APPLICATION FORM – Internet Version (issued September 2002)

**NATO SCIENCE PROGRAMME**
**Cooperative Science and Technology Sub-Programme**
**COLLABORATIVE LINKAGE GRANT**
**NATO Scientific Affairs Division, Bd. Leopold III, B-1110 Brussels, Belgium**
**fax +32 2 707 4232 : e-mail science@hq.nato.int**

Enter Scientific Area **PST**
Enter Research Topic of Special Interest if applicable: **Detectors, Sensors, etc.**
1.PROJECT TITLE: **NEW FERROELECTRIC - RELAXOR OXYDES FOR MICROELECTRONIC APPLICATIONS**
2. PRINCIPAL INVESTIGATORS
(i) Project Coordinator from a NATO country:
Surname/First Names(s)/Title: **DIEGUES Ernesto, Professor** Date of Birth: **1948**
Institute and Address: *Dpto. Física de Materiales , Univ.* 91.397.4977 / 8579
*Autónoma de Madrid 28049 Madrid*
Telephone : **+33 (0) 3 22 82 78 83**, Fax **: +33 (0)3 22 82 78 91**, E-mail: ernesto.dieguez@uam.es,

Signature:________________________________________
(ii) Project Coordinator from a Partner country:
Surname/First Name(s)/Title **ELAATMANI Mohamed, Professor** Date of Birth: **1952**
Institute and Address: ***Laboratory of the Mineral Solid state Chemistry,***
***Faculty of Sciences Semlalia, University of Cadi Ayyad,***
***B.P.2390, Marrakech, MOROCCO***

Telephone : **+212-(0)44-43-46-49**, Fax **: +212-(0)44-43-67-69**, E-mail: elaatmani@ucam.ac.ma,

Signature: ________________________________________
(iii) Other Principal Investigators

| Surname | First Name | Title | Institute and Address | Telephone/Fax/E-mail | Date of Birth |
|---|---|---|---|---|---|
| **Lukyanchuk** | Igor | Prof. | ***LPMCs, University of Picardy 33, rue Saint-Leu - 80039 Amiens France*** | : +33 (0) 3 22 82 78 83 / 91 lukyanc@ferroix.net | 1964 |
| **Saber** | **Mohamed** | Prof. | *Physics Department, Faculty of Sciences BP4010 Meknes Morocco* | *+212-55-538870 / 6808* saber@fsmek.ac.ma | *1958* |

3. SCIENTIFIC CODES and Percentage of discipline content

| | | | | | |
|---|---|---|---|---|---|
| **321** | **30 %** | **323** | **40 %** | **223** | **30 %** |

4. PROJECT KEYWORDS (*maximum 15*) **Ferroelectric, Relaxors, Sensors, Phase transitions, Ceramics, Modeling**
5. SUPPORT REQUESTED: (a) and (b) – for visits abroad (*add separate page if necessary*) US$

| *Name* | *Destinations* | *Duration* | (a) *Travel* | (b) *Living* |
|---|---|---|---|---|
| I. Luk'yanchuk | Amiens – Marrakech – Meknes | 14 days | 500 $ | 980 $ |
| M. El Marssi | Amiens – Marrakech | 7 days | 500 $ | 490 $ |
| V.Bermudez | Madrid – Marrakech | 7 days | 500 $ | 490 $ |
| D. Mezzane | Marrakech – Amiens | 7 days | 500 $ | 700 $ |
| M. Elaatmani | Marrakech – Madrid | 7 days | 500 $ | 700 $ |
| M. Saber | Meknes – Amiens | 7 days | 500 $ | 700 $ |
| I. Luk'yanchuk | Amiens – Meknes | 7 days | 500 $ | 490 $ |
| D. Mezzane | Marrakech – Amiens | 14 days | 500 $ | 1400 $ |
| J. L. Dellis | Amiens – Marrakech | 7 days | 500 $ | 490 $ |
| E.Diéguez | Madrid – Amiens | 7 days | 400 $ | 700 $ |
| A. Ainane | Meknes – Amiens | 7 days | 500 $ | 700 $ |
| M. El Marssi | Amiens – Marrakech | 7 days | 500 $ | 490 $ |
| M. Elaatmani | Marrakech – Amiens | 7 days | 500 $ | 700 $ |
| | | Sub-Totals | 6500 $ | 9030 $ |
| Other Expenditure - Partner countries<br>**Marrakech:** Furnace (T°max=1400C°)+ programmeur of Temperature (for ceramics elaboration with a good quality:density,compacity,porosity,retrait,etc) | | | | 4470 $<br>**20.000 $** |

## 6. RESEARCH PLAN

### *a. Summary*

**Project joints** NATO (Amiens-France, Madrid-Spain) and Mediterranean (Marrakech and Meknes, Morocco) material-research teams.

**Scientific objective.** Study of ferroelectric and relaxor properties of the recently synthesized family complex oxides of tungstate bronze (TKWB) type.

Fundamental importance. Ferroelectric transition involves a large variety of internal degrees of freedom (elasticity, nano-clustering, ion tunnelling) that leads to various intermediate incommensurate and relaxor phases of yet unknown structure.

