Quenching of Majority-Channel Quasiparticle Excitations in Cobalt

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The low-energy electronic excitations in cobalt are studied by a theoretical method that includes many-body effects and a realistic description of the band structure. Angle-resolved photoemission spectra measured on a thick film of hexagonal close-packed Co on Cu(111) agree well with calculated spectral functions. Because of many-body effects no sharp quasiparticle peaks exist for binding energies larger than 2 eV and in this energy region the spectrum is essentially incoherent. The many-body corrections are much stronger in the majority-spin channel and drastically affect the spin polarization of the spectra.

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The current interpretation of photoemission spectra of metals relies on the Landau Fermi liquid theory and the underlying concept of weakly interacting quasiparticles [1]. The fundamental assumption is that the low-energy excitations of the interacting electrons can be put into a one-to-one correspondence with those of noninteracting electrons with renormalized properties (energy and mass). Transition metals are the simplest systems exhibiting nontrivial electron-electron correlations; in the prototypical case of nickel a quasiparticle band structure has been unambiguously identified in a large energy window below the Fermi energy: local correlations between electrons in the partially filled d band do modify the energy position and dispersion of low-energy excitations with respect to single particle results, but the spectral functions for one electron removal still present well-defined quasiparticle peaks down to 9 eV below E_f [2–4].

We show that the existence of a well-defined quasiparticle band structure over a relatively wide energy range below E_f is not a general feature of transition metals and that in the case of cobalt severe energy renormalization is accompanied by a removal of d band spectral weight from the region below 2 eV binding energy. This fact is associated with the damping of quasiparticle excitations and the corresponding increase of the incoherent part of spectral function in this energy region.

Photoemission spectroscopy is a direct experimental tool to check the validity of the Fermi liquid scheme. The state of interacting electrons with one removed particle $\hat{c}^n_{\mathbf{k}\sigma}|\Psi_0\rangle$ is in general not an eigenstate but a complicated combination of a large number of them; its norm is distributed over many energies with density given, at energy ω, by the hole spectral function $A^-_{\mathbf{k}\sigma}(\omega)$. If $A^-_{\mathbf{k}\sigma}(\omega)$ exhibits a sharp maximum of width 2Γ, centered at $\omega = \epsilon$ the components of $\hat{c}^n_{\mathbf{k}\sigma}|\Psi_0\rangle$ around ϵ oscillate at common frequency and their phases remain coherent over a time $1/\Gamma$. Only in these cases the time development of the system with one removed particle can be interpreted as the propagation of a quasiparticle with a reasonably well-defined energy and a sufficiently long lifetime, and the spectra measured in a photoemission experiment can be reduced to a (quasiparticle) band structure.

For a detailed comparison of photoemission spectra to such quasiparticle spectral functions, we have performed angle-resolved photoemission experiments on Co(0001). Ultrathin Co films of up to 24 monolayers (ML) thickness deposited on Cu(111) were used as samples. These films show the hexagonal close-packed (hcp) structure with the Co(0001) surface exposed. They produce well-defined Fermi surface contours in angle-resolved photoemission experiments [5], which is not the case on Co(0001) single crystal surfaces [6]. The latter problem is related to the photoelectron momentum smearing in the magnetic stray fields above the Co(0001) surface which has the easy magnetization axis normal to the surface, while it is in plane for Co/Cu(111) for films thicker than 2 ML [7].

The film growth and photoemission experiments were carried out in a modified Vacuum Generators ESCALAB 220 [8]. Co was evaporated from a hot filament onto Cu(111) that has been cleaned by standard procedures. Sample cleanliness and Co coverages were measured by x-ray photoelectron spectroscopy. The film structure was verified to be hcp by x-ray photoelectron diffraction and the high crystalline quality can be judged from the fine contours measured in photoemission Fermi surface maps [5], as well as from the sharp (1×1) spots in the low-energy electron diffraction pattern. Scanning tunneling microscopy images show a compact but fairly rough film typical for room-temperature-grown Co films on Cu(111) [9]. Angle-resolved photoemission data were measured using monochromatized He Iα radiation for excitation, with a photon energy of 21.2 eV. The spectrometer energy and angular resolution were set to 50 meV and 1.5°, respectively.

