Quasiparticle Lifetime in Macroscopically Uniform Ag/Fe(100) Quantum Wells

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Epitaxial Ag(100) films have been grown on highly perfect Fe(100) substrates by deposition at low temperatures followed by annealing. Precise control of the deposition process results in film thicknesses of exactly N atomic layers (N an integer up to 39) over a macroscopic distance ($\sim$1 mm). The elimination of atomic layer fluctuation, which has been generally believed to be impossible at such large thicknesses, allows us to observe atomic-layer resolved quantum well states. Such states exhibit extremely narrow linewidths, which provide a direct measure of the quasiparticle lifetime. [S0031-9007(98)07992-7]

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Quantum well states in thin metal overlayers have been observed by photoemission in a number of systems [1,2]. Much effort has been directed toward analyzing the peak positions as a function of layer thickness, which can be related to the band structure of the overlayer material [3,4]. The observed line shape and linewidth, in contrast, have been largely ignored, even though such information is potentially extremely useful. Unlike bulk single crystals for which the measured photoemission linewidths are a complicated convolution of initial and final state lifetimes, thin films, if well made, should exhibit linewidths that are a direct measure of the quasiparticle lifetime, a quantity of basic importance in solid state physics [5]. The problem is that films made in the laboratory are generally not uniform on the atomic scale and exhibit rather large linewidths caused by effects related to structural imperfection [6]. A film with a nominal thickness of N atomic layers typically consists of multiple thicknesses including N, $N \pm 1$, $N \pm 2, \ldots$, over different domains. Such atomic layer fluctuation can be caused by kinetic effects associated with film growth, defects, and steps on the substrate surface, etc., which are hard to avoid or eliminate. Worse yet, most samples have such small domains and a high defect density that random lateral confinement and scattering cause further smearing of the quantum well peaks [7]. The result is that peaks derived from individual thicknesses can no longer be resolved, and the broad line shape becomes quite useless for lifetime analysis. Thickness fluctuation is one of the most important issues in thin film science, and its control has posed a great challenge for film thicknesses larger than just a few monolayers.

The present work shows that uniform epitaxial Ag films with a single thickness of N up to 39 can be prepared on Fe(100) over a macroscopic distance ($\sim$1 mm), which is by far the largest film thickness ever reported for any system that is uniform over a macroscopic distance. This result is made possible by choosing a highly perfect Fe(100) whisker as the substrate, by depositing the Ag overlayer at a low temperature (100 K), and by choosing an appropriate annealing temperature [8]. Angle resolved photoemission from these uniform films shows extremely narrow quantum well peaks. The elimination of thickness fluctuation allows us to examine the lifetime widths of these quantum well states as a function of layer thickness and binding energy. The lattice mismatch between fcc Ag and bcc Fe is only about 0.7% in the (100) plane, and this system has attracted much interest in recent years as a prototypical metal epitaxial system [9–14]. However, previous studies of this system were generally limited to rather thin layers, and the broad line shapes did not allow a lifetime analysis.

Our photoemission experiment was carried out at the Synchrotron Radiation Center, University of Wisconsin, in Stoughton, Wisconsin. Photoemission spectra were taken using light from the 4-m Normal Incidence Monochromator. Photoelectrons emitted normal to the surface were detected with a hemispherical analyzer. Fe whiskers were used as substrates. These were prepared by numerous cycles of Ar^+ ion sputtering at energies between 0.5 and 1.5 keV starting at room temperature and ending at 550 °C. After each sputtering cycle the sample was annealed at 550 °C for 5 min. This treatment led to samples exhibiting the very pronounced but extremely contamination sensitive Fe(100) surface state [15]. An effusion cell was used to deposit Ag on the substrate surface at a temperature of 100 K. The growth rate, about 0.5 monolayer (ML) per minute, was monitored by a quartz thickness monitor. The film was then annealed to 300 °C for 90 sec and cooled back down to 100 K for the photoemission measurements. The resulting film thickness was often not an exact integral multiple of monolayers. To make up the difference to the next integral multiple, regrowth of Ag was carried out at 100 K followed by annealing as described above. The above preparation procedure yielded the best films. A great deal of effort was spent on experimenting with other sample preparation conditions, for example, growth at room temperature or higher temperatures, but invariably the resulting films were "rough," i.e., exhibiting multiple

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thicknesses. Once a rough film was created, no amount of annealing or regrowth could revive it, and the only way to return to a uniform film was to start fresh from a bare Fe(100) surface.

