One-Dimensional Spin-Polarized Quantum-Wire States in Au on Ni(110)

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Au chain structures have been prepared on Ni(110). Au6*s*, *p*-derived features in photoemission spectra are identified as quantum-wire states due to their strong dispersion along the chains and absence of dispersion perpendicular to the chains in agreement with our *ab initio* calculation of the electronic structure. Spin analysis reveals that the states have minority-spin character showing that the confinement of electrons in the chain structure depends on the electron spin.

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Research on quantum confinement in metallic heterostructures has intensified very much since the link was found between long-range oscillatory magnetic coupling phenomena in magnetic-nonmagnetic multilayers [1] on the one hand and the periodic appearance of structures in photoemission and inverse photoemission spectra on the other hand [2]. The so-called "quantum-well model of oscillatory magnetic coupling" is based on the confinement of electrons in a metallic layer brought about by spin-dependent band gaps in the neighboring layers. These metallic quantum wells are similar to the ones known in semiconductor structures; however, due to larger offsets of the relative band gaps (several eV) a spatially narrower confinement of electrons can be achieved. The model has been supported by a number of subsequent experiments on bilayers and trilayers revealing the layer-thickness dependence, dispersion parallel to the film plane, spin character, and a second, small period of the appearance of quantum-well states [2–9].

As in these experiments confinement occurs only in the direction perpendicular to the metal-metal interfaces giving rise to two-dimensional quantum-well states, it appears challenging to attempt confinement also within the plane with the aim of producing a *quantum wire* of atomic dimensions. Research interest has been concentrating on steps as natural one-dimensional structures at clean metal surfaces, and these have therefore been studied first by photoelectron spectroscopy. In this way, the one-dimensional equivalent of surface states, i.e., step states, has been identified in photoemission spectra [10,11]. A one-dimensional dispersion could be shown for image-potential states [12].

Deposited metals adsorb preferentially at steps. Taking advantage of this, a growth method termed "step decoration" has been used to grow quasi-one-dimensional structures. For Cu/W(110), a binding-energy shift in Cu3*d* photoemission has been attributed to the formation of Cu stripes [13]. Although much progress has been made concerning the quality of such stripes, e.g., using scanning tunneling microscopy (STM) [14], a one-dimensional anisotropy of their electronic structure has never been shown. The developments up to this point have been documented in more detail in Ref. [15].

We report in this Letter on a surface alloy that has long-range one-dimensional geometry. By the so-called "dealloying" phase separation during the growth of Au on Ni(110), chains of impressive coherence and a narrow interchain distance (a few surface lattice constants) are observed by STM [16]. The structural characterization of Ref. [16] reveals, in brief, that surface alloying of Au occurs up to a coverage of 0.4 monolayers (ML). Above 0.4 ML, 42% of the Au atoms form dimer-trimer chains out of a Au-Ni surface alloy as a result of a first-order phase transition. These chains are oriented along the [001] direction as seen from our model in Fig. 1. For increasing coverage, the interchain distance decreases until 0.93 ML are deposited [16]. We show below with photoemission that Au/Ni(110) is the first example of a metal-on-metal system where one dimensionality of the chains can be seen from the measured angle dispersion. The characteristic dispersion (strong parallel to the chain orientation and absent perpendicular) is observed for Au5*d* and Au6*s*, *p* states. Analogous to the case of spin-polarized quantum-well states in multilayer structures, electrons of different spin character are confined to a different degree.

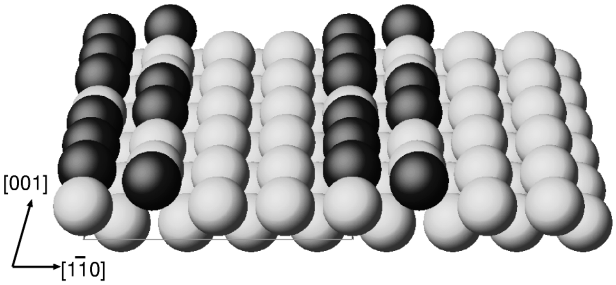

FIG. 1. Geometry of Au/Ni(110) used for the *ab initio* band structure calculation according to the structural model of Ref. [16]. Au atoms (black) for dimer-trimer chains along [001] on top of the Ni substrate atoms (white).

