PHYSICAL REVIEW B VOLUME 35, NUMBER 4 1 FEBRUARY 1987

Evidence for free-electron-like Stoner excitations in Fe

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(Received 17 October 1986)

An analysis of spin-polarized electron-energy-loss experiments in Fe is described which suggests that (1) free-electron-like Stoner excitations are far more probable than the usual type and (2) exchange events involving relatively large energy losses are much more likely than 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 very recently have the techniques to probe the Stoner excitation spectrum been developed.[2-6] The methods have utilized electron-energy-loss spectroscopy[3] (EELS) 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).

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 carry out a detailed theoretical analysis of the spin-dependent scattering process. The analysis presented here concludes that free-electron-like Stoner excitations (FESE) contribute prominently to the scattering process. All of the experimental papers, Refs. 3–5 and I, 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 dominant contribution of FESE over DESE, is completely unexpected and requires a reassessment of the significance of I.

A FESE is an electron-hole pair excitation consisting of a d hole of 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 state which makes the dominance 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 and the polarization of the inelastic reflected beam was measured for energies from 0 to 4.5 eV below the incident energy and at angles 10° and 15° 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, $\overline{F}^{\sigma}$ (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 $\overline{N}^{\sigma}$ (nonflip) denotes the cross section for a spin-σ electron in and a spin-σ electron scattered out.

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 at least an order of magnitude less than the number of unfilled minority states and are 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 $\epsilon\sigma'$ 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 got 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 $0.5<\omega<4.5$ 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 far more probable than direct scattering.

The scattering amplitudes that describe these processes are given in Table I. For example, $f_{\sigma'}\equiv\mathscr{A}((i\sigma,d\sigma')\rightarrow(f\sigma,\varepsilon\sigma'))$ 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

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
$i\sigma$ $d\sigma'$	$f\sigma$ $\epsilon\sigma'$	$\mathcal{A}((i\sigma,d\sigma')\rightarrow(f\sigma,\epsilon\sigma'))$	$f_{\sigma'}$
$i\sigma$ $d\downarrow$	$f\sigma$ $d^*\downarrow$	$\mathcal{A}((i\sigma,d\downarrow)\rightarrow(f\sigma,d^*\downarrow))$	$F_{\downarrow}$
$i\sigma$ $d\sigma'$	$\epsilon\sigma$ $f\sigma'$	$\mathcal{A}((i\sigma,d\sigma')\rightarrow(\epsilon\sigma,f\sigma'))$	$g_{\sigma'}$
$i\downarrow$ $d\sigma'$	$d^*\downarrow$ $f\sigma'$	$\mathcal{A}((i\downarrow,d\sigma')\rightarrow(d^*\downarrow,f\sigma'))$	$G_{\sigma'}$

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.

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,

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

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

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

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

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

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

where

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

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

The data from I is shown in Fig. 1 for the scattering angles $\theta=10°$ and 15°. The flip and nonflip scattering cross sections are plotted versus energy loss ω. Values of Δ are found to be very small ($\Delta \ll \bar{F}^{\sigma},\bar{N}^{\sigma}$) and positive as required with an average value of 0.26 corresponding to an average value of $A(\omega)=0.05$. Because $\sum|G_{\sigma}|^2$ involves a sum over the states $d\sigma$ one expects $\sum|G_{\downarrow}|^2=R_d\sum|G_{\uparrow}|^2$ where R_d is the ratio of the number of minority to majority spin d states; $R_d\simeq\frac{3}{5}$. Since Δ is small, $\sum|G_{\sigma}|^2$ is small and any error introduced by this approximation will have only a very small effect on the values obtained for $\sum|g_{\sigma}|^2$ and D. These quantities as well as $\sum|G_{\sigma}|^2$ are now evaluated using Eqs. (2a) and (3) and the experimental values for $\bar{F}^{\sigma}$ and $\bar{N}^{\sigma}$. Results are shown in Fig. 2 for scattering angles $\theta=10°$ and 15°.

A number of interesting points are evident: (a) the exchange terms $\sum|g_{\sigma}|^2$ which include FESE are dominant; (b) $\sum|g_{\downarrow}|^2\approx R_d\sum|g_{\uparrow}|^2$ as should be the case; (c) the ratio of direct terms D to the exchange terms is $D/E\approx0.1$, where $E\equiv\sum|g_{\downarrow}|^2+\sum|g_{\uparrow}|^2+\sum|G_{\downarrow}|^2+\sum|G_{\uparrow}|^2$; (d) the DESE term $\sum|G_{\uparrow}|^2$ is very small, i.e., $\sum|G_{\sigma}|^2/\sum|g_{\sigma}|^2\simeq0.1$. Thus the observed scattering is primarily exchange scattering rather than direct scattering as is usually assumed. Furthermore, the contribution of DESE to the scattering is quite small.

