Free-electron-like Stoner excitations in Fe

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An analysis of spin-polarized electron-energy-loss experiments in Fe is described which (1) identifies the contribution of d-electron Stoner excitations, the usual type of Stoner excitation, (2) suggests that free-electron-like Stoner excitations are more probable than d-electron Stoner excitations, and (3) indicates that exchange events involving large energy losses are as likely as direct scattering.

Of the two types of elementary magnetic excitations in ferromagnets, spin waves and Stoner excitations, only the former have been experimentally investigated in detail.[1] Unlike the case of spin waves, Stoner excitations have not been amenable to study by neutron diffraction and only recently have the techniques to probe the Stoner excitation spectrum been developed.[2–6] The methods have utilized the electron-energy-loss spectroscopy[3] combined with polarization of the incident electron beam,[4] or with polarization detection of the scattered beam,[5] and most recently with both a polarized incident beam and polarized detection[6] (denoted by I). The results of I represent a revision as well as an extension of previous work.[7] With the aid of a simple model for the electronic structure of Fe we analyzed[8] the contributions of various scattering processes to the measured cross sections using the early data.[7] The new data are very different from the old data and the purpose of this paper is to update our previous analysis[8] and present the new results. Many of our previous conclusions are modified or changed.

Due to the many possible scattering processes besides the Stoner type none of the experiments observe the Stoner excitation cross section directly but rather they measure the magnitudes of combinations of scattering amplitudes. *In order to draw even semiquantitative conclusions regarding the Stoner excitation spectrum from the data a theoretical analysis is required.* Because I gives more complete information than has previously been available it is possible for the first time, to our knowledge, to carry out a detailed theoretical analysis of the spin-dependent scattering process. The analysis presented here concludes that free-electron-like Stoner excitations (FESE) make a substantial contribution to the scattering process. All of the experimenters,[3–6] interpret their results in terms of d-electron Stoner excitations (DESE), the usual type which differ from FESE in that they produce a relatively large asymmetry between the scattering of up and down spin electrons. Thus the principal result of our analysis, the important contribution of FESE, is completely unexpected.

A FESE is an electron-hole pair excitation consisting of a d hole of a given spin and an electron in a free-electron-like state of opposite spin. In the DESE the electron is in a d state. The density of states for creating a FESE is far smaller than for DESE due to the small ratio of empty free-electron-like states to empty d states which makes the observation of FESE scattering all the more surprising.

In I, an incident electron beam of about 20 eV with spin parallel ($\uparrow$) or antiparallel ($\downarrow$) to the majority spin direction of Fe was scattered from the Fe(100) surface. The polarization of the inelastic reflected beam was measured for energies from 0 to 7.0 eV below the incident energy and at angles 0° to 70° off the specular direction. With the assumption that the measured electrons suffer elastic specular scattering preceded or followed by an inelastic event the experiment gives the cross sections for four different inelastic scattering processes involving a definite energy loss ω and momentum transfer q. In I, $\bar{F}^{\sigma}$ (flip) denotes the cross section for the scattering event in which an incoming electron of energy E_0 has spin σ and the outgoing detected electron has energy $E_0-\omega$ and spin opposite to σ, and $\bar{N}^{\sigma}$ (nonflip) denotes the cross section for a spin σ electron in and a spin-σ electron scattered out.

In a flip event (with cross section $\bar{F}^{\sigma}$) an electron with energy E_0 and spin σ is scattered into an empty spin σ state and an electron of opposite spin is scattered from the Fermi sea to energy $E_0-\omega$. Consider a model which assumes that an electron with energy E_0 is scattered only into empty d states, i.e., only DESE and no FESE are possible. For this model a very rough estimate of the flip cross sections is obtained from $\bar{F}^{\downarrow}\approx n_{\downarrow}^{h}n_{\uparrow}^{e}|M|^2$ and $\bar{F}^{\uparrow}\approx n_{\uparrow}^{h}n_{\downarrow}^{e}|M|^2$ where n_{σ}^{h} is the number of d holes of spin σ, and n_{σ}^{e} is the number of electrons of spin σ, and M is the screened Coulomb matrix element for the scattering event. Thus, $\bar{F}^{\downarrow}/\bar{F}^{\uparrow}\approx(n_{\downarrow}^{h}/n_{\uparrow}^{h})(n_{\uparrow}^{e}/n_{\downarrow}^{e})$ where $n_{\uparrow}^{e}/n_{\downarrow}^{e}\simeq 1.7$ from the magnetization of Fe. Band-structure calculations[9] give $n_{\downarrow}^{h}/n_{\uparrow}^{h}\simeq 10.0$ or $\bar{F}^{\downarrow}/\bar{F}^{\uparrow}\simeq 17.0$ whereas the results of I give $\bar{F}^{\downarrow}/\bar{F}^{\uparrow}\simeq 2.0$–$4.5$. In other words a model that assumes only scattering into empty d states requires $n_{\downarrow}^{h}/n_{\uparrow}^{h}\simeq 1.2$–$2.6$, a ratio that is incompatible with band-structure calculations. A model that allows for FESE as well as DESE is required.