The evolution of normal emission spectra with Co film thickness is shown in Fig. 1. The spectrum from clean Cu(111) (top curve) is dominated by strong emission from the Cu $3d$ band in the binding energy range from 2 to

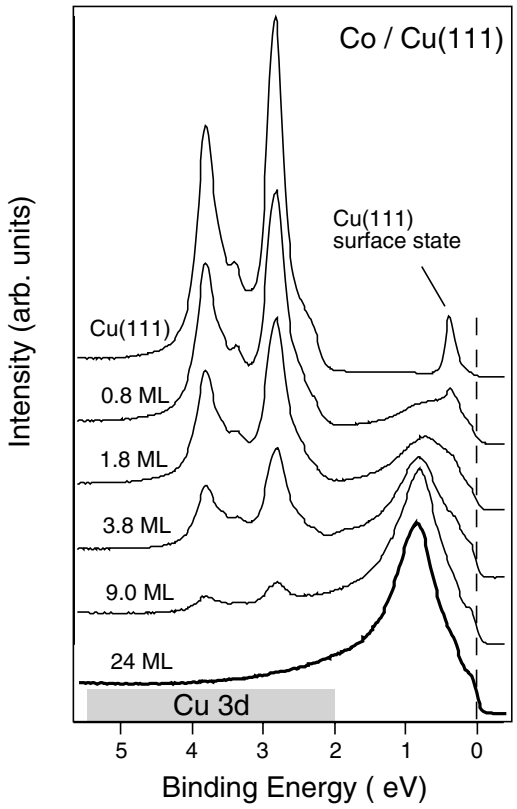

FIG. 1. He Iα (21.2 eV) excited photoemission spectra from Co layers deposited on Cu(111) as a function of film thickness in monolayers (ML). The spectra were taken in normal emission. The Cu 3d spectral region is indicated, as well as the Shockley surface state on clean Cu(111).

5.6 eV, and by emission from the well-known Shockley surface state at a binding energy of 0.4 eV. Upon Co deposition, the Cu 3d band is strongly attenuated, maintaining more or less its spectral shape. In the 24 ML spectrum (bottom curve) it has completely vanished, confirming that the Co film is compact and no Cu atoms are floating on top of the film. The Cu(111) surface state disappears between 1 and 2 ML Co coverage, a fact that is consistent with the complex morphology of the initial film growth in this system [9]. Simultaneously, a broad peak centered at 0.8 eV binding energy grows in, showing two weak shoulders in the leading edge near the Fermi energy. The shape and position of this peak agrees well with normal emission spectra measured by Himpsel and Eastman on a Co(0001) single crystal at 21 eV photon energy [10]. The shoulder at 0.3 eV binding energy has been identified as a surface state of Λ_1 symmetry [11]. These data thus confirm that the 24 ML film is an excellent sample representing the photoemission spectrum of Co(0001). There are no distinct spectral features from Co at binding energies higher than 2 eV that may be identified with quasiparticle excitations.

This is confirmed by the analysis of the polar dispersion plot along the $\overline{\Gamma}\overline{M}$ direction of the surface Brillouin zone [12] shown in Fig. 2(b): For all polar angles, dispersing features are observed only for binding energies of less than 2 eV. This is quite surprising since the single particle band structure of ferromagnetic hcp cobalt exhibits a whole manifold of states that extends to higher energies [see Fig. 2(d)]. It is furthermore in drastic contrast to the quasiparticle dispersion seen on clean Cu over a broad energy range [Fig. 2(a)] as measured under identical experimental conditions.

