Direct and Exchange Contributions in Inelastic Scattering of Spin-Polarized Electrons from Iron

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I report on the first complete experimental characterization of elastic and inelastic two-electron scattering processes in a ferromagnet. By explicit measurement of the spin polarization of primary and scattered electrons the contributions of all four partial scattering rates are determined. The Stoner continuum in Fe at finite q is observed for the first time.

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Elementary excitations in ferromagnetic crystals are classified into collective excitations (spin waves) and electron-hole pair excitations with spin flip (Stoner excitations). Spin waves have been the realm of neutron scattering,[1] while the region of the Stoner excitations has in general been out of reach for experimental reasons. Despite its importance for the theory of ferromagnets the "Stoner continuum" has therefore largely remained *terra incognita* in energy-momentum space. Recently, it has been shown that Stoner excitations can be probed via exchange processes in inelastic scattering of low-energy spin-polarized electrons.[2–4] In particular, for Ni it was shown[2] that the average exchange splitting and its abundance distribution over the Fermi surface can be obtained to first order from intensity asymmetries of the specular beam (corresponding to momentum transfer $q=0$) on reversal of the spin orientation of the incident electrons. This was possible because one particular inelastic scattering channel (among four) plays a dominant role for the structure of the asymmetry function at $q=0$ in Ni. At arbitrary wave vector and in other materials all four channels, to be discussed below, have to be taken into account. This was not possible by the experimental technique(s)[2,3] used previously, since the spin states of primary electrons and scattered electrons were not known simultaneously. In this Letter, I report on a new experiment, which, for the first time, allows one to obtain all the necessary information at finite **q**, and demonstrates the general applicability of spin-polarized inelastic electron scattering for the investigation of Stoner excitations.

When exchange processes are taken into account, the dielectric theory of electron-energy-loss spectroscopy is no longer applicable and the two-electron character of the energy transfer process has to be considered explicitly. Let us assume that we have a source of completely polarized electrons and a perfect spin analyzer for the scattered electrons. Depending on the spin orientation of the primary electrons [$\uparrow$ (up) means parallel to the majority spin orientation] we may observe the four different processes (a)–(d) presented in Fig. 1, where we assume the spin of an individual electron to be conserved in the scattering process. For incident up electrons we may find either scattered electrons with down spin [with the intensity $F^\uparrow$, process (a)] or with up spin [with the intensity $N^\uparrow$, process (c)]. Conversely, for primary down-spin electrons we may find up-spin electrons with the rate $F^\downarrow$ [process (b)] or down-spin electrons with rate $N^\downarrow$ [process (d)]. The transition rates pertinent to (a) and (b) are called "flip rates" ($F^\uparrow, F^\downarrow$)[5] because the detected electron has its spin opposite to the incident one. Correspondingly, processes (c) and (d) are described by the "nonflip rates" $N^\uparrow$ and $N^\downarrow$, respectively. For each of the nonflip rates there are two possible electron-hole pair configurations near the Fermi level. For the ones shown in parentheses the primary

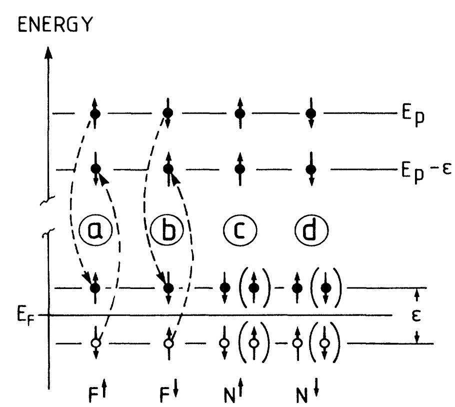

FIG. 1. Schematic representation of two-electron inelastic scattering processes with the spins included. The final states in processes (a) and (b) represent electron-hole pairs with opposite spins (Stoner excitations). Because of the exchange processes, indicated by dashed lines, the inelastically scattered electron with energy $E_p - \epsilon$ appears to have its spin flipped with respect to the primary electron of energy E_p. The corresponding transition rates are termed "flip rates" ($F^\uparrow, F^\downarrow$), in contrast to the "nonflip rates" ($N^\uparrow, N^\downarrow$) where the scattered electron has the same spin as the primary electron.