Angle-resolved photoelectron spectra have been measured with a VG Escalab photoemission chamber at 1×10^{-10} mbar with a combined energy resolution of 150 meV and an angle resolution of 2° using linearly polarized synchrotron light from the 3m NIM-1 monochromator at BESSY I. Spin-resolved photoemission at 300 meV and 2° resolution has been done at the TGM-1 monochromator. In both setups, light incidence angle was about 30° with respect to the fixed analyzer direction leading to mixed $(s + p)$ polarization in normal emission. Sample preparation was performed *in situ* according to Ref. [16] using a quartz microbalance to adjust the Au evaporation rate (0.1 ML/min) and, in addition, low-energy electron diffraction. We conducted *ab initio* calculations of the electronic structure for a dimer-trimer chain on a $p(3 \times 4)$ Ni(110) surface as shown in Fig. 1 (for clarity, four unit cells are displayed). This structural model has been given by Nielsen *et al.* It does not include those Au atoms (58%) that do not join the chain structure and instead are alloyed into the Ni surface layer since their position is not observed by STM [16] and therefore unknown. We employed the full-potential linearized augmented plane wave method for thin film geometry [17] in the local spin density approximation including spin orbit coupling inside the atomic spheres with the spin direction in the Ni plane. We used approximately 100 augmented plane waves per atom and eight $\vec{k}$ points in the irreducible part of the Brillouin zone for self-consistency.

Figure 2 shows photoemission spectra of the valence band for clean Ni(110) and Au coverages between 0.1 and 2 ML. The spectra permit a clear distinction of different coverage regions as labeled in the figure: In region I, two peaks at high binding energy (5.7 and 4.2 eV) are seen. These peaks are characteristic of the surface-alloy-formation stage. In region II, an extra structure appears at 3.0 eV, which becomes narrow and sharp at 0.6 ML. In addition, another peak occurs at lower binding energy (1.9 eV). Below we focus on this thickness and spectrum and show that they are characteristic of the one-dimensional Au chain. Towards 0.9 ML, the structures in the spectra become broader and are believed to be due to the formation of a two-dimensional Au film.

Figure 3a shows for 0.6 ML the angle dependence in the $[1\bar{1}0]$ azimuth; i.e., **k** is varied perpendicular to the chains. (Here, **k** denotes the electron-wave-vector component in the surface plane.) We concentrate on the lower binding-energy region since we observe only here a peak that is not present for lower and for higher Au coverage. This structure remains at constant binding energy of 1.9 eV. For angles along the [001] azimuth, i.e., **k** variation parallel to the chains as displayed in Fig. 3b, the structure disperses by almost 1 eV. This can be explained by a confinement of Au electrons perpendicular to the chains and no confinement along the chains as expected for a quantum wire.

