Spin-Flip Stoner Excitations in a Ferromagnet Observed by Inelastic Spin-Polarized Electron Scattering

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(Received 12 September 1983)

It is shown by electron-energy-loss spectroscopy with spin analysis that electrons inelastically scattered from a ferromagnet ($Fe_{82}B_{12}Si_6$) can have a high spin polarization as a result of exchange scattering. A maximum of the spin polarization occurs around 2.2-eV energy loss corresponding to the ferromagnetic exchange splitting. This experiment offers new possibilities for studying spin-flip Stoner excitations in ferromagnets.

PACS numbers: 75.30.Et, 71.70.Gm, 75.50.Kj, 79.20.Kz

Over the last decade electron-energy-loss spectroscopy (EELS) on metal surfaces (in the low-energy-loss regime) has mainly concentrated on vibrational spectroscopy of adsorbates.[1] Little attention has been paid to the electronic excitations of the substrate which are present as a continuous background. Only very recently have there been some studies on low-energy excitations on clean metal surfaces.[2,3] In electron scattering from a ferromagnetic sample the excitation of especially magnons and electron-hole pairs is expected to lead to spin-polarization effects because of the imbalance of the number of spin-up and spin-down electrons in the target. Bringer *et al.*[4] and Yin and Tosatti[5] have recently suggested that important spin-polarization effects should occur for electron-hole pair excitations. Glazer and Tosatti[6] have further pointed out that spin-flip excitations across the Stoner gap can be generated and can be observed both in spin-polarized and in ordinary EELS, and they should be characterized by a strong primary-electron energy dependence. Stoner excitations are fundamental in the theory of itinerant-electron ferromagnetism, although they have never been observed directly. In this Letter we show that Stoner excitations are readily observed by inelastic electron scattering with spin analysis. A ferromagnetic glass, $Fe_{82}B_{12}Si_6$, was studied, since it is easily magnetized and has a large ferromagnetic exchange splitting between spin-up and spin-down states.

The experiments were performed with the apparatus previously described.[7] The sample, a ribbon of $80\times9\times0.5$ mm^3, was mechanically clamped together to form a circular loop with the ends overlapping a few millimeters (see Fig. 1). In this way it forms a closed magnetic circuit and it can readily be magnetized by a current pulse through a coil wrapped around it. This sample geometry has also been used successfully in a recent experiment involving scattering of primary spin-polarized electrons.[8] The sample was cleaned by 1.5-keV Ne-ion bombardment, initially for several hours, until a sharp Fermi edge developed in the He I photoemission spectrum and no contamination was detected. The source of the primary electrons is a commercial LEED/Auger electron gun. The overall energy resolution (source and energy analyzer) is 650 meV (full width at half maximum). The electrons impinged on the surface at 30° off normal. The scattered electrons were analyzed for normal exit from the sample. The scattering angle is thus 150°. The spin polarization of the scattered electrons is due to magnetic effects only, because spin-orbit–induced polarization effects were averaged out by reversing the magnetization of the sample.

Figure 2 (lower panel) shows the typical, rather smooth intensity distribution curve in the range of energy loss from 0 to 12 eV for 180 eV primary beam energy. No significant structure is observed in this energy range. The two upper panels of Fig. 2 show the measured spin-polarization spectra for two different primary energies. For the *elastically* scat-

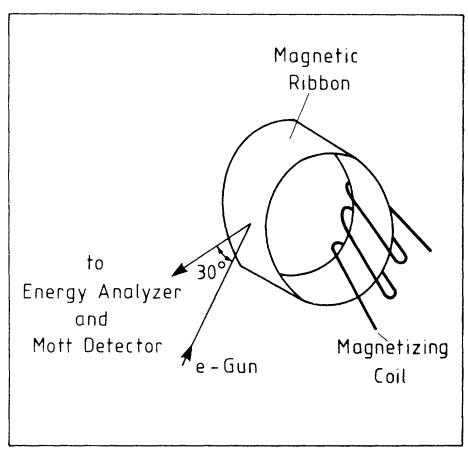

FIG. 1. Schematic of the scattering geometry.

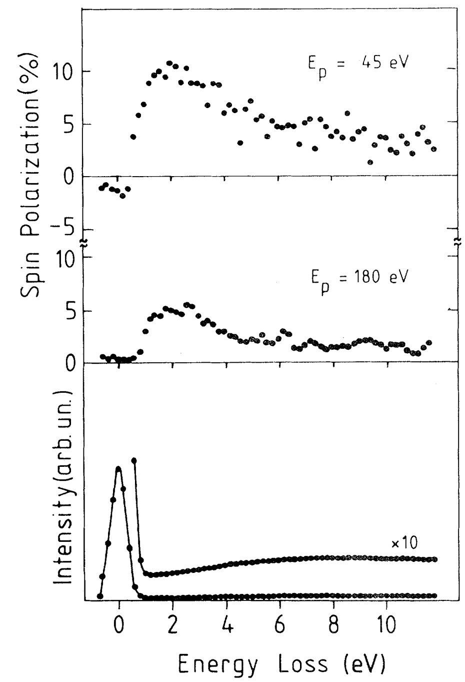

FIG. 2. Energy distribution curve (lower panel) and spin-polarization spectra for two different primary energies (upper panels) in the energy-loss range 0–12 eV.