On the other hand, if interference terms in Eq. (2) are kept very similar conclusions to (a)—(d) above can be reached by a different line of reasoning based on the energy dependence of the scattering amplitudes. Equation (2b) gives

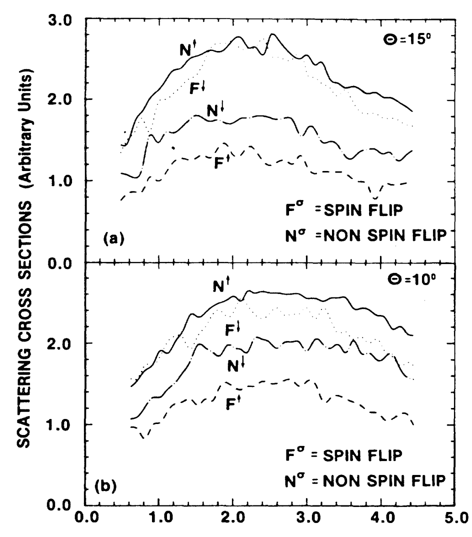

FIG. 1. Spin-dependent cross sections versus energy loss from Ref. 2 for scattering angles $\theta=10°$ and 15°. Spin-flip events are denoted by $\bar{F}^{\sigma}$ and nonflip events by $\bar{N}^{\sigma}$ where σ refers to the spin of the incident electron.

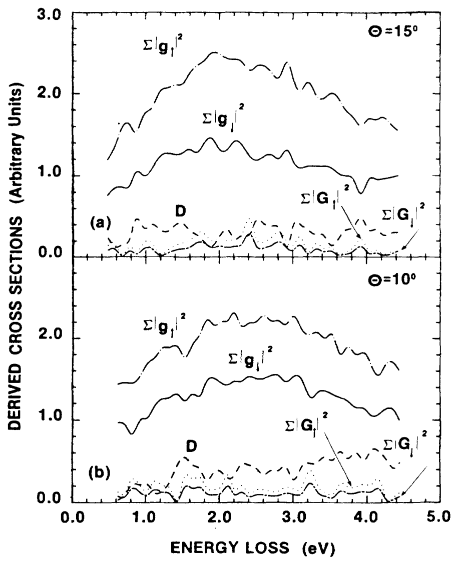

FIG. 2. Partial cross sections versus energy loss as determined by data analysis. See the text for definitions of the cross sections.

$$\bar{N}^{\sigma}=\sum|g_{\sigma}|^2-2\,\mathrm{Re}\sum f_{\sigma}g_{\sigma}^{*}+D$$
$$+\delta_{\sigma,\downarrow}\left[\sum|G_{\downarrow}|^2-2\mathrm{Re}\sum F_{\downarrow}G_{\downarrow}^{*}\right]. \tag{4}$$

The dependence of the Stoner type of terms $\sum|G_\sigma|^2$, $\sum F_\downarrow G_\downarrow^*$ on energy loss ω is expected to be quite different from that of the other terms D, $\sum|g_\sigma|^2$, $\sum f_\sigma g_\sigma^*$ due to the fact that the former terms involve creation of a d hole and a d^* electron while the latter correspond to creation of a d hole and an electron in a free-electron-like state ϵ. In both cases the excitation energy of the electron-hole pair is ω. It is expected[3] that the Stoner-type terms will peak at values of ω roughly equal to the exchange splitting. For example, if the matrix elements entering the scattering probabilities $\bar{F}^{\sigma}$ and $\bar{N}^{\sigma}$ are assumed to be independent of momentum then it follows[7] that these probabilities are related to the joint densities of states $\rho_{d\sigma}\rho_{d\downarrow}(\omega)$ and $\rho_{d\sigma}\rho_{\epsilon}(\omega)$. Thus $\sum|f_\sigma|^2$, $\sum|g_\sigma|^2$, and $\sum f_\sigma g_\sigma^*$ are proportional to $\rho_{d\sigma}\rho_\epsilon(\omega)$ while $\sum|F_\downarrow|^2$, $\sum|G_\sigma|^2$, and $\sum F_\downarrow G_\downarrow$ are proportional to $\rho_{d\sigma}\rho_{d\downarrow}(\omega)$.

