# Stoner Transitions and Spin-Selective Excitations in bcc Cobalt

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Spin-selective excitations (Stoner excitations) have been unambiguously identified by comparisons between spin-polarized-electron energy-loss spectra and calculated transition density of states. Analysis of the specular beam for polarized electrons incident on thin films of bcc Co shows three sharp, highly polarized loss features (background corrected asymmetry near 100%). Comparison with the theoretical Stoner and spin-nonflip transition density of states shows excellent agreement, permitting identification of the associated loss mechanisms.

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Collective excitations (spin waves) have been experimentally verified and characterized by neutron scattering for quite some time.[1] Much more recently, experimental techniques to probe single-particle excitations, especially spin-flip Stoner transitions (electronic excitations resulting in an electron in the conduction band and a hole of the opposite spin in the valence band), have been developed.[2-7] Spin-polarized-electron energy-loss (SPEELS) measurements on single-crystal Ni and polycrystalline alloys of Fe have yielded evidence for the first observations of Stoner transitions,[4-6] but the analysis of these measurements has been impaired by a lack of comparison with theoretical calculations of the Stoner transition density of states (DOS) and by an intrinsic complication arising from the existence of possible spin-nonflip excitations (direct excitations) which can have the same incident polarization dependence and loss character as the Stoner transition.[8] (Stoner DOS have been calculated for spin-wave measurements[9] and very recently for use in SPEELS analysis,[7] but with only limited success.) This Letter reports on SPEELS measurements on bcc Co which is a nearly ideal ferromagnetic material for the observation of Stoner excitations. For the first time, multiple, sharp, loss features are observed. Comparison of the rich experimental spectra with calculated Stoner DOS derived from calculations of the bcc Co band structure[10,11] show excellent agreement with the SPEELS features allowing for an unambiguous differentiation between Stoner and spin-nonflip transitions. The calculated band structure of bcc Co shows that it is a good material for Stoner transition measurements; a completely band-saturated ferromagnetic (i.e., one with completely filled majority states) with a large mean exchange splitting of $\simeq 1.6$ eV. Although the bcc structure of Co has not been synthesized in bulk form, atomically clean and well characterized thin films of limited thickness have been epitaxially deposited on GaAs. In contrast, the ferromagnetic materials previously examined by SPEELS are not ideal—Fe is a band-unsaturated ferromagnet, possessing a significant number of unfilled majority states, and Ni has a small exchange splitting.

In these experiments, a $\simeq$40-Å film of bcc Co(110) grown epitaxially on GaAs(110) is used as the target. The film is characterized by Auger-electron spectroscopy, reflection high-energy electron diffraction, and vibrating-sample magnetometry with similar results as to those previously described.[12-14] The film can be completely magnetized along the magnetically easy [001] direction by poling *in situ* with a solenoid. The electron energy-loss measurements are made with a $\simeq$30% polarized $e^-$ beam, generated by photoemission from a GaAs source,[15] incident at 45° to the sample normal and along the [100] direction as shown in Fig. 1. The specular beam, which is emitted at 90° from the electron-source axis and along the [010] direction, is energy analyzed by a hemispherical detector with a 4° acceptance angle.

The experimental loss spectrum for the two incident polarization directions is shown in Fig. 2(a). The elastic peak [with a measured full width at half maximum (FWHM) of 0.6 eV] has been removed. The width of the two lowest-lying loss peaks (centered at 1.6 and 3.1 eV) is $\simeq$0.6 eV, the instrument resolution. Therefore, the underlying loss features themselves are sharp, in contrast to the broad Stoner features previously reported for Ni,[5] Fe,[6] and Fe alloys[4] (where only one loss feature was observed). A third, much broader feature peaks at 4.4

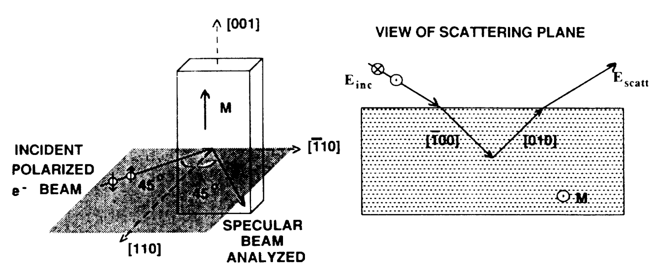


FIG. 1. The scattering geometry of SPEELS measurement. Polarized electrons are aligned (spin-up) or antialigned (spin-down) with the magnetization direction. The corresponding view of the scattering plane with the incident and reflected electron directions are also indicated.

