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Low-energy interband absorption in bcc Fe and hcp Co

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We have examined the electronic structure of bcc Fe and single-crystal hcp Co by using optical absorptivity and thermoreflectance techniques for $0.2 \leq h\nu \leq 5$ eV. The optical conductivities σ were calculated by Kramers-Kronig analyses. A prominent structure was observed in σ for Fe at 2.37 eV and a shoulder was observed near 0.8 eV; the latter structure was the dominant feature in the thermoreflectance spectrum. These were discussed in terms of minority-spin band interband absorption and spin-flip interband transitions. The anisotropic optical conductivities of hcp Co were discussed in terms of recent energy-band calculations.

INTRODUCTION

The electronic structure of the ferromagnetic transition metals is generally less well understood than those of the nonmagnetic transition metals. Their optical spectra are less structured, in the case of Fe and Ni, or not known for single crystals, in the case of hcp Co. In the following we present the first optical spectra for single-crystal Co that reveal several anisotropic interband structures below 5 eV. We also present thermoreflectance data for Fe that identify a low-energy interband structure that had been unobserved previously but that had been predicted by several recent band calculations. Our intent is to improve the incompletely-understood connection between the optical properties and the electronic structure, and our emphasis is on the low-energy optical behavior (0.2–5 eV) as revealed through absorptivity measurements at 4.2 K and thermoreflectance measurements at 140 K. An accurate first-principles model of the electronic structure of these metals is indispensible if we are to understand their diverse properties. Great progress[1-9] has been made, but such fundamental quantities as the exchange splitting $\delta\epsilon_{ex}$ continue to be controversial.

Studies of the optical properties are important in the process of formulating a model of the electronic structure. The determination of the dielectric function $\tilde{\epsilon}(\omega)=\epsilon_1(\omega)+i\epsilon_2(\omega)$ is the primary goal of an experimental optical study since ϵ_2 carries information about the photon-induced electric dipole transitions from an occupied initial state $|i\rangle$ to an empty final state $|f\rangle$, each with wave vector k. ϵ_2 can be calculated from a first-principles model of the band structure and comparisons made between the predictions of the model and the experimental results.

The importance of optical studies has, of course, been known for some time and both Fe and Co have been studied by several other groups. For Fe Moravec *et al.*[10] measured the reflectance at near-normal incidence at 300 K and determined the dielectric function between about 3 and 27 eV. Jones *et al.*[11] investigated the infrared optical properties, and Yolken and Kruger[12] determined the optical constants in the visible. Blodgett and Spicer[13] included reflectance measurements as part of their photoelectron spectroscopy (PES) studies of Fe. Johnson and Christy[14] determined the dielectric function between 0.6 and 6.4 eV. Bolotin *et al.*[15] reported ellipsometric measurements. From these studies, it was clear that Fe had strong interband structure about 2.4 eV, but that a precise k-space interpretation of that broad structure was not possible because the band structure of ferromagnetic Fe offered too many possible interband transitions at that energy. If optical studies were to be helpful in understanding the electronic structure of Fe, then, further information must be wrung from the 2.4-eV feature or additional infrared structure must be identified, structure which could be related to bands in a particular part of the Brillouin zone involving either minority-spin bands or bands of mixed spin. This was part of the motivation of this study.

Optical studies of Co have been relatively few.

Johnson and Christy[14] and Yu *et al.*[16] studied thin films while Kirillova and Charikov[17] studied polycrystalline samples. The work with the films showed only weak, broad structure in the dielectric function. The work on polycrystals revealed additional interband structure, but gave no information about the anisotropy of the optical properties and dielectric function. We felt that a careful study of the optical anisotropy of this interesting metal was needed.

This paper stems from our on-going interest in the transition metals, their optical properties, and their electronic structure. In the following sections, we briefly discuss the experimental techniques employed in our efforts, present the data, discuss the procedures used in extracting the dielectric function, and interpret the results in terms of recent band calculations. [For additional insight into the electronic structure of Fe and Co, the reader is referred to recent PES and angle-resolved PES (ARPES) studies.[18]]