In order to estimate the magnitudes of the terms in Eqs. (2a) and (4), we note that the shapes of $\rho_{d\sigma}\rho_\epsilon$ and $\rho_{d\sigma}\rho_{d\downarrow}$ as functions of ω are quite different. The ratio $(\rho_{d\sigma}\rho_{d\downarrow})/(\rho_{d\sigma}\rho_\epsilon)$ is obtained for Fe using the calculated densities of states.[8] This ratio peaks in the interval $1.5<\omega<2.5$ eV as expected.[5] The cross section $\bar{F}^{\uparrow}$ involves only a term of the type $\rho_{d\sigma}\rho_\epsilon$. Thus we examine $\bar{F}^{\downarrow}/\bar{F}^{\uparrow}$ and $\bar{N}^{\sigma}/\bar{F}^{\uparrow}$ as functions of ω and expect to find a peak due to contributions from terms of the type $\rho_{d\sigma}\rho_{d\downarrow}$ to $\bar{F}^{\downarrow}$ and $\bar{N}^{\sigma}$. Values of $\bar{F}^{\downarrow}/\bar{F}^{\uparrow}$ are shown in Fig. 3 for $\theta=10^\circ$ and 15°. The ratios $\bar{N}^{\sigma}/F^{\uparrow}$ are also independent of ω. There is no evidence for the presence of $\rho_{d\sigma}\rho_{d\downarrow}$ type terms so $\sum|F_\downarrow|^2$, $\sum|G_\sigma|^2$, and $\sum F_\downarrow G_\downarrow^*$ are neglected in Eq. (6). Now Δ defined in Eq. (4) has the value

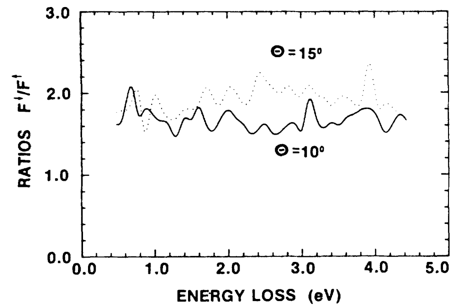

FIG. 3. Values of $\bar{F}^{\downarrow}/\bar{F}^{\uparrow}$ versus energy loss for scattering angles of $\theta=10^\circ$ and 15°.

$$\Delta=2\,\mathrm{Re}\sum f_\uparrow g_\uparrow^*-2\,\mathrm{Re}\sum f_\downarrow g_\downarrow^* .$$

Assuming $\sum f_\downarrow g_\downarrow^*\simeq R_d\sum f_\uparrow g_\uparrow^*$ allows $\sum|g_\sigma|^2$, D, and $\sum f_\sigma g_\sigma^*$ to be determined from the experimental data for $\bar{F}^{\sigma}$ and $\bar{N}^{\sigma}$. The values of $\sum|g_\sigma|^2$ are essentially as in Fig. 2 while D is two to three times larger. The previous conclusions again follow; exchange scattering is substantially larger than direct scattering and DESE scattering is very small.

The suppression of direct transitions must be due either to screening of the Coulomb interaction or matrix-element effects. The Coulomb matrix element for direct scattering goes like $1/q^2$ where q is the momentum transfer whereas the momentum transfer in the exchange process is much larger than q; thus the direct process is normally larger than the exchange event. However, the matrix element for direct scattering is also proportional to $1/|\epsilon(q,\omega)|$. Optical data shows that $|\epsilon(0,\omega)|$ is very large for small ω and this suggests that strong screening of $|\epsilon(q,\omega)|$ may account for the small values of the direct scattering. It is hoped that future theoretical calculations will pay special attention to these points. Furthermore, the possibility that the dominant scattering in an EELS experiment can be exchange in character must have a profound effect on the microscopic interpretation of EELS in the case where the energy loss is due to electronic excitation.

The small contribution of the DESE to the scattering follows from the small values of the scattering asymmetry, $A(\omega)$, reported in I; $A(\omega)\le 0.05$. Preferential scattering of spin-$\downarrow$ electrons is due to (a) the Pauli principle and (b) the large number of spin-$\downarrow$ empty d states relative to empty spin-$\uparrow$ d states into which a spin-$\downarrow$ electron can scatter. If a majority spin electron is excited in the latter process the result is a DESE. Thus, a very small value of $A(\omega)$ implies a small contribution of DESE to the scattering.

While the small contribution of DESE scattering is consistent with previous estimates[7] which find it to decrease rapidly with increasing energy E_0 and to be quite small for energies $E_0 > 5$ eV, a microscopic explanation of such a strong energy dependence is missing.

As the beam energy is increased it is expected that because of the large momentum transfer involved event the direct process will gain importance relative to exchange events while for low beam energies, $E_0 < 5$ eV, the usual type of DESE should become an important scattering mechanism. It is also suggested that other systems be investigated for the presence of FESE.

We are indebted to Professor M. L. Cohen for many valuable suggestions concerning the presentation of this data analysis. This work was supported in part by the Office of Naval Research.

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