Ferromagnetic Stoner excitations detected by electron-energy-loss spectroscopy

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Spin-flip Stoner excitations have been detected in $Fe_{80}B_{15}Si_5$ using simple electron-energy-loss spectroscopy via the strong energy dependence of the exchange process involved. The Stoner-excitation contribution, obtained as the difference between energy-loss spectra taken with low-energy and high-energy primary electrons, is in good agreement with the results of spin-polarized electron-energy-loss spectroscopy and with theoretical predictions.

The exchange splitting between the d bands in a ferromagnetic transition metal is bridged by the so-called Stoner excitations, i.e., spin-flip electronic transitions from the majority, up spin band, to the minority, down spin band. Despite their importance for the understanding of ferromagnetism, these excitations have been experimentally inaccessible for a long time. Very recently, progress has been made by two of us[1] (GT), who, based on the earlier approach of Soe Yin and Tosatti,[2] have identified theoretically an exchange mechanism by which an external electron couples directly to the Stoner excitations, and by Hopster, Raue, and Clauberg,[3] who have unambiguously detected them, via this mechanism, in $Fe_{82}B_{12}Si_6$. Independently, Kirschner, Rebenstorff, and Ibach[4] have also reported direct evidence for Stoner excitations in Ni. [Earlier electron-energy-loss spectroscopy (EELS) data of Colavita *et al.*[5] and of Ibach and Lehwald[6] were probably also pertinent, but had been interpreted in terms of dipole transitions.] In the spin-polarized electron-energy-loss (SPEELS) experiments, the Stoner excitations are reflected as a peak of spin polarization of the inelastically scattered electrons,[3] or as an enhanced inelastic scattering rate for incoming spin down electrons,[4] when the energy loss roughly equals the Stoner gap. What happens is that a spin-down primary electron (energy E_p) can fall into an empty down d band at an energy, say, $E_F+\Delta/2$, and very efficiently kick out another d-band electron of spin up and energy $E_F-\Delta/2$ to a final energy $E_p-\Delta$ (see Fig. 1). Obviously, the same process is not available for a spin-up primary, which leads one to expect an excess of up spins at $E_p-\Delta$, as indeed is seen.

This idea, which is the essence behind GT's model calculation, actually leads to other interesting predictions. The first is that the (momentum-integrated) total cross section of this process should be a strongly decreasing function of the primary electron energy E_p. This was observed experimentally over an extended energy range in Ref. 3. The second prediction is that Stoner excitations should be observable also in simple electron-energy-loss spectroscopy (EELS), *without any spin analysis at all.* In the GT model the calculated EELS spectrum develops a hump around $\omega=\Delta$, whose magnitude strongly decreases at high primary energies, which is the typical signature of an exchange process.

In this Brief Report we report fresh EELS data on ferromagnetic glasses, which demonstate conclusively that Stoner excitations are indeed directly detectable even without spin analysis, as predicted by GT. The resolution of our EELS apparatus is only 0.5 eV full width at half maximum (FWHM), which rules out Ni for a test case. It is, however, well suited for iron, or an iron-based ferromagnetic glass. For the purpose of direct comparison with the SPEELS data of Hopster *et al.*,[3] we have therefore used a ferromagnetic glass, $Fe_{80}B_{15}Si_5$, which is virtually identical to $Fe_{82}B_{12}Si_6$ used by Hopster *et al.* The sample is cleaned by Ar^+ sputtering to reduce the level of surface contamination (mostly C) down to ~ 0.05 monolayers. The EELS geometry used is an incidence angle of 60°, exit normal to the surface. No angular resolution has been attempted, and our results are therefore momentum-integrated loss spectra. Several checks have been made to ensure the absence of geometrical effects, owing to, e.g., residual magnetization.

