DOE/ER/45081--T1

DE92 015661

FINAL TECHNICAL REPORT
TO THE
DEPARTMENT OF ENERGY
ON
HIGH PRESSURE AND SYNCHROTRON RADIATION STUDIES OF SOLID STATE ELECTRONIC INSTABILITIES

(CONTRACT NO. DE FG05-84ER45081, RUTGERS ACCOUNT NO. 4-24792
4-25477

For the Period
May 1, 1984 - April 30, 1987

From

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Date Submitted April 1992

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I. Introduction

The publications and presentations that this grant contributed toward are listed in section III under the groupings of: refereed articles; book/conference papers; and abstracts and presentations. The descriptions in section II below refer to the numbers of the citations in the first two of these groupings from section III. Articles published after the termination of the contract have been included if the work leading to them was initiated during the contract.

II. Research Article Synopses

Below the research articles facilitated by this grant is divided into into sub-areas and a brief explanation of the work in these areas is provided.

1. Eu and General Valence Instabilities

Eu based systems provide an excellent proving ground to study rare earth valence/magnetic instabilities of the class where the 4f levels are energetically narrow (weakly hybridized). The sensitivity and convenience of Eu-Mossbauer spectroscopy to valence changes and the strong magnetism of the Eu^{2+} state are partially responsible for the pedagogical character of Eu-studies. Moreover relatively simple theoretical modeling of the valence instabilities in such narrow 4f level systems can elucidate a host of interesting phenomena. The work supported by this grant has made significant contributions in this area, some of which are outlined below.

Journal Articles 3, 10, 25, 31
Conference/Book Articles 15, 16

Conference/Book paper 5 described the extensions of a phenomenological model for valence instabilities (including the Eu case). This theoretical framework allows one to understand the interplay of entropy, the intervalence hybridization-strength and excitation-energy, and cooperative intersite valence and magnetic interactions in mixed valent systems. In particular this theoretical framework was shown to generate a phase diagram like that observed in previous work by our lab. Moreover the model predicted a number of interesting generic effects for unstable valence materials.

Article 3, although brief, make an important correlation between the Mossbauer effect and L_3 x-ray absorption methods of Eu-valence state determination (mentioned below). This article addressed the question of establishing a good absolute estimate of valence - the most widely used theoretical parameter in the characterization of the valence/magnetic instability. The important proposal that Eu-magnetic response and magnetic order could persist in the initial stages of valence mixing was made. This point is still under discussion in the literature.

Articles 14 and 31 address the interesting interplay of ferromagnetism, valence mixing, and the metal-insulating metamorphosis in EuO under pressure.

Article 25 and Book/Conference Paper 6 discuss X-ray diffraction studies of the lattice response near both first order and continuous valence transitions for $EuPd_2Si_2$ based materials.

2. Ce Problem: L_3 Spectroscopy Emphasis; Bulk Property Emphasis

Articles: 4,5,7,9,11-13,19,21,27-29,31-33

The Ce-4f orbitals are more extended and more strongly hybridized than those of all the other rare earths. Under this grant our group perused extensive studies of Ce systems in which such hybridization effects make spin fluctuations the dominant mechanism in determining the Ce-demagnetization process. The Ce valence state is intimately coupled to the spin fluctuation effects and hence a spectroscopy which gives insight into this important parameter is of great value in understanding this problem.

L_3 X-ray absorption spectroscopy provides one of the most direct and convenient means of probing relative valence states in mixed-valence systems. The work under this grant enabled the exploitation of this technique ; to explore the valence state-environment coupling, to identify interesting systems to pursue in depth with bulk property measurements, and to use in conjunction with bulk measurements to understand the physics. Here our groups application of L_3-spectroscopy and such bulk studies to the Ce Problem are reviewed.

L_3Spectroscopy Emphasis

Articles 7 [on intermetallics], 9 [on amorphous materials] and 19 [on ion implanted materials] use Ce-L_3-valence measurements to identify the response of the Ce-valence to systematic variations in the host electronic-structure. These studies were intended : to quantify and systematize observations on valence state variations in compound series; to identify new systems (to be studied more extensively) lying in an interesting valence state ranges; and to motivate/provide guidelines-for first principles calculations of the mixed-valent-atom to host environment coupling.

It should be noted that the L_3 studies of intermetallic compounds in article 7 indicated a theretofore unsuspected recurrence of a Ce-valence typical of heavy fermion behavior in 3d-row 1:2:2 compounds. Article 12 describes detailed L_3, structural, and transport measurements confirming a unique reentrant trend in the Ce magnetic/valence state this series of compounds. Articles 27-29 focused in on (and expand via alloy techniques) a particular portion of this series . Here the use of the systematic L_3-results were used to identify these materials as lying interesting regime.

