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Crossover from Negative to Positive Spin Polarization in the Photoyield from Ni(111) near Threshold

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The measured spin polarization of the photoelectrons emitted from Ni(111) shows a crossover from negative to positive values at 270 ± 90 meV above photothreshold. This is in constrast to the case of Ni(100), where a crossover at about 75 meV occurs. A single-particle photoemission calculation with an exchange splitting of 0.33 eV, and which does not invoke emission from surface states, is in good agreement with the experimental results for both faces. These new results corroborate the empirical band structure obtained by Eastman and co-workers from angle-resolved photoemission data.

In this Letter, we report measurements of the electron spin polarization (ESP) of photoemitted electrons from a well-characterized single-crystal Ni(111) surface for photon energies $\hbar\omega$ ranging from threshold φ to about 1 eV above φ. We find that the spin polarization changes from negative values (− 45% at threshold, i.e., preferential direction of the magnetic moment of the emitted electrons antiparallel to the magnetization) to positive values at an energy $E_c = 270 \pm 90$ meV above threshold. A photoemission calculation based on the Stoner-Wohlfarth-Slater (SWS) theory and using an exchange splitting of 0.33 eV is in good agreement with the experiment. This allows us to extract the value of the exchange splitting and the value of the Stoner gap for the ⟨111⟩ direction. In agreement with the interpretation of recent angle-resolved ultraviolet photoemission spectra[1] (ARUPS), surface-state emission is found to be not necessary to interprete the experimental ESP data for both (100)[2] and (111) surfaces studied to date in contrast to the suggestion by Dempsey and co-workers.[3]

Ni is accepted to be the prototype "strong" ferromagnet i.e., the majority-spin d band is full and the Fermi level crosses only the minority-spin d band. Primarily two related quantities characterize the electronic structure of Ni: the magnetic exchange splitting Δ and the "Stoner gap" δ, the latter being the energy differences

between the top of the majority-spin d band and the Fermi level. Self-consistent local-density calculations predict values of Δ at the top of the d band ranging from about 0.6 to 0.9 eV and values δ of about 0.4 to 0.55 eV.[4,5] Experimentally it has been very difficult to obtain information on those parameters. However, as discussed by Wohlfarth,[6] the ESP spectrum for Ni should show a rapid variation with photon energy close to threshold. In particular the SWS theory indicates that the polarization should cross over from negative to positive, the crossover energy E_c being sensitive to the value of δ. In fact, the ESP measurements of Eib and Alvarado on Ni(100),[2] previously mentioned, have been interpreted with $\Delta = 0.33$ eV and $\delta = 20$ meV at X_5, the top of the d bands along the Γ–X direction. Another method of determining Δ and δ is offered by angularly resolved photoemission spectroscopy, as first pointed out by Heimann and Neddermeyer[7] and by Dietz, Gerhardt, and Maetz.[8] Quite recently Eastman and co-workers[1] deduced a value of $\Delta = 0.31 \pm 0.05$ eV and of δ in the range 100–250 meV for the $\langle 111 \rangle$ direction. Evidently the experimental results determined from ARUPS[1] disagree with the self-consistent band-structure calculations.[4,5]

The Ni(111) single-crystal sample used in the present experiment is a cylinder of 3 mm diameter and 18 mm length, the shape being so chosen because of the favorable demagnetizing factor. It was spark cut and mechanically polished with a surface orientation better than 0.5°. The crystal was cleaned and characterized by a conventional procedure involving low-energy electron diffraction (LEED) and Auger tests in a preparation chamber attached to the main photoemission chamber through an ultrahigh-vacuum gate valve. The atomically clean sample is then placed into the photoemission chamber operating at a base pressure 1.5×10^{-10} Torr (with the gate valve to the Mott detector also open). An axial magnetic field of 2200 Oe is applied to the sample in order to align the Weiss domains. Photoelectrons are emitted by monochromatized light at a constant wavelength resolution $\Delta\lambda \approx 30$ Å at normal incidence (i.e., s-polarized light). The light source is a 500-W high-pressure Xe lamp and the monochromator is a 30-cm–focal-length Czerny-Turner type. Details of the electron optics and of the spin analyzer (Mott detector) have been previously described.[9] The count rates of the Mott detector with use of a gold foil with thickness of 220 μg/cm^2 are about 1 sec^{-1} at $h\nu \sim 5.2$ eV and about 20 sec^{-1} for $h\nu = 5.5$ eV. The present photocurrent intensities reaching the Mott detector are estimated to be at least an order of magnitude larger than previously reached in similar experiments.[2] We believe this to be primarily due to a significantly improved electron-optical design. The accumulation of the data in the available spectral range takes less than 2 h. This is of importance since we observed that contamination effects gave rise to changes of the ESP.

