(10) has to be kept.

[13]In some recent papers, the interference effect and its strong energy dependence seem to be underestimated in spite of the CL theory. See, e. g., W. J. Pardee, G. D. Mahan, D. E. Eastman, R. A. Pollak, L. Ley, F. R. McFeely, S. P. Kowalczyk, and D. A. Shirley, Phys. Rev. B 11, 3614 (1975), and D. R. Penn, Phys. Rev. Lett. 38, 1429 (1977), where the role of intrinsic versus extrinsic plasmon production is discussed, but where the experimental data do not mention the photoelectron or exciting radiation energy.

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# Experimental Exchange-Split Energy-Band Dispersions for Fe, Co, and Ni

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(Received 24 July 1979)

Using angle-resolved photoemission with single-crystal Fe(111) and Co(0001), we have determined accurate exchange-split energy-band dispersions $E(\vec{k})$ along symmetry lines. For Fe, Co, and Ni, respectively, occupied $d$-band widths are 3.1 eV ($P$), 3.8 eV ($L$), and 3.4 eV ($L$,$X$) while exchange splittings are ~ 1.5 eV ($P$), ~ 1.1 eV ($\Gamma$), and 0.3 eV (near $L$). Comparison with theory shows that state-of-the-art *ab initio* calculations describe the strong ferromagnets Fe and Co much better than Ni.

A long-standing goal has been the determination of accurate exchange-split energy-band dispersions for the itinerant-electron ferromagnets Fe, Co, and Ni.[1-14] As often noted,[2,4] magnetic exchange splittings and energy-band dispersions $E(\vec{k})$ are fundamental for understanding various physical properties involving $d$-band electronic structure,[1-4] collective itinerant-electron ferromagnetism,[5,6] transition-metal surfaces, etc. Recently, the temperature-dependent exchange splitting and energy-band dispersions for Ni have been determined.[13,14] For Fe and Co, however, exchange splittings and energy-band dispersions remain inconclusive or absent despite several angle-resolved photoemission studies[9-12] and other ultraviolet and x-ray photoemission studies.[7,8] Rather, contradictory conclusions have been reached, i.e., (i) that angle-resolved photoemission spectra measure a density of states[12] [nondirect-transition model, in which case $E(\vec{k})$ dispersions cannot be determined] and (ii) that direct transitions (with band dispersions[10]) are sometimes of primary importance but also more complicated effects, e.g., surface/polarization effects are sometimes predominant.[9-11]

In this paper, we show that direct transitions are of primary importance for normal photoemission from Fe(111) and Co(0001) and determine accurate exchange-split energy-band dispersions $E(\vec{k})$ for bcc Fe along the $\Gamma$–$P$–$H$ symmetry line and for hcp Co along the $\Gamma$–$A$–$\Gamma$ line. These results together with experimental $E(\vec{k})$ dispersions for Ni (Refs. 13 and 14) and Cu (Ref. 15) permit us to systematically compared with state-of-the-art *ab initio* band calculations.[1] This comparison shows that the ratio of the theoretical to experimental occupied $d$-band width is about 1.1, 1.2, 1.45, and 1.1 for Fe, Co, Ni, and Cu, respectively, while the ratio of the theoretical to experimental exchange splitting $\delta E_{ex}$ is about 1.0, 1.2, and 2.2 for Fe, Co, and Ni, respectively. General conclusions are as follows: (1) Fe, Co, and Ni can all be described by a Stoner-Wohlfarth-Slater (SWS) spin-split energy-band model, and (2) state-of-the-art *ab initio* one-electron band calculations describe Fe quite well even though it has a large exchange splitting and high Curie temperature (strong ferromagnet), in marked contrast with Ni.

Experimentally, we have used a display-type electron spectrometer[14,15] and an ultrahigh-vacuum toroidal-grating monochromator together with radiation from the 240-MeV storage ring at the University of Wisconsin–Madison. This system was used for this study in the energy range $5 \leq h\nu \leq 30$ eV with an overall energy resolution of ~100–150 MeV and angular resolution of $\delta\theta = 4°$ (full angle). Fe(111), Fe(100), and Co(0001) crystals were prepared by Ar-ion etching and annealing, checked with use of Auger spectroscopy and low-energy electron diffraction, and measured in working vacuums in the $10^{-11}$-Torr range.