Figure 1 illustrates the experimental resolution of individual layer thicknesses. Photoemission spectra taken at a photon energy of 13 eV are shown for layer thicknesses of 38.0, 38.5, and 39.0 ML (1 ML = 2.04 Å). These layers were prepared by first growing a nominally 38 ML thick film in one step and adding very small amounts of Ag at low temperatures with subsequent annealing steps until a uniform layer showing just one set of quantum well peaks was obtained. The corresponding spectrum is shown as the bottom trace in Fig. 1. Subsequently 0.5 ML of Ag was added to yield the 38.5 ML spectrum, which exhibits two sets of quantum well peaks. One set is at the same position as the 38 ML case, and the newly emerged set is at somewhat lower binding energies and must correspond to a thickness of 39 ML. This is verified by adding another 0.5 ML to yield the 39 ML spectrum. The set of peaks corresponding to 38 ML is now completely suppressed, and only the 39 ML peaks remain. The same discrete layer behavior has been seen for many different starting thicknesses. If the overlayer or substrate is not optimally prepared, several layer thicknesses can be simultaneously present, giving rise to a large number of peaks. Films with even lower qualities exhibit broad line shapes, where peaks from individual thicknesses can no longer be detected. Since the sample area probed in our photoemission experiment is about $1 \times 0.5\ \mathrm{mm}^2$, the above result establishes that our film is uniform on an atomic scale over a macroscopic distance. Clearly, growth at low temperatures is the key to the making of such uniform films. It is likely that the reduced diffusion at low temperatures allows the film to build up uniformly before annealing to restore the atomic order. In contrast, growth at higher temperatures involves a large diffusion length, and kinetic effects related to the interaction of diffusing atoms and atomic steps often lead to the formation of surface roughness.

With thickness fluctuation eliminated, the quantum well peak widths become a direct measure of the quasiparticle lifetime. Shown in Fig. 2 are spectra for 12 and 38 ML films. The peaks are extremely narrow, and, in fact, narrower than any previous photoemission measurements of Ag bulk states. The 12 ML spectrum shows just two quantum well peaks and a well-defined Fermi edge. From an analysis of the Fermi edge, a system resolution of 30 meV is deduced. A fit to the quantum well peaks using Voigt line shapes (convolution of Gaussian and Lorentzian) yields a Gaussian component very close to the system resolution. This confirms that the intrinsic line shape of the quantum well peaks is Lorentzian in nature. In our final analysis, the Lorentzian widths are deduced from the fit by setting the Gaussian width equal to the system resolution obtained from the Fermi edge.

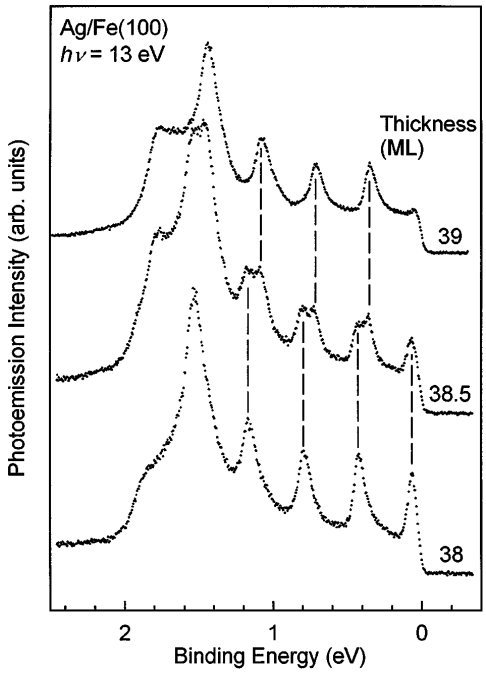

FIG. 1. Normal emission spectra from the valence band region of a 38 ML (bottom), 38.5 ML (center), and 39 ML (top) Ag film on Fe(100) taken at a photon energy of 13 eV. The quantum well peak positions are indicated by vertical lines. The spectrum for the 38.5 ML film shows two sets of quantum well peaks indicating the simultaneous presence of areas covered by 38 and 39 ML of Ag.