Although some aspects of the model used here are phenomenological, the results yield order-of-magnitude effects and hence the conclusions appear to be insensitive to details of the model. The model assumes that the occupied states are d-like and all states above the Fermi energy, E_F, are free-electron-like with the exception of the unoccupied minority spin d states located in the vicinity of the E_F. The number of unfilled majority spin d states is an order of magnitude less than the number of unfilled minority states and is neglected. The majority and minority spin free-electron-like states are assumed to be identical. In this model inelastic scattering takes place as follows: an electron from the incident beam in the state i and the ground-state electron in the state d interact via a screened Coulomb interaction and scatter producing electrons in the detected state f and in the state ϵ or d^* (denoting a free-electron-like state or excited d state). In a "direct" scattering event the electron in the state $i\sigma$ is scattered to the observed final state $f\sigma$ and the electron in $d\sigma'$ is scattered to the $\epsilon\sigma'$ or in the case that $\sigma'=\downarrow$ the electron in $d\sigma'$ can also be scattered to $d^*\downarrow$. In an "exchange" process the electron in $i\sigma$ is scattered to either $\epsilon\sigma$ (or in the case $\sigma=\downarrow$ it can go to $d^*\downarrow$) while the ground-state electron $d\sigma'$ scatters to $f\sigma'$. In I the energy ω lost by the beam electron i is 0.5 eV $<\omega<7.0$ eV. In direct scattering the beam electron i loses energy ω while in the exchange event it loses on the order of $E_0-E_F\simeq 20$ eV. It is found that exchange scattering is as important as direct scattering.

The scattering amplitudes that describe these processes are given in Table I. For example $f'_\sigma \equiv A[(i\sigma,d\sigma')\rightarrow(f\sigma,\epsilon\sigma')]$ is the amplitude for the direct event in which the electron in state $i\sigma$ is scattered to $f\sigma$ and the ground-state electron $d\sigma'$ is scattered to $\epsilon\sigma'$.

All of the nonzero amplitudes are shown in Table I where use has been made of the fact that the only empty d states are minority spin. The amplitudes $f_{\sigma'}$, $F_{\sigma'}$, and $g_{\sigma'}$ in Table I are independent of σ because the majority and minority free-electron-like states are assumed to be identical.

In Table I, $F_{\sigma'}$ and $F_{\sigma'}$ denote direct scattering events and $g_{\sigma'}$ or $G_{\sigma'}$ denote exchange events. The subscript σ' denotes the spin of the d hole created in the excitation process and small f,g refer to processes in which the excited electron is in the conduction state ϵ while F,G, denote an excited electron in a d state. Thus, $G_\uparrow$ is the amplitude for creating a DESE and $g_{\sigma'}$ is that for a FESE.

TABLE I. Definitions of the scattering amplitudes that contribute to the measured cross sections $\bar{N}^\sigma,\bar{F}^\sigma$.

Initial state	Final state	Scattering amplitude	Nomenclature
$i\sigma$ $d\sigma'$	$f\sigma$ $\epsilon\sigma'$	$A[(i\sigma,d\sigma')\rightarrow(f\sigma,\epsilon\sigma')]$	$f_{\sigma'}$
$i\sigma$ $d\downarrow$	$f\sigma$ $d^*\downarrow$	$A[(i\sigma,d\downarrow)\rightarrow(f\sigma,d^*\downarrow)]$	$F_\downarrow$
$i\sigma$ $d\sigma'$	$\epsilon\sigma$ $f\sigma'$	$A[(i\sigma,d\sigma')\rightarrow(\epsilon\sigma,f\sigma')]$	$g_{\sigma'}$
$i\downarrow$ $d\sigma'$	$d^*\downarrow$ $f\sigma'$	$A[(i\downarrow,d\sigma')\rightarrow(d^*\downarrow,f\sigma')]$	$G_{\sigma'}$

