Solid State Chemistry and Physics

The *Solid State Group* in the *Chemistry Department* has developed during the past years a broad range of expertise in complementary areas of solid state science and focused, under a global approach, on new materials with unconventional electrical and magnetic properties. The nature of the group is multidisciplinary combining a wide range of expertise ranging from the synthetic chemistry of either molecular materials with transition metal complexes or intermetallic compounds, to many different specialised solid state characterisation techniques. Among the more relevant facilities developed, operated, maintained and used by in the group are a high temperature laboratory with crystal growth techniques (Czochralski, Bridgman, float zone), X-ray diffraction both in single crystal and powders, EPR spectroscopy, a Mössbauer spectroscopy laboratory, magnetisation measurements by Faraday and extraction techniques and AC-susceptibility measurements, electron transport measurements, heat capacity, in a broad range of temperature and magnetic fields. The use of all these specialised characterisation techniques, often requiring low temperatures down to 0.3 K and high magnetic fields up to 18 T, lead to the development of a strong expertise in cryogenics. The group was the main promoter of the installation at ITN in 1993 of a helium liquefier, that since then remains the only one operational in Portugal providing helium also to many users outside ITN, under the supervised of the group.

The rare combination of the preparative chemistry expertise with the specialised solid state physics techniques enables the group to deal with different problems of modern materials science. Due to the valuable techniques developed the group often act as a partner of many national and international research projects. Due to strategic reasons the group has centered his activities in selected type of materials:

- Molecule based conducting and magnetic materials.
- Intermetallic compounds with uranium and lanthanides
- Oxides including high temperature superconductors.

The specific ongoing research projects are described in more detail in the following sections.

The molecular conductors have since the discover of the first organic metals in 1973 remained one of the most active areas of research of modern materials science, and the group has a long and established tradition of research in this field. However, in order to take profit from some common molecular precursors and synthetic procedures, the interests of the group have recently been extended to molecular magnets. The group intends to further develop its expertise in molecular design and chemical synthesis to crystal engineer new molecular materials with desired electrical and magnetic properties.

The research on intermetallic compounds was initiated by 1992 in structures thought to be good candidates for hard magnets and, due to specific characteristics of ITN as a nuclear laboratory, containing mainly U and Fe. The M¬ssbauer spectroscopy was used as a valuable tool to probe the role of iron atoms in the magnetic properties of these materials in complement to all other techniques. The group intend to extend its activities to compounds with lanthanides, whose role can be also studied by M¬ssbauer spectroscopy using non-commercial sources to be activated in the RPI. More recently the interests of the group in intermetallic extended to other f-element compounds where more exotic properties derived from strongly correlated electrons are observed.

As a strategic effort to extend the expertise of the group in the study of bulk materials to artificially confined structures, and wishing to take profit from possible synergies with valuable surface characterisation techniques available at ITN, in 1999 it was started a project aiming at to install facilities for the preparation of thin films with uranium.

The research in the field of superconductors was initiated soon after the discover of high Tc materials, during a period where almost all main solid state laboratories in the world were involved in this subject. Presently the research in this field is focused on the use of the excellent low temperature and high magnetic field facilities existing in the group, to study the vortex motion and pinning mechanisms in thin films and multilayers of top quality made in other laboratories.

Magnetic Molecular Materials based on Dithiolate Complexes*

V. Gama, I. C. Santos, S. Rabaça[1], D. Belo[1], R.T. Henriques[2]

Objectives

The objective of this work consisted on the synthesis and study of new molecular materials based on dithiolate anionic complexes. Metamagnetic and ferri or ferromagnetic materials were expected. Studies on the correlation between the crystal structure and the magnetic properties as well as on the effect of the spin variation on the magnetic properties of the materials were the main goal in this work.

Results

Several new charge transfer salts based on decamethylmetalloceniums and on dithiolate monoanionic complexes were prepared. In these compounds four distinct types of crystal structures were observed to exist, consisting of linear chain arrangements of three different stacking sequences: ··ADADAD··, ··(DD)A(DD)A·· and ··(DD)[AA](DD)[AA]··. For the Ni and Pt dithiolates acceptors the nature of the dominant intrachain magnetic coupling was observed to be dependent on the stacking sequence and on the decamethylmethalocenium metallic element. For the simple alternated sequence a FM coupling was observed for the Fe and Mn $[M(Cp^*)_2]^+$ salts and an AFM coupling was observed for the $[Cr(Cp^*)_2]^+$ salts. The motive (DD)A(DD)A, in most cases was observed to lead to a FIM or to AFM behaviors. In the case of the sequence ··(DD)[AA](DD)[AA]··, the magnetic behavior was observed to be dominated in most cases by an AFM coupling in the [AA] dimmers.