In Fig. 1 we present selected angle-resolved energy distribution curves (AREDC's) for normal emission for Fe(111), Co(0001), and Ni(111). The pronounced spectral changes with photon energy $h\nu$ indicate that direct interband transitions are of primary importance. Consequently, these normal-emission spectra directly probe electron-

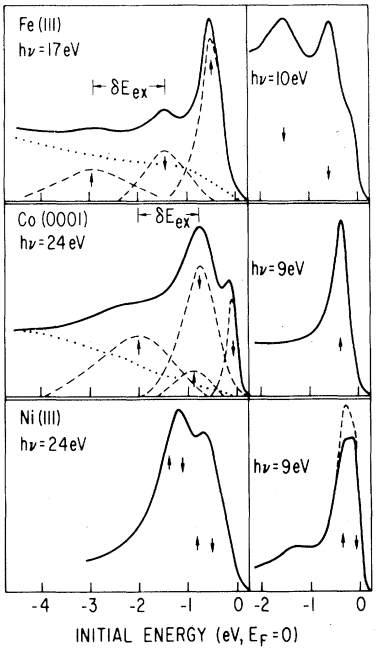


FIG. 1. Normal-emission spectra for Fe(111), Co(0001), and Ni(111) with use of $s$-polarized radiation. Arrows indicate spectral peak locations while exchange splittings for Fe and Co are indicated (see text).

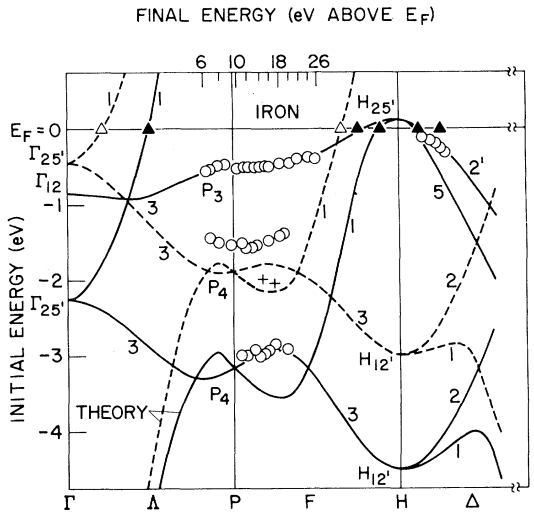


FIG. 2. Experimental $E(k_\perp)$ dispersions for Fe along $\Gamma$–$P$–$H$ ($\Lambda$ direction) and near $H$ along the $\Delta$ direction. Only $\Lambda_3/F_3$ symmetry bands are seen for normal emission with $s$ polarization. (The crosses denote weak features.) Solid and dashed lines denote the majority and minority bands calculated by Callaway and Wang[2] while the solid and open triangles denote Fermi-surface crossings determined from de Haas–van Alphen data.

ic states along the symmetry line perpendicular to the surface (the crystal momentum parallel to the surface $\vec{k}_\parallel = 0$ with the crystal momentum perpendicular to the surface $k_\perp$ controlled by $h\nu$ together with the final-state dispersions $E(k_\perp)$,[13,14] which we have determined to be simple (i.e., nearly free-electron-like) for our experimental conditions.[14]

From extensive data with both $s$- and mixed $s/p$-polarized radiation in the range $5 \lesssim h\nu \lesssim 30$ eV, we have determined experimental exchange-split energy-band dispersions $E(\vec{k})$ for Fe as shown in Fig. 2 and for Co and Ni as shown in Fig. 3. In Fig. 2, the solid and dashed lines denote the majority-spin and minority-spin bands as given by the *ab initio* calculation by Callaway and Wang[2] (von Barth–Hedin local exchange-correlation potential) while the full and open triangles denote Fermi surface crossings of majority and minority bands,[2] respectively, as determined from de Haas–van Alphen data. The energy scales at the top of Figs. 2 and 3 give the final-state energies corresponding to the $k_\perp$ scale. As previously done for Ni,[13,14] we have used experimental conditions for Fe(111) and Co(0001) such that there is one nearly-free-electron-like final state of primary importance.[16] Values of $k_\perp$ are estimated to be accurate to ~5% of $\Gamma$–$P$–$H$ for Fe(111) and of $\Gamma$–$A$–$\Gamma$ ($\equiv \Gamma$–$L$) for Co(0001).