The apparative asymmetry was calibrated by use of an Al foil in the Mott detector (Al has a spin asymmetry ≈ 0). The error bars quoted in Fig. 1 represent the statistical error and the photon energy resolution. The photothreshold was determined from Fowler plots to be $\varphi = 5.2 \pm 0.07$ eV. Since the uncertainty of the work function also determines the accuracy of the crossover energy E_c, it is important to compare the present value with the values given in the literature for Ni(111). With use of photoemission techniques[1,10] the latter span the range 5.15–5.4 eV in agreement with our determination.

As a check before doing the Ni(111) experiment we have first measured the crossover energy of the (100) surface to $E_c < 100$ meV and thereby experimentally confirmed the data of Ref. 2. The detailed spectrum of the ESP or Ni(111) is presented in Fig. 1. The spin polarization of the photoyield is large and negative at threshold

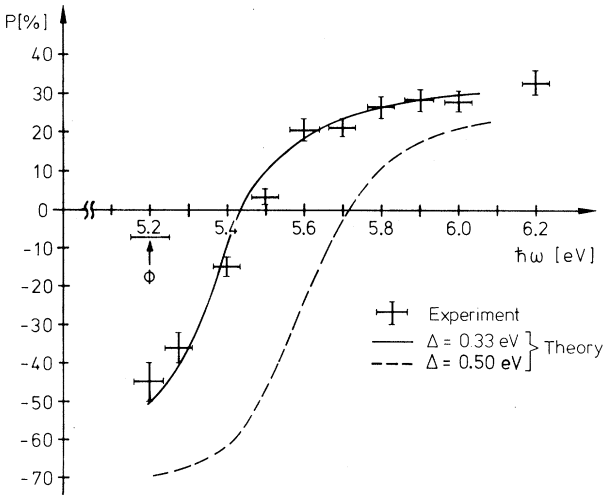

FIG. 1. Experimental and calculated electron spin polarization of the photoyield from Ni(111). The bars indicate the statistical error and the photon energy resolution of the experimental results. The values of the magnetic exchange splitting Δ used in the calculations are indicated. φ is the work function.

($\Phi \cong 5.2$ eV), changes sign at about $h\nu - \Phi \simeq 270$ meV, and reaches values of about +30% at about 1 eV above threshold.[11] The change in sign from negative to positive to positive spin polarization and the crossover point E_c are the crucial features of these data. As found in the calculations,[12] E_c is sensitive to the value of the Stoner gap δ (which is determined by Δ) along the $\langle 111 \rangle$ direction. For $h\nu \gtrsim \Phi$, one expects emission from the $\Lambda_3^{\downarrow}$ (minority-spin band) into evanescent LEED states to dominate. This determines the negative spin polarization. The $L_3^{\uparrow}$ majority-spin d-band edge has been experimentally located[1] between 50 and 250 meV below the Fermi energy. We note that in contrast to the suggestion of Himpsel and Eastman[13] the presence of a surface state of Λ_1 symmetry on the Ni(111) surface can be neglected within the present context because it does not contribute significantly to the photocurrent for s-polarized light.