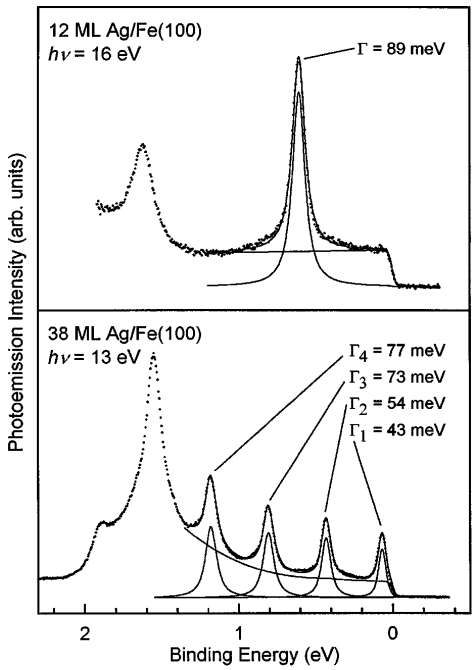

FIG. 2. Examples of line shape analysis for 12 and 38 ML Ag films on Fe(100).

These Lorentzian widths are shown in the figure. The background function used in the fit is also shown; it is taken to be a linear function for the 12 ML case and a cubic function for the 38 ML case (the background appears different for the two spectra due to the use of different photon energies).

Figure 3(a) summarizes the observed trends for the quasiparticle lifetime width as a function of binding energy and layer thickness. A total of 70 peak fits was performed for different layers and for spectra taken at different photon energies. The majority of data points in the plot represent average values deduced from several spectra. The error bar, based on variations among the different fits, is less than 5% and not shown. The graph shows data from Ag films of 1, 2, 3, 10, 11, 12, 28, and 38 ML thickness. Additional data for thicknesses in the range from 5 to 9 ML are suppressed from the graph in order to maintain clarity, but follow the same trends described below. The straight lines are linear fits to the data; they represent the behavior of films in different thickness groups. The top line is a fit to the peak widths obtained from 1–3 ML thick films, and the next line is a fit for 10–12 ML thick films. The bottom two lines are fits to results from 28 and 38 ML thick films, respectively.

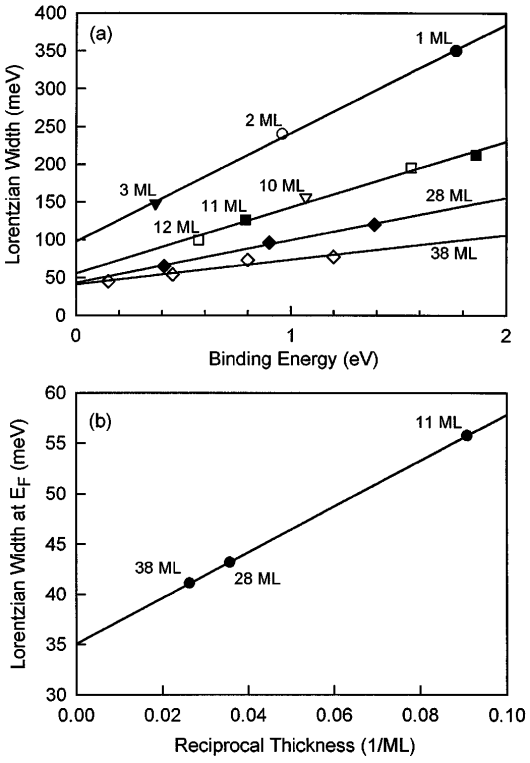

FIG. 3. (a) Observed Lorentzian linewidths of quantum well states as a function of binding energy. Each symbol represents a different layer thickness as indicated. The straight lines are fits to the data in different thickness groups (1–3, 10–12, 28, and 38 ML). The intercepts at the vertical axis represent linewidths at the Fermi level. (b) Linewidths at the Fermi level vs the reciprocal layer thickness. The straight line is a fit, and its intercept at the vertical axis shows a width of 36 meV at the Fermi level for infinite layer thickness (no boundary loss).