The raw EELS data, normalized to the elastic peak inten-

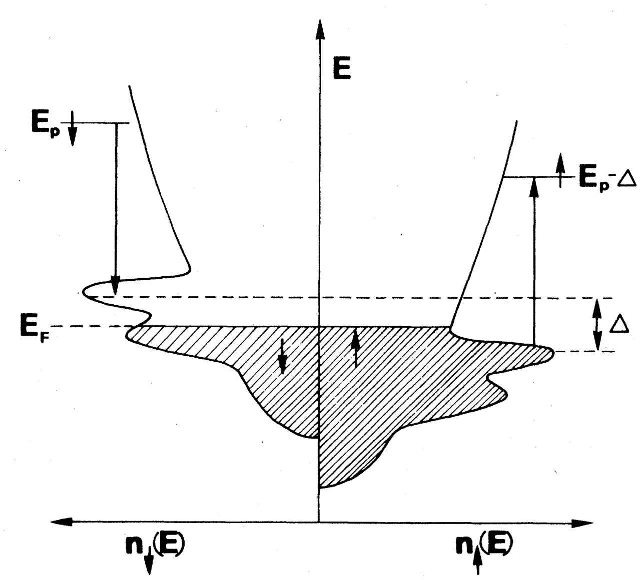

FIG. 1. Stoner excitation by electron-energy-loss process in a ferromagnetic material.

sities, are shown in Fig. 2. There is a generalized decrease of loss intensity with increasing primary energy E_p, which is probably to be attributed to elastic scattering. The feature of interest here is the broad shoulder centered around 2 eV. This feature, though not very prominent, is unique, in that it depends much more strongly upon E_p than the rest of the spectrum. In order to elucidate more quantitatively the primary energy dependence of this low-energy feature, we have rescaled out data so that they all coincide at some higher energy loss, chosen somewhat arbitrarily to be $\omega \simeq 8$ eV, a value where the shape of the loss spectrum is essentially independent of primary energy. We can then use the loss spectrum for some very high primary energy E_∞, as a reference, and plot directly the difference spectra $D(E_p,E_\infty,\omega)=\tilde{I}(E_p,\omega)-\tilde{I}(E_\infty,\omega)$, where $\tilde{I}$ are the rescaled data. The results, shown in Fig. 3 for $E_\infty = 1000$ eV and $E_p = 45$ eV, are now very suggestive. At low primary energies, some extra loss mechanism exists, which is peaked around 2 eV, exactly where the Stoner excitations are known to lie.[1,3] There is a close similarity, furthermore, between our difference spectrum $D(\omega)$ and the spin-polarization spectrum $S(\omega)$ measured by Hopster *et al.*[3] Direct comparison with the calculations of GT is also made, by taking from their work the difference between the loss spectra with and without *d* bands, for $E_p = 45$ eV. This comparison, given in Fig. 3, is also satisfactory, given the crudeness of the subtraction made. This result demonstrates that spin-flip excitations can be clearly brought out, at least in $Fe_{82}B_{12}Si_6$, even without spin analysis, by just taking the difference of EELS spectra taken at low and high primary energies.

As a final check of this point, we have studied in detail the primary energy dependence in comparison with both

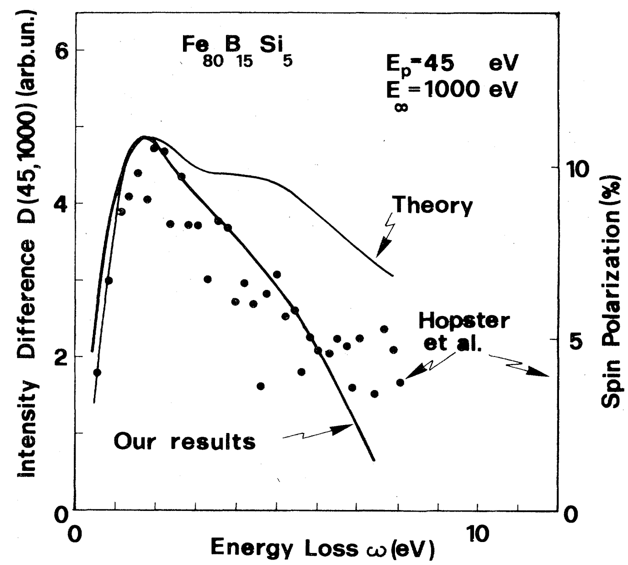

FIG. 3. Difference between the normalized energy-loss spectra at $E_p = 45$ eV and $E_p = 1000$ eV. Thick solid line: our results; dots: spin polarization spectrum of Hopster *et al.* (Ref. 3); thin solid line: contribution of Stoner excitations to the energy loss at $E_p = 45$ eV calculated by Glazer and Tosatti (Ref. 1).

