# High-Resolution Spin-Polarized Electron-Energy-Loss Spectroscopy and the Stoner Excitation Spectrum in Nickel

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We report on the first experimental determination of the spectrum of Stoner excitations in Ni at $q \approx 0$. The spectrum consists of a broad distribution covering the range from $\sim 0.1$ to $\sim 0.5$ eV.

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The spectrum of electron-hole pair excitations with holes and electrons in bands of opposite spin (Stoner excitations) is of particular importance for the microscopic understanding of magnetism. The energetic position and the shape of the Stoner spectrum at zero momentum provides a measure of the mean exchange splitting and the distribution function over the Brillouin zone. This Letter reports on the first experimental determination of the Stoner spectrum at $q \sim 0$ using momentum- and energy-resolved inelastic exchange scattering of spin-polarized electrons. The spectrum for Ni has a broad distribution in energy [0.3 eV full width at half maximum (FWHM)] and is centered around 0.3 eV. While the spectrum is consistent with the average exchange splitting as determined in previous photoemission experiments, the width of the distribution provides evidence that the exchange splitting for bands near the Fermi energy is not constant over the Brillouin zone.

Energy-loss processes of the exchange type have been observed by measuring the dependence of the loss spectrum of spin-polarized electrons on the spin orientation relative to the sample magnetization. The apparatus is based on a high-resolution energy-loss spectrometer,[1] in which the thermal emitter was replaced by a GaAsP photocathode. The cathode emits spin-polarized electrons when irradiated by circularly polarized HeNe laser light. The polarization vector stands normal to the scattering plane and is reversed by reversing the handedness of light. This part of the apparatus is described by Kirschner, Oepen, and Ibach.[2] The sample has the shape of a transformer core, with the (110) face on the front side.[3] The magnetization vector is aligned with the $[\bar{1}10]$ direction and with the polarization vector. The surface is cleaned by standard procedures and controlled by low-energy electron diffraction (LEED) and Auger electron spectroscopy. The measurements were made at room temperature in the (0,0) beam, which means that we observe excitations of small momentum transfer $q$. The overall resolution of the electron spectrometer system was $\sim 35$-meV FWHM. Further details are described by Rebenstorff.[4]

Typical energy-loss spectra for spin-up and spin-down orientation are shown in Fig. 1(a). The spin-averaged intensity curve is very similar to that published by Ibach and Lehwald.[5] As observed previously, the loss intensity decays rapidly when going off the specular direction. For a quantitative analysis of the spin-dependent features we have measured the asymmetry directly. The asymmetry $A(\epsilon)$ is defined as the normalized intensity difference at loss energy $\epsilon$ when reversing the primary polarization:

$$A(\epsilon) = \frac{1}{|P_0|} \frac{I^{\uparrow}(\epsilon) - I^{\downarrow}(\epsilon)}{I^{\uparrow}(\epsilon) + I^{\downarrow}(\epsilon)}. \quad (1)$$

$P_0$ is the polarization of the electron beam. A typical result is shown in Fig. 1(b).

The asymmetry changes sign upon reversal of the magnetization, demonstrating the magnetic origin of the intensity variations. In addition there is a small, magnetization-independent, asymmetry contribution due to spin-orbit coupling in elastic and inelastic scattering, which causes the imperfect symmetry of the curves in Fig. 1(b) with respect to the zero line. The spin-orbit–induced asymmetry is, however, sufficiently small over the whole energy-loss range (of the order of a few percent) that no corrections have been made. The large structure centered around $\epsilon = 0.3$ eV was found to be independent (within the experimental uncertainty) of the incident electron energy between $E_0 = 5$ and $\sim 18$ eV,[6] and also independent of the scattering angle $\theta$. In Fig. 1(c) we show the results at $\theta = 75°$ averaged over various incident energies and the two magnetization orientations. The structure below about 70 meV is a remnant of the elastic ($\epsilon = 0$) spin-dependent scattering caused by spin-orbit and exchange interaction. It is due to multiple-scattering LEED processes[7] and does not necessarily average out. In the following we focus

