Resonant Photoemission and Linear Dichroism in Gd*

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Abstract

Magnetic Linear Dichroism in Angular Distributions (MLDAD) from Photoelectron Emission was used to probe the nature of Resonant Photoemission. Gd 5p and Gd 4f emission were investigated. Using novel theoretical simulations, we were able to show that temporal matching is a requirement for "True" Resonant Photoemission, where the Resonant Photoemission retains the characteristics of Photoelectron Emission.

Discussion

Resonant Photoemission is the phenemonon where the emission of 4f and 5p electrons from rare-earth metals and their compounds is strongly enhanced because a second emission channel opens up, e.g the photon has just enough energy to excite a 4d electron to an unoccupied 4f level. In a generic picture, the indirect channel of the resonant photoemission is interpreted as due to a process where a 4d electron in the initial state is first excited to the unoccupied 4f level, forming a tightly coupled, bound intermediate state, 4d core hole plus 4f electrons. Then a decay via autoionization occurs into the final state, thus producing a final state identical to that obtained by a direct photoemission process for the ejected electron. [1] The transition rate is greatly enhanced if the excited state decay is by a Coster-Kronig or a super-Coster-Kronig [(s)CK] process. [2,3] The key question is whether these processes are coherent or incoherent: Is it truly "resonant photoemission" or merely the incoherent addition of a second emission channel? Should the overall intensity be treated as a squaring of the sum of the amplitudes (coherent) or summing of the squares of the amplitudes (incoherent)? A true "resonant photoemission" process should be coherent, involving interference terms between the direct photoemission and indirect photoemission channels. Possibly, incoherence would give rise to the loss of photoemission characteristics in the process, with a domination of Auger-like properties.

To this problem we have applied the new photoelectron spectroscopy technique of magnetic linear dichroism in angular distributions (MLDAD). [4-7] This technique is related to but distinct from the techniques of magnetic x ray circular dichroism (MXCD) in photoelectron spectroscopy and x ray absorption. [8-14] The key is that while large dichroic effects in ferromagnets can be observed with MXCD-photoemission and MXCD-absorption, the large MLDAD effects in ferromagnets is solely a photoemission, not an absorption-driven, process. This is because the chirality which gives rise to magnetic sensitivity is due to the vectorial configuration in MLDAD as opposed to the intrinsic chirality of circularly polarized x rays in the MXCD techniques. In absorption, where there is an essential averaging over all emission angles, the vectorial chirality is lost. Thus, MLDAD is the ideal measurement to distinguish between photoemission and absorption processes. Angle-resolved photoemission in a magnetic

system should show an MLDAD effect: x ray absorption and thus auger-like emission will show no MLDAD effect. Experimental details can be found elsewhere. [15-19] Theoretical spectra were calculated in intermediate coupling using Cowan's relativistic Hartree-Fock code.[20] Radiative transitions were taken into account to first order and (s)CK transitions to infinite order. [21-23] Line broadening of the photoelectron state and experimental resolution were included by a convolution with a Lorentzian and a Gaussian, respectively. For the 4f emission the interference effects between the different photoemission final states and between direct and resonant channel were fully included. The interference term was excluded in the 5p calculation.

The study included an extensive theoretical and experimental data set, collected "on" and "off" resonance. Some of the spectra can be seen in Figure 1. These spectra, coupled to results not shown here [17-19], lead to the following conclusions. The Gd 4f resonant photoemission is confirmed to be photoemission-like, because it shows an MLDAD effect on resonance. The Gd 5p resonant emission is shown to be dominated by Auger-like contributions, owing to the absence of an MXLD effect at resonance. The experiment results are confirmed by the theoretical simulations. It appears that temporal channel matching is a requirement for channel interference and the persistence of photoemission effects. The Coster-Kronig decay that occurs in the 5p emission occurs on a time scale of about 10^{-15} sec. [2] The super-Coster-Kronig delay of the 4f should be significantly faster. [2,3] This would speed up the indirect channel, bringing it nearer to the time duration of x ray absorption (10^{-17} sec or less) that dominates the

