* GRANT WEB-site: www.nato.ferroix.net

Final Report (updated August 2003) **CLG Ref. No.** PST.CLG.980055

NATO SCIENCE PROGRAMME
Cooperative Science and Technology Sub-Programme
COLLABORATIVE LINKAGE GRANT
NATO Public Diplomacy Division, Science Programme, Bd. Leopold III, B-1110 Brussels, Belgium
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GRANT CLOSURE

A. SCIENTIFIC REPORT

1. Project title (max 10 words)

NEW FERROELECTRIC - RELAXOR OXYDES FOR MICROELECTRONIC APPLICATIONS

Scientific Area

PST

2. Principal investigators

(a) NATO-country Project Coordinator (grantholder)

Surname/Initials/Title	Institute and address	Telephone, Fax and E-mail
DIEGUES Ernesto, Professor	*Dpto. Física de Materiales , Univ. Autónoma de Madrid 28049 Madrid*	*Tel./FAX* 91.397.4977 / 8579 E-mail: ernesto.dieguez@uam.es

(b) Partner-country Project Coordinator

Surname/Initials/Title	Institute and address	Telephone, Fax and E-mail
ELAATMANI Mohamed, Professor	*Lab. Mineral Solid State Chemistry* *Fac. of Sci. Semlalia, Univ. Cadi Ayyad* *B.P.2390, Marrakech, MOROCCO*	*Tel./FAX* +212-(0)44-43-46-49 / 67-69 E-mail: elaatmani@ucam.ac.ma

(c) Other Principal Investigators if any (please give names only)

Lukyanchuk Igor, Professor
Saber Mohamed, Professor

3. Publications resulting from the project (continue on additional page if necessary)

No.	Please list the publications from the entire grant and enclose one set of reprints and manuscripts accepted for publication if available	NATO support acknowledged	
		Yes	No
1	**Y. Gagou, Ch. Muller, M.-A. Fremy, D. Mezzane, E. Elkaïm and P. Saint-Grégoire;** *Structural study of ferroelectric and paraelectric phase in PbK2LiNb5O15*; Physica Status Solidi, B241, 2629 (2004)	**Yes**	
2	**M. Oualla, M. Elaatmani, M. Daoud 1, D. Mezzane, I. Luk'yanchuk and A. Zegzouti** *Study of New Rare Earth Family Pb1.6K1.2R0.2Nb5O15 (R = La, Nd, Sm, Eu and Gd) of Tetragonal Tungsten Bronze - Type Ferroelectrics*, Solid State Communication, 130, 777 (2004)	**Yes**	
3	**Y.Gagou , M. Elmarssi, M.-A. Frémy , N. Aliouane, D. Mezzane, P. Saint-Grégoire,** *Mechanism of the Phase Change in PbK2LiNb5O15: Dielectric, structural, and Raman scattering studies,* http://xxx.lanl.gov/abs/cond-mat/0601516, submitted to Europ. Phys. J. (2005)	**Yes**	
4	**A. Tabyaoui, A. Ainane, M. Saber and I. Lukyanchuk,** *Phase transition property of ferroelectric superlattice with three alternative layers from Ising model in a transverse field* Physica Scripta. 72, 265 (2005)	**Yes**	
5	**R. Ait Benhamaou, M. Daoud, A. Zegzouti, M. Elaatmani, E.Dieguez** *A synthesis and characterization of lanthanum potassium niobate oxide with tetragonal*	**Yes**	

	tungsten bronze (TTB) type structure Moroccan journal of condensed mater physics (2006), to be published		
6	**M. Saber, I. Lukyanchuk, M. Madani , A. Tabyaoui and A. Ainane,** *The transverse spin-1/2 Ising order-disorder superlattice,* Chin. J. Phys. 2006, to be publ.	**Yes**	
7	**D. Mezzane,Y. Gagou, M. ElMarssi, J.-L.Dellis, M. Elaatmani, I. Lukyanchuk** *Phase diagram of rare-earth TTB ferroelectric compounds Pb2(1-x) K(1+x)Gd(x)Nb5O15* Submitted, (2006)	**Yes**	