EXPERIMENTAL TECHNIQUES AND RESULTS

Two different experimental methods were used in these studies. A calorimetric technique was used to determine the normal-incidence absorptivity spectra $A(h\nu)$ ($A = 1 - R$, where R is the reflectance) at 4.2 K for $0.2 \leq h\nu \leq 4.4$ eV. This technique has been discussed in detail elsewhere[19]; it has been used in earlier studies of other metals and alloys (transition metals, lanthanides, actinides).[20-22] It is conceptually quite straightforward: monochromatic radiation (polarized for Co) is focused onto a sample at near-normal incidence so that part of the beam is absorbed by the sample and part is reflected onto an absorber which is coated with Au-black and which absorbs all of the incident radiation. The experimental chamber is immersed in liquid helium so that both sample and absorber have low heat capacities. The absorbed radiation (~ microwatts) produces measurable temperature increases (mK); Joule heating with matched metal film resistors attached to the two elements is used to duplicate those increases. This technique is particularly suited to (but not limited to) infrared studies of metals because their reflectivities (absorptivities) are large (small) and A can be determined with an accuracy of ~1% of A. This accuracy (e.g., $A = 0.1068 \pm 0.001$ for Fe at 0.3 eV) is equivalent to measuring the reflectance to about 0.1%. Hence, calorimetric absorptivity measurements have much greater sensitivity for infrared structure than can be expected for reflectance measurements. For further details about the technique, see Ref. 19.

An ac modulation technique was used to determine the temperature dependence of the reflectance of Fe at 140 K. In these thermoreflectance measurements, the temperature of the sample was cyclically varied and the small ac component of the reflectance was determined. A thin Fe crystal was attached to a thin-film heater (evaporated Cr on sapphire) and was thermally driven at 2 Hz by square-wave heating pulses. This technique is not a new one, but it has been used infrequently in studies of metals. It offers the principal advantage that interband transitions which involve bands cut by E_F or bands which have a large deformation potential are emphasized more than others. For example, a Fermi-surface transition has the temperature dependence of the Fermi function and a characteristic line shape. This selective enhancement may make it possible to resolve a complicated structure into two or more components or distinguish a critical point transition from a Fermi-surface transition. On the other hand, if large volumes of k space contribute to a feature in ϵ_2, it may be that the temperature dependence of that feature is nondescript or defies simple line-shape analysis. In that case, thermoreflectance results may be disappointingly uninformative. Details of the thermoreflectance technique can be found elsewhere.[23,24]

Bulk samples were used in both kinds of measurements employed in these studies. A high-purity polycrystalline rod of Fe was sectioned with a diamond saw. The sample was mechanically polished through 1 μm alumina abrasive to obtain a specular surface, then annealed for strain relief. Finally, it was electropolished[25] immediately before either set of measurements and then quickly transferred to the respective vacuum system. The cobalt single crystal was generously loaned to us by Ignatiev[26] and had previously been used in low-energy electron-diffraction studies of Co. Like Fe, it was electropolished[25] immediately before study.

The results of the absorptivity studies for Fe are shown in Fig. 1. Also shown in Fig. 1 are the results of Moravec *et al.*[10] to indicate the excellent agreement in the region of overlap (e.g., Moravec *et al.* reported $R \simeq 0.59$ at 3 eV and we measured 0.586). Comparison with other existing optical data for Fe shows general agreement concerning the absorptivity maximum at 2.05 eV; our curve shows the feature more clearly defined than does that of Johnson and Christy[14] presumably because they studied a vacuum deposited film; our results are comparable to those of Yolken and Kruger[12] in the energy range of their measurements.

The absorptivity spectra for hcp Co are shown in Fig. 2. The orientation of the electric field

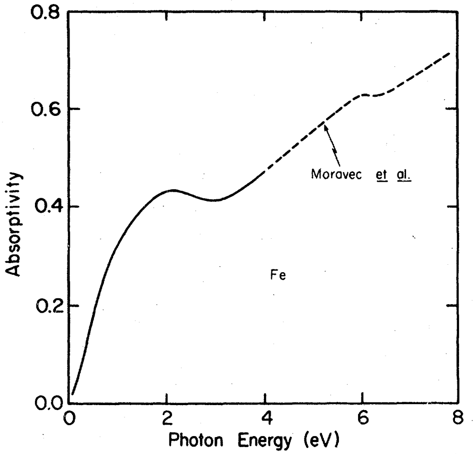

FIG. 1. Optical absorptivity of polycrystalline Fe at near-normal incidence and 4.2 K. The data above 4 eV are those of Moravec *et al.* (Ref. 10). The absorptivity spectrum below ~2 eV displays only a hint of structure near 1 eV.