To understand the origin of this disappearance of spectral weight we have explicitly included electron-electron correlation and calculated self-energy corrections, hole propagator, and spectral functions. We have applied the three-body scattering (3BS) approximation [13–15], a method which augments a realistic band structure calculation with many-body correlations. 3BS is most appropriate to treat the short-range intrasite interactions of narrow band systems that are not simply accounted for by other methods [16]. The electron-electron interaction is included via on-site screened Coulomb and exchange integrals U and J giving rise to a multiband Hubbard-like Hamiltonian

$$\hat{H} = \sum_{\alpha\sigma i} \epsilon_{i\alpha}\hat{n}_{i\alpha\sigma} + \sum_{\alpha\beta\sigma i\neq j} t_{i\alpha,j\beta}\hat{c}^{\dagger}_{i\alpha\sigma}\hat{c}_{j\beta\sigma} + \frac{1}{2}\sum_{\alpha\beta\sigma i}\left[(U-J)\hat{n}_{i\alpha\sigma}\hat{n}_{i\beta\sigma} + U\hat{n}_{i\alpha\sigma}\hat{n}_{i\beta-\sigma}\right]. \quad (1)$$

Here α, β run over the d orbitals and the one-body terms $\epsilon_{i\alpha}, t_{i\alpha,j\beta}$ are related to the single particle band eigenvalues $\epsilon^n_{\mathbf{k}}$ [3].

According to the 3BS approach the interacting many-body state is expanded on the configurations obtained by adding single electron-hole (e-h) pairs to the ground state of the single-particle Hamiltonian. The response of the interacting system to the creation of one hole is then described in terms of interactions between configurations with one hole plus one e-h pair, giving rise to multiple h-h and h-e scattering. The Faddeev theory [17] is used to determine the total scattering matrix to get the self-energy $\Sigma_{n\mathbf{k}}(\omega)$, and the hole spectral function given by

$$A^{-}_{n\mathbf{k}}(\omega) = -\frac{1}{\pi}\,\mathrm{Im}\frac{1}{\omega - \epsilon^n_{\mathbf{k}} - \Sigma_{n\mathbf{k}}(\omega)}. \quad (2)$$

Further details on the method have been given elsewhere [3,18,19].

The *ab initio* local density approximation band structure of bulk hcp cobalt has been calculated with the linear muffin-tin orbital method [20] and augmented with the inclusion of on-site correlation according to the 3BS scheme. Figure 2(c) shows the hole spectral function along the $\overline{\Gamma}\overline{M}$ direction calculated within the 3BS approach (U = 2.1 eV and J = 0.8 eV [21]). No sharp peak exists below 2.0 eV in agreement with experimental findings, and the spectral functions exhibit only a uniform background. Because of many-body effects the components of $c^{\hat{n}}_{\mathbf{k}\sigma}|\Psi_0\rangle$ remain then coherent only over a very short time ($1/\Gamma$ = $\mathrm{Im}[\Sigma_{n\mathbf{k}}(\omega)]^{-1} \ll 1$) and the time evolution of the state