Type of scattering	Excited electron in free-electron state (ϵ)	Excited electron in d state (d)
Direct	f	F
Exchange	g	G

In order to analyze the experiment we include all contributions to the observed cross sections. The scattering cross sections for the flip and nonflip events, $\bar{F}^\sigma$ and $\bar{N}^\sigma$, can be calculated in terms of the amplitudes given by Table I. For example,

$$\bar{F}^\downarrow=\sum|A[(i\downarrow,d\uparrow)\rightarrow(\epsilon\downarrow,f\uparrow)]|^2+\sum|A[(i\downarrow,d\uparrow)\rightarrow(d^*\downarrow,f\uparrow)]|^2, \tag{1}$$

where the summation is over the states $d\uparrow,\epsilon\downarrow$ and $d\uparrow,d^*\downarrow$ such that energy and momentum are conserved. Use of Table I now yields

$$\bar{F}^\sigma=\sum|g_{\bar{\sigma}}|^2+\delta_{\downarrow,\sigma}\sum|G_\uparrow|^2, \tag{2a}$$

$$\bar{N}^\sigma=\sum|f_{\bar{\sigma}}|^2+\sum|f_\sigma-g_\sigma|^2+\sum|F_\downarrow-\delta_{\downarrow,\sigma}G_\downarrow|^2, \tag{2b}$$

where $\bar{\sigma}$ denotes the spin state opposite to σ.

Equation (2) will be solved with two different assumptions which yield very similar results. First, interference terms will be neglected in which case Eq. (2b) is replaced by

$$\bar{N}^\sigma=\sum|g_\sigma|^2+D+\delta_{\downarrow,\sigma}\sum|G_\downarrow|^2, \tag{3a}$$

where

$$D=\sum|f_\uparrow|^2+\sum|f_\downarrow|^2+\sum|F_\downarrow|^2. \tag{3b}$$

All the direct transitions are contained in D and the DESE contribute only to $\bar{F}^\downarrow$. The quantity $\Delta=\bar{N}^\downarrow+\bar{F}^\downarrow-\bar{N}^\uparrow-\bar{F}^\uparrow$ is the unnormalized asymmetry and from Eq. (3),

$$\Delta=\sum|G_\uparrow|^2+\sum|G_\downarrow|^2. \tag{4}$$

The cross section $\sum|G_\uparrow|^2(\sum|G_\downarrow|^2)$ corresponds to an electron with energy E_0 falling into an empty $d_\downarrow$ state and scattering a $d\uparrow(d\downarrow)$ electron into a state with energy $E_0-\omega$. Similarly $\sum|g_\uparrow|^2(\sum|g_\downarrow|^2)$ corresponds to an electron falling into an empty free-electron-like state and scattering a $d\uparrow(d\downarrow)$ electron to energy $E_0-\omega$. Therefore, we assume

$$\frac{\sum|G_\uparrow|^2}{\sum|G_\downarrow|^2}=\frac{\sum|g_\uparrow|^2}{\sum|g_\downarrow|^2}\equiv\beta, \tag{5}$$

where it is expected that $\beta\approx n_\uparrow^e/n_\downarrow^e$. Equations (2), (4), and (5) give

$$\Delta=\left(1+\frac{1}{\beta}\right)\sum|G_\uparrow|^2, \tag{6a}$$

$$\bar{F}^\downarrow=\beta F^\uparrow+\sum|G_\uparrow|^2. \tag{6b}$$

Given the experimental values for $\bar{F}^\sigma,\bar{N}^\sigma$ Eqs. (6) can be solved for β and $\sum|G_\uparrow|^2$. Equations (2) and (3) then yield $\sum|g_\uparrow|^2$ and D.

The data from I are shown in Fig. 1 for the scattering angles $\theta=10°$, 30°, and 50° off specular. The flip and