vector $\vec{E}$ relative to the c axis of the crystal is indicated by $\vec{E}\|\hat{c}$ (dashed curve) or $E\perp\hat{c}$ (solid line). Comparison of our results with those of Yu *et al.*[16] or Johnson and Christy[14] shows remarkably good overall agreement in magnitude above about 2 eV—better agreement, in fact, than is usually found in the literature. At lower energy, our spectra reveal anisotropic features that could only be observed with single-crystal samples. Comparison with the results of Kirillova and Charikov[17] reveals that their absorptivity magnitudes were lower than those of the others.

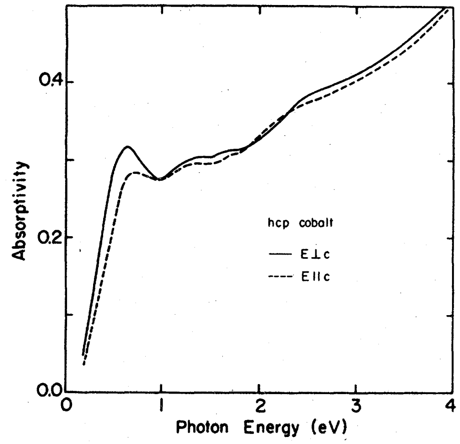

FIG. 2. Optical absorptivity of single-crystal hcp Co at 4.2 K. The orientation of the electric field vector relative to the c axis of the crystal is given by $\vec{E}\|\hat{c}$ or $\vec{E}\perp\hat{c}$.

The results for Fe shown in Fig. 1 indicate that the absorptivity climbs smoothly from 0.0610 at 0.2 eV to a maximum value of 0.433 at 2.05 eV with only a suggestion of a shoulder near 0.9 eV. Above the maximum at 2.05 eV, the absorptivity displays a minimum at 3.0 eV and a structure at about 6.1 eV (as shown in the data of Moravec *et al.*). In effect, the absorptivity results indicate that optical transitions have a low-energy onset below 0.2 eV (the data of Ref. 11 indicate the onset to be below 0.1 eV) and numerous interband features overlap at higher energy–precluding the chance of sharp structure in the dielectric function. This absence of sharp structure is rather typical for transition metals, but it is usually not quite so severe.

The absorptivity spectra for Co are more structured than that of Fe. The interband onset again occurs below 0.2 eV (the absorptivity is increasing sharply at 0.2 eV indicating strong interband absorption). For $\vec{E}\|\hat{c}$, the infrared maximum in A occurs at 0.71 eV and additional features are observed at about 1.30, 1.70, and 2.3 eV (the exact energies are difficult to determine because the features are broad). For $\vec{E}\perp\hat{c}$, the low-energy maximum in A occurs at 0.63 eV and is followed by structure at 1.35, 1.65, and 2.5 eV. Above about 3 eV, the spectra for $\vec{E}\|\hat{c}$ and $\vec{E}\perp\hat{c}$ run nearly parallel. At 3.5 eV, the weighted average of our absorptivity spectra is 0.46; Johnson and Christy measured about 0.47 and Yu *et al.* reported about the same for Co films (data extracted from journal-sized figure for the latter).

The interpretation of optical data is better made through considerations of structure in ϵ_2 than through A or R. This is because the absorptivity or reflectance is a complicated function of the dielectric constants, viz.,

$$R=[(n-1)^2+k^2]/[n+1)^2+k^2]$$

and $\epsilon_2=2nk$ and $\epsilon_1=n^2-k^2$. It is ϵ_2 which is the measure of dipole absorption strength, and it is ϵ_2 which can be calculated from a microscopic model of the bands.

