Free electronlike Stoner excitations in Fe

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Methods have recently developed to probe the Stoner excitation spectrum which has not been amenable to study by neutron diffraction. The experiments have utilized energy loss spectroscopy combined with spin polarization of the incident electron beam or with spin polarization detection of the scattered beam, or both beam spin polarization and polarization detection [J. Kirschner, D. Rebenstorff, and M. Iback, Phys. Rev. Lett. **53**, 698 (1984) (denoted by I)]. Due to the many possible scattering processes the experiments do not measure the Stoner excitation cross section directly but rather measure the magnitudes of combination of scattering amplitudes. To draw even semiquantitative conclusions regarding the Stoner spectrum requires theoretical analysis. Because Glazer and Tosatti [Solid State Commun. **52**, 905 (1984)] give more complete information than previous experiments it is possible for the first time to carry out a detailed theoretical analysis. The analysis concludes that free electronlike Stoner excitations (FESE) make a much larger contribution to the scattering than d electron Stoner excitations (DESE), the usual type.

Recently experiments that probe the Stoner excitation spectrum have been reported.[1-4] The most complete one,[4] I, makes possible a theoretical analysis that concludes that free electronlike Stoner excitations (FESE) scattering is much larger than d electron stoner excitations (DESE) scattering. A FESE is an electron hole pair excitation consisting of a d hole and an electron of opposite spin in a free electronlike state. In a DESE the electron is in a d state. The dominance of FESE scattering over DESE in I is completely unexpected because (a) all the experimental papers interpret their results in terms of DESE and (b) the density of states for creating a FESE is much smaller than for a DESE due to the small density of free electronlike states relative to d states. It is also found that exchange scattering events dominate nonexchange events.

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 electronlike 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 state is very small and they are neglected. The majority and minority spin free electronlike 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 electronlike 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 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 $< \omega <$ 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 where E_0 is the energy of the incident beam.

The scattering amplitudes that describe these processes are given in Table I. For example $f'_\sigma \equiv A[(i\sigma,d\sigma') \to (f\sigma,\epsilon\sigma')]$ 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 electronlike 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 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) denote the cross

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

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

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section for a spin σ electron in and a spin σ electron scattered out.

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, $\overline{F}^{\sigma}$ and $\overline{N}^{\sigma}$, can be calculated in terms of the amplitudes given by Table I.

$$\overline{F}^{\downarrow} = \sum |A\,[(i\downarrow, d\uparrow) \to (\epsilon\downarrow, f\uparrow)]|^2$$

$$+ \sum |A\,[(i\downarrow, d\uparrow) \to (d^*\downarrow, f\uparrow)]|^2, \qquad (1)$$

where the summations are over the appropriate states $d\uparrow, e\downarrow$ and $d\uparrow, d^*\downarrow$, respectively and are such that energy, momentum, etc., are conserved. Use of Table I yields

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

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

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

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

where

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

All the direct transitions are contained in D and the DESE contribute only to $\overline{F}^{\downarrow}$.

The quantity $\Delta \equiv \overline{N}^{\downarrow} + \overline{F}^{\downarrow} - \overline{N}^{\uparrow} - \overline{F}^{\uparrow}$ is the unnormalized asymmetry and from Eq. (2a) and Eq. (3), $\Delta = \Sigma |G_{\uparrow}|^2 + \Sigma |G_{\downarrow}|^2$.

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 ($\Delta \ll \overline{F}^{\sigma}, \overline{N}^{\sigma}$) and positive as required with an average value of 0.26 corresponding to an average value of $A(\omega) = 0.05$. Because $\Sigma |G_{\sigma}|^2$ involves a sum over the states $d\sigma$ one expects $\Sigma |G_{\downarrow}|^2 \simeq R_d \Sigma |G^{\uparrow}|^2$ where R_d is the ratio of the number of minority to majority spin d states; $R_d \simeq 3/5$. Since Δ is small, $\Sigma |G_{\sigma}|^2$ is small and any error introduced will have only a very small effect on the values obtained for $\Sigma |g_{\sigma}|^2$ and D. These quantities as well as $\Sigma |G_{\sigma}|^2$ can now be evaluated using Eqs. (2a) and (3a) and the experimental values for $\overline{F}^{\sigma}$ and $\overline{N}^{\sigma}$. Results are shown in Fig. 2 for scattering angles $\theta = 10°$ and $15°$.