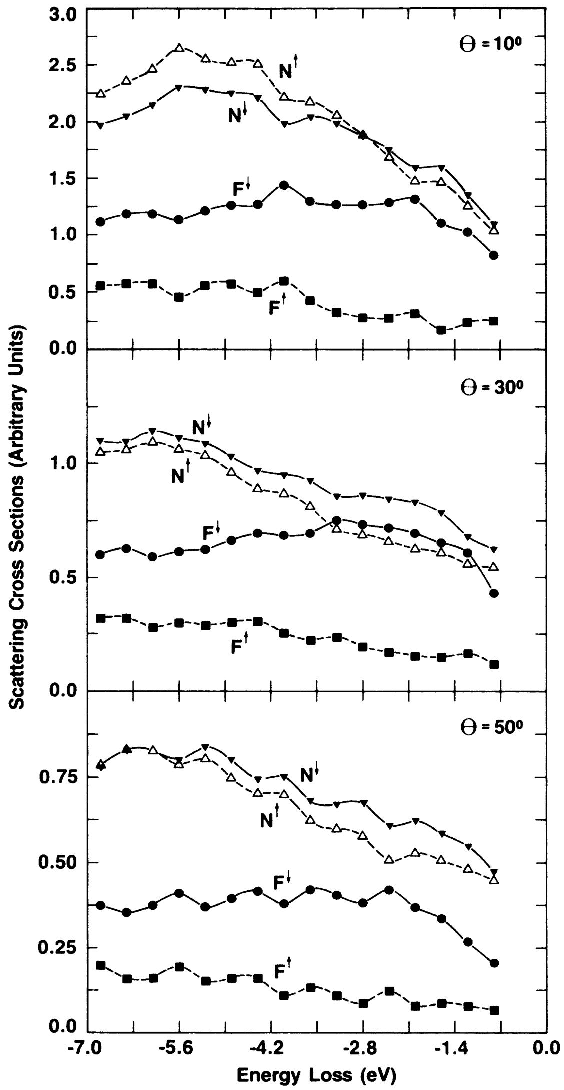

FIG. 1. Spin-dependent cross sections vs energy loss from Ref. 6 for scattering angles $\theta = 10°$, 30°, and 50°. Spin-flip events are denoted by $\bar{F}^{\sigma}$ and nonflip events by $\bar{N}^{\sigma}$ where σ refers to the spin of the incident electron.

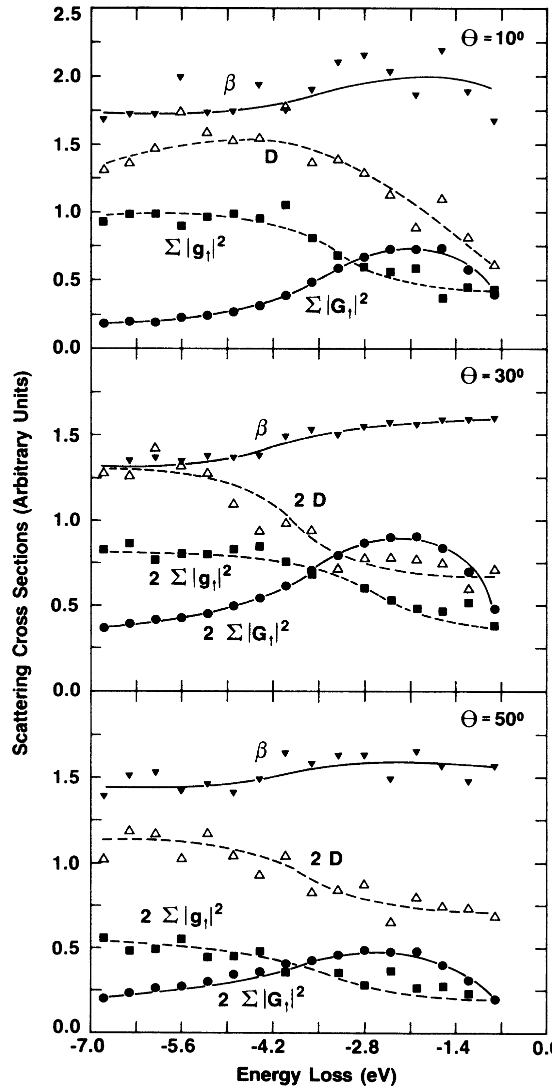

FIG. 2. Partial cross section vs energy loss as determined by data analysis. The cross section for d-electron Stoner excitations is $\sum|G_\uparrow|^2$, the cross section for free-electron-like Stoner excitations is $\sum|g_\uparrow|^2$, and the cross section for direct scattering is D. The quantity β is defined in Eq. (5) and it is expected that $\beta \approx n_\uparrow^e/n_\downarrow^e = 1.7$.

nonflip scattering cross sections are plotted versus energy loss ω. In I the data are plotted in such a way that for each energy loss the sum of the cross sections is equal to one and the total intensity versus energy loss is given separately. The result is that this normalized cross section representing $\bar{F}^{\downarrow}$ shows a pronounced peak at energy losses in the vicinity of 2 eV, the exchange splitting. However, the normalized cross sections have no physical significance and so we have plotted the total cross sections. It is clear from Fig. 1 that $\bar{F}^{\downarrow}$ does not show a well-defined structure at 2 eV whereas the density of states for exciting DESE is expected to. The earlier data of Ref. 7 is quite different from that shown in Fig. 1, for example $F^{\downarrow}$ was significantly larger than $N^{\downarrow}$ in Ref. 7 whereas the reverse is now reported in I as shown in Fig. 1.