In Fig. 3 we have plotted the energy bands of Co along the (0001) direction using a fcc Brillouin-zone (BZ) diagram. Symmetry labels for both hcp and fcc BZ's ([111] direction) are given. The line $\Gamma\Delta A\Delta\Gamma$ in the hcp BZ corresponds to the line $\Gamma\Lambda L$ in the fcc BZ. States at $\Gamma$ in the hcp BZ correspond to states at $\Gamma$ or $L$ in the fcc BZ because of an extra hcp lattice vector. However, if the hcp bands are folded back about $A$ in Fig. 3 as is usually done for hcp bands, dipole selection rules forbid transitions from initial states of one kind of symmetry to final state of the other kind, i.e., from initial states at $L$ to final states at $\Gamma$ and vice versa in the corresponding fcc BZ (to be discussed in a separate publication). Therefore, transitions for normal emission from hcp Co(0001) can be classified in terms of a fcc BZ; in fact, as seen in Fig. 3, the bands for Co(0001) show an overall similarity with those of Ni(111) when plotted in this manner.

Using the $E(\vec{k})$ dispersions in Figs. 2 and 3, one can readily describe the nature of the spectral peaks in Fig. 1. For Fe(111) with $s$ polarization and normal emission, only transitions of $\Lambda_3/F_3$ symmetry along the $\Gamma$–$P$–$H$ line are allowed by

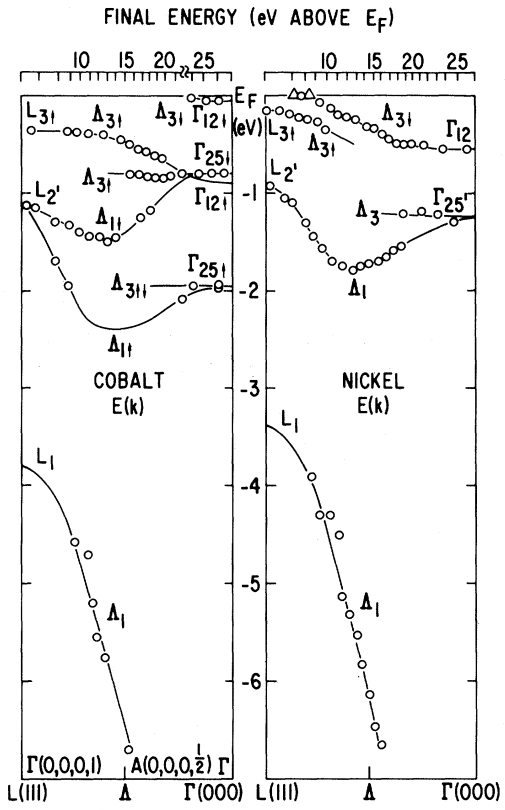


FIG. 3. Experimental $E(k_\perp)$ dispersions for Co(0001) along $\Gamma$–$A$–$\Gamma$ (equivalently $\Gamma$–$L$) and for Ni(111) along $\Gamma$–$L$. The solid lines denote band dispersions based on the experimental data together with shapes based on theoretical bands (Ref. 1).

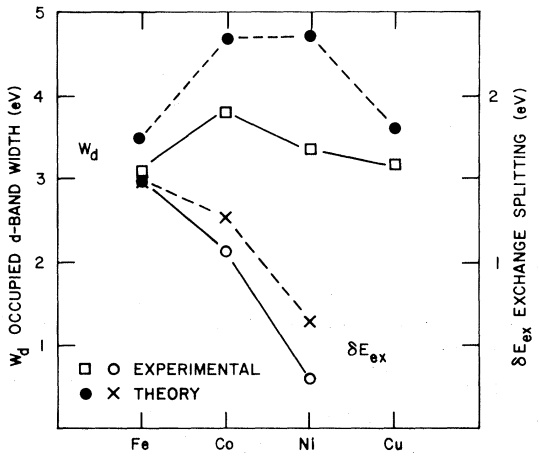


FIG. 4. Occupied $d$-band widths (eV) and magnetic exchange splittings (293 K). The widths are at the symmetry points $P$, $L$, $L$, and $X$, and at $X$ for Fe, Co, Ni, and Cu, respectively, while the exchange splittings are at $P$, $\Gamma$, and near $L$ for Fe, Co, and Ni, respectively. Theoretical widths are taken from Ref. 1. For Co, experimental exchange splittings are 0.9 ($\Gamma_{12}$) and 1.2 ($\Gamma_{25'}$) eV. An average value is given in Fig. 4. Experimental accuracies are about ± 0.2 eV.

symmetry selection rules.[17] As seen from Fig. 2, there are three such bands; in Fig. 1, the upper two are seen for $h\nu = 10$ eV and all three are seen for $h\nu = 17$ eV.[18] For $h\nu = 17$ eV, the lower two peaks correspond to the lower spin-split $\Lambda_3/F_3$ symmetry bands near $P$ and show an exchange splitting $\delta E_{ex} \simeq 1.5$ eV. These two exchange-split peaks have estimated widths of ≃1 and 1.7 eV, respectively, with the lower-energy peak having increased broadening as expected. We have also measured off-normal-emission data for Fe(111) and find steeper spin-split band dispersions perpendicular to the $\Lambda$ axis near $P$, which are in good agreement with the calculated dispersions.[1,2]

