spheroid would be much less symmetric about the central component of the band (which is common to both π- and σ-polarized radiation) than that computed for a prolate spheroid.

We believe that the discrepancy between computed and measured intensities shown in Fig. 3 is significant. The first-order Bragg reflection band is somewhat weaker, and the second order is stronger, than predicted. Altering the initial and final aximuth of the dielectric ellipsoid or its principal values only made the fit worse. Assuming an error in pitch measurement did not help either. Thin regions near the surfaces with anomalous dielectric properties might account for the discrepancy.

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PHOTOEMISSION STUDIES OF FERROMAGNETIC AND PARAMAGNETIC NICKEL*

D. T. Pierce and W. E. Spicer
Stanford Electronics Laboratories, Stanford University, Stanford, California 94302
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High-resolution photoemission spectra obtained from ferromagnetic nickel (295°K, $0.47T_C$) and paramagnetic nickel (678°K, $1.07T_C$) give no evidence of a change in the position of the d-electron peak near the high-energy cutoff of the photoelectron distribution within the experimental uncertainty of ±0.05 eV. On the other hand, a calculation of the spectra based on a band structure with an exchange splitting of 0.37 eV for ferromagnetic Ni and zero splitting for paramagnetic Ni predicted peak position changes of 0.1 to 0.2 eV.

We report the first high-resolution photoemission measurements above and below the Curie temperature of a ferromagnetic metal in the $3d$ transition series. An attempt was made to detect any differences between the electronic structure of ferromagnetic and paramagnetic Ni by obtaining photoemission spectra of 295°K where the magnetization of nickel is 95% of its maximum value and at 678°K where the magnetization is zero. Any effect of spin ordering on photoemission spectra would be most apparent for such a large difference in magnetization rather than for a small difference in magnetization at temperatures just above and just below the Curie temperature ($T_C = 631$°K). The strongest structure in the photoelectron energy-distribution curve (EDC) is a peak near the high-energy cutoff due to electrons originating in d bands near the Fermi energy E_F. The position of this peak was found to be the same at both 295 and 678°K within the experimental uncertainty of ±0.05 eV.

In contrast to the experimental results, our calculations of EDC's for ferromagnetic and paramagnetic Ni predicted 0.1- to 0.2-eV differences in the position of the main peak. The difference in energy between electron states of opposite spin is manifested in the exchange splitting of the spin-up and spin-down bands which has been estimated for Ni by Wohlfarth[1] as 0.35 ± 0.05 eV and by Phillips[2] as 0.5 ± 0.1 eV. We calculated photoemission energy-distribution curves using the interpolated energy bands of Hodges, Ehrenreich, and Lang[3] with an exchange splitting of 0.37 eV at E_F. We assumed the paramagnetic state could be represented by unsplit bands as is conventional in paramagnetic band calculations[4,5] and models of magnetism where the exchange splitting is proportional to the magnetization.[6,7]

The experimental results reported here were obtained from a sample which was prepared by electron-gun evaporating a Ni film ~1500-Å thick onto a single-crystal Ni substrate. The pressure rose from a base pressure of 1×10^{-11} Torr to 1×10^{-9} Torr during the initial stages of evaporation (2 min) but remained less than 5×10^{-10} Torr during the rest of the evaporation (17 min). Lack of surface contamination was established by the absence of a low-energy peak of scattered

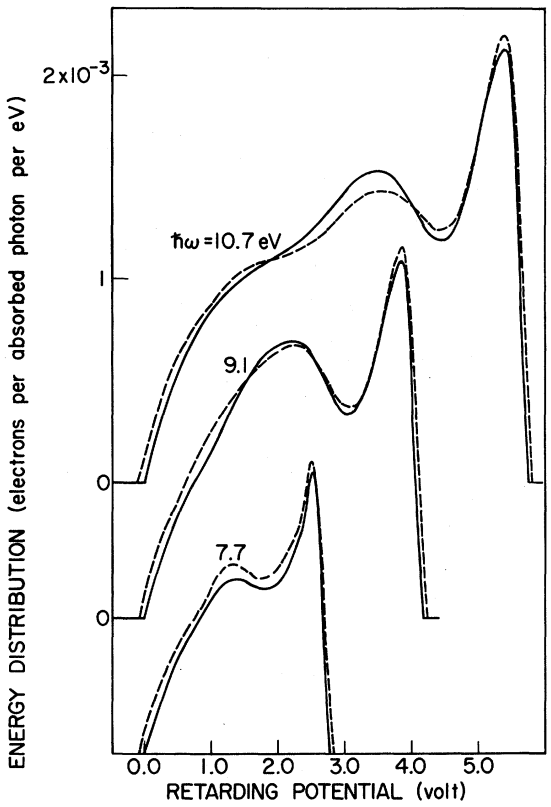


FIG. 1. Typical experimental EDC of photoemitted electrons from ferromagnetic nickel (295°K, full curves) and paramagnetic nickel (678°K, dashed curves) in the photon energy range 7.7 to 10.7 eV. The curves are plotted against the retarding potential applied to the collector.

electrons even at photon energies of 11.6 eV. The experimental results from this sample are in agreement with studies of two other different samples: (1) a heat-cleaned Ni single crystal, and (2) a Ni film electron-gun evaporated epitaxially onto a cleaved NaCl substrate.