 

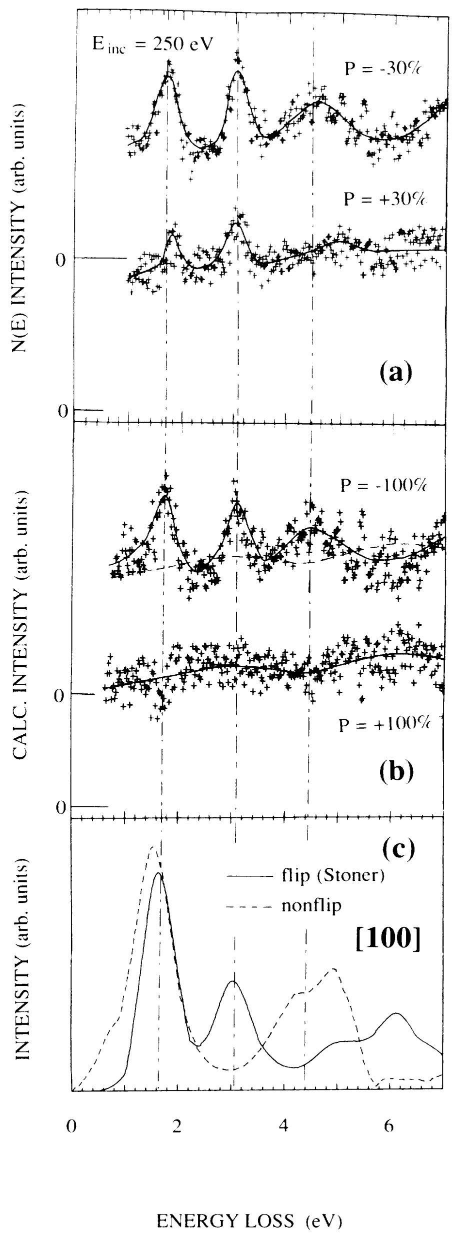


FIG. 2. (a) The loss spectra for an incident 250 eV, 30% polarized elecron beam for polarization direction aligned with the magnetization (+30%) and antialigned (−30%). The −30% spectra has been offset, the zero line is indicated. The elastic peak has been removed for clarity. The solid lines represent Gaussian least-squares fits to the appropriate ranges of the three major peaks. (b) The calculated loss spectra for incident completely (±100%) polarized beams, extracted from the measured data. The solid line represents a Gaussian least-square fit. The dashed line in the −100% spectra is the fit to the +100% spectra, suggesting the presence of a spin-independent loss background. (c) The calculated spin-flip (Stoner; solid line) and spin-nonflip (direct; dotted line) DOS along the Γ-*H* or [100] direction for bcc Co, broadened by convolution with a 0.6-eV FWHM Gaussian to represent the instrument resolution.

eV. The magnetic character of the loss features is demonstrated by reversal of the sample magnetization direction, which transposes the observed polarization dependence.

Because the incident beam polarization, $P_0$, is only ±30%, the polarization dependence can be extracted by calculation of the anticipated loss spectrum for an incident ±100% polarized beam. These extrapolated spectra, $I_+$ for an incident +100% beam and $I_-$ for an incident −100% beam, are calculated from

$$I_+ = I(+P)\frac{(1+|P|)}{2|P|} - I(-P)\frac{(1-|P|)}{2|P|},$$

and

$$I_- = I(+P) + I(-P) - I_+,$$

with $P_0 = (N\uparrow - N\downarrow)/(N\uparrow + N\downarrow)$ where $N\uparrow(N\downarrow)$ is the number of incident spin-up (spin-down) electrons and $I(+P)$ $[I(-P)]$ is the scattered intensity for incident spin-up (spin-down) polarization direction. These calculated spectra are shown in Fig. 2(b). It is now clear that the loss features are due only to the incident spin-down electrons. The polarization of the features is near −100%, an order of magnitude larger than previously reported values.[4-6] Also note the presence of a spin-independent loss background [shown by the dashed line in Fig. 2(b)]. Figure 2(c) shows the calculated Stoner DOS (solid line) and a spin-nonflip DOS (dotted line) for bcc Co expected for our experimental conditions. Details of this calculation are given later.

