Zeitschrift: Helvetica Physica Acta
Band: 62 (1989)
Heft: 6-7

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DOI: https://doi.org/10.5169/seals-116116

EXCHANGE EXCITED f-f TRANSITIONS IN THE ELECTRON-ENERGY-LOSS SPECTRA OF RARE-EARTH METALS

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Abstract: Dipole-forbidden, exchange excited f-f transitions are found to characterize the Electron-Energy-Loss Spectra (EELS) of rare-earth metals taken at low primary energy. The transitions are identified by comparison with optical and photoemission data. The intensities of the f-f peaks and of the plasmon peak resonate when the primary energy is close to the 4d binding energy E_{4d}.

Exchange scattering has been recently invoked to explain reflection EELS from solids, mainly the spin polarized ones [1]. In the exchange process, in contrast to dipole scattering, the primary electron is captured in an empty level of the solid, kicking out from the sample the electron which is detected. This process should be most effective at low primary energies E_p [1]. In this paper we show that the EELS of Rare-Earth Metals (REM) are characterized by f-f transitions excited via this process.

The measurements were performed at constant resolution (0.5 eV). Polycrystalline samples were cleaned by Ar-ion sputtering. Cleanliness was checked by Auger and XPS. Due to the 12^{o} acceptance angle of the hemispherical analyzer the spectra must be considered as momentum-integrated.

Fig.1 shows representative data for Dy (f^9 configuration). In panel (a) the low energy part of the EELS (continuous lines) taken at low E_p show sharp peaks that have no correspondence in the bulk (dashed line) and surface (dotted line) loss functions. These structures are highly insensitive to oxidation. The energies of the EELS peaks agree well with the weak lines observed in the optical absorption spectra of the trivalent ion [panel (b)]; these lines are due to f-f transitions very weakly allowed in an environment

without inversion symmetry. The structures in the EELS can also be related to the shake-up features of the XPS of Ho (f^{10} configuration) [panel(c)]. The reason is that the photoemission process leaves the f^{n+1} metal in a f^n configuration, with the same spectrum of final states as in the EEL process, the only difference being the energy scale, due to the difference in the nuclear charge. A similar line of reasoning holds for the bremsstrahlung isochromat spectra from the f^{n-1} metal.

An enhancement of the intensities of the f-f peaks and of the plasmon peak is observed when the E_p is close to E_{4d}. Fig.2(a) and (b) show the processes involved in the resonant enhancement of the f-f peaks. As far as the plasmon peak resonance is concerned, we suggest that the excitation mechanism depicted in Fig.2(c) should be active along with the normal dipole-allowed one [Fig.2(d)]. Both processes have a resonant counterpart [Figgs.2(e) and 2(f), respectively]. In process (f), however, the plasmon is excited by the switching on of the d core hole, and no plasmon satellite can be observed in the optical absorption data of the REM.

A fuller account of the results will appear elsewhere [2].

[1] See, e.g., C.J. Bocchetta, E. Tosatti, and S. Yin, Z. Phys. B 67, 89 (1987), and references therein.

[2] F. Della Valle and S. Modesti, Phys. Rev. B (in press).

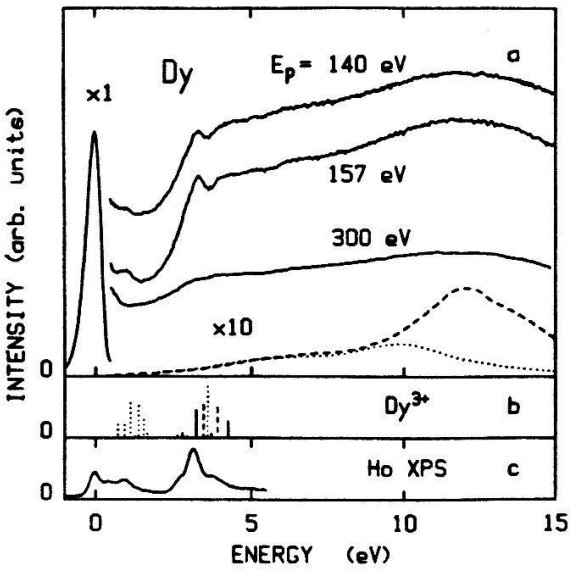

Fig.1

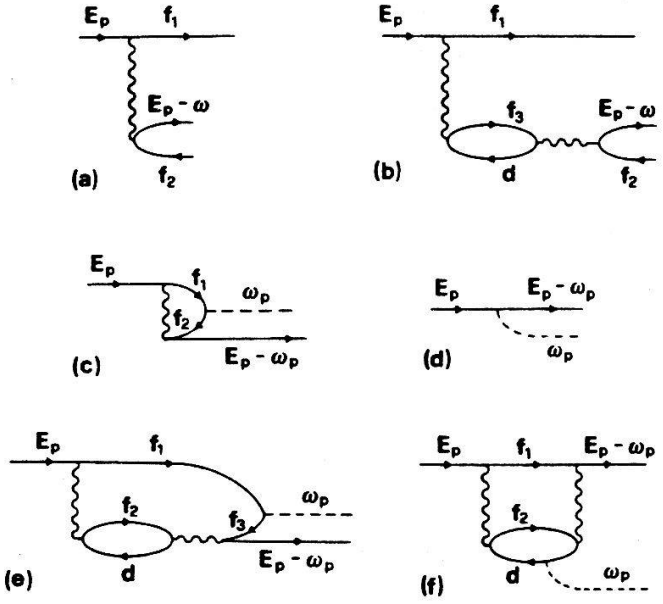

Fig.2