Energy distributions of photoemitted electrons were measured at six to 15 photon energies over the range 7.7 to 10.7 eV. In Fig. 1 we show typical EDC's at 295°K ($0.47T_C$, full curve) and 678°K ($1.07T_C$, dashed curve). The curves at 295°K were taken after the curves at 678°K. In examining the EDC's we focus on the sharp peak near the high-energy cutoff for several reasons: (1) This is the sharpest available experimental structure; (2) it can be followed over the widest range of photon energy (more than 3 eV); (3) this peak is strong in all of the calculated EDC's; and (4) the peak corresponds to a well-defined peak in the density of states. As can be seen from Fig. 1, there is no change in the position of the leading peak in going from ferromagnetic to paramagnetic nickel. Thermal broadening of the Fermi surface is directly observed in an increase of 0.05 to 0.1 eV in the high-energy cutoff of the high-temperature EDC's.

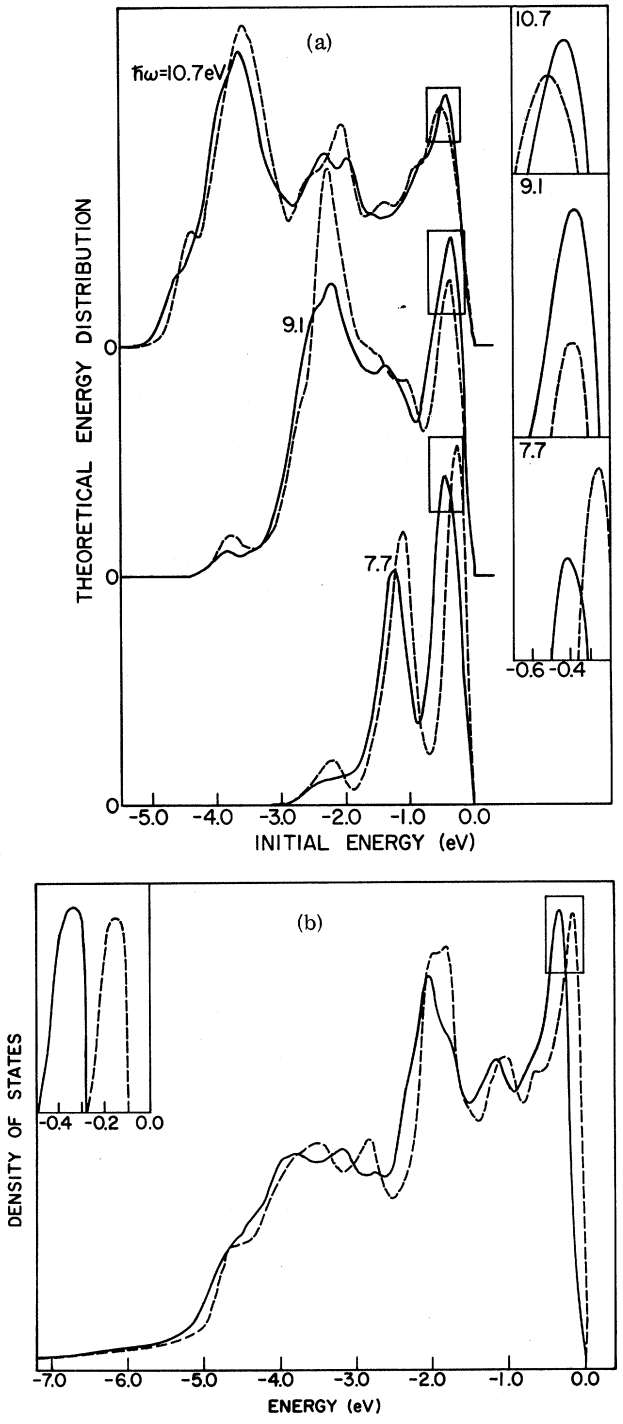
A new high-resolution energy analyzer which has a screened field-free drift region to improve spherical symmetry was developed for this experiment. Details of the operation of this screened emitter configuration are reported elsewhere.[8,9] The estimated maximum energy distortion due to the analyzer would shift a monoenergetic peak in an EDC to lower energy by an amount 1.4% of the electron's initial kinetic energy E_0 and broaden it to 2.8% E_0 full width at half-maximum. Due to the improved resolution, the d-electron peak near the high-energy cutoff of the EDC's was considerably sharper than in prior investigations.[10]