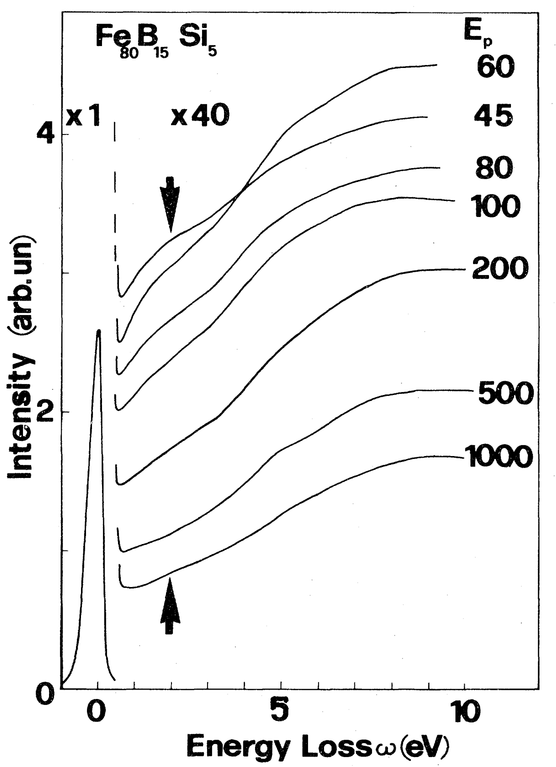

FIG. 2. Energy-loss spectra of $Fe_{80}B_{15}Si_5$ for different primary energies E_p, normalized to the elastic peak intensity. Note that no shift has been introduced. The enhanced loss near 10 eV for $E_p = 60$ eV could be due to the fact that at this E_p surface sensitivity is maximum (the surface plasmon energy of $Fe_{80}B_{15}Si_5$ is ~ 11 eV). The arrows emphasize the Stoner excitation region.

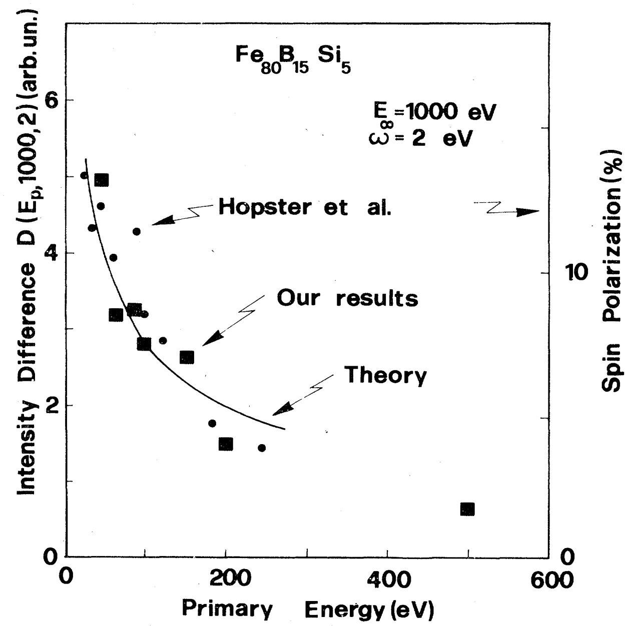

FIG. 4. Squares: intensity of the 2-eV peak in $D(E_p,E_\infty,\omega)$ vs E_p; solid line: calculated primary energy dependence of the total intensity of the 2-eV hump (from Ref. 1); dots: spin polarization of the 2.2-eV loss in $Fe_{82}B_{12}Si_6$ vs E_p (from Ref. 3). The theoretical curve has been normalized to experiment at $E_p = 100$ eV.

spin-polarized data and theory. Figure 4 shows the behavior of the peak value of the difference spectrum of Fig. 3 for increasing primary energies. A strong decrease is found which is again similar to that found by Hopster *et al.*[3] for their spin-polarization peak, and is close to the theoretical expectations of GT. In essence, the higher the primary energy, the more "distinguishable" the electron is and the less probable the exchange scattering with the Fermi sea of Fig. 1 becomes.

In conclusion, we have shown for the first time that ferromagnetic Stoner excitations can be detected by simple electron-energy-loss spectroscopy and can be identified, making use of the strong primary energy dependence of the exchange process involved.

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