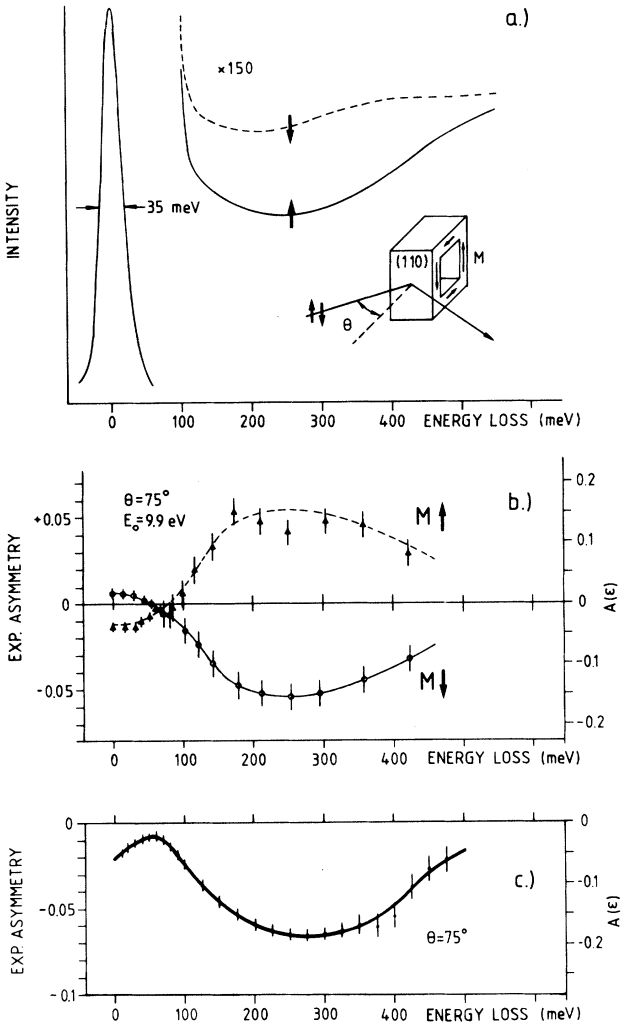


FIG. 1. (a) Typical electron-energy-loss spectra for spin-up and spin-down orientation. The difference between the spin-down and spin-up curves is corrected for the finite polarization of the incident beam. The scattering geometry is depicted in the inset. (b) Asymmetry as a function of loss energy for the two orientations of the magnetization $M$ and for a particular impact energy $E_0$ and scattering angle $\theta$. The experimentally observed asymmetry is smaller than $A(\epsilon)$ as defined with Eq. (1) since the incident beam is not completely polarized (polarization $P_0 \sim 35\%$). (c) Asymmetry spectrum averaged over different energies $E_0$. The asymmetry peaks at 0.28 eV with a FWHM of 0.32 eV.

the discussion on the broad asymmetry peak, which represents the Stoner excitation spectrum. We note that in previous inelastic polarized electron scattering experiments with iron-based metallic glasses similar asymmetry features had not been observed.[8]

The theory of electron-energy-loss spectroscopy has been developed in several steps.[1,9-12] However,

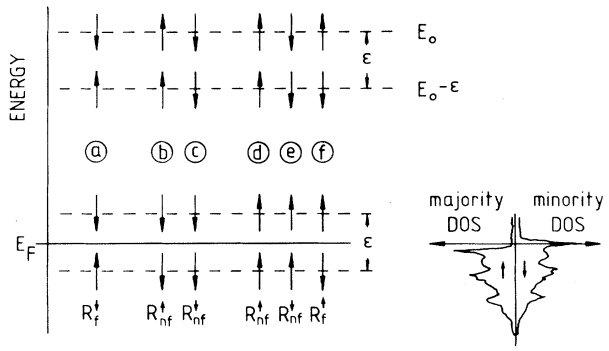


FIG. 2. Inelastic two-electron scattering processes and corresponding partial scattered intensities $R$. The inset shows the spin-split density of states for ferromagnetic Ni.