The absorptivity spectra were Kramers-Kronig (KK) analyzed to obtain the dielectric functions. The Fe results were extended to 30 eV (200 eV) by using the published $R(E)$ data of Moravec *et al.* (the absorption-coefficient results of Sonntag *et al.*[27]); those for Co were extended to 30 eV with the unpublished reflectance data of Olson[28] (polycrystalline Co), then to 200 eV with Sonntag's results. Further, Johnson and Christy[14] determined $\tilde{\epsilon}$ through analysis of thin film transmission and reflectance data; our extrapolation above 200 eV, which was of the form $R=R_0E^{-\beta}$, could then be

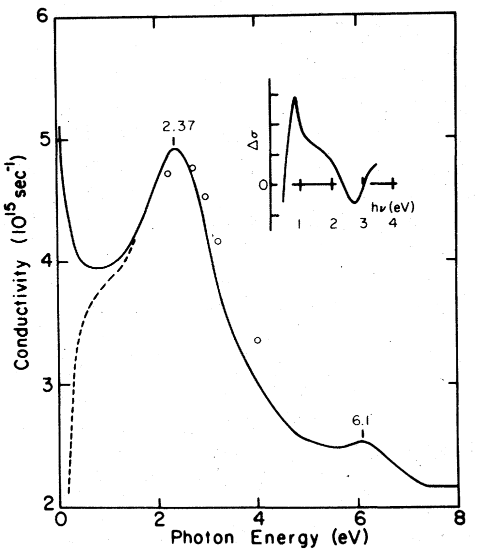

FIG. 3. Optical conductivity of Fe derived from the data of Fig. 1. The open circles represent the results of Johnson and Christy (Ref. 14). Note that the ordinate does not extend to zero. The dashed curve represents a qualitative estimate of the interband conductivity (see text). The inset shows the differential conductivity obtained from the results of Fig. 5 and shows the temperature dependence of the structure near 0.8 eV to be greater than that of the higher-energy interband structures.

adjusted to bring our magnitudes of $\tilde{\epsilon}$ into agreement with their results. Finally, Drude forms were assumed for the infrared behavior, and the Drude parameters were adjusted so that the extrapolation matched experiment in magnitude and slope.

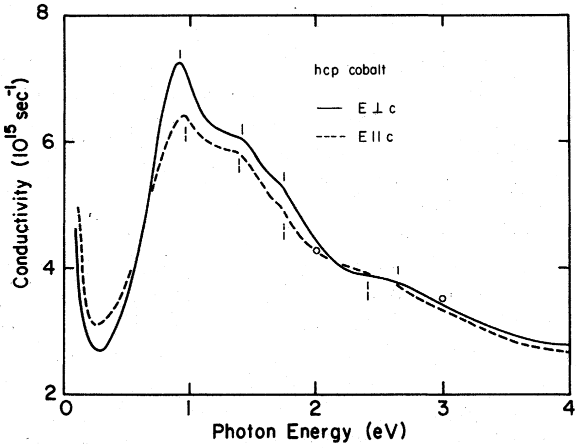

FIG. 4. Optical conductivity of hcp Co derived from the data of Fig. 2. The open circles represent the results of Ref. 14.

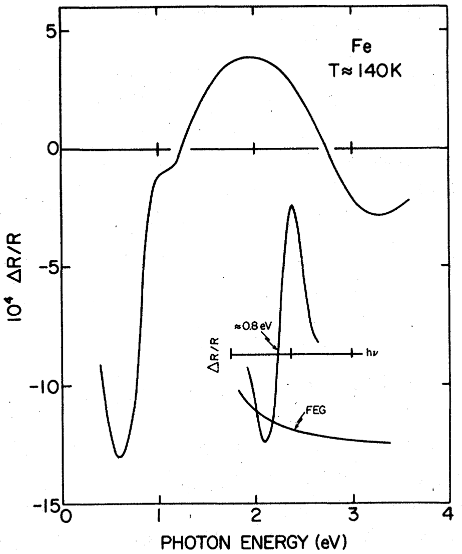

FIG. 5. Thermoreflectance spectrum of bulk Fe at about 140 K. The insert shows a qualitative deconvolution of the $\Delta R/R$ spectrum into a free-electron-gas modulation and an interband structure.

The results of the KK analyses are presented on Figs. 3 and 4 for Fe and Co, respectively. We have chosen to display the optical conductivity $\sigma = \epsilon_2 E/4\pi\hbar$ rather than ϵ_2 because σ shows low-energy structure more clearly than does ϵ_2. In Fig. 3, we have further emphasized the low-energy structure by subtracting the Drude term used in the KK analysis. This Drude term is not unique, of course, and the subtraction must be made cautiously. It serves to reveal, however, a shoulder in the conductivity near 0.8 eV which tends to be obscured by the free carrier and higher energy absorption. The open circles in Figs. 3 and 4 represent the conductivities as determined by Johnson and Christy.