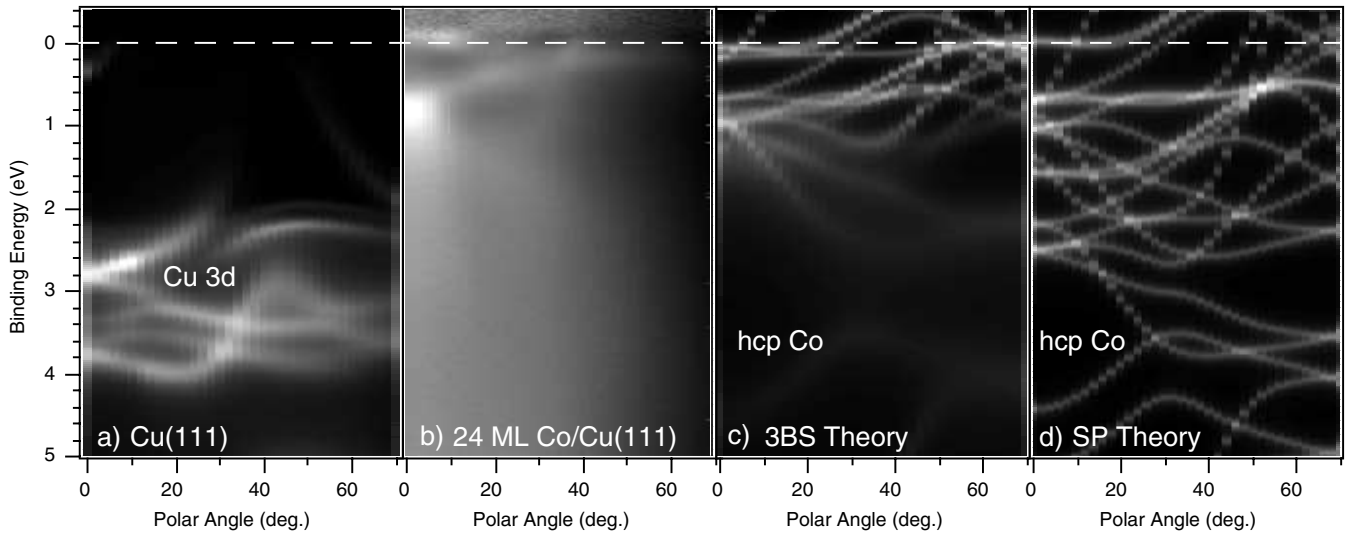

FIG. 2. (a) He Iα excited photoemission dispersion plot from Cu(111) measured along the $\overline{\Gamma}\overline{M}$ direction of the surface Brillouin zone: for each polar angle, the column in the plot represents the photoemission spectrum in a linear grey scale. (b) The same for the 24 ML film of hcp Co on Cu(111). The data have been normalized to equal average intensity along each horizontal line. (c) Quasiparticle spectral function for hcp Co, calculated for the same wave vectors using the three-body scattering approach. (d) Corresponding single-particle band structure.

with one removed particle cannot be reduced to a quasiparticle propagation. The dominance of incoherent excitation, i.e., the large value of $\mathrm{Im}[\Sigma_{n\mathbf{k}}(\omega)]$ in this energy region, is the main point here. Why are these effects so much stronger in cobalt than in nickel, for instance, in spite of the similar strength of the screened on-site e-e interaction? The point is that the importance of correlation effects is related not only to the strength of the e-e interaction but also to the band occupation. In the extreme case of a completely filled band, where no e-h pairs can be added, correlation effects are absent, irrespectively of the strength of the e-e interaction. This is the case of copper, the only metal of the $3d$ transition series where band theory gives a full account of photoemission data [22]. In all the other cases we expect self-energy corrections to increase — for given U's and J's — with an increasing number of unoccupied d states [23]: in the same way as in atoms, where the complexity of the multiplet structure depends on the shell occupation, the number and mixing of configurations is larger when the valence band has a high density of holes. This is particularly evident in the 3BS approach where the e-h and e-e scattering efficiency depends directly on the number of empty d states necessary for the creation of excited configurations [18].

The same physical argument explains why many-body interactions affect differently states of different spin. Because of the relatively small number of empty states in the majority spin band, the creation of a majority spin hole will be followed by scattering processes involving mainly opposite spin electron-hole pairs; conversely the creation of a minority spin hole will correspond to scattering between parallel spin particles. The different strength of the interaction among opposite spin particles (proportional to U) and parallel spin ones (proportional to $U - J$) gives rise to spin-dependent self-energy renormalizations which affect spin-up states more than spin-down ones. This is shown in Fig. 3 reporting spin-dependent spectral functions.