Results for β, $\sum|G_\uparrow|^2$, $\sum|g_\uparrow|^2$, and D are shown in Fig. 2. From Eq. (5), β is expected to be approximately $n_\uparrow^e/n_\downarrow^e \simeq 1.7$, $\sum|G_\uparrow|^2$ is the cross section for DESE, $\sum|g_\uparrow|^2$ is the cross section for FESE, and D is the sum of the three cross sections for direct scattering. A number of interesting points are evident (a) the FESE cross section $\sum|g_\uparrow|^2$ is relatively large; (b) the average of β over all energy losses and angles is 1.59, close to $n_\uparrow^e/n_\downarrow^e$; (c) the FESE and DESE cross sections are of the same magnitude as the average cross section for direct scattering $D/3$; (d) the asymmetry Δ is restricted by Eq. (4) to be positive and it is, i.e., $\sum|G_\uparrow|^2 > 0$ (Ref. 10); (e) the ω dependence of $\sum|g_\uparrow|^2$ and $\sum|G_\uparrow|^2$ correlates roughly with joint density of states $\rho_{d\uparrow}\rho_{\epsilon}(\omega)$ and $\rho_{d\uparrow}\rho_{d\downarrow}(\omega)$ which measure the density of states available for making a hole in a $d\uparrow$ state an an electron in a free-electron-like $\downarrow$ state or in a $d^*\downarrow$ state, respectively.[11] As ω increases $\rho_{d\uparrow} \times \rho_{\epsilon}(\omega)$ increases and then levels off at $\omega \simeq 2$–3 eV while $\rho_{d\uparrow} \times \rho_{d\downarrow}(\omega)$ peaks at $\omega \approx 2$ eV.

On the other hand, if interference terms in Eq. (2) are kept it is still possible to estimate the relative magnitudes of the cross sections for DESE and FESE. Equation (2a) and Eq. (5) yield $\sum|g_\uparrow|^2$ and $\sum|G_\uparrow|^2$ if β is known

$$\sum|g_\uparrow|^2=\beta\bar{F}^\uparrow , \tag{7}$$

$$\sum|G_\uparrow|^2=\bar{F}^\downarrow-\beta\bar{F}^\uparrow . \tag{6'}$$

It is expected that $\sum|G_\uparrow(\omega=-7.0\text{ eV})|^2\approx 0$ because $\sum|G_\uparrow|^2$ should have an ω dependence roughly proportional to the joint density of states $\rho_{d\uparrow}\times\rho_{d\downarrow}(\omega)$. Equation (6) then gives

$$\beta=\bar{F}^\downarrow(\omega=-7.0\text{ eV})/\bar{F}^\uparrow(\omega=-7.0\text{ eV}) . \tag{8}$$

Equations (6'), (7), and (8) yield values for $\sum|g_\uparrow|^2$ and $\sum|G_\uparrow|^2$ that are similar to those found when interference was neglected. Generally $\sum|g_\uparrow|^2$ is larger and $\sum|G_\downarrow|^2$ smaller, the difference being 20% or less. Values for β are increased by roughly 20%. No statements can be made regarding D, which measures direct scattering, in the case that interference is included.

We have shown that a model that only allows DESE cannot explain the data of I and FESE must be included. It is found that DESE contribute to the scattering and their cross section has a similar dependence on energy loss to that expected from the appropriate joint density of states for creation of DESE. The corresponding statement holds for FESE. However, it is found that on average the cross section for FESE is somewhat larger than that for DESE, a very surprising result in view of the fact that the joint density of states for creation of DESE is much larger than that for FESE (due to the larger number of empty d states compared to empty free-electron-like states).

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[10] I gives data for specular scattering and for scattering angles of 5°, 10°, 20°, 30°, 40°, and 50°. The only negative values of Δ are found for large energy loss ($\omega > 4.4$ eV) in the 5° data, in which case $|\Delta|$ is small.

[11] It is suggested in Ref. 6 that the appropriate density of states is that with momentum corresponding to the scattering angle.