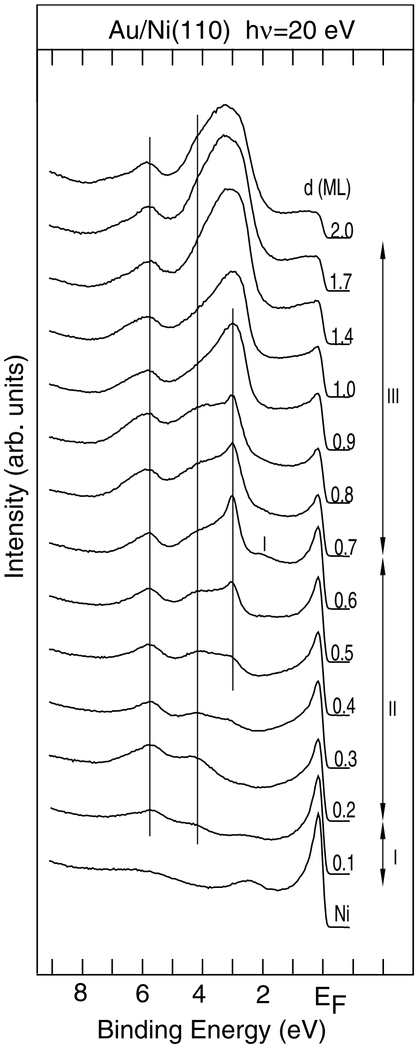

FIG. 2. Thickness-dependent photoemission spectra of Au/Ni(110) at 20-eV photon energy and normal emission. The characteristic chain structure occurs for $d = 0.6$ ML.

In addition, we have performed spin analysis of the spectrum at $-8°$ emission angle in Fig. 3b. The result is displayed in Fig. 4 as comparison of the spin-polarization spectrum between the Ni substrate before and after preparation of the Au chains. Its large and negative spin polarization shows that the quantum-wire state is of minority spin.

The large dispersion and the low binding energy of the quantum-wire peaks suggest that they are derived from Au$6s,p$ states. That Au$6s,p$ states have a highly anisotropic dispersion and must therefore be confined to the one-dimensional structure is surprising in view of the band structures of Au and Ni. The d states of Au and Ni are clearly separated in energy whereas s,p states are overlapping. This fact is easily seen from the calculated partial densities of states on Au and Ni atoms in Fig. 5b. The densities of states, dominated by d states, are for Ni high above and for Au below an energy position of about -3 eV. The s,p states are, however, generally extended over the whole energy range, and in order to assign them we use the band dispersion displayed for states

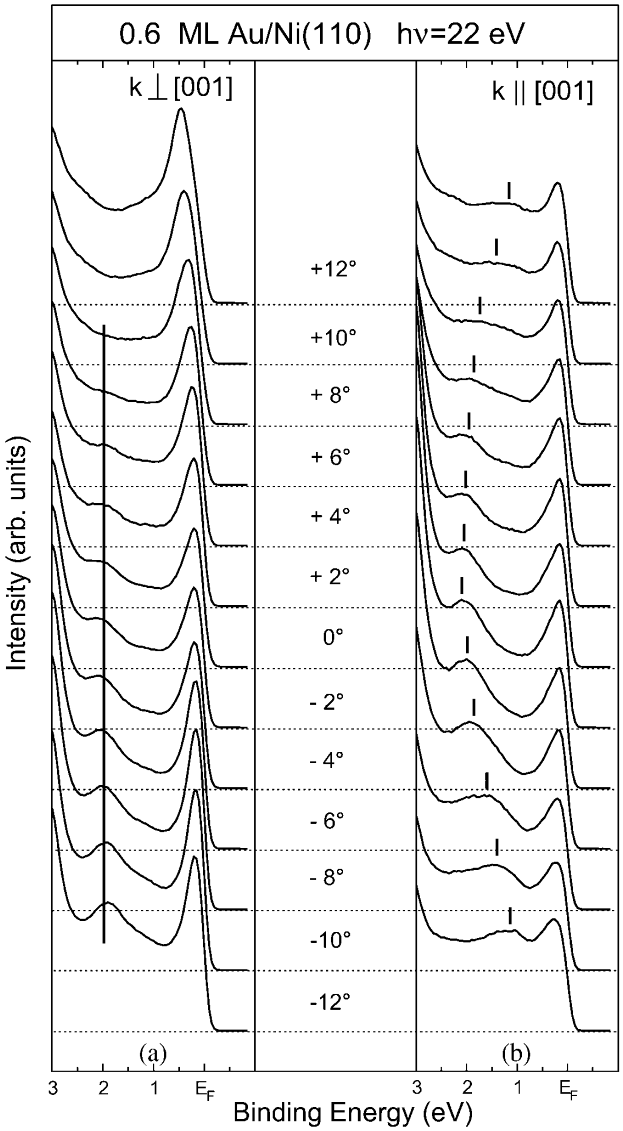

FIG. 3. Angle-resolved photoemission spectra of 0.6 ML Au/Ni(110) taken at 22-eV photon energy. (a) The electron emission angle Θ is varied perpendicular to the chains leaving the quantum-wire peak at fixed binding energy. (b) For angle variation parallel to the chains a finite dispersion is observed.