4. Abstract of the work accomplished and the results obtained

1. Y. Gagou, Ch. Muller, M.-A. Fremy, D. Mezzane, E. Elkaïm and P. Saint-Grégoire;
Structural study of ferroelectric and paraelectric phase in PbK2LiNb5O15;
Physica Status Solidi, B241, 2629 (2004)

The structures of PbK2LiNb5O15 showing the ferroelectricity below about 640 K have been studied in the paraelectric and ferroelectric phases by means of synchrotron X-ray powder diffraction. The data are analyzed with a Rietveld refinement method. It is found that the paraelectric structure and the ferroelectric one are of tetragonal and orthorhombic symmetry with *P*4/*mbm* and *Pba*2, respectively. The *Pba*2 structure gives a polar displacement along *c*-axis, whose direction is consistent with that deduced from dielectric measurements. The refined chemical occupancies of the cations Pb, K and Nb give the site-situation of these ions in the tunnels with square sections and pentagonal sections in each phase.

2. M. Oualla, M. Elaatmani, M. Daoud 1, D. Mezzane, I. Luk'yanchuk and A. Zegzouti
Study of New Rare Earth Family Pb1.6K1.2R0.2Nb5O15 (R = La, Nd, Sm, Eu and Gd) of Tetragonal Tungsten Bronze - Type Ferroelectrics,
Solid State Communication, 130, 777 (2004)

A new ferroelectric rare earth family Pb1.6K1.2R0.2Nb5O15 with R ¼ La, Nd, Sm, Eu and Gd (PKRN) of tetragonal tungsten bronze type ferroelectrics was synthesized. The ferroelectric transition with nonuniform distribution of critical temperature over a ceramic sample was found from dielectric measurements. According to X-ray diffraction measurements, the ferroelectric phase has an orthorhombic symmetry. The transition temperature was shown to decrease weakly with increasing radius of the rare earth ion R.

3. Yaovi Gagou , Mimoun Elmarssi, Marie Angèle Frémy (L2MP), Nadir Aliouane, Daoud Mezzane, Pierre Saint-Grégoire,
*Mechanism of the Phase Change in PbK2LiNb5O15: Dielectric, structural, and Raman scattering studies***,** http://xxx.lanl.gov/abs/cond-mat/0601516, submitted to Europ. Phys. J. (2005)

Experiments reveal that PbK2LiNb5O15 which belongs to the tetragonal tungsten bronze family presents paraelectric and ferroelectric phases and a complex structural change between them. High and low temperature phases are of symmetry P4/mbm and Pba2 respectively, so that this change is also of ferroelastic type. As presented here, crystallographic results hint at a displacive character of the ferroelectric ordering but show a more complex behaviour, with a clear order-disorder mechanism which accompanies the appearance of ferroelasticity. To complete our knowledge of this material, we have performed Raman experiments which exhibit a low frequency mode, but no clear soft mode is observed.

4. A. Tabyaoui, A. Ainane, M. Saber and I. Lukyanchuk,
Phase transition property of ferroelectric superlattice with three alternative layers from Ising model in a transverse field
Physica Scripta. **72**, 265 (2005)

Theoretical temperature, polarization and dielectric susceptibility in ferroelectric Superlattices with three alternative layers described by a transverse spin−1/2 Ising Model are studied using the effective field theory with a probability distribution technique. We discuss an L layer superlattice of simple cubic symmetry with nearest-neighbour interactions. We derive the phase diagram, the polarization profiles and the dielectric susceptibilities. In such superlattices, the critical temperature can shift to either lower or higher temperature compared with the corresponding bulk value. The superlattice dielectric longitudinal susceptibility diverges at the superlattice critical temperature.

5. R. Ait Benhamaou, M. Daoud, A. Zegzouti, M. Elaatmani, E.Dieguez
A synthesis and characterization of lanthanum potassium niobate oxide with tetragonal tungsten bronze (TTB) type structure
Moroccan journal of condensed mater physics (2006), to be published

The main aim of this study is to lower the reaction and sintering temperatures of K2LaNb5O15 in order to improve the quality of this material. The K2LaNb5O15 compound has been synthesized by soft chemistry (co precipitation) and its physico-chemical properties were studied. This method permits the synthesis of chemical purity, fine powder materials with good crystallization at low temperatures.