electron and the one to be ejected from below E_F form a triplet state. For low primary energy we expect the matrix element for triplet scattering to be small, since the direct and the exchange amplitudes in the matrix element cancel for pure s-wave scattering. We emphasize that in the flip processes there is no real spin reversal, but that the apparent spin flip is due to exchange processes. For example, in case (a) the primary up-spin electron transfers its energy (via Coulomb interaction) to a minority electron in an occupied band and finds itself a place in an empty majority-type band above the Fermi level. The energetic difference ϵ between the two band states corresponds to the measured energy loss, while their difference in $\mathbf{k}$ vectors is reflected in the momentum change $\mathbf{q}$ of the detected electron. The momentum transfer is measured with respect to the specular beam. (The momentum needed to turn around the electron is provided by the crystal as a whole.) The configuration around the Fermi level after the scattering process is precisely that of a Stoner excitation: an electron-hole pair with opposite spins and definite momentum. If the corresponding flip rates $F^{\uparrow(\downarrow)}$ are measured separately it is, furthermore, possible to distinguish between the Stoner configuration "majority-hole with minority-electron" (from $F^{\downarrow}$) and the configuration "minority-hole with majority electron" (from $F^{\uparrow}$).

It is clear from the above that an experiment aimed at a complete determination of all four scattering channels must comprise a source of polarized electrons, a single-domain magnetic sample, an energy- and momentum-resolving spectrometer, and a spin analysis of the scattered electrons. The apparatus built for this purpose will be described in detail elsewhere; here only the essential ingredients are briefly mentioned. The primary beam is formed from photoelectrons from GaAsP(100), excited by circularly polarized HeNe laser light. Use has been made of a previous observation[6] that by adjustment of the vacuum level, fairly monochromatic electrons can be generated (less than 50 meV FWHM has been reported[6,7]). The longitudinally polarized electron beam hits an Fe(110) crystal which is magnetically shortcircuited by a toroidal iron yoke. In this way magnetic stray fields are suppressed, which allows work at low energies where exchange interaction is strongest. A description of these components may be found in an earlier paper.[8] The scattered electrons are accepted by an energy-, momentum-, and spin-resolving spectrometer system, similar to the one used recently in spin-polarized photoemission.[9] It consists of a transport lens, cylindrical-mirror type analyzer, and a LEED spin detector.[10] The angular resolution is about $\pm 2°$, and two components of the polarization vector are measured simultaneously. The overall energy resolution of the experiment is about 0.4 eV FWHM which is dictated by intensity. The polarization and magnetization vectors both lie in the scattering plane, which coincides with a mirror plane of the crystal. In this way, spin-orbit polarization effects are effectively suppressed.[11]

For each orientation of the primary spin polarization relative to the magnetization of the sample (upper index) the spin detector yields two intensities $j_{\uparrow}^{\uparrow(\downarrow)}(\epsilon)$ and $j_{\downarrow}^{\uparrow(\downarrow)}(\epsilon)$ of the two spin-sensitive diffracted beams (lower index). If source and detector were ideal, these four intensities would yield directly the desired four spin-dependent scattering rates. The nonideal behavior of source and detector can be described by (2×2) matrices $\mathbf{Q}$ and $\mathbf{D}$, respectively. With these and the unknown transition-rate matrix $\mathbf{R}(\epsilon)$ the resultant intensity matrix $\mathbf{J}(\epsilon)$ is given by