Both the damping of quasiparticle states induced by $\mathrm{Im}[\Sigma_{n\mathbf{k}}(\omega)]$ and the energy renormalization due to $\mathrm{Re}[\Sigma_{n\mathbf{k}}(\omega)]$ are stronger for majority spin states; the effect of self-energy renormalization on majority-spin states close to E_f is an overall upward shift of the order of 0.8 eV while minority-spin states are almost unaffected, resulting in spectral functions that are rather similar for both spins, and thus in a rather weak spin polarization in photoemission spectra in this energy range. The spectral functions are in excellent agreement with spin-resolved normal emission spectra from 20 ML films of hcp Co on Cu(111) [24] and on W(110) [25]. In spectra measured at or near the same photon energy (21.2 eV) a relatively narrow peak is observed in the minority channel at binding energies of about 0.5 eV. The majority channel presents a broader peak at slightly higher (0.1–0.2 eV) binding energy with a high-binding-energy tail. Correlation effects reduce the separation between majority- and minority-spin structures close to E_f thus correcting the discrepancy between the exchange splitting measured in photoemission and the prediction of band theory.

In conclusion we have shown that the d-band occupation is a key parameter to quantify the effectiveness of many-body interactions and that the relatively large number of d holes in cobalt makes its photoemission spectrum incoherent over a large energy window below E_f. e-e correlations are responsible for strong spin-dependent energy renormalization close to E_f that affects the exchange splitting of quasiparticle states; this is expected to be a

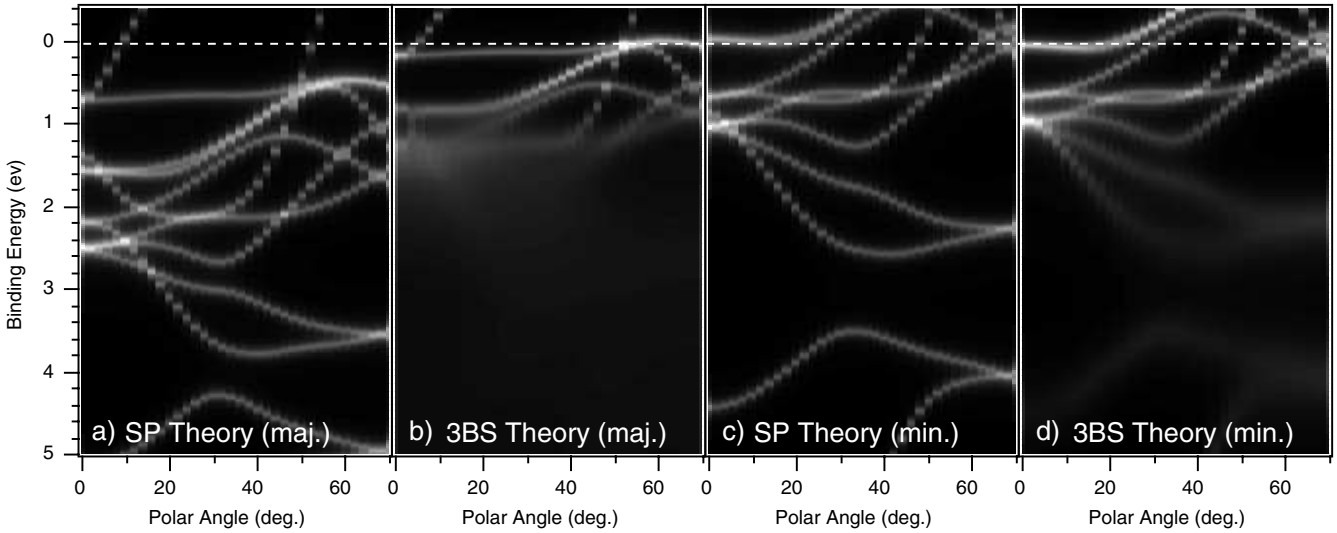

FIG. 3. Comparison of spin-resolved single-particle band structure and three-body scattering quasiparticle spectral function for the same wave-vector region as in Fig. 2. (a) and (b) represent majority-spin excitation, (c) and (d) minority-spin excitation.

general feature of all ferromagnets and indicates a profound interplay between magnetism and many-body effects and how these phenomena appear in photoemission spectra.

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