Article 32 traced the Ce-valence instability (via L_3-measurements) through a series of intermetallic compounds. The suggestion was made that a critical host-electron count can parameterize the occurrence of the valence instability through three systems.

Bulk Property Emphasis

Articles 27-29 dealt with a new facet of the mixed valent/heavy fermion field where the interplay of Ce-spin fluctuations (on a low energy scale) interplay with strong host Mn-magnetism on a much larger energy scale. The rebirth low-energy scale Ce-nonmagnetic effects in the Mn-antiferromagnetic phases of these materials is, to date, unique in this field. Also the consistency of Ce-L_3 based valence variations, upon both temperature and alloy-concentration variation, with the Anderson model was seen in these materials.

Article 5 stressed that the band filling effect induced by Th^{4+} substitution for Ce in Ce-compounds dramatically reduces spin fluctuations and Ce-valence mixing. Specifically for two 1:2:2 compounds the facts that Th substitution rapidly reduces the Ce-spin fluctuation temperature and drives the Ce-valence toward the magnetic 3+ valence were shown. L_3 and transport measurements were correlated here.

3. Transition Metal Compound Electronic Structure.

Journal Articles 13, 22, 24, 30.

Within this grant period our lab began the study of the electronic structure of transition metal compounds. These studies were motivated, in part, by the systematic response of the rare earth valence state to varying transition metal component in intermetallic compounds. These studies emphasized x-ray absorption measurements to probe the unfilled d-states above the Fermi energy (ϵ_F) . Photoemission experiments, to complete the below ϵ_F d-state picture, were also employed.

Article 23 illustrated how the "white line" (WL) feature at the L_2 and L_3 x-ray absorption edges of the 5d-row elements reflects the variation of the 5d-orbital occupancy. The breakdown of the nearly linear WL area to 5d-hole count relation near the beginning of the 5d-row was attributed to atomic many body affects. Finally the application of this method to estimate the chemical-bonding-induced changes in the 5d-hole count in TAl_2(T=Au and Pt) compounds was discussed.

Article 24 combined ultraviolet photoemission measurements with $L_{2,3}$ x-ray absorption spectroscopy (XAS) measurements into a picture of the states both above and below ϵ_F in TAl_2 compounds. The idea that T-Al hybridization shifts T-5d states up in energy (into antibonding states) thereby inducing more 5d-holes than in elemental-T was underscored.

Article 30 extended the study of T-X hybridization to a wide class of compounds involving T=5d row transition metal and X=p-block elements. The strength of the T-X hybridization was proposed to be reflected by the degree of bonding-induced 5d-hole creation. Systematic $L_{2,3}$-XAS measurements of a wide range of compounds were correlated into a picture of the strength of covalent bonding in these solids.

4. Electronic Structure-Phonon Coupling Studies

Journal Articles 6, 8
Conference/Book Article 3

The rare earth-4f orbitals couple strongly to the phonon and elastic properties of a solid in a number of ways. Firstly changes in the 4f occupation cause a large atomic volume increase which, in a solid, entails elastic energy. Secondly the energy levels associated with the 4f orbital can couple to elastic degrees of freedom which possess the proper energy and symmetry. Through this grant our lab was involved in the following collaborative efforts to explore such effects.

Article 6 was a Raman study of a unique resonant coupling of phonons to a crystalline electric field level in $CeAl_2$. This unusual coupling induces a splitting of this level beyond that expected by the static crystal symmetry alone. Raman studies of other dialuminides were also discussed.

Article 8 proposed a unique Raman electronic excitation involving the intervalence excitation energy. Although our lab now favors a more sophisticated explanation of these experiments the observation of such an electronic excitation in a metallic system is a very interesting effect. (This article is mentioned in this section along with the other Raman studies in this collaborative effort.)

5. High Temperature Superconductivity and Oxide Materials

Articles 15-18,20,23, and 26
Late in the funding period of this grant an effort in the area of high T_c superconductivity was initiated which lead to a number of publications. These articles fell into two groups. The first delt with x-ray absorption studies which appeared soon after the fields' inception and were part of the first steps toward understanding the electronic and structural properties of these materials. These studies encompassed related oxide materials also. The second dealt with bulk studies of a number of high-T_c material types to help establish he character of their superconductivity.

6. Novel materials collaboration with Chemistry

With the help of this grant our lab initiated a collaborative program with the Rutgers Chemistry Department. Here the common thread was new/novel materials and the linkage between structure and physical properties. Articles 1 and 18 were part of this effort.

III. Contract Supported Publications/Presentations

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8/18/92