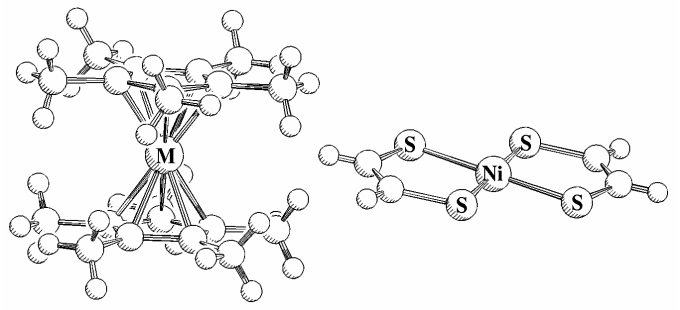

Fig. 1. Perspective view of the molecular structure of $[M(Cp^*)_2][Ni(edt)_2]$, M = Fe, Cr

Metamagnetic behaviors were observed in the compounds $[Fe(Cp^*)_2][Ni(edt)_2]$, $[Mn(Cp^*)_2][Ni(bdt)_2]$, $[Mn(Cp^*)_2][Ni(tds)_2]$ and $[Fe(Cp^*)_2][Ni(\alpha tpdt)_2]$, with T_c = 4.5, 2.3, 2.1 e 2.6 K and H_c = 4 kG, 200 G, 60 e 600 G. A FIM ordering was observed a temperature of 2 K in the case of $[Mn(Cp^*)_2][Pt(bdt)_2]$.

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Further work

We plan to focus the subsequent studies on the compounds presenting the most simple linear chain motives, ··ADADAD·· and ··(DD)[AA](DD)[AA]··. In these compounds we intend to study in more detail effects such as the competition between the intra and interchain coupling or the intradimmer interactions induced by minor changes on the dithiolate complexes.

* Funding: Contract PBIC/C/QUI/2204/95 (~7×10^6 PTE).
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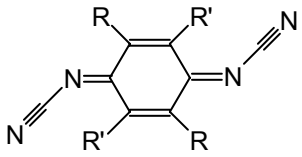
Molecular Magnetic Materials based on the Conjugated Polynitriles RR'-DCNQI*

V. Gama, R. T. Henriques[1], M. T. Duarte[1], D. Simão[1], I. Catarino[2], G. Bonfait[3], J. Soares[4], R. Meira[5]

Objectives

By the synthesis and characterization of new magnetic molecular materials, the coordination polymers $M(RR'\text{-}DCNQI)_2 \cdot ySolv$, where DCNQI = N,N'-dicyanoquinone diimine, M = Ni, Co, Fe, Mn, V, we pretend to achieve a better understanding of molecule based magnets.



The $M(RR'\text{-}DCNQI)_2 \cdot ySolv$ compounds were chosen, with the aim of obtaining magnetic materials ordering at high temperatures. Depending on the preparation both disordered and crystalline materials can be obtained, and these series of compounds constitute a broad basis in order to perform a consistent study concerning essentially the four main goals, cited below.

I- To correlate the magnetic behavior and magnetic ordering with the structure.

II-To study of the effect of weak intermolecular interactions, as Van der Walls and hydrogen bonding, due to variations on the substituents R, R' on the crystal structure.

III-To correlate the degree of disorder with the magnetic properties.

IV-To perform a detailed study on the influence of the different substituents (R, R'), due to the spin and charge distributions, on the magnetic behavior.

Results

This is a recent work that started in April 1999, and so far the main results consist on the synthesis of and chemical characterization of the disordered materials $M(DM\text{-}DCNQI)_2 \cdot ySolv$, with M = Ni, Co, Fe, Mn; Solv = MeCN and DCM; $y \approx 2$. The precursor compounds, $M(BF_4)_2 \cdot 6MeCN$ for the synthesis of the crystalline materials could be accomplished in this period.