Figure 3 shows a simplified band structure for a band-saturated ferromagnet. To help visualize the core-hole excitations, only four bands are displayed (two majority spin bands and the two corresponding minority spin bands). Figure 3(a) shows an incident spin-down (minority spin) electron scattering into an unoccupied minority state and ejecting a spin-up (majority spin) electron with an energy reduced from the incident energy by an amount equal to the exchange splitting energy, Δ, of the bands. This interaction is a spin-flip or Stoner excitation, as an examination of the initial and final states will show. By our fixing the experimental conditions so that only the specular beam is measured (near zero momentum transfer, $q \sim 0$), the interband, interspin excitation is assured to be a vertical transition. This transition is labeled as a primary Stoner excitation since the electron promotion occurs between the conjugate minority and majority bands and the energy of the excitation is the band spin-splitting (or Stoner) energy. A transition between two opposite spin bands with *different* band indexing is labeled as a secondary Stoner transition [Fig. 3(b)] because it is still a spin-flip transition, but the transition energy depends on the relative band positioning. It is the presence of the secondary Stoner transitions which result in additional peaks in the Stoner DOS beyond the primary Stoner peak centered at energy Δ.

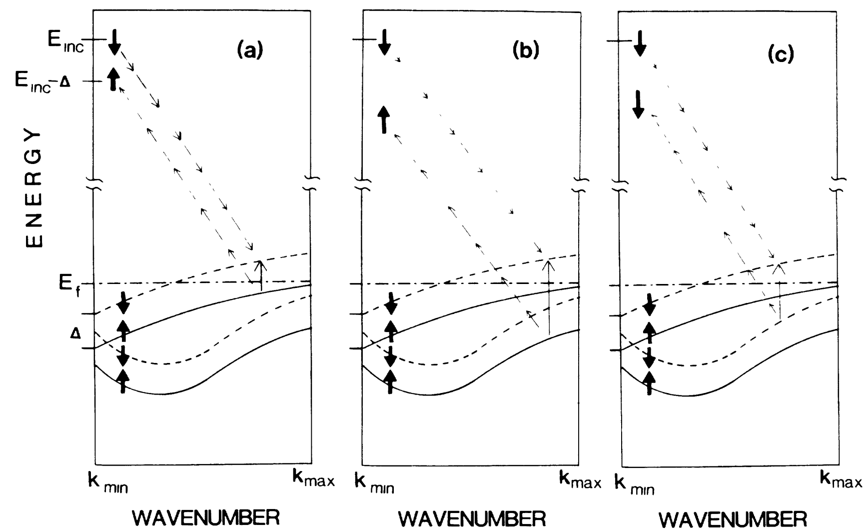


FIG. 3. (a) Diagram of a primary Stonder excitation. An incident minority spin electron scatters into an unoccupied minority band, exciting a majority electron, from the conjugate majority band, to an energy $E_p - \Delta$. Experimental arrangement constrains the transition to being vertical, $q \sim 0$. (b) Diagram of a secondary Stoner excitation involving opposite spin bands with different band indexing. (c) Diagram of exchange portion of spin-nonflip direct excitation involving bands with same spin.

There is a complicating transition[8] still to be addressed; the spin-nonflip exchange[16] portion of the direct transition shown in Fig. 3(c). Again, a spin-down electron scatters into an unfilled minority band, but now an electron from a different filled *minority* band is ejected. Because of the experimental configuration, the transition is also vertical, but is now a spin-nonflip excitation. (The effects of cross terms in the scattering cross section[8] of the direct excitation are thought to be small,[17] and have not been included.) Although this is a spin-nonflip excitation, this transition occurs only for an incident spin-down electron. Therefore, this spin-nonflip excitation and the Stoner excitation have the same incident polarization dependence and are indistinguishable in a SPEELS measurement utilizing only a polarized electron source. To differentiate between these two excitations, spin analysis of the loss spectrum can be added as was recently implemented by Kirschner and co-workers.[6,7]

To calculate the Stoner DOS, first an augmented-plane-wave spin-polarized band-structure calculation of bcc Co with potential functions due to Hathaway[10] is performed. The augmented-plane-wave results are then fitted to a nonorthogonal Slater-Koster Hamiltonian that reproduced the augmented-plane-wave eigenvalues to better than 1 mRy accuracy. Details of this procedure are given in Ref. 11. This method generates very efficiently the $E(k)$ in various directions of the Brillouin zone for a large number (100) of $k$ points necessary for the evaluation of the Stoner DOS. The transition energies for both spin-flip and spin-nonflip near-vertical transitions (allowing for the momentum spread of the spectrometer) between occupied and unoccupied bands can then be extracted.