with substantial charge density on Au atoms (>15% in Fig. 5a). Although at somewhat lower energy than in the experiment, a band originating at -2.7 eV at $\overline{\Gamma}$ is easily identified and shows a considerable dispersion of about 0.6 eV along the chains and negligible dispersion perpendicular [18]. The experimental dispersion in Fig. 3b also amounts to about 0.5 eV between 0° and the zone boundary at 8°. Moreover, we can analyze this dispersing band further and find, e.g., at $\overline{X}$ with 48% s, p character within Au muffin-tin spheres, a distribution that is typical for s, p states in this energy range [19].

In this way, the calculated results confirm also that (i) the chain-substrate interaction is small enough and (ii) the chain-chain interaction is sufficiently reduced in the present system and in the model (chain-chain separation of 10.0 Å) in order to cause a one-dimensional band dispersion. We investigated the influence of the height (h) of the chain above the Ni surface and found that above 0.5 Å the dispersion of the bands is constant within 0.01 eV. Closer to the surface the bands parallel to the [001] direction had 0.05–0.1 eV less dispersion and were still almost flat perpendicular to [001]. Aware of the fact that the apparent height of an STM image can be quite different from the true atomic positions, we have chosen the structural model of Ref. [16] in Fig. 5 since a small change in dispersion does not affect our interpretation.

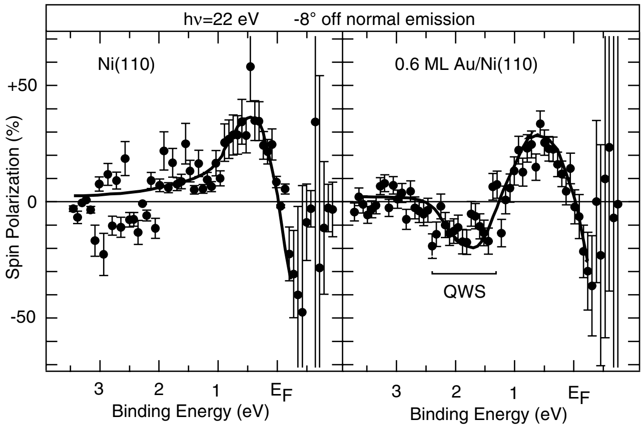

FIG. 4. Spin-polarization spectra show that the quantum-wire state is of minority spin.

We want to discuss the peak broadening seen in Fig. 3b at $-8°$ and 1.5 eV. The periodicity of the zigzag structure (3 Ni lattice constants) leads to Bragg reflection gaps expected at ±8.3° and 1.5 eV. Although our calculation does not predict the size of this gap precisely enough, this is the most likely explanation for this broadening [20].

The minority-spin character shows that the confinement of the electrons to the Au chain depends on their spin. Spin-dependent hybridization of electronic states can lead to a stronger chain-substrate interaction for one spin direction and a weaker chain-substrate interaction for the other one, here minority spin, which is confined more strongly to the Au chain. Analysis of the spin character in the calculation shows that the dispersing band in Fig. 5a is of minority spin. Figure 5b shows already that this band is located near a 0.5-eV wide minority-spin bulk band gap extending around -3 eV. However, two Ni layers as substrate are too thin to observe Ni bulk band gaps in a **k**-resolved manner. Comparison to a 13-layer ferromagnetic Ni(110) band structure [21] shows that away from normal emission the measured energy positions of the quantum-wire state lie in a bulk band gap of even symmetry and minority spin. Keeping in mind that in the case of two-dimensional multilayers the spin character of quantum-well states is understood as a result of spin-dependent bulk band gaps in the magnetic material [2–9], this result indicates that also in the present system the confinement in one dimension may