$$\mathbf{J}(\epsilon)=\mathbf{D}\mathbf{R}(\epsilon)\mathbf{Q}, \tag{1}$$

with

$$\mathbf{J}(\epsilon)=\begin{pmatrix} j_{\uparrow}^{\uparrow}(\epsilon) & j_{\uparrow}^{\downarrow}(\epsilon) \\ j_{\downarrow}^{\uparrow}(\epsilon) & j_{\downarrow}^{\downarrow}(\epsilon)\end{pmatrix}, \quad \mathbf{R}(\epsilon)=\begin{pmatrix} N^{\uparrow}(\epsilon) & F^{\downarrow}(\epsilon) \\ F^{\uparrow}(\epsilon) & N^{\downarrow}(\epsilon)\end{pmatrix},$$
$$\mathbf{D}=\frac{1}{2}\begin{pmatrix} 1+A & 1-A \\ 1-A & 1+A\end{pmatrix}, \quad \mathbf{Q}=\frac{1}{2}\begin{pmatrix} 1+P & 1-P \\ 1-P & 1+P\end{pmatrix}, \tag{2}$$

where A is the polarization sensitivity of the detector [$A=-0.27\pm 0.02$ (Ref. 10)] and P the effective polarization of the source [$P=P_0 \sin\theta$ with $P_0 = 0.35 \pm 0.03$ (Ref. 6)]. This system of linear equations can be solved by

$$\mathbf{D}^{-1}\mathbf{J}(\epsilon)\mathbf{Q}^{-1}=\mathbf{R}(\epsilon), \tag{3}$$

which yields the desired flip and nonflip transition rates as a function of the energy loss ϵ.

Experimental results, after application of the above corrections, are shown in Fig. 2 for $\theta = 55°$ (corresponding to 10° off the specular beam), and in Fig. 3 for $\theta = 60°$ (15° off specular). These scattering conditions were chosen in order to demonstrate the feasibility of measurements far out in the Brillouin zone. The contribution from higher-order processes of the type "elastic scattering with momentum transfer plus energy loss with $q \sim 0$" can be neglected because the loss probability is small. The data representation is such that the sum of all rates gives the total scattered intensity. Each of the shaded areas represents the contribution from the process indicated.

First, we note that in the loss region the sum of the nonflip rates is nearly equal to the sum of the flip rates. This shows that the Stoner excitations (i.e., those involving a "spin flip") play a substantial role in the energy-loss process, also at finite momentum transfer. Spin-polarized electron-energy-loss spectroscopy apparently is a suitable technique to study them.

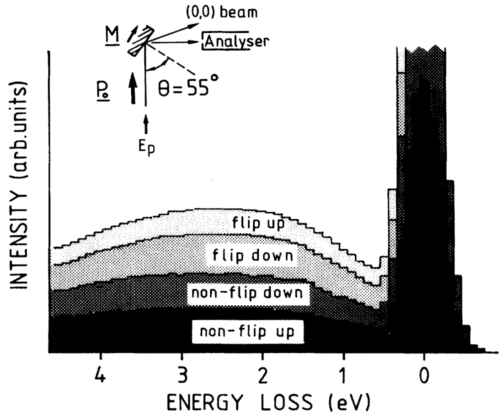

FIG. 2. Flip and nonflip rates as a function of energy loss. The inset shows the scattering geometry. The sample magnetization is along [100]. The momentum transfer is $|\mathbf{q}| \sim 0.8$ Å^{-1} at $E_p \sim 21$ eV.

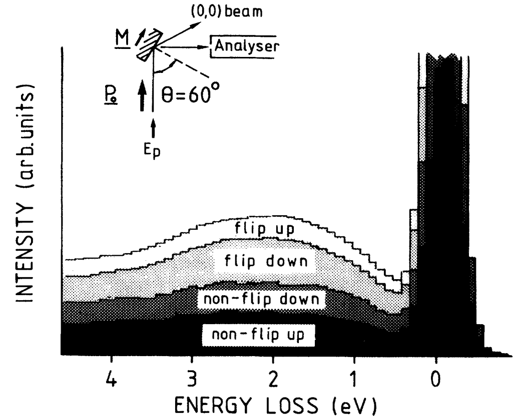

FIG. 3. Flip and nonflip rates like in Fig. 2 for $\theta = 60°$, corresponding to $|\mathbf{q}| \sim 1.2$ Å^{-1}. The total intensity has been scaled to equal that in Fig. 2 at about 2-eV energy loss.