We have studied the temperature dependence of the exchange splitting for Fe and find a decrease in $\delta E_{ex}$ of about 0.3 eV from 293 to 973 K ($T_c$ = 1043 K). This reduction occurs with the minority-spin bands shifting to lower energy about 2–3 times as rapidly as the majority-spin bands shift to higher energy. These shifts, which are opposite to those seen for Ni,[13] are consistent with the band picture of Fe.

Fe Co(0001) and Ni(111) with $s$ polarization and normal emission, only bands of $\Lambda_3$ symmetry can be excited.[17] At $h\nu = 9$ eV, one sharp peak is seen for Co at $E_i = -0.35$ eV that corresponds to the $\Lambda_{3\downarrow}$ band at $k_\perp \simeq 0.75$ the distance from $\Gamma$ to $L$. At $h\nu = 24$ eV, the three resolved peaks for Co correspond to $\Lambda_3$-band transitions at $k_\perp \simeq 0.2$ with energies nearly equal to the $\Gamma_{12\downarrow}$, $\Gamma_{25'\downarrow}$ and $\Gamma_{25'\downarrow}$ critical-point energies, respectively.

For Co, the band ordering at $L$ is inverted from that predicted,[1] with $L_{3\downarrow}$ being well above $L_2'$. The $L_{3\downarrow}$ critical point at $E_i = -0.35$ eV corresponds to the Stoner gap and lies nearer $E_F$ than predicted[1] ( e.g., −0.53 eV). As seen in Fig. 3, a small portion of the upper $\Lambda_{3\downarrow}$ band lies just below $E_F$ near $\Gamma$, resulting in an electron pocket with a minimum at $\Gamma_{12\downarrow}$ (~ −0.05 eV). This feature has not been identified in previous band calculations (which have had too large exchange splittings[1,3]) but would appear to account for observed de Haas–van Alphen frequencies.[3]

Comparisons of experimental occupied $d$-band widths and exchange splittings at selected ($\vec{k}$) points with state-of-the-art calculations is given in Fig. 4. Experimental values for Fe, Co, and

Ni are taken from Figs. 2 and 3. Similar $E(\vec{k})$ data has been obtained for Cu (Ref. 15) and is given for comparison. Theoretical values in Fig. 4 are those of Moruzzi, Williams, and Janak, who have performed *ab initio* self-consistent Kohn-Korringa-Rostoker calculations using the local-density theory of electronic exchange and correlation. In summary, Ni and Co (to a lesser extent) show significant deviations from state-of-the-art ground-state calculations.[1-4] Using the experimental exchange splittings $\delta E_{ex}$ in Fig. 4 and Bohr-magneton data ($\mu_s = 2.1\mu_B$, and $0.55\mu_B$ for Fe, Co, and Ni, respectively), we observe that a phenomenological measure[19] of the intra-atomic exchange interaction $U^{d-d}$, namely $U^{d-d} \simeq \delta E_{ex}/\mu_s$, is approximately constant ($U^{d-d} \simeq 0.6$–$0.7$) for Fe, Co, and Ni. This value of $U^{d-d}$ is smaller ($\lesssim \frac{1}{2}$) than a previous estimate based on electron-energy-loss and soft-x-ray spectra.[19]

We wish to acknowledge A. Ignatiev for supplying the Co crystal, the excellent support of J. J. Donelon and A. Marx, and the staff of the Synchrotron Radiation Center. This work was supported in part by the U. S. Air Force Office of Scientific Research under Contract No. F44620-76-C-0041.

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## Thermodynamic and Resistive Transitions of Thin Superconducting Films

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(Received 13 February 1979)

In amorphous, nongranular, thin films of $Nb_3Ge$, the temperature dependence of the resistance and the current-voltage relation have been investigated at temperatures below the experimentally determined thermodynamic transition. This temperature-dependent resistance is compared to several theories which predict flux-flow resistance below the BCS transition temperature due to thermally excited vortex pairs.

In this Letter we report measurements of the temperature dependence of the electrical resistance and heat capacity of amorphous, nongranular, $Nb_3Ge$ films deposited on thin polished-sapphire substrates. The heat capacities were measured with an ac technique.[1] We have found the