A number of interesting points are evident (a) the exchange terms $\Sigma |g_{\sigma}|^2$ which include FESE are dominant, (b) $\Sigma |g_{\downarrow}|^2 \approx R_d \Sigma |g_{\uparrow}|^2$ as should be the case, (c) the ratio of direct terms D to the exchange terms is $D/E \approx 0.1$ where $E \equiv \Sigma |g_{\downarrow}|^2 + \Sigma |g_{\uparrow}|^2 + \Sigma |G_{\downarrow}|^2 + \Sigma |G_{\uparrow}|^2$, (d) the DESE term $\Sigma |G_{\uparrow}|^2$ is very small, i.e., $\Sigma |G_{\sigma}|^2 / \Sigma |g_{\sigma}|^2 \simeq 0.1$. 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.

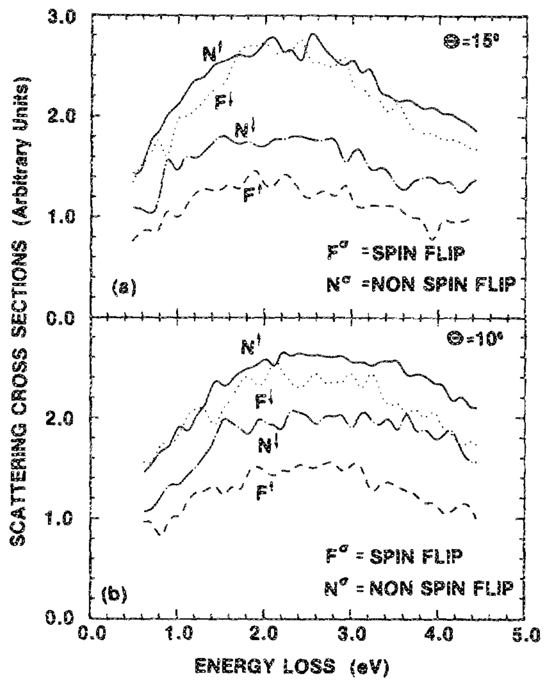

FIG. 1. Spin dependent cross sections vs energy loss from Ref. 4 for scattering angles $\theta = 10°$ and $15°$. Spin flip events are denoted by $\overline{F}^{\sigma}$ and nonflip events by $\overline{N}^{\sigma}$ where σ refers to the spin of the incident electron.

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

$$\overline{N}^{\sigma} = \Sigma |g_{\bar{\sigma}}|^2 + D - 2\,\mathrm{Re}\,\Sigma f_{\sigma} g_{\sigma}^*$$

$$+ \delta_{\sigma,\downarrow} (\Sigma |G_{\downarrow}|^2 - 2\,\mathrm{Re}\,\Sigma F_{\downarrow} G_{\uparrow}^*), \qquad (4)$$

where D is given by Eq. (3b). The dependence of the Stoner type of terms $\Sigma |G_{\sigma}|^2, \Sigma F_{\downarrow} G_{\uparrow}^*$ on energy loss ω is expected to be quite different from that of the other terms D, $\Sigma |g_{\sigma}|^2$, $\Sigma f_{\sigma} g_{\sigma}^*$ 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 electronlike 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 probabilities $\overline{F}^{\sigma}$ and $\overline{N}^{\sigma}$ are assumed to be independent of momentum then it follows[5] that these probabilities are related to the joint densities of states $\rho_{d\sigma} \times \rho_{d\uparrow}(\omega)$ and $\rho_{d\sigma} \times \rho_{\epsilon}(\omega)$. Thus $\Sigma |f_{\sigma}|^2$, $\Sigma |g_{\sigma}|^2$, and $\Sigma f_{\sigma} g_{\sigma}^*$ are proportional to $\rho_{d\sigma} \times \rho_{\epsilon}(\omega)$ while $\Sigma |F_{\downarrow}|^2$, $\Sigma |G_{\sigma}|^2$, and $\Sigma F_{\downarrow} G_{\uparrow}^*$ are proportional to $\rho_{d\sigma} \times \rho_{d\downarrow}(\omega)$.