The results of the thermoreflectance measurements are shown in Fig. 5. Above about 1 eV, $\Delta R/R$ is broad and slowly varying; below 1 eV, a strong derivativelike line shape is observed. The character of the latter is shown qualitatively in the insert of Fig. 5. Not shown is an assessment of contributions from higher-energy interband transitions since their contribution to $\Delta R/R$ is impossible to estimate.

The $\Delta R/R$ spectrum can be Kramers-Kronig analyzed to show the temperature dependence of the dielectric function, $\Delta\tilde{\epsilon}$. The extrapolations outside the energy range of the experimental data are less important in modulation spectroscopy than in the treatment of reflectance data.[29] In the ultraviolet, $\Delta R/R$ was brought smoothly to zero at 4.5 eV; in the infrared, a combination of a

Drude term, with its $\Delta\tilde{\epsilon}$ determined from the temperature dependence of the resistivity[30] and from the effect of thermal expansion[31] on the plasma frequency, and a harmonic oscillator, the resonance frequency of which was shifted with temperature, was used. The strength of this oscillator and its temperature shift were adjusted to match the experimental $\Delta R/R$ at 0.4 eV. The resultant $\Delta\sigma$ spectrum is shown in the insert of Fig. 3.

DISCUSSION

The energy bands of a ferromagnetic material are difficult to calculate since both spin-orbit and exchange effects should be included in the Hamiltonian and their operators do not commute. The extent to which spin-orbit effects can be neglected in the 3*d* transition metals is not completely known (splitting approximately 0.06 eV for Fe and Co compared with exchange splitting of 1–2.5 eV). Its inclusion lifts accidental degeneracies in the majority and minority bands and mixes the spin character of the bands. Early calculations treated ferromagnetic Fe and Co as metals with two separate sets of paramagnetic bands rigidly shifted by the exchange splitting. More recent calculations[2-4] have shown that the exchange splitting is energy and *l* dependent with the splitting (in Fe) of *s* states at the bottom of the bands (Γ_1) amounting to only (10–20)% of that of *d* states at E_F.

Callaway and co-workers[2] recently calculated the energy bands of Fe from first principles, including both exchange splitting and spin-orbit effects with several values for the exchange-correlation potential. Wakoh and Yamashita,[3] Yasui *et al.*,[4] and Moruzzi *et al.*[5] have reported extensive calculations for Fe but without spin-orbit effects. These calculations show that the Fermi level lies very close to the top of the majority-spin *d* bands (near H'_{25}), a point dictated by the de Haas–van Alphen measurements of Gold *et al.*[32] and Baraff[33] (majority-spin hole pockets). Its position in the minority bands is very similar to that found in the V- and Cr-group bcc metals. Baraff has compared the Fermi-surface de Haas–van Alphen character of Fe to that of W.

In the following interpretation of the optical properties of Fe and Co, we will assume that the spin mixing due to $L\cdot S$ effects is small and that spin-flip transitions do not give rise to the observed optical structures. The optical features can be interpreted without appeal to spin-flip transitions, but spin-flip transitions cannot be completely rejected based on the optical studies.

The nearly filled majority-spin bands should play little role in the optical properties, but the minority bands resemble those of other bcc transition metals, and should give rise to structures similar to those of, for example, Cr (E_F in Fe lies relatively lower in Fe than in Cr). In studies of V, Nb, and Ta we have suggested[20-22] that strong optical transitions occurred between the third and the fifth bands along Σ, namely, $\Sigma_1 \rightarrow \Sigma_1$; these transitions occurred at about 1.5–2 eV. Features were also observed in the Cr-group metals which could be related to the transitions. We should then expect analogous structure in ferromagnetic Fe. In studies of Cr, Mo, and W, we found that the shift in E_F (relative to its position in the bands of the vanadium group) introduced structure at low energy which could be related to the newly occupied bands along $\Gamma(\Delta)H$. The location of the Fermi level within the minority bands of Fe suggests that we should be alert for structure in the dielectric function due to $\Gamma(\Delta)H$ transitions.