none of the models neglecting the electron spin can account for the observed asymmetry. Whereas the electronic and magnetic fields involved are too small to flip the spin of the scattered electron directly, an exchange process is possible[12,13] which involves two electrons with their respective spins being conserved. For example, an incident electron with minority-spin orientation may drop into an empty minority-spin state above the Fermi level while the energy is being released to an electron of the occupied majority-spin band. The latter electron then emerges at a kinetic energy equal to the energy of the incident electron minus the energetic difference between the spin-split states. The interaction is of purely Coulombic nature and the spins of the individual electrons remain unchanged during the excitation process.[13] The possible configurations before and after the excitation are listed in Fig. 2. The different channels are assumed to be independent of each other and are designated by partial loss intensities.[12] For example, the process described above [(*a*) in Fig. 2] is characterized by the "flip" intensity $R_f^{\downarrow}$ for an incident down-spin electron (i.e., parallel to the minority-spin orientation). The term "flip" means that the emerging electron has the spin opposite to the incident electron. The subscript "nf" in $R_{nf}^{\uparrow}$ [process (*b*)] denotes the case of the emerging electron having the same spin orientation as the incident up-spin electron. The nonflip intensities $R_{nf}^{\uparrow(\downarrow)}$ describe transitions within a particular spin system. They are basically the same as observed in optical absorption. The processes (*d*), (*e*), and (*f*) all are characterized by an up-spin electron in the final state of the sample. Except for the *s*-*p* hybridized bands with low density of states there are no empty majority states available in Ni and we therefore neglect these transitions as being weak. With the remaining two

nonflip intensities, $R_{nf}^{\uparrow}$ and $R_{nf}^{\downarrow}$, from processes ($b$) and ($c$) and one "flip" intensity $R_f^{\downarrow}$ from process ($a$) the asymmetry is

$$A = \frac{R_{nf}^{\uparrow} - R_{nf}^{\downarrow} - R_f^{\downarrow}}{R_{nf}^{\uparrow} + R_{nf}^{\downarrow} + R_f^{\downarrow}}. \quad (2)$$

The two nonflip contributions are not known *a priori*, but they should be proportional to the joint optical density of states. The calculation by Wang and Callaway[14] shows the joint density of states to be a slowly varying function on the present energy scale. The large, energy-dependent feature in the asymmetry therefore cannot be caused by the nonflip contributions. Furthermore, at low loss energies ($\sim 100$ meV) the flip rate $R_f^{\downarrow}$ is close to zero since there are very few empty majority-spin states near the Fermi level (Fig. 2). Experimentally we find that the asymmetry at 100 meV which must be caused by the remaining nonflip transitions is indeed small. This is in line with the results of a calculation by Rendell and Penn[15] of the asymmetry of the inelastic mean free paths in Ni. Considering nonflip processes (in our language), they found the asymmetry to be small, of the order of 1%. As the asymmetry is small where it is solely determined by the nonflip rates and as the latter are weakly energy dependent we conclude that the difference $R_{nf}^{\uparrow} - R_{nf}^{\downarrow}$ of the nonflip contributions in the energy range of interest here is small. This does not mean that the rates themselves are small. As they enter into the denominator of Eq. (2) they may considerably reduce the asymmetry caused by flip processes $R_f^{\downarrow}$.

With the nonflip processes contributing a small and weakly energy-dependent part of the asymmetry the energetic structure of the asymmetry [Eq. (2)] essentially is determined by $R_f^{\downarrow}$ [process ($a$) in Fig. 2]. The magnitude of the flip rate may roughly be estimated from the experimental data in Fig. 1 to be of the order of 20% of the sum of the two nonflip rates. With $R_{nf}^{\uparrow} \simeq R_{nf}^{\downarrow} = \frac{1}{2} R_{nf}$ and $A \ll 1$, Eq. (2) then simplifies to

$$A \approx \frac{-R_f^{\downarrow}}{R_{nf} + R_f^{\downarrow}} \approx -\frac{1}{R_{nf}} R_f^{\downarrow}.$$

$$\approx -\text{const} \times R_f^{\downarrow}. \quad (3)$$

The sign of the asymmetry should be negative, in agreement with the experiment. The expression for the asymmetry [Eq. (2)] is applicable to our experimental approach. If the experiment is performed with unpolarized primary electrons and polarization analysis after scattering, such as in the experiment by Hopster, Raue, and Clauberg,[16] the expression for the polarization $P$ reads[12]

$$P = \frac{R_{nf}^{\uparrow} - R_{nf}^{\downarrow} + R_f^{\downarrow}}{R_{nf}^{\uparrow} + R_{nf}^{\downarrow} + R_f^{\downarrow}}. \quad (4)$$

