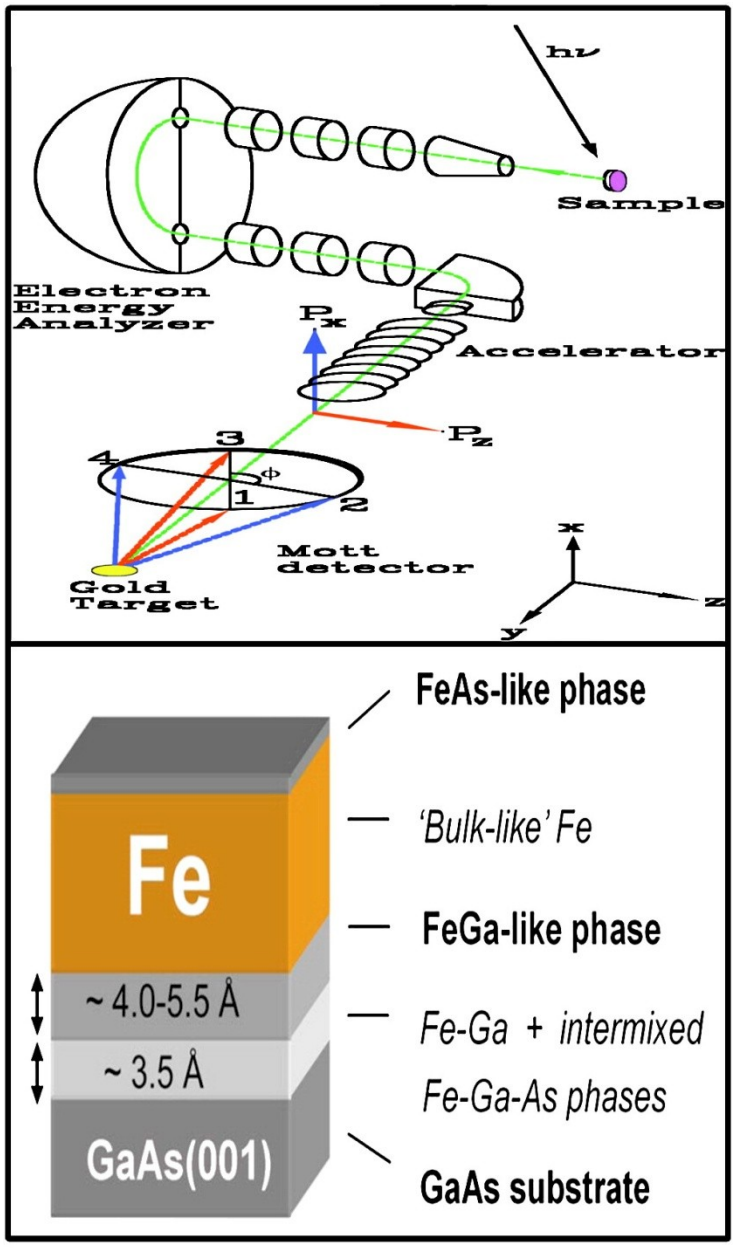


Figure 2.

Top: the spin-specific bands and Brillioun Zone of α-Fe(001) from Kisker et al. [7]. The band designations follow Kisker et al. H'25 and Δ'25 correspond to the bands marked by "5," with minima at Γ and binding energies in the range of about − 2 eV to 0 eV. (See Ref. [7] for details.) Solid (broken) lines are spin up (down). The cross-cutting lines are downshifted final states, for each specific photon energy, as described in the text.Bottom: the spin integrated spectra from 20 Å of Fe/GaAs(001), also referred to as 0°. The inset contains the corresponding results for Fe grown upon a vicinal surface of GaAs, with a 3 degree offset from the [001] direction of GaAs. In the inset, the photon energy range is 170–120 eV. The Fermi Edge is at 0 eV.


hν
Sample
Electron
Energy
Analyzer
P_x
Accelerator
P_z
3
4
1
2
Mott
detector
Gold
Target
x
z
y
FeAs-like phase
'Bulk-like' Fe
Fe
FeGa-like phase
~ 4.0-5.5 Å
Fe-Ga + intermixed
Fe-Ga-As phases
~ 3.5 Å
GaAs(001)
GaAs substrate

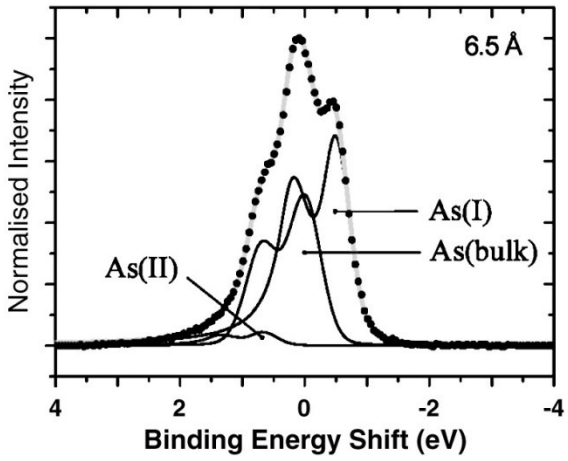

6.5 Å
Normalised Intensity
As(I)
As(bulk)
As(II)
4
2
0
-2
-4
Binding Energy Shift (eV)

Figure 3

Top panel: the experimental diagram. Middle panel: a model of Fe growth on GaAs, developed in Ref. [4]. Bottom panel: high-resolution, core-level photoelectron spectroscopy (spin integrated) of the As 3d levels in the system composed of 6.5 Å of Fe deposited upon GaAs(001), from Ref. [4]. The fine structure is strong evidence of multiple chemical sites for the As and thus strong intermixing. Please see Ref. [4] for a discussion of the different sites. Ref. [4] J.D.W. Thompson, J.R. Neal, T.H. Shen, S.A. Morton, J.G. Tobin, G.D. Waddill, J.A.D. Matthew, D. Greig and M. Hopkinson, J Appl Phys 104 (024516) (2008) and references therein.[4] also includes the corresponding spectra for the As 3d and Ga 3d states for a wide range of coverages.