In Fig. 1 we also present results of what can be called a rigorous single-particle calculation of the spin polarization of the photoyield from Ni(111). The theory, whose detailed results will be published elsewhere,[12] uses the same ingredients as previously used for the calculation of the spin polarization of the photoyield from the Ni(100) face.[14] We only mention here that the key parameters of the theory are being fixed as follows: The inverse lifetimes of the hole and of the hot electrons are taken to be 0.05 and 0.5 eV,[15] respectively, and the Fermi energy is determined by fitting to the experimental Fermi wave vector along the $\langle 111 \rangle$ direction. The only adjustable parameter in this theory is the exchange splitting Δ. For $\Delta = 0.33$ eV, which was obtained from the empirical fit to the Ni(100) spin-polarization data,[2,14] the *same* calculation can be shown to reproduce well the two-bump structure observed by Eastman and co-workers for off-normal emission near the Fermi energy in their angle-resolved photoemission studies (see Fig. 1 of Ref. 1). In Fig. 2 the results of a new calculation for Ni(100) (in Ref. 14, an inverse hot electron lifetime of 2.0 eV has been used) are presented along with a schematic representation of the experimental data of Ref. 2 for purpose of comparison. It is seen that the *same* parameter $\Delta = 0.33$ eV which fits well the spin-polarization data for Ni(100) also gives very good agreement with the data from the (111) face.

Despite the excellent agreement between single-particle calculations and experiments, a major question remains. Both angle-resolved uv photo-

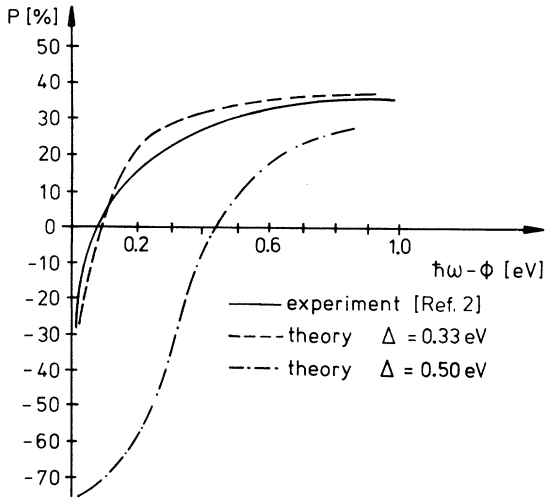

FIG. 2. Experimental (data from Ref. 2) and theoretical spin polarization of the photoyield from Ni(100). See also caption to Fig. 1.

emission studies and spin polarization measurements imply *independently* that the value of the exchange splitting of Ni is of the order of $\frac{1}{3}$ eV. This value is about $\frac{1}{2}$ as large as the one predicted by *ab initio* self-consistent local-density calculations[4,5] which seem otherwise to be in good agreement with other physical properties of Ni (e.g., Fermi-surface studies). Wang and Callaway[4] report that the Kohn-Sham-Gaspar exchange potential gives a splitting at X_5 of 0.88 eV and that the von Barth–Hedin local potential (which includes some correlation) gives a splitting of 0.63 eV. As is clear from Figs. 1 and 2, neither splitting is small enough to explain the ESP data. Why?

One answer to this question is that further refinements of the density functional are required. More specifically we expect that an improved description of correlation would lead to a reduction in Δ. Electron-gas calculations[16] show that correlation corrections in random-phase approximation and beyond lessen the spin dependence of the potential. Moreover, the local-density functional is not aware of the presence in Ni of a system of low-density holes at the top of the d band which provides an additional source of correlation, reducing Δ further.[17] Thus an effectively single-particle theory with a small exchange splitting can be justified.

An alternative answer has been provided by Anderson and by Edwards.[18] In it the potential acting on the hole created by the photon is irreduc-

ibly energy dependent and hence defies a single-particle description. Edwards and Hertz claim that the splitting appropriate to excitations in Ni is less than the band splitting (calculable in density-functional theory) because of electron-magnon interactions.[19] It is fair to conclude that more work on the importance of density functional and many-particle corrections in Ni is needed.

Experimentally work is in progress on the ESP spectrum of Ni(110) which we hope will provide more data on the empirical band structure. Measurements of the temperature dependence of the ESP spectrum of Ni and a study of Co are also planned for the near future.

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