One sees that Eq. (4) is almost identical to Eq. (2), except for a sign change of $R_f^{\downarrow}$ in the numerator. As a consequence, the spin polarization caused by flip processes should be positive, which was indeed observed by Hopster, Raue, and Clauberg.[16] The two approaches thus complement each other, and, when combined to the same system, could yield quantitative information on the transition rates involved, provided that both experiments are $q$ resolved.

The final state in the scattering process in Ni, involving the flip rate $R_f^{\downarrow}$, is characterized by a hole below the Fermi level and an electron in a band of opposite spin above the Fermi level. This configuration is identical with that of a Stoner excitation, in which a majority-spin electron is excited into a minority-spin state while reversing its spin. The flip rate $R_f^{\downarrow}$ therefore is a measure of this process, and its energy dependence reflects the spectrum of Stoner excitations. The asymmetry would be directly proportional to the $q$-dependent "Stoner density of states"[17] if the matrix elements were independent of $\epsilon$ and $k$. As the measurements are done in the specular beam apart from a small, weakly energy-dependent contribution from nonflip processes, the measured asymmetry $A(\epsilon)$ reflects the spectrum of Stoner excitations *at* $q \sim 0$. The differential cross section peaks near $q \sim 0$, similar to classical "dipole" losses of electrons, though for a different reason. The reason here is that Stoner excitations involve transitions between bands which are nearly parallel over larger fractions of the Brillouin zone which makes the transition rates for a particular loss energy peak at $q = 0$. If one were to assume the bands to be rigidly split, the Stoner density of states for $q = 0$ would have been a $\delta$ function at the energy of the exchange parameter $\Delta$. The broad energy distribution of the asymmetry, however, suggests that the exchange splitting is $k$ dependent. With the exchange splitting being wave-vector dependent, this peak broadens into a distribution over $\epsilon$, the maximum of which may be associated with the exchange splitting $\langle\Delta\rangle$ averaged over the Fermi surface. From our experimental results we find $\langle\Delta\rangle \approx 0.3$ eV, in agreement with the majority of experimental data.[3,18-23] The distribution is about 0.3 eV wide (FWHM), and is thus consistent with all experimental splittings reported in the literature. These experimental data were, however, obtained by photoemission spectroscopy.

There, the energy difference between hole states in occupied spin-up and spin-down bands is measured. The large discrepancy between the exchange splitting as determined by photoemission and most *ab initio* band-structure calculations based on the local-spin-density functional[24,28] has been attributed to large self-energy corrections applicable to photoemission.[29,30] On the other hand, as pointed out recently by Olés and Stollhoff,[31] the neglect of anisotropic exchange and correlation in the local-spin-density functional scheme may lead to too large exchange splittings. Including anisotropy they found $\Delta_{e_g}=0.27$ eV and $\Delta_{t_{2g}}=0.50$ eV, in reasonable agreement with a semiempirical calculation by Cooke, Lynne, and Davis,[17] who found $\Delta_{e_g}\sim 0.1$ eV and $\Delta_{t_{2g}}\sim 0.4$ eV. As noted by Liebsch,[30] the self-energy should be corrected for the amount of correlation already contained in the band calculation. Our results suggest that the exchange splitting as measured in photoemission is less subject to self-energy corrections than is currently assumed.

We have shown that elementary excitations in ferromagnets may be studied by spin-polarized electron-energy-loss spectroscopy. It unveils regions in $\omega$-$q$ space that have not been accessible to neutron scattering and other techniques.

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