The observed linewidth or lifetime of the quasiparticle depends on two independent parameters: the thickness of the film and the binding energy. The thickness dependence is significant and can be understood as a boundary effect. The formation of quantum well states in the Ag film is a result of electron confinement by a "hybridization gap" in the Fe substrate [9–14]. However, this is not a real, absolute band gap, and the confinement is not perfect. The electron reflection coefficient at the Ag-Fe boundary is expected to be less than unity, and the loss of amplitude upon reflection will contribute to the time decay of the quantum well state, and hence its lifetime broadening. The situation is very similar to the Fabry-Perot interferometer, where a finite finesse due to less than perfect reflectivity of the end mirrors causes the cavity modes to broaden. This boundary effect is expected to diminish at large thicknesses, and this is indeed observed experimentally.

The measured width also depends on the binding energy E, and the data seem to suggest an approximately linear dependence for a given thickness over the entire energy range probed in this experiment. Phonon scattering should contribute a term nearly independent of E, while Auger decay should contribute a term proportional to E^2 near the Fermi level based on a phase space argument [16]. This E^2 dependence is often taken to be an important test of the Fermi liquid behavior, but its validity is limited to a very small energy range and thus cannot be verified in the present experiment. The increase in linewidth for increasing E as observed in Fig. 3 is qualitatively consistent with the Auger decay mechanism.

Extrapolating the four straight lines in Fig. 3(a) to $E = 0$ yields linewidths of 97, 55, 43, and 41 meV, which represent the linewidths at the Fermi level and are thus without the Auger contribution. There is still a thickness dependence due to the finite finesse effect mentioned above, whose contribution should decay as the inverse thickness asymptotically. Figure 3(b) shows a plot of these linewidths at the Fermi level as a function of $1/t$, where t is the average thickness for each straight line in Fig. 3(a). The $N = 1$–3 case is ignored in this plot, because the $1/t$ asymptotic behavior is not expected to hold for small thicknesses. By extrapolation to $1/t = 0$ as indicated by the straight line fit, we can deduce a limiting width of 36 meV for infinite finesse (no boundary effect). It is also clear from the fit that the 38 ML case is fairly close to the asymptotic limit.

What is left in this 36 meV width is the phonon contribution plus anything else that has not yet been accounted for. Based on our temperature dependent study of the linewidth in the range of 100–400 K (data not shown here), we can extrapolate down to 0 K and deduce a phonon contribution of about 13 meV at $T = 100$ K [17]. Thus, a linewidth of 23 meV remains at the Fermi

level at $T = 0$. Ideally, a perfect film with no boundary loss should show a zero linewidth at $T = 0$, assuming that the system behaves as a Fermi liquid [18]. There is little doubt that Ag behaves as a Fermi liquid. The 23 meV residual width in our case translates into a longitudinal quasiparticle coherence length of 370 Å. This is a fairly large length, but not infinite. The reason for this finite coherence length is likely scattering by residual defects in the film such as trace impurities and point defects. After all, no materials can be made truly perfect. Notice that the lattice match between Fe and Ag is actually not perfect, which could be a source of strain and defects. The 23 meV residual width is by far the narrowest determined by photoemission to this date for any bulk state in a three dimensional material.

In conclusion, this work reports the successful preparation of uniform thin films of Ag on Fe(100) with a single thickness of N atomic layers for N up to 39 over a macroscopic distance. This level of layer control is unprecedented and allows us to observe layer resolved quantum well peaks in angle resolved photoemission spectra. The peaks are much sharper than previously reported in similar systems due to the elimination of thickness fluctuation, and a detailed analysis of the quasiparticle lifetime can be made. The peak widths are seen to depend on film thickness and binding energy and are explained in terms of elementary processes including Auger decay, reflection loss at the interface, and phonon scattering.

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[18] A key issue here is that the quasiparticle (described by a valence state) is coherent over a large distance. In contrast, core hole excitation involves a highly localized charge, and the photoemission line shape can be significantly affected by electron hole excitations (x-ray singularity) and a Franck-Condon lattice response giving rise to a Gaussian contribution to the line shape. See, for example, P. H. Citrin, G. K. Wertheim, and Y. Baer, Phys. Rev. B **16**, 4256 (1977). In our case, the valence charge is distributed over a very large volume, and therefore the x-ray singularity and Franck-Condon effects as discussed by Citrin, Wertheim, and Baer are negligible. The paper by McDougall, Balasubramanian, and Jensen cited in Ref. [17] contains an excellent discussion of the Fermi liquid response to a valence excitation.