The uncertainty in comparing the position of a peak in EDC's at two different temperatures is not limited by shifts due to analyzer resolution since the analyzer geometry and ambient magnetic field remain the same at both temperatures. Rather, the limiting factor is the upper limit which can be set on the remote possibility of a change in the collector work function. The work function of the sample under study as determined from Fowler plots[11] was 4.87 ± 0.04 eV at 678°K and 4.97 ± 0.02 eV at 295°K. A corresponding shift of 0.1 eV appears in the trailing edge of the EDC. Taking into account the uncertainty of the sample work functions and the consistency of the intercepts of the leading and trailing edges of the EDC, we place an upper limit of ± 0.05 eV on the uncertainty in comparing the position of the leading peak in high-temperature and room-temperature EDC's. If all work functions were known exactly, a shift in the peak position about 0.01 eV could be detected.

In order to predict what changes might be searched for in photoemission spectra obtained above T_C, we made calculations of EDC's for nickel assuming direct transitions with constant matrix elements similar to the calculations for copper by Smith.[12] The interpolated band structure was sampled at over 10^5 points in the 1/48 of the Brillouin zone. The Fermi function was used in order to account for the differences in thermal smearing at 295 and 678°K. The calculated energy distributions of photoexcited electrons were convolved with a reasonable threshold function[13] to yield the energy distributions of photoemitted electrons. In order to compare better with the experiment, these in turn were con-

volved with a broadening function[14] to give the EDC's displayed in Fig. 2(a). The main features of the calculated EDC's are in qualitative agreement with the experimental curves throughout the energy range investigated.

One can see a number of significant changes in the calculated EDC's above and below the Curie temperature. As mentioned earlier, however, we will concentrate on the changes in the leading peak since this peak appears in all of the calculated and measured curves. In the calculated EDC of Fig. 2(a) the leading peak of the paramagnetic nickel curve nearly lines up in energy with the peak of the ferromagnetic nickel curve at a photon energy $\hbar\omega = 9.1$ eV. But the leading peak in the calculated paramagnetic curve is shifted 0.16 eV higher in energy for $\hbar\omega = 7.7$ eV and 0.09 eV lower in energy for $\hbar\omega = 10.7$ eV.



Significant shifts in the position of the leading peak would also be predicted by calculations of EDC's based on the nondirect transition model. Although it has recently been questioned,[12,15-17] the nondirect model successfully explains much previous work.[10,13,18,19] In the nondirect model (where $\vec{k}$ conservation is not taken to be an important selection rule) it is found that in the absence of structure in the final density of states experimental EDC's reflect the optical density of states.[19] The optical density of states obtained from analysis of the photoemission data on the nondirect model has been found to be quite similar to the band density of states in previous studies of noble and transition metals.[13,18] On the basis of this model we would expect changes in the band density-of-states curves, such as those in Fig. 2(b) from Hodges, Ehrenreich, and Lang,[3] to indicate expected changes in experimental EDC's in going from ferromagnetic to paramagnetic nickel. In order to compare better with the experiment, the calculated band density-of-states curves have been convolved with a broadening function.[14] From Fig. 2(b) we would expect the position of the leading peak in an EDC to change by 0.18 eV.

On the assumption of a band structure in which the exchange splitting of the bands goes to zero at T_C, we have predicted changes in the position of the leading peak in the EDC's corresponding to changes in the energy bands near the Fermi level. The changes, if they occurred, would be

FIG. 2. Expected shifts in the position of the leading peak of the EDC's in going from ferromagnetic nickel (full curves) to paramagnetic nickel (dashed curves) calculated assuming (a) direct transitions between interpolated bands and (b) the nondirect model of the optical excitation process. In the ferromagnetic case the spin-up and spin-down bands are split 0.37 eV at the Fermi level. The inserts, which are 3× magnifications of the leading peaks, show shifts in position that would be clearly observable with our experimental uncertainty of ±0.05 eV. The zero in energy of the curves is taken at the Fermi level.

clearly observable with our experimental uncertainty of ±0.05 eV. None of these changes were detected. The absence of peak position changes in a previous photoemission study of Gd[20] is not so surprising since the magnetic moment is carried principally by the highly localized unpaired $4f$ electrons. In nickel, however, where the moment is carried by the itinerant d electrons, the absence of EDC peak-position changes calls into question the assumption that the exchange splitting vanishes above the Curie temperature. An explanation of our experimental results may be found in a model discussed by Harrison.[21] In this qualitative picture, increasing the temperature above T_C causes disordering of the local moments formed from resonant states on each atom, and changes in EDC's would only be expected to be of the order of $k_B T_C$, i.e., they would lie at the limit of our experimental resolution. The apparent conflict between our results and the results of a neutron-scattering[22] and optical-absorption[23] experiments is recognized and will be discussed in a more complete report of this work.

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