Further work

The magnetic and structural characterization of the disordered materials will be carried on and the first crystalline materials are being prepared.

* Funding: Contract PRAXIS/P/QUI/12063/1998 (25444×10^3 PTE).

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Molecular Metals based on Transition Metal Chalcogenates*

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Objectives

Study the physical properties of new families of conducting charge transfer solids based in several organic donors and series of transition metal dithiolates and diselenates, as a part of the general aim of establishing correlations between structure and physical properties of molecule-based conductors. In particular it is aimed to study the influence of magnetic metal ions (in the chalcogenate complexes) upon the electrical transport.

Results

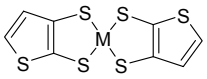
The series most deeply studied was the $(dt\text{-}ttf)_2M(mnt)_2$ (dt-ttf= dithiophenetetrathiaulvalene) for which the M=Au, Ni and Pt are conductors, although with different behaviours, the first being the first organic spin-ladder system (the gold bisdithiolate complex is diamagnetic), with poorer electrical conductivity than the other two, where electrons are more delocalised in the dt-ttf stacks[1]. The M=Co and Fe analogs are semiconductors. The spin ladder behaviour of the Au compound was confirmed by muon spin resonace[2].

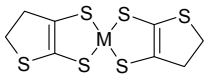
Other families of compounds prepared and studied comprise:

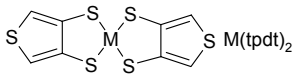
The $M(dcdmp)_2$ series (dcdmp= dicyanodimercaptopyrazine) which were combined with the donors bedt-ttf (*bis*-ethylenedithio-tetrathiafulvalene), tmtsf (tetramethyltetraselenafulvalene) and ttf (tetrathiafulvalene) [3]

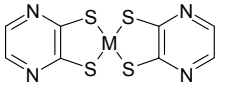
The $M(qdt)_2$ series combined also with the previous donors as well as with perylene [4];

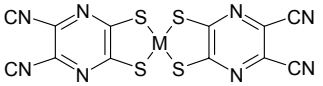
The new series of metal complexes with thiophenedithiolate ligands, (tpdt), α-thiophenedithiolate (α-tpdt) and dithiophenedithiolate (dtpdt), was recently prepared and the first conducting materials obtained either with the neutral complexes or with ttf type donors [5].











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Further work

To achieve one of the targets, the metal complexes with tpdt, α-tpdt and dtpdt are to be combined to the homologous donors, i.e., those for which the central metal is replaced by the ethylene fragment of the tetrathiafulvalene, dt-ttf for the first and bet-ttf for the third (the corresponding donor to α-tpdt was not synthesised so far) or alternatively cross-combined.

* Funding: Contract PRAXIS/2/2.1/QUI/203/94 (30×10^6 PTE), "*Multisulfured based charge transfer solids*", ICCTI-CSIC.

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Uranium based thin Films*

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Objectives

This project aims at the installation of a sputtering system dedicated to the preparation of thin films and multilayers containing uranium and to the study of the physical properties of these films with emphasis on the magnetic and magnetotransport properties, as a function of structure, film thickness, degree of crystallinity, number of layers, etc.

The comparison of the properties of these films and multilayers with those of materials based on transition elements is expected to shade light on the role of the uranium atoms in the magnetic properties

Results

This project started recently (September 1999) with external funding enabling just the design of a sputtering chamber with 3 sources and the purchase of DC and RF generators.

Further work

The sputtering system is expected to be operational by June 2000 and after this date deposition tests with uranium and iron will start.

We plan to start by preparing UFe_2 and $UFe_{10}Si_2$ thin films. The quality of the films will be investigated by Rutherford back scattering, and complemented by other methods, namely X-ray diffraction and Mössbauer spectroscopy. The physical characterization will be made by magnetization, Mössbauer-effect, electrical-resistivity and magnetoresistance measurements.

In a second step U/Fe multilayers will be prepared and studied.

* Funding: Contract PRAXIS/P7CTM712068/1998 (18×10^6 PTE).