Secondly, we see that the two nonflip rates $N^{\uparrow}$ and $N^{\downarrow}$ are similar in both cases. Since electron and hole have the same spin, this means that the electron-hole pair production within each of the two spin systems is to first order independent of the spin of the incoming electron. A closer inspection, however, reveals that the nonflip up rate is somewhat larger than the nonflip down rate in both figures. This means that an incoming electron of majority type has, to second order, a larger probability to lose energy in a nonflip process than a minority electron. This can be made plausible by recalling from above that the configurations set in parentheses in Fig. 1 will occur with less probability. If we neglect them completely,[12] $N^{\uparrow}$ will be favored over $N^{\downarrow}$ simply because there are more empty minority states available than majority states. This simple argument "explains" what we see by indicating a general trend, but the details of this second-order effect are expected to depend on the primary energy, the energy loss, and the momentum transfer.

Finally, we turn to the flip rates in Figs. 2 and 3, which represent the Stoner excitations. They have a broad peak between 2 and 2.5 eV energy loss, i.e., approximately at the value of the average exchange splitting in Fe. The spectra extend down to zero (below 0.5 eV the data are unreliable because of the large quasielastic peak), and also far above 4 eV. This demonstrates that the Stoner continuum covers a large energy range at finite $\mathbf{q}$, whereas for $q \sim 0$ in Ni it was found to be quite narrow (~ 0.3 eV FWHM). This behavior is consistent with expectation, since for large momentum transfer $\mathbf{q}$ the transitions between the nearly parallel exchange-split bands become less predominant over those between different bands farther away from the Fermi level. Comparing flip up ($F^{\uparrow}$) and flip down ($F^{\downarrow}$) rates in Figs. 2 and 3, we find the flip down rates consistently to be larger than the flip up rates. This is again because there are more minority states than majority states above the Fermi level (and more majority electrons below, of course), which favors electron-hole pairs of the type "minority electron with majority hole." Pairs of the opposite type occur less frequently, but they are not negligible because in Fe a considerable part of the majority bands extends beyond E_F. Furthermore, the experiment shows that their relative contribution depends on the momentum transfer $\mathbf{q}$. By contrast, in Ni at $q \sim 0$, this rate $F^{\uparrow}$ could be neglected altogether,[2] since Ni is a saturated ferromagnet with very few empty d-band states of majority type.

Since the energy loss of the electrons is small relative to the primary energy the data in Figs. 2 and 3 essentially represent "constant-q scans" in ϵ-q space, even at fixed scattering angle. The flip rates may therefore be taken as vertical cuts through the Stoner continuum in Fe, at $|\mathbf{q}| \sim 0.8$ Å^{-1} (Fig. 2) and $|\mathbf{q}| \sim 1.2$ Å^{-1} (Fig. 3), i.e., about halfway to the Brillouin-zone boundary. The spectral functions of the corresponding flip rates differ notably in magnitude and shape. These reflect qualitatively the different structure of the Stoner continuum for different constant-q scans, which may result from structure in the "Stoner densities of states" i.e., joint densities of states with finite momentum transfer and opposite spin character. A theoretical Stoner density of states for Fe, calculated by Cooke, Lynn, and Davis,[13] is not directy comparable to the experimental data since the momentum transfer is different. However, the width of this Stoner density of states and the energetic position are consistent with the present data. It is hoped that more theoretical data will become available.

Perhaps, spin-polarized electrons may become for

Stoner excitations what neutrons have been for spin waves.

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