6. M. Saber, I. Lukyanchuk, M. Madani , A. Tabyaoui and A. Ainane
The transverse spin-1/2 Ising order-disorder superlattice
submitted to Chin. J. Phys. (2005)

Using the effective field theory with a probability distribution technique, we apply the Ising Model in a transverse field to analyse the properties of KH2PO4/KD2H2PO4 superlattice. The on-site polarization and dielectric susceptibility, their mean values, the susceptibility and the macroscopic pyroelectric coefficient are calculated for possible comparison with experimental data. We show for thick layer

superlattice, two peaks in the mean dielectric susceptibility and pyroelectric coefficient, as they had two phase transitions whereas thin-layer superlattices, show one peak behaviour.

7. D. Mezzane,Y. Gagou, M. El Marssi, J.-L.Dellis, M. Elaatmani, I. Lukyanchuk
Phase diagram of rare-earth TTB ferroelectric compounds Pb2(1-x) K(1+x)Gd(x)Nb5O15
submitted to (2006)

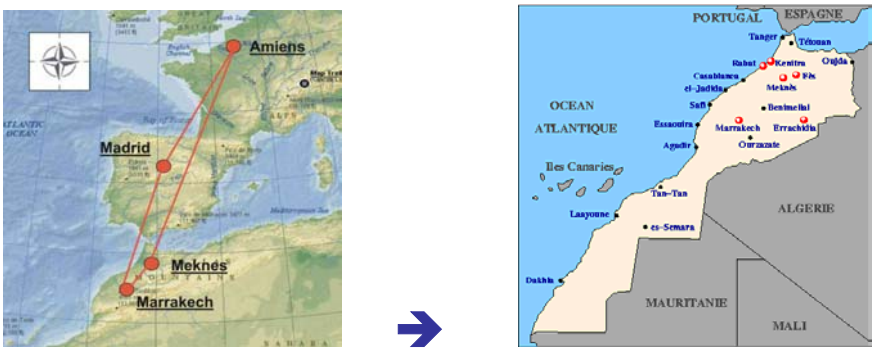
A new family of rare-earth ferroelectric TTB compound with general formula Pb2(1-x) K1+xGdxNb5O15 (0<x<1) are elaborated by solid state reaction. Structural and electrical properties are investigated using X-ray diffraction, Raman-spectroscopy and dielectric measurement. The symmetry of the ferroelectric phase and the transition temperature strongly depend of Pb. For the high lead concentration (x < 0.35) the ferroelectric phase is described by the symmetry group Cm2m. For the low lead concentration (x > 0.35) the symmetry group is Pba2. It is shown that transition temperature decreases with the lead concentration: for x=0.1 Tc=350°C and for x=0.4, Tc=220°C

Remarks: **IMPACT OF THE COLLABORATIVE LINKAGE GRANT** PST.CLG.980055:

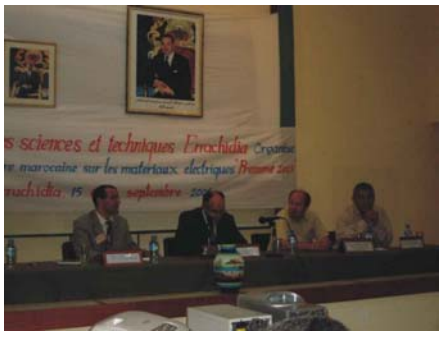
1. Scientific program was mostly completed (see abstracts). We consider the achieved results as the good point for further collaboration.