The Stoner DOS for the zero-momentum-transfer case is calculated from

$$Y(E) \propto \sum_{\mu,\nu} \int\int_{BZ} M\delta[E - E_\mu(k) + E_\nu(k')]\delta(k-k')dk\,dk',$$

where $\mu,\nu$ index the unfilled minority and filled majority spin bands, $M$ is the transition matrix element, and the integration is over the entire Brillouin zone. The spin-nonflip transition DOS can be similarly obtained by use of the filled minority spin bands, indexed by $\nu$, in the calculated DOS equation. To reflect experimental conditions, the momentum $\delta$ function was relaxed to include the momentum spread admitted by the spectrometer (4° acceptance angle). Earlier calculations[7] have taken the transition matrix element to be constant over the entire Brillouin zone resulting in a Stoner DOS with equal contributions from all points in the zone. However, the transition matrix element in a SPEELS measurement is strongly coupled to the incident electron direction. The matrix element is largest when the incident electron momentum and the band electron momentum are parallel. In this situation the physical overlap of the electron wave functions, and therefore the transition matrix element, is maximum.

Because of this transition-matrix-element $k$ dependence, a realistic approximation restricts the sum over the Brillouin zone to only that portion along the electron momentum direction. To allow for loss-then-reflection and reflection-then-loss possibilities, the incident and scattered electron propagation directions must both be included. We have constructed our experimental arrangement so that the incident and scattered electron propagation directions are both along ⟨100⟩ axes.

For direct comparison with our experiment, the calculated Stoner and spin-nonflip DOS along the ⟨100⟩ (or

Γ-$H$) direction are broadened to simulate the 0.6-eV FWHM instrumental response. The Gaussian-broadened Stoner DOS (solid curve) and spin-nonflip DOS (dotted curve) for an incident minority spin electron are shown in Fig. 2(c) aligned with the experimental data. (Summing over of the entire Brillouin zone results in a single peak at the mean exchange splitting energy superimposed on a rather featureless background.) Because of the very small number of unfilled majority states, transitions involving unfilled majority states (both spin-flip and spin-nonflip) are reduced in intensity by 2 orders of magnitude. It is clearly seen that the experimental loss feature at 1.6 eV corresponds to features in the DOS containing both Stoner and direct contributions. Calculations for Fe predict a similar compound feature which is even further complicated by the unfilled majority state transitions of the unsaturated ferromagnet.[18] (Experimental evidence for both spin-flip and spin-nonflip excitations has already been observed in SPEELS measurements done with spin analysis of the loss features of Fe.[6,7])

The broad peak at $\simeq$4.4 eV is identified as a spin-nonflip transition, although contributions from Stoner transitions may be present. The intermediate, highly polarized loss feature at 3.1 eV is now unambiguously seen to be a Stoner excitation. The slight differences are well within the uncertainties of the band-structure calculations. Furthermore, long-range dipole scattering, shown to be a dominant loss channel for specularly reflected electrons in Fe,[7] is not seen to be the dominant loss mechanism for bcc Co, but may contribute to the slowly varying loss background upon which the spin-flip excitations are superimposed. Preliminary Stoner DOS calculations[18] for Ni and Fe show much broader transitions which indicate possibilities for extension of previous experimental work.[5-7]

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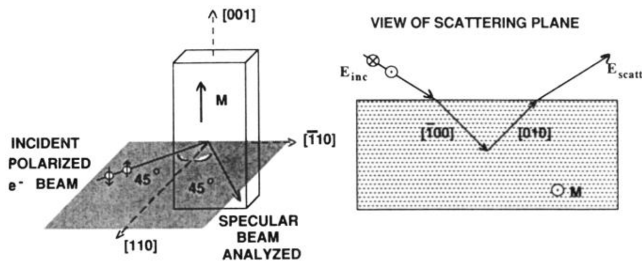


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