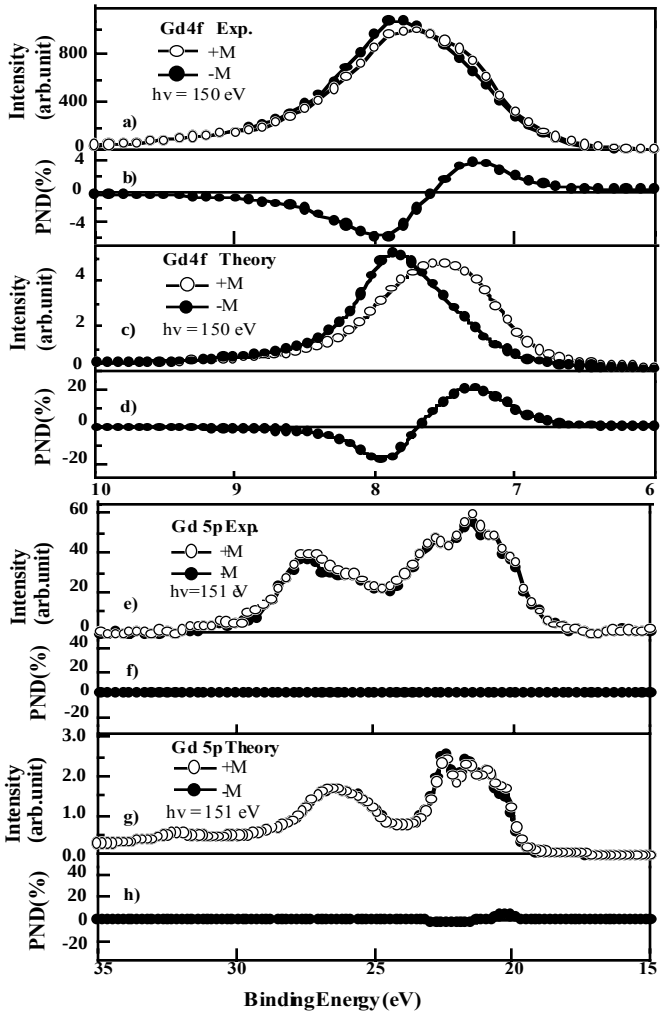

Figure 1. A series of experimental and theoretical 4f and 5p photoemission spectra (for the two opposite magnetization directions) and normalized difference curves. The photon energy was 150 eV for the 4f and 151eV for the 5p; (a) Gd 4f, photoelectron spectra, experimental; (b) Gd 4f, photoelectron spectra difference, experimental; (c) Gd 4f, photoelectron spectra, theory; (d) Gd 4f, photoelectron spectra difference, theory; (e) Gd 5p, photoelectron spectra, experimental; (f) Gd 5p, photoelectron spectra difference, experimental; (g) Gd 5p, photoelectron spectra, theory; (h) Gd 5p, photoelectron spectra difference, theory. EDC is energy distribution curve. The spectra in (a), (c), (e), & (g) are EDC's, where the photon energy is held constant and the kinetic energy is scanned. PND stands for peak normalized difference, where the dichroism difference at each binding energy is divided by the sum of the two intensity maxima, one from each pair. The photon energies of 150 eV and 151 eV are "on" resonance.

direct photoemission channel. Thus, not only must the energies of the two channels match but also the time duration, in order to observe "true resonant photoemission".

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References

[1]J.L. Dehmer, et al, Phys. Rev. Lett 26, 1521 (1971); J. Sugar, Phys. Rev. B 5, 1785 (1972), A. F. Starace, Phys. Rev. B 5, 1773 (1972); L.C. Davis, and L.A. Feldkamp, Phys. Rev. A 17, 2012 (1978); F. Gerken, et al Phys. Rev. Lett. 47, 993 (1981); Extensive references by J. Allen in Synchrotron Radiation Research, ed. R.Z. Bachrach (Plenum Press, New York, 1992), Vol. 1., p. 253, Giant Resonances in Atoms, Molecules, and Solids, eds. J.P. Connerade, J.-M. Esteva, and R.C. Karnatak, NATO ASI series B (Plenum Press, New York, 1987).
[2]B. Feuerbacher, B. Fitton and R.F. Willis, ed., "Photoemission and the Electronic Properties of Surfaces", John Wiley & Sons, New York, See Fig. 5.1 on Page 115.
[3]T.A. Carlson, "Photoemission and Auger Spectroscopy," Plenum Press, New York.
[4]Ch. Roth, et al, Phys. Rev. Lett. **70**, 3479 (1993); Solid State Commun. 86 647 (1993); F.U. Hillebrecht, et al, Phys. Rev. B53, 12182 (1996).
[5]F. Sirotti and G. Rossi, Phys. Rev. **B 49**, 15 682 (1994); G. Rossi, et al, Solid State Commun. **90**, 557 (1994).
[6]W. Kuch, et al, Phys. Rev. B51, 609 (1995).
[7]F.O. Schumann, et al, Phys. Rev. Lett. 79, 5166 (1997).
[8]L. Baumgarten, et al, Phys. Rev. Lett. **65**, 492 (1990).
[9]J.G. Tobin, et al, Phys. Rev. Lett. 68, 3642 (1992).
[10]C.M. Schneider, et al, Phys. Rev. **B 45**, 5041 (1992); C.M. Schneider et al, Appl. Phys. Lett. **63**, 2932 (1993); Venus et al, J. Phys. Cond. Matt. **5**, 1239 (1993).
[11]B.T. Thole and G. van der Laan, Phys. Rev. **B 44**, 12424 (1991).
[12]K. Starke, et al, Phys. Rev. **B 48**, 1329 (1993).
[13]K. Starke, et al, Phys. Rev**. B 55**, 2672 (1997); E. Arenholz, Ph.D. Thesis, "Magnetic Dichroism in Photoemission from Lanthanide Materials: Basic Concepts and Applications", Wissenschaft and Technik Verlag, Berlin, 1996.
[14]J.G. Tobin, et al, Surf. Sci. Lett. 395, 227 (1998).
[15]J.G. Tobin, et al, J. Appl. Phys. **79**, 5626 (1996); J. Vac. Sci. Tech., B 14, 3171 (1996).
[16]J.D. Denlinger et al. Rev. Sci. Instrum. **66**, 1342 (1995).
[17]W.J. Gammon, et al, J.Vac. Sci. Technol. A **15**, 1 (1997), W.J. Gammon, M.S. Thesis, Virginia Commonwealth Univ., 1994, Unpublished..
[18]S.R. Mishra, et al, J. Vac. Sci. Tech. A 16, 1348 (1998).
[19]K.W. Goodman et al, MRS Symp. Proc. **475**, 493 (1997).
[20]R.D. Cowan, The Theory of Atomic Structure and Spectra, University of California Press, Berkeley (1981).
[21]G. van der Laan, et al, Phys. Rev. B 46, 9336 (1992).
[22]G. Van der Laan, Phys. Rev. B 51, 240 (1995).
[23]G. van der Laan, et al, Phys. Rev. B 56, 3244 (1997).

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