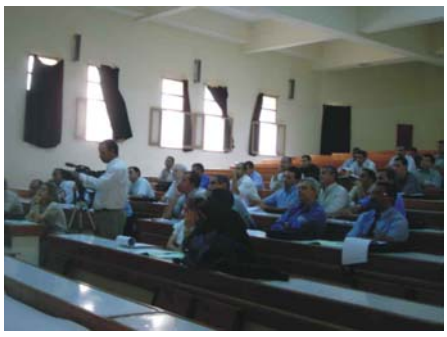
2. The NATO collaborative Mediterranean Network Marrakech-Amiens-Madrid-Meknes initiated the creation of the Moroccan material research network "Moroccan Electronic Materials" MEM (see Fig.) that unifies more then 9 scientific institution of Morocco. The objective of this Network is the creation of the horizontal collaborative links in Morocco and transferring the material research technologies from Europe to North Africa.

The working web-site of the MEM network is: www.reseau-mem.org

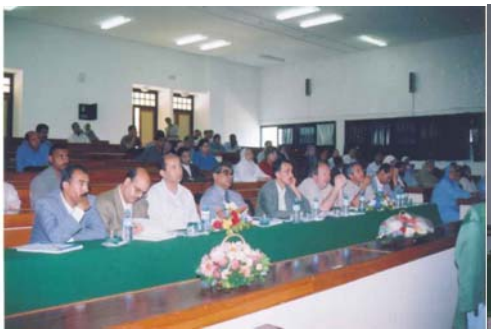


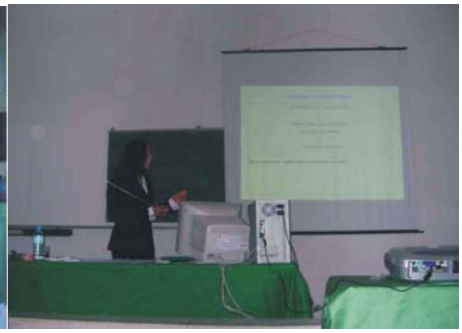
Two coordination meetings of the NATO linkage grant (Marrakech, 2004, Errachidia 2005) were transformed to the meetings of the MEM research network. The second one (PREMME-1, photo below) held in the small University of Errachidia in the Sahara desert region, became the national Moroccan congress with participation of researches from several European and Maghrebian countries. The additional information is available at www.reseau-mem.org



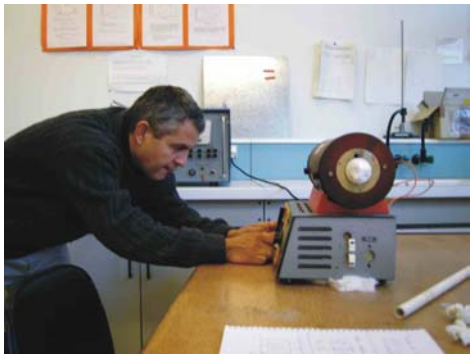


3. Two Moroccan PhD students: A. Tabyaoui and M. Oualla were formed during NATO-grant collaboration. The NATO linkage grant did help the formation of joint PhD commission (photo: PhD commission of A. Tabyaoui, Meknes, 2004)





4. Research equipment, not accessible in Morocco was bought in France and then transferred by Air-Cargo to Marrakech. The advantages of the European market and flexibility of the NATO grant were used to negotiate the cheapest prises. Another part of equipment (like crystal growth oven in photo) was donated by French participants to University of Marrakech.



5. Program of further developing of the created research Network was discussed in the Closure Meeting (Amiens, 2006)

a) Continue the research in the field of smart electroceramics and nano- deposited films using the advantages of delocalization and joint research experience of the created Network. The corresponding emerging Moroccan industrial societies will be involved.

b) Expand the created network Moroccan Electronic Materials MEM (www.reseau-mem.org) to other Mediterranean and Maghrebian countries. Several researches from Algeria and Mauritania were already contacted. Organize with this purpose in Marrakech in April-May 2007 the next regular meeting of the network MEM in the format of International Congress with active participation of researches from other Mediterranean countries. At the same time, apply for the satellite NATO Advanced Research Workshop, "Smart Materials for the Electronics, Communication and Security - **SMECS**" that will help to invite the leading scientists to present them the created research structure MEM and to stimulate the technology transfer from NATO countries to Mediterranean dialogue countries.

c) Apply for the National and European foundations for the grants to maintain and develop the created research network. Special emphasis in the projects will be given to design of gas sensors and detectors for the sake of application in the Environmental Security Control. Prepare and apply the NATO project "Science for Peace" involving the French, Moroccan, Spanish and Mauritanian partners.