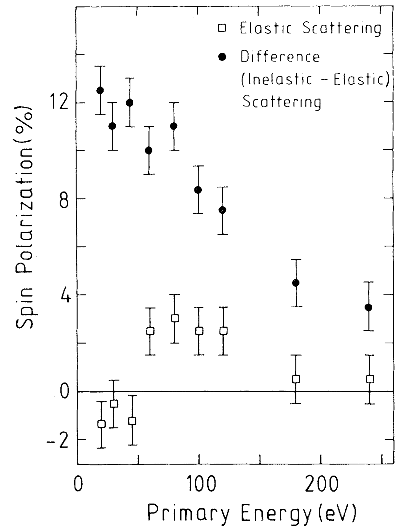

FIG. 3. Spin polarization for elastic scattering (squares) and difference between the maximum around 2.2-eV energy loss and the elastic polarization (circles) as a function of primary energy.

tered electrons we find spin-polarization values of a few percent, positive or negative, depending on the primary energy.[9] In the regime of *inelastic* scattering the spin polarization shows a maximum between 2- and 2.5-eV energy loss falling off smoothly towards higher energy loss. The maximum value of the spin polarization for inelastic scattering depends on the primary energy while the general shape of the spin-polarization curve is the same over the range of primary energies investigated. This insensitivity to primary energy indicates that surface effects are not important in the present experiment. In Fig. 3 we show the spin polarization for elastic scattering, and the difference between the maximum value around 2.2-eV energy loss and the value for elastic scattering. The error bars have different origins for the elastic and inelastic case. For the inelastic case they represent the statistical error of the difference measurement, whereas for the absolute value for the elastic case they are related to apparatus asymmetries arising upon reversal of the magnetization of the sample. This is one reason why we have plotted the difference. Another reason why the difference is the more relevant quantity is that it is generally thought that the inelastic scattering through large angles consists of a small-angle inelastic scattering event preceded or followed by large-angle elastic scattering. In this way the spin polarization for elastic scattering is the value to which the inelastic spin polarization should be referenced. Note that although the experiment is performed with angle resolution the diffuse elastic scattering from the glassy sample does not allow us to obtain angular information on the inelastic scattering only. This information can only be obtained on a single-crystalline sample.

Over the energy-loss range of interest the main energy-loss mechanism is electron-hole pair excitation. These processes can take place with or without spin flip. In Fig. 4 we show the major exchange process in a spin-polarized density-of-states diagram. A primary spin-down electron falls into the empty part of the minority-spin density and excites a spin-up electron from below E_F. The opposite process, i.e., spin flip for a primary spin-up electron, has comparatively low probability because, as indicated by spin-polarized photoemission,[10] the majority-spin states are nearly filled. Therefore a positive spin polarization is expected to result, in accordance with the experimental observation. The process shown in Fig. 4 is expected to have the highest weight for an energy loss where the primary electron falls into the maximum of the minority-spin density just above E_F and excites an electron from the maximum of the majority-spin density of states. This energy loss corresponds then to the ferromagnetic exchange splitting. The observed maximum in the spin polarization around 2.2-eV

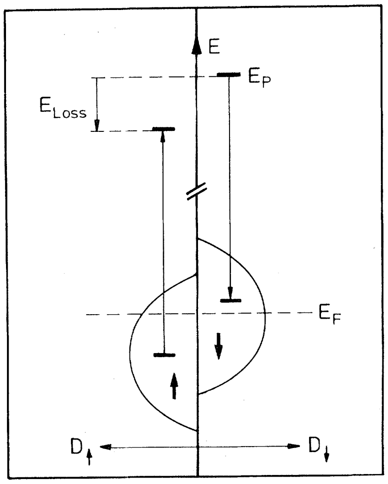

FIG. 4. Inelastic spin-flip process in a spin-polarized density-of-states diagram.

energy loss is actually in good agreement with the exchange splitting of Fe measured by photoemission.[11] Of course the observed value for the spin polarization depends on the relative strength of the spin-flip to nonflip excitations. In a model calculation Glazer and Tosatti[6] find a rather good agreement for the shape of the spin-polarization curve. They also emphasize that spin-flip processes (via exchange) are distinguished by a strong dependence on primary energy. With increasing primary energy the exchange process becomes less probable, in agreement with the experimental data.

Finally we mention two recent related experiments. Mauri, Allenspach, and Landolt[12] observed spin-polarization effects (due to exchange scattering) in core-level excitations. Kirschner, Rebenstorff, and Ibach[13] showed that spin-flip excitations in single crystalline Ni can also be observed by scattering of spin-polarized *primary* electrons, an experimental approach complementary to our technique. These results demonstrate the general importance of exchange scattering in EELS. Therefore we conclude that spin-polarized inelastic electron scattering obviously offers a new possibility for studying the electronic structure of ferromagnetic materials, a technique complementary to angle-resolved spin-polarized photoemission and inverse photoemission.

In summary, we have identified spin-flip Stoner excitations in a ferromagnet by spin-polarized electron-energy-loss spectroscopy with spin analysis. We have shown that it is possible to measure exchange splittings also in amorphous ferromagnets, a case where photoemission fails to do so because of the rather structureless density of states. Angle- as well as temperature-dependent measurements on single crystals appear to be feasible. These experiments promise to yield new insights into the electronic structure of ferromagnets (e.g., also for ferromagnetic alloys).

We thank H. J. Güntherodt for providing us with the sample, M. Campagna and G. Güntherodt for continuous interest and support, and J. Kirschner for communicating results prior to publication. We acknowledge helpful discussions with S. Bader, A. Bringer, R. Feder, J. Glazer, and E. Tosatti. This project has been supported in part by the Deutsche Forschungsgemeinschaft through SFB125.

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[11]The interpretation as a pure density-of-states effect according to Fig. 4 should be taken *cum grano salis.* In a single crystal the excitations (for small energy loss) have to be interpreted in terms of almost vertical transitions ($q=0$) in a band structure since in EELS small–momentum-transfer processes are strongly favored. How this picture has to be modified for an amorphous sample is not quite clear. But since there is considerable short-range order also in the glassy state the interpretation in terms of an (in this case rather ill-defined) band structure as was done in Ref. 6 can still be expected to apply.

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