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Intermetallics Derived from the $ThMn_{12}$ type structure *

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Objectives

This project aims at a detailed understanding of the role of the f elements (U and lanthanides) and d-elements, in the magnetic properties of intermetallic compounds derived from the $ThMn_{12}$-type structure. Special attention is given to the effects of spin orbit coupling on the f-element, spin exchange interactions between the f-element and the transition metal sublattices and the effect of variations of the atomic distribution by different sites on the magnetic properties and its anisotropy.

Results

An ^{57}Fe Mössbauer spectroscopy study on UFe_4Al_8 single crystal, in an external magnetic field, $\mathbf{B_{ext}}$, confirmed previous magnetization and magnetoresistance data which showed that before reaching saturation the magnetization is blocked perpendicularly to $\mathbf{B_{ext}}$. Furthermore, this study revealed that this blocking is a metastable state with a relaxation time of a few hours, at 4.2 K [1].

A consistent description of the Mössbauer spectra of UFe_xAl_{12-x} ($4 \le x \le 5.8$) was obtained for the first time. This model showed that the magnetic moments of the Fe atoms, μ_{Fe}, depend not only on the crystallographic site occupied by the Fe atoms but also on the number of Fe nearest neighbours. Since the distribution of the Fe atoms on each crystallographic site with different μ_{Fe} is random, it cannot be detected by neutron diffraction techniques [2].

A second magnetic transition below the magnetic ordering temperature was observed for the first time in UFe_xAl_{12-x} ($4 < x < 5$) intermetallics. A new magnetic phase diagram was proposed [3].

A simple and inexpensive apparatus for fast specific heat measurements between 15 K and 300 K was built attached to a usual Gifford Mac-Mahon cryocooler. Testing the system with a copper sample measured at velocities up to 200K/h, matched the tabulated values within 3%. This apparatus was used to measure specific heat of the UFe_xAl_{12-x} compounds [4].

A detailed investigation of the Y-Fe-Al and U-Pt-Sn phase diagrams was initiated. In the first case the stability range of the $ThMn_{12}$ type phase was found to extend from x=3.5 to 6, however without congruent melting[5]. YFe_2Al_{10} was found to crystallise in a new structure type.

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Further work

The dependence of the magnetic properties on the preparation of the AFe_xAl_{12-x} intermetallics (arc melting or pulling from the melt, thermal treatments, etc.) will be investigated.

Proceed with a detailed study of the magnetic structures already detected in the magnetic phase diagram of the UFe_xAl_{12-x} ($3.8 \le x \le 5.8$) and the characterisation of the magnetic phase transitions by specific heat measurements.

The distribution of μ_{Fe} in the RFe_xAl_{12-x} systems (R = Y, Lu) will be investigated by Mössbauer spectroscopy and compared with those observed in the U analogs.

* Funding: Contracts PRAXIS/PFIS/10040/1998 (4.8×10^6 PTE). Feb. 1999 - Feb 2001. PRAXIS/2/2.1/QUI/202/94 (10×10^6 PTE, 5×10^6 PTE for ITN).Oct 96-Oct 99.

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Strongly-correlated 5f-electron systems*

L. C. J. Pereira, A. P. Gonçalves, J. C. Waerenborgh, I. Catarino[1], G. Bonfait,[2]
M. Kuznietz[3], M. Almeida, P. A. S. Silva[4], M. Godinho[4].

Objectives

This project aims at establishing correlations between the crystal structure of intermetallic compounds with 5f elements and the unusual physical properties (heavy Fermion, spin fluctuations etc.) derived from the existence of narrow 5-f bands nearby or at the Fermi level.

Results

Several members of the family U_2T_2X, for which single crystals have been grown, where studied in detail. U_2Pt_2In was characterised as a non-Fermi liquid by measurements of electrical resistivity and specific heat at low temperatures [1]. In U_2Co_2Sn and U_2T_2In (T = Ni, Pd, Pt) the magnetic structure was investigated by neutron diffraction.. In the first case polarised neutrons enabled the determination of magnetisation density maps [2].

The $(U_{1-x}Th_x)_2Pt_2In$ alloys were prepared in order to study the electronic response of the uranium sublattice to the dilution by thorium.

