Photon Energy Dependence of Spin Polarization of Photoelectrons from Cesiated Co

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Photoelectrons from cesiated Co films with different work functions exhibit a positive spin polarization with magnetic moments preferentially parallel to the magnetization at photon energies from 2.8 to 5.5 eV. The photoelectron polarization spectra show that the existing band-structure calculations with either a direct or nondirect transition interpretation of the photoemission process are inadequate.

In the Stoner-Wohlfarth-Slater (SWS) band theory of ferromagnetism, the electrons move in an average Hartree-Fock field that is different for electrons of opposite spin. The advantage of the SWS band theory is that it can make distinct predictions such as the sign of the electron spin polarization (ESP) as a function of energy and the temperature dependence of the d-band peak position in photoemission. For the strong ferromagnets Ni and Co, a predominance of minority spins is predicted at the Fermi level E_F, so that the measured spin polarization of photoelectrons from these states should be negative (preferential direction of the magnetic moment antiparallel to the direction of magnetization). However, the polarization of photoelectrons emitted from states within 0.4–0.8 eV of E_F was found to be positive even for Co and Ni.[1,2] Tunneling experiments also showed a positive polarization of electrons from states within 1 meV of E_F for Ni.[3] Futhermore, the temperature dependence of the Ni d-band peak position did not show the expected behavior,[4,5] and the d-band widths of Fe, Co, and Ni obtained from high-energy photoemission experiments[6] are narrower than the widths obtained from band calculations. These discrepancies between the experimental findings and the predictions of the SWS band theory have been variously attributed to the need for more refined band calculations, to the method of interpreting the data, and to the inadequacy of the one-electron approximation.

We present here measurements of the photoelectron spin polarization from cesiated Co with the aim of specifying more clearly the discrepancies with the band theory, which may then in turn lead to a better understanding of photoemission and ferromagnetism in 3d metals. A crucial test of the band theory is the occurrence of negative ESP. For Ni, the expected photoelectron spin polarization was calculated by Smith and Traum[7] and by Wohlfarth.[8] The negative ESP appears for electrons from an energy range near E_F that is difficult to detect by photoemission techniques because it is so narrow. Futhermore, the predictions rely on fine details of the calculated Ni band structures which have d-band widths which may be too wide.[6] Therefore, Ni is not favorable for this type of test of the SWS band theory.

Co was then chosen because the negative polarization is expected over a wider energy range as a result of the larger magnetic moment compared to that of Ni. In contrast to the previous ESP measurement on Co,[2] this work shows the photon energy dependence of the ESP for a series of different work functions. These new data enable us to conclude that the SWS band theory with the present band calculations is inadequate. Many-body effects, such as have been suggested by Anderson,[9] Baltensperger,[10] and Doniach,[11] appear to be important to understand the experimental results.

The Co films were evaporated by electron gun from 99.999%-pure Co onto substrates held at approximately 150°C or onto substrates at liquid-nitrogen temperatures, which were subsequently heated to 150–200°C. During evaporation the pressure rose to 1×10^{-8} Torr and returned to a pressure of $<3\times10^{-10}$ Torr a few minutes after evaporation. The work function of the sample was varied from 2.5 to 3.4 eV with Cs from a zeolite source. The apparatus has been described previously.[12] A superconducting coil produced the saturation magnetic field[13] of 17 kG perpendicular to the sample surface. The photoelectrons are accelerated to 100 keV and the spin asymmetry is detected by Mott scattering from a gold foil.[14]

The measured spin polarization P is defined as

$$P=(I_\uparrow-I_\downarrow)\ (I_\uparrow+I_\downarrow),$$

where $I_\uparrow$ ($I_\downarrow$) represents the photoelectron current due to majority (minority) spins. At a high enough photon energy so that the band density of states is observed, one expects to measure a polariza-

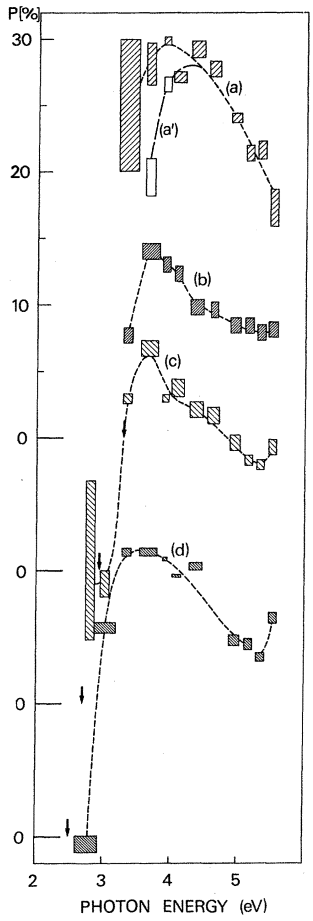

FIG. 1. The spin polarization of photoelectrons from four cesiated Co films is shown as a function of photon energy. The photoelectric thresholds (marked by arrows) are curve a, 3.3; curve b, 2.95; curve c, 2.7; and curve d, 2.5 eV. The polarization scale of each successive curve is shifted 10% for clarity. The vertical dimension of the fields corresponds to a polarization uncertainty of ±1 standard deviation, and the horizontal dimension to the maximum spectral uncertainty. The two points on curve a' were measured $6\frac{1}{2}$ h after film preparation.

tion $P = n_B/n = 17.3\%$ for Co since $n_B = 1.56\mu_B$/atom and $n = 9$ electrons/atom.