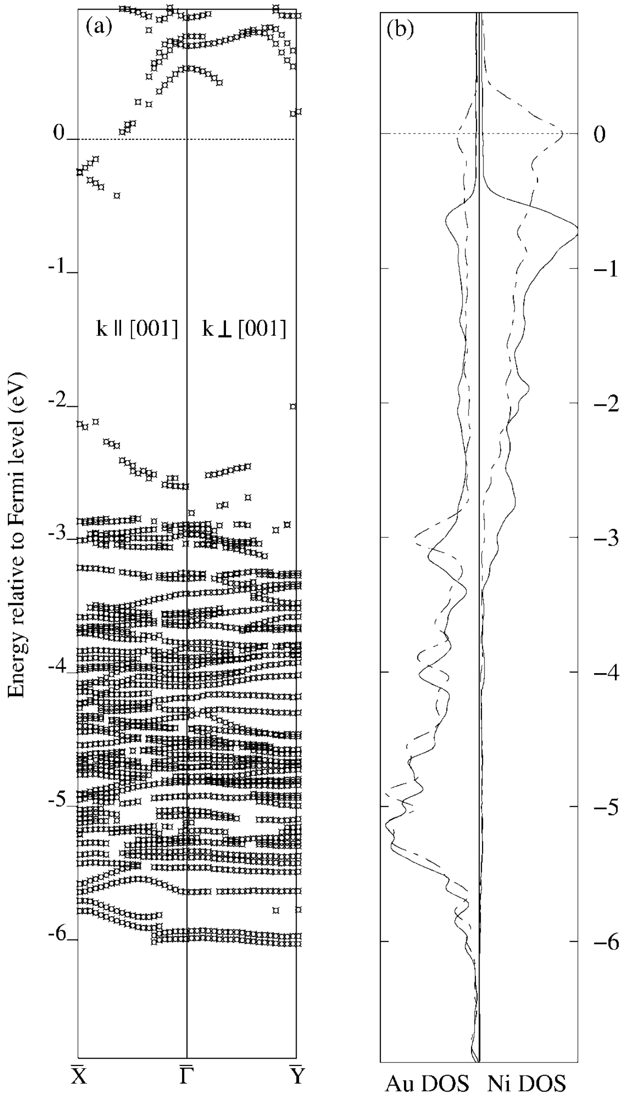

FIG. 5. Results from the *ab initio* calculation. (a) E vs **k** band dispersions for states with substantial charge density on Au atoms. A strongly anisotropic dispersion is observed. [$\overline{X}$ and $\overline{Y}$ refer to the $p(3 \times 4)$ structure.] (b) Partial densities of states for Au (left) and Ni (right). Solid lines are for majority spin and dashed lines for minority spin.

be imposed by spin-dependent bulk band gaps which lead to the different degree of confinement for the two spin directions.

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[17] See, e.g., M. Weinert and S. Blügel, in *Magnetic Multilayers,* edited by L. H. Bennett and R. H. Watson (World Scientific, Singapore, 1993).

[18] The position of alloyed Au atoms beneath the chains cannot be specified by STM and has been neglected in our model. Note also that our Au chain is embedded in only two layers of Ni and that this might affect energy positions of the Au states.

[19] For bulk Au the density of states has, e.g., at E_F, 36% s, p character within muffin tins according to D. A. Papaconstantopoulos, *Handbook of the Band Structure of Elemental Solids* (Plenum, New York, 1986).

[20] Truly one-dimensional metals show Tomonaga-Luttinger-liquid behavior [F. D. Haldane, J. Phys. C **14**, 2585 (1981)], in particular, a power-law behavior of the intensity at the Fermi energy and spin-charge separation in the spectral function. Whether in the present system chain-substrate interaction is sufficiently weak and intrachain Coulomb interaction sufficiently high to lead to observable effects, is at present unclear and might be an interesting matter for follow-up studies. With our method, Au states at the Fermi energy are experimentally not accessible due to dominating Ni-substrate emission, and the various peak broadenings or splittings observed far from the Fermi energy can well be explained on the basis of Bragg scattering.

[21] J. Redinger (unpublished).