A non Fermi Liquid behaviour was also established in $U_{3-x}Ni_3Sn_{4-y}$ obtained as single crystals for the first time [3]. Single crystals of $UNi_{1.9}Sn$ were also obtained for the first time and characterised [4]

In the borocarbide $Dy_2Ni_2B_2C$, one of the rare systems with both superconducting and antiferromagnetic statesat low temperatures, the effect of replacing U for Dy on both superconductivity and magnetic order was investigated. A rapid decrease of both the superconducting transition and Néel temperature was found, the last one attributed to different preferred orientation of magnetic moments, leading to a directional frustration [5].

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Further Work

Future work in this project will concentrate in the study by magnetisation and specific heath measurements of the alloys like $(U_{1-x}Th_x)_2Pt_2In$ that have been prepared, in order to probe the effect of Fermi level changes on the heavy fermion properties and on possible magnetic ordering in these systems.

* Funding: Contracts PRAXIS XXI, PRAXIS/P/FIS/12070/1998. (6.5million escudos). Feb. 1999 - Feb 2002.
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Vortex Motion in High Tc Superconductors*

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Objective

This project aims at the study of the vortex motion and pinning mechanisms in high Tc Superconductors, with a strong emphasis on magnetotransport measurements, in complement to magnetisation and other characterisation techniques.

Results

In the High Tc Superconductors, the scaling relation between the Hall (r_H) and longitudinal (r) resistivities, $\rho_H = \rho^2$, and the effect of pinning on the Hall conductivity have been subject of some controversy. We have studied this scaling relation at high magnetic field in unidirectionally twinned YBCO thin film specially patterned in order to inject the electrical current along $(\rho^{//},\rho_H^{//})$ or perpendicularly $(\rho^{\perp},\rho_H^{\perp})$ to the twin boundaries (TB). The scaling relations $\rho_H^{//} = (\rho^{//})^{\beta}$ and $\rho_H^{\perp} = (\rho^{\perp})^{\beta}$ were observed, with an exponent close to 2 for current parallel to the TB's and higher than 2 for perpendicular current, contradicting the models for purely anisotropic pinning, and reflecting the existence of isotropic pinning centers in the film. A different type of scaling relation was also observed: $\rho_H^{//} = (\rho^{//}\rho^{\perp})^{\nu}$ and $\rho_H^{\perp} = (\rho^{//}\rho^{\perp})^{\nu}$, with an exponent n close to 1 for the parallel current configuration, and higher for the perpendicular case. The anisotropy of the exponent n results from the TB contribution to pinning and is consistent with a pinning dependent Hall conductivity [1,2].

A high precision rotative sample holder was built to measure the angular dependence of the magnetoresistance in $YBCO_n/PBCO_m$ multilayers (n and m are the number of unit cell of YBCO and PrBCO respectively) in magnetic fields up to 16 T and with an angular resolution of 0.01°. Measuring r(T) at various angle and various magnetic field allowed to determine the activation energy U(Q,B) as a function of the angle Q between the c-axis and the magnetic field B. r(Q) at various magnetic field allowed, using the "standard" scaling law for anisotropic superconductors r(B,Q)=r(Bred) where $B_{red}=B(\sin^2Q+\cos^2Q/\Gamma^2)^{1/2}$, to determined the anisotropy parameter G(Q,B). We showed that, near Q=0, this parameter presents an unexpected field and angle dependence, in agreement with our U(Q,B) results. This dependence was interpreted as a consequence of vortex shape effects in the YBCO layers [3,4]. Let us note that these results were obtained only due to the high precision and resolution of our home-made rotative sample-holder.

$YBCO_n/PBCO_m$ multilayers grown by the new Pulsed Injection CVD developed in LMGP have been characterized by X-Ray (LMGP), magnetoresistance (Dpt. Química, ITN), magnetization (Dpt. Física, FCUL) and RBS (Dpt. Física, ITN). The results show a good cristallinity through the whole thickness and that critical currents, irreversibility lines and pinning energies are similar to those measured in multilayers obtained by other techniques [5].

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Further works

The effect of the TB will be studied with same type of experiments on single crystals in order to separate the contribution of the isotropic pinning centers and the TB pinning centers. The dependence of $\Gamma(\Theta,B)$ will be studied varying the thickness of the YBCO and PBCO layers.

* Funding: "*Vortex Motion in High Tc Superconductors*" PRAXIS, 2/2.1/QUI/410/94. (10×10^6 PTE)
"*Characterisation of superconducting thin films*", ICCTI-French Embassy.

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