Figure 1 shows P as a function of photon energy $h\nu$ for four representative Co films with work functions 2.5, 2.7, 2.95, and 3.30 ± 0.1 eV that were determined from Fowler plots of the photoelectric yield. The initial-state energies of the photoemitted electrons range from E_F to a maximum of 3 eV below E_F. The striking feature of these spectra is that P, even as near as 0.1 eV from threshold (curve a), is not negative, in contrast to the expectations for Co in the Hartree-Fock approximation. It is interesting to note the maximum which occurs at the photon energy of approximately 3.7 eV, independent of the work function; this indicates an influence of unoccupied electron states on the transition strength for the majority electrons. Surface contamination may lower the ESP near threshold as indicated by the two points of curve a' which were measured $6\frac{1}{2}$ h after film deposition.

The possibility of understanding experimental ESP spectra in terms of the SWS band theory has been examined by Smith and Traum.[7] Their calculation showed that the positive polarization observed in the early experiments[1] on Ni can be explained if k conservation (direct transitions) is important in the optical excitation process. But the magnitude of the polarization and the fact that it remained unchanged on varying the optical excitation depth from 0.4 to 0.8 eV are not explained. Depending on photon energy and work function, the calculation[7] predicts an ESP for Ni of up to ±100%; similar effects are expected for Co in the framework of the SWS band theory. There is a predominance of minority spins near E_F over an energy interval of the ground state that can be inferred from the various band calculations.[15-17] While this energy interval is not known accurately, it is approximately 3 times larger in Co than in Ni. It is extremely unlikely that k conservation would produce positive ESP near threshold over the 0.8-eV range of work functions covered by the experimental spectra in Fig. 1.

Wohlfarth[8] has suggested that the positive ESP of Ni could be explained in terms of a density-of-states model (nondirect optical transition) if the energy difference Δ between the top of the majority spin d bands and E_F is very small. Figure 2 shows the ESP predicted by the SWS theory for two model Co band structures, assuming that the polarized electron currents are given by

$$I_{\uparrow,\downarrow} = \int_{h\nu}^{E_F} - \Phi n_{\uparrow,\downarrow} T_{\mathrm{eff}} dE,$$

where $n\uparrow$ ($n\downarrow$) is the density of majority (minority) spin electrons, T_{eff} is the semiclassical escape function,[18] and Φ is the work function. Curve a of Fig. 2 is calculated using the density-of-states curve of Wong, Wohlfarth, and Hum,[15] which has an exchange splitting $\Delta E_{ex} = 1.35$ eV and $\Delta = 0.46$ eV. Curve b results from the same density of states but with $\Delta E_{ex} = 1.05$ eV and $\Delta = 0.15$ eV, which have been suggested by Wohlfarth[16] as the best values for a consistent picture of Co and Ni and Fe. Comparison with the experimental spectra shows that there is no way to understand the

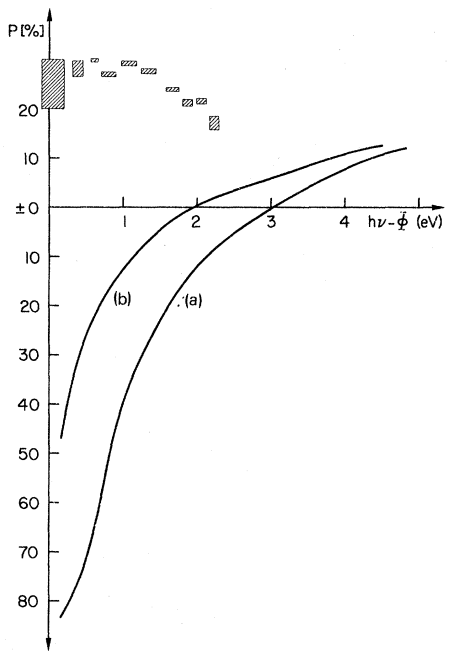

FIG. 2. The photoelectron spin polarization of Co predicted using the density-of-states model: curve *a*, the Co band structure calculated by Wong, Wohlfarth, and Hum (Ref. 15); curve *b*, with the best values of Δ and ΔE_{ex} suggested by Wohlfarth (Ref. 16). Curve *a* of Fig. 1 is also shown as an example of the experimental results.

measurements within this framework.