In order to estimate the magnitudes of the terms in Eqs. (2a) and (4) we note that the shapes of $\rho_{d\sigma} \times \rho_{\epsilon}$ and $\rho_{d\sigma} \times \rho_{d\downarrow}$ as functions of ω are quite different. The ratio $(\rho_{d\sigma} \times \rho_{d\downarrow})/(\rho_{d\sigma} \times \rho_{\epsilon})$ is obtained for Fe using the calculated densities of states.[6] This ratio peaks in the inverval 1.5 eV $< \omega <$ 2.5 eV as expected.[3] The cross section $\overline{F}^{\uparrow}$ involves only a term of the type $\rho_{d\sigma} \times \rho_{\epsilon}$. Thus we examine $\overline{F}^{\downarrow}/\overline{F}^{\uparrow}$ and $\overline{N}^{\sigma}/\overline{F}^{\uparrow}$ as functions of ω and expect to find a peak due to contributions from terms of the type $\rho_{d\sigma} \times \rho_{d\downarrow}$ to $\overline{F}^{\downarrow}$ and $\overline{N}^{\sigma}$.

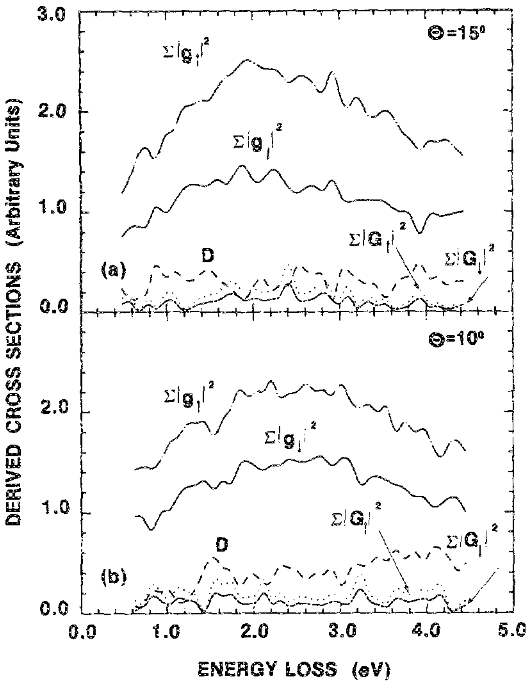

FIG. 2. Partial cross sections vs energy loss as determined by data analysis. See the text for definitions of the cross sections.

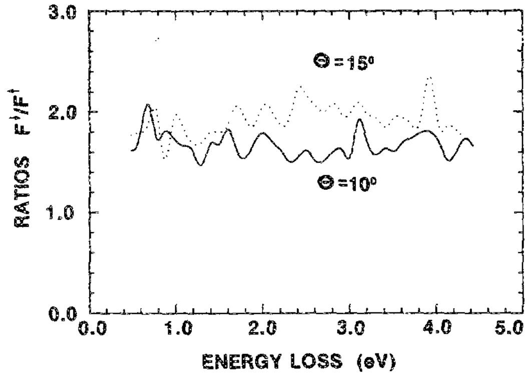

FIG. 3. Values of $\overline{F}^\downarrow/\overline{F}^\uparrow$ vs energy loss for scattering angles of $\theta = 10°$ and 15°.

Values of $\overline{F}^\downarrow/\overline{F}^\uparrow$ are shown in Fig. 3 for $\theta = 10°$ and 15°. The ratios $\overline{N}^\sigma/\overline{F}^\uparrow$ are also independent of ω. There is no evidence for the presence of $\rho_{d\sigma} \times \rho_{d\downarrow}$ type terms so $\Sigma|F_\downarrow|^2$, $\Sigma|G_\sigma|^2$, and $\Sigma F_\downarrow G_\uparrow^*$ are neglected in Eq. (6). Now Δ has the value

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

Assuming $\Sigma f_\downarrow g_\uparrow^* \simeq R_d \Sigma f_\uparrow g_\uparrow^*$ allows $\Sigma|g_\sigma|^2$, D, and $\Sigma f_\sigma g_\sigma^*$ to be determined from the experimental data for $\overline{F}^\sigma$ and $\overline{N}^\sigma$. The values of $\Sigma|g_\sigma|^2$ 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, it is hoped that future theoretical calculations will pay special attention to these points.

The small contribution of the DESE to the scattering follows from the small values of the scattering asymmetry, $A(\omega)$ reported in I; $A(\omega) \leqslant 0.05$. Preferential scattering of spin ↓ electrons is due to (a) the Pauli principal and (b) the large number of spin ↓ empty d states relative to empty spin ↑ d states into which a spin ↓ 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[5] 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.

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