6. All the participants are deeply grateful to the NATO Programme for Security Through Science for the outstanding possibility to organize the mutual collaboration links. We express the hope of the successful collaboration in a future.

(from left to right)

Igor Lukyanchuk [1], Daoud. Mezzane [2], Mohamed Elaatmani [2], Ernesto Dieguez [3], Mohamed. Saber [4], Abdelmajid Ainane [4], Mimoun El Marssi [1], Jean.-Claud Picot [1], Yaouvi. Gagou [1], Jean.-Luc Dellis [1]

[1] University of Amiens, France;
[2] University of Marrakech, Morocco;
[3] University of Madrid, Spain;
[4] University of Meknes, Morocco

B. FINANCIAL REPORT

Award, in Euro (Belgian francs if awarded before 2002): **11000**

		Currency **EUR**
INCOME:	Payment received	**11000**
	Bank interest earned	0
	TOTAL INCOME	**11000**
EXPENDITURE: ***(Include details below)***	1. Travel expenses	**3136**
	2. Living expenses	**5758**
	3. Other Expenditure *(Partner country only, as included in application form)*	**2106**
	TOTAL EXPENDITURE	**11000**
	OUTSTANDING BALANCE**	**0**

DETAILS OF TRAVEL AND LIVING EXPENSES (*)

(*) According to bilateral agreement between Amiens and Marrakech certain expanses and flights were paid by home/guest institutions. This allowed to extend the NATO visiting program. The corresponding expanses are notes as "0" in the financial report

Name	**From/To**	**Reason**	**Period from/to**	**Currency** (*specify*): **EUR**	
				Travel Expenses	**Living Expenses**
Y. Gagou	Amiens - Marrakech	Installation Impedance Specr.	10.12 - 17.12 2003	239	240
M. Saber	Meknes-Amiens	Theor. Modelling Discussion	07.01 - 20.01 2004	30	550
I. Luk'yanchuk	Amiens - Marrakech	Coordination	29.02 - 07.03 2004	277	240
I. Luk'yanchuk	Amiens - Marrakech	Sci. discuss. PhD committee,	04.06 - 16.06 2004	241	390
D. Mezzane	Marrakech - Amiens	Experiment: Laser deposition	16.09 - 07.10 2004	0	1100
M. Elaatmani	Marrakech - Amiens	Coordination,Mater. Ellaborat.	16.09 - 07.10 2004	0	1100
J.L.Dellis	Amiens - Marrakech	Imp. Spectr. measurements	01.12 - 08.12.2004	439 (package)	
J.C. Picot	Amiens - Marrakech	Install. of new equipment	13.05 - 20.05 2005	266	0
E.Diéguez	Madrid - Marrakech	Coordination,Mater. Ellaborat.	13.09 – 18.09 2005	456	200
M. El Marssi	Amiens - Marrakech	Dielectric measurements	18.12 – 27.12 2005	411	0
Y. Gagou	Amiens - Marrakech	Article preparation.	22.12 – 26.12 2005	374	0
A. Ainane	Meknes-Amiens	Theory, article preparation	18.01 - 24.01 2006	401	339
D. Mezzane	Marrakech - Amiens	Closing, report preparation	27.01 - 16.02 2006	0	1050
E.Diéguez	Madrid - Amiens	Closing, report preparation	31.01 – 05.02 2006	221.15	329.5
			Sub-Totals	**3136**	**5758**

Description of Other Expenditure (only applicable if included in application - Partner countries only)	**Other Exp**
Equipment was bought in Amiens and then sent to Marrakech by air-cargo, it includes the accessories for impedance spectroscopy and ceramic elaboration, automation of experiment: thermo-powers, electric connectors, PC-cards, temperature regulations, and related tools.	**2106**
Total	**11000**

Both the NATO-country and the Partner-country Coordinators should indicate their agreement to this report, by signing below.

Signature and Date:
NATO-country Coordinator

Signature and Date:
Partner-country Coordinator