Various processes which might explain the positive ESP, such as inelastic electron-magnon scattering, were considered previously[1,12] and found to be negligible. The difference between the present experiment and the previous work[2] is the Cs on the surface of the Co film. The Cs is present as an ion and, at lower work functions, also as an atom. In principle, both can alter the photoelectron spin polarization. The Coulomb field of the Cs ions will attract mainly *s* electrons and reduce the *s*-electron screening at the Co sites. This in turn will shift the energy of the *d* states and might reduce the magnetization in the first two layers of the Co film.[19]

There can be spin exchange between the photoelectrons from the metal and the Cs atoms; little can be said about the details since the Cs atoms may themselves be polarized through exchange interactions or may form molecules on the surface. The Cs atoms can also be photoionized. The yield is comparable to the yield from Co metal at photon energies near threshold, but should be negligible at slightly higher photon energies. Unpolarized light was used to rule out any possibility of obtaining polarized electrons from the Cs atoms as is known to be possible when free Cs atoms[20] or Cs metal[21] is irradiated with circularly polarized light (Fano effect[22]).

The ESP of +21% obtained with clean Co and with polychromatic light exciting electrons up to 0.8 eV below E_F is consistent with the present results on cesiated Co shown in Fig. 1. It is also reassuring to note that the spectra do not show a strong dependence on the amount of Cs and that the asymptotic value at higher photon energies is consistent with the ultimate saturation value of 17.3%. Therefore, our conclusions about the inapplicability of the SWS band theory remain unaffected.

Anderson[9] has suggested many-body corrections to the Hartree-Fock approximation which may explain the photoemission results. In this picture, strong interparticle interactions require a renormalization which reduces the density of spin-down quasiparticles at E_F; moreover, a spin-down quasiparticle is equally likely to be a bare spin-down electron or a spin-up electron plus a "spin wave." Baltensperger[10] has shown that because of the correlation of electrons with opposite spin, it is energetically favorable for electron pairs to form a coherent state in such a way that it costs more energy to excite a spin-down electron that a spin-up electron. This theory qualitatively explains the positive polarization, the small temperature dependence of the *d*-band peak, and the narrow *d*-band width observed in photoemission. In the model of Doniach,[11] the Hartree-Fock ground state is not altered, but it is impossible to see the ground state in photoemission because of the spin-dependent relaxed orbital correction around the resulting hole. The relaxed orbital correction conteracts the exchange splitting, and one can obtain a positive ESP near E_F and little temperature dependence of the *d*-band peak.

At present it is not possible to decide which of the many-body theories is appropriate.

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Dynamic Properties of a One-Dimensional Heisenberg Magnet*

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A form for the relaxation shape function for a Heisenberg paramagnet is proposed which has the merit of satisfying certain sum rules and limits. It is shown by comparison with exact infinite-temperature calculations by Carboni and Richards, and inelastic neutron scattering measurements by Hutchings *et al.* on $(CD_3)_4NMnCl_3$, to provide a very good description of an antiferromagnetic linear chain at all temperatures.

Various measurements on one-dimensional magnetic systems[1,2] have created renewed interest in the static and dynamic properties of a Heisenberg linear-chain magnet. Most previous theories have been for either zero or infinite temperature. Exceptions are the exact calculation by Fisher[3] of static two-spin correlation functions for a classical Heisenberg chain, and McClean and Blume's[4] study of dynamic properties, which is based on an integro-differential equation for the relaxation function [of Eq. (4), see below] discussed previously by several authors[5] for simple-cubic Heisenberg magnets. Numerical calculations have been performed for finite-length spin-$\frac{1}{2}$ chains by Carboni and Richards[6] and for long classical Heinsenberg chains by Blume, Watson, and Vineyard.[7] Also, Richards[8] has recently found a spectrum for the collective mode by linearizing the equations of motion in second order. Here we propose a new, simple theory which gives both good agreement with the calculations of Carboni and Richards and with neutron scattering measurements by Hutchings *et al.*[2] on the linear-chain antiferromagnet $(CD_3)_4NMnCl_3$ at temperatures between 1.9 and 40°K. Apart from a multiplicative factor of $\sqrt{\frac{3}{2}}$, Richards's dispersion relation is found to compliment our calculation.

The magnetic energy of the system is described by the Heisenberg exchange Hamiltonian

$$\mathcal{H}=\tfrac{1}{2}J\sum_{i\delta}\vec{S}_i\cdot\vec{S}_{i+\delta}, \tag{1}$$

where J is the exchange parameter between adjacent spins.

The inelastic partial-differential neutron cross section for scattering from a paramagnet with wave-vector change $\vec{k}$ and energy change ω is