Resonant Photoemission versus incoherent superposition of Auger and Photoemission signals

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The enhancement of the 6-eV satellite structure in the valence-band photoemission (PE) spectra of Ni metal, when the photon energy is varied across the M_{III} absorption threshold, has been assigned to a strong Fano-type resonance [1-2]. Such a resonance is understood as an interference between two different channels leading to the same final state: the direct photoionization of $3d$ valence-band states and an $M_{2,3}M_{4,5}M_{4,5}$ Super-Coster-Kronig (SCK) Auger decay of the core-excited $3p^5 3d^{10}$ intermediate state. Necessary for the occurrence of a Fano-type resonance is a coherence between the two channels. It has been shown [3] that the PE spectra of Ni and CuO at the L_{III} thresholds have to be described as a superposition of the Auger and the PE signals. This finding deviates from the established picture of resonant PE and is explained as a loss of coherence between the two channels caused by the itinerant character of the intermediate state. It has to be tested if the same mechanism causes the enhancement of the 6-eV satellite in the PE spectrum of Ni metal at the M_{III} threshold. We present the results of a comparative study of resonant electron-emission processes at the M_{III} threshold of Ni metal (Ni $3p \rightarrow 3d$ resonance) with those occurring at the Ce $N_{IV,V}$ threshold of the alloy CeGd (Ce $4d \rightarrow 4f$ resonance) [4]. The intensities of the various spectral components were measured as a function of the electron-emission angle Θ relative to the $\vec{E}$-vector of the incident light. In case of a PE process, the intensity should show a $cos^2\Theta$-behavior, while an Auger signal ought to be isotropic in case of a polycrystalline sample.

The measurements were performed at the TGM-1 beamline of BESSY. The photoelectrons were collected at different angles with respect to the $\vec{E}$-vector. Polycrystalline samples were intentionally used to avoid angular dispersion effects and were cleaned by scraping with a diamond file.

Fig. 1 shows valence-band PE spectra of Ni metal, taken at (a) $h\nu$=75 eV (non-resonant) and (b) $h\nu$=67 eV (on-resonance), and of CeGd taken at (c) $h\nu$=122 eV (Ce $4d \rightarrow 4f$ resonance) for two different angles Θ. The Ni non-resonant spectra were normalized to equal intensities of the Auger signals (dashed-double-dotted curve). This is justified on the basis of an isotropic Auger emission for a polycrystalline sample. The resulting intensity angular variations of the non-resonant valence-band (dashed curve) and satellite (dashed-dotted) emission are given for several values of Θ in the inset of Fig. 1 by open symbols. The dashed curve in the inset represents a $cos^2\Theta$ behavior. Both intensities vary roughly as $cos^2\Theta$ as expected for a PE process. The Ni on-resonance spectra in Fig. 1(b) were normalized to valence-band intensities equal to those in the non-resonant spectra. This leads to an approximately isotropic behavior of the on-resonance 6-eV feature of Ni metal as given in the inset by the filled triangles, contrasting the $cos^2\Theta$-behavior of the 6-eV PE satellite in the non-resonance spectra (open triangles). Thus, the 6-eV peak on-resonance behaves like an Auger signal and cannot be described as resonant PE.

To insure that the Auger-like angular character of the satellite emission is not a general property of resonant PE, Fig. 1(c) shows PE spectra of the polycrystalline alloy CeGd, taken in Ce $4d \rightarrow 4f$-resonance at $h\nu$=122 eV; the spectra were taken at the same angles Θ as those of Ni metal and the same normalization procedure was applied.

At a BE of 2 eV, the resonantly enhanced emission from the Ce $4f$ states is observed, while the weaker emission at $\simeq$8-eV BE represents non-resonant PE from Gd $4f$ states, which can be used for internal calibration. When increasing the angle Θ from 0° to 60°, the intensity of both resonant Ce-$4f$ and non-resonant Gd-$4f$ emissions decrease proportional to $cos^2\Theta$ in exactly the same way. The internal calibration given by the ratio of intensities of the Ce-$4f$ and Gd-$4f$ PE signals, which does not change from Θ=0° to Θ=60°, proves in addition that the resonantly enhanced Ce-$4f$ PE signal has the same angular dependence as that of the non-resonant Gd-$4f$ PE signal, i.e. the angular dependence of a PE process.

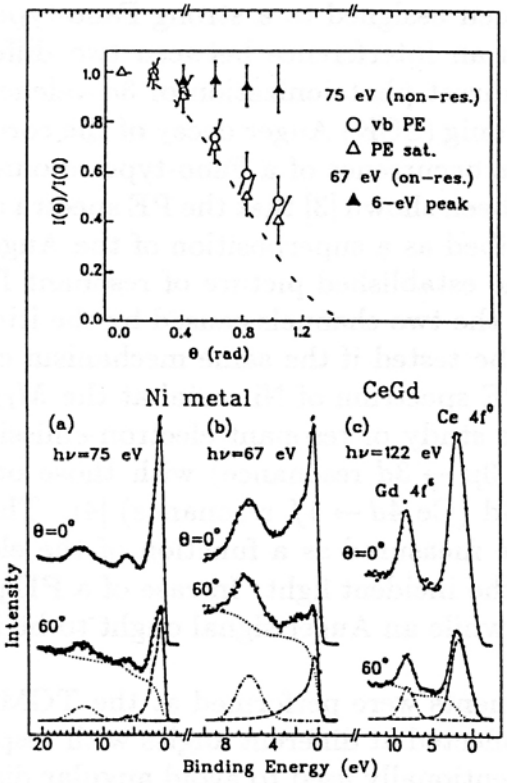

Fig. 1: Photoelectron spectra of Ni metal taken at (a) $h\nu$=75 eV (non-resonant) and (b) $h\nu$=67 eV (on-resonance), and of CeGd at $h\nu$=122 eV (Ce $4d \rightarrow 4f$ resonance) for two different angles Θ. The inset shows the angular dependences of the intensities of the valence-band signal (open circles) and the 6-eV satellite (open triangles) in the non-resonance spectra, and of the 6-eV feature in the on-resonance spectra (filled triangles).

We have shown that the resonant enhancement of the 6-eV satellite in the PE spectrum of Ni metal in the $3p \rightarrow 3d$-resonance is almost completely due to an incoherent superposition of the valence-band PE signal with an $M_3M_{4,5}M_{4,5}$ Auger emission. This clear difference to the true resonant PE behavior in rare-earth systems is probably caused by the weaker localization of the $3d$ states in Ni metal [4], where a strong hybridization of the core-excited intermediate state with conduction-band states leads to delocalization and thereby to a loss of coherence.

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