Two structures are clearly apparent in the optical spectra for Fe (Figs. 1, 3, and 5) and these can readily be interpreted by analogy to the nonmagnetic bcc metals. First, the lower-energy structure can be related to transitions along $\Gamma(\Delta)H$, namely, $\Delta_5(\downarrow) \rightarrow \Delta_2(\downarrow)$. Hence, we would argue that the calculated Δ-band separation[2] (~1.1 eV) is larger than indicated by experiment (~0.8 eV) [unless the maximum in the interband strength arises from off-symmetry parts of the zone near $\Gamma(\Delta)H$]. We also conclude that the calculations of Duff and Das[6] have significantly overestimated the Δ-band separation (~2 eV). Reducing the magnitude of their exchange splitting from about 3.5 eV to a magnitude closer to that proposed by Callaway *et al.* would improve the agreement with our data. Pessa *et al.*[18] recently argued for the same reduced exchange splitting based on their PES findings; they estimated that $\delta\epsilon_{ex} \leq 1.9$ eV.

The second optical structure is the broad feature in the conductivity at 2.37 eV. This undoubtedly arises from contributions from numerous bands. The involvement of the bands along Σ can be inferred from studies of the V- and Cr-group metals, but other pairs of minority bands probably contribute as well. The slowly varying, undifferentiated thermoreflectance spectrum indicates that these occur over significant volumes of *k* space and that critical point or Fermi-surface transitions are not dominant.

Band-structure calculations for Co have been fewer in number than for Fe due, in part, to the added difficulties associated with the hcp crystal structure and the existence of fewer experimental studies of the electronic structure.

Comparison of the energy bands calculated by Singal and Das,[7] Batallan *et al.*,[8] and Ishida[9] shows that there is still significant controversy over the band character of hcp Co. As is the case for Fe, much of that controversy involves the handling and magnitude of the exchange splitting. There is better agreement about the d-band width of the majority or minority bands (about 4 eV by Singal and Das, 4.2 eV by Ishida, and 4.75 eV by Batallan *et al.*). Each of these calculations shows the minority d bands cut by E_F with complicated Fermi-surface crossings and numerous interband transitions possible for $h\nu < 5$ eV.

Singal and Das recently reported a detailed study of the band structure, cohesive energy, and related properties for hcp Co. Their results indicated an average exchange splitting of ~3.5 eV. The implications of this large exchange splitting, as far as the ferromagnetic cobalt band structure is concerned, is that the calculated majority d bands lie ~5 eV below E_F and overlap very little with the minority bands. The predicted overall occupied d-band width is about 7.5 eV which must be compared with recent PES experimental results which give an occupied d-band width of ≈4 eV. Further, Heimann *et al.* used their PES results to argue that $\delta\epsilon_{ex} \approx 1.1$ eV. The calculations of Battalan *et al.* and Ishida give results which qualitatively agree with each other and with the experimental PES results, and our qualitative interpretation is based on them. Because a k-independent exchange splitting was used, we regard these bands as illustrative but not definitive. Heimann *et al.* came to that same conclusion when they attempted to interpret their normal-emission PES results for hcp Co.

The optical conductivity spectra of hcp Co. (Fig. 4) show interband structure between about 0.3 and 3.5 eV as well as an additional interband feature at about 5.5 eV (as reported earlier by other workers). As shown in Fig. 4, the center of strong low-energy interband absorption is near 1.3 eV for both polarizations. The calculated bands offer many possible pairs of bands which may be contributing to this strong absorption feature. There are pairs of bands which have a high joint density of states (they appear to be parallel in some parts of the zone) that are separated by energies ranging from 1 to 2 eV and extend throughout the Brillouin zone. States in the uppermost d band (e.g., the band extending from A_3 to H_3) would be likely candidates for final states in these transitions. A more detailed interpretation of the various optical features does not appear warranted at this point because of the inherently qualitative character of existing bands. The observed anisotropy, as in all noncubic solids, arises from the electric dipole matrix elements, which have not been calculated.

There remains the possibility that contributions to interband absorption arise from spin-flip transitions. In the above paragraphs we have suggested alternate origins of the observed optical structures, but we cannot rule out the possibility that spin-flip transitions are occurring. It is possible that the optical conductivity structures at 0.28 eV in Ni, 2.37 eV in Fe, and 1.3 eV in Co do involve spin-flip transitions. Recent ARPES measurements[34] have shown that $\delta\epsilon_{ex}$ is 0.31 eV in Ni, and it appears that the predicted exchange splitting in Fe and Co is approximately at the energy of the optical structure. Unfortunately, optical spectroscopy is unable to identify clearly some feature which can be related to the exchange splitting.

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