Importance for applications. Several electro-mechanical characteristics are better then that for traditionally used perovskite materials that make them attractive for technology application in RAM, *sensors, detectors*, transducers, nonlinear electro-optic switches. Environment-friendly lead-free composition.

**We suppose** understand the mechanisms of ferroelectric transitions, the structural organization of the involved intermediate phases and evaluate the eligibility of these compounds for microelectronics.

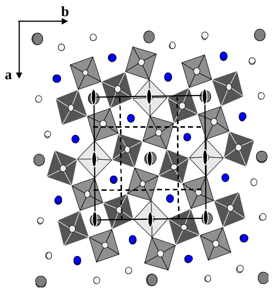
**Long-term objective.** Install the NATO - Mediterranean well-structured research network able to solve the complex problems of material science of ferroelectrics and related materials. This distributed network is integrated in the research structures of the NATO and Mediterranean countries, takes an advantage of the experimental setup and experience of each laboratory. It includes the complete material research cycle: elaboration – characterization – modeling – evaluation for application and will give the impact on the joint formation of the PhD students "by research".

### *b. Full description*

**State-of-art:**

Relaxor based ferroelectrics are of extreme importance as a key part of memory-storage devices, high performance actuators, sensors, high power ultrasounds transducers, underwater acoustics, advanced robotics micro-biomechanical and thermal sensors [Lines]. The special emphasis in modern researches is given to search of environment-friendly lead-free materials. The contemporary industrial usage of ferroelectric-relaxor materials is based on traditional perovskite materials in which the arrangement $MO_6$ octahedra in cubic network allows the easy structural engineering.

There is however a large variety of perspective complex oxides composed from the similar structural units $MO_6$, but having more complicated layer-structure [Landolt]. E.g. the SBT oxide ($SrBi_4Ti_4O_{15}$) has large advantages for technology: negligible fatigue, low leakage current, stability in nano-scale environment, [Scott]. Another BNN oxide ($Ba_2NaNb_5O_{15}$) can be used as an efficient second harmonic generator, stable to focused laser radiation [Lines].




Complex oxydes: Projection of $MO_6$ octahedra on plane (001)

Because of the complicated structure a little has yet been learned about the fundamental nature of phase transition in complex oxygen octahedral oxides. Similar to the perovskite materials, the polarization there is driven by displacement of the central ion in $MO_6$ octahedra, but the new degrees of freedom are involved in the ferroelectric transition. Coupling of the polarization with crystal elasticity and with subsystem of small-size ions that can tunnel through the vertical cavities provide the reach phase diagram with a series of the intermediate phases of yet unknown nature and a diffusive ferroelectric phase transition of the relaxor type with enhanced dielectric constant in a wide temperature region [Lines]. The later property makes the complex oxides attractive for the application as sensors, actuators etc.

**Objectives and motivation:**

We propose to investigate the ferroelectric and relaxor properties of the specified below new group of complex oxides of tungstate bronze (TKWB) type to evaluate their eligibility for microelectronics. Mostly lead-free oxides will be studied. At the same time the fundamentally important structural and dynamical properties of ferroelectric phase transition and of intermediate relaxor / incommensurate states will be understood.

**Network structure**

The proposed network has the appropriate experts structure (1/3 – technology, 1/3-experiment, 1/3-modeling) and the necessary experience and hard equipment to ensure the complete cycle of material research. The human resources are equally distributed between partner countries:

- Team "**Marrakech**": Lab.Min.Solid St.Chem.,& Fac. Sci.. Techn. Univ. Cadi Ayyad, Marrakech, MOROCCO
- Team "**Meknes**": Dept. of Phys., Univ. Meknes, MOROCCO
- Team "**Amiens**": Lab. Phys. Cond. Mat., Univ. Picardie, Amiens, FRANCE
- Team **"Madrid"**: Lab. Cryst. Growth., Autonomia Univ. Madrid, SPAIN

Moroccan participants are responsible for the elaboration and preliminary characterization of the ferroelectric-relaxor materials by locally-available experimental techniques and for theoretical and numerical modelling and design of the studied materials.

NATO participants provide more elaborate crystal growth and characterisation, based on the non-available in Morocco experimental technique, modelling and expert evaluation of possibility to use these materials in nanoscopic mono- and multilayered structures, integrated to microelectronic devices

**Materials to be studied:**

The participants of the current project dispose the family of the complex oxygen octahedral ferroelectric-relaxor materials of tungstate bronze (TKWB) structure with yet unknown properties.

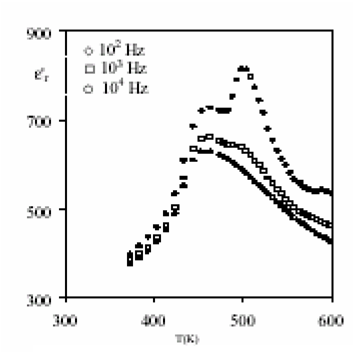
Large variety of Niobium (Titanium, Tantalum) - derivatives of $Ba_2NaNb_5O_{15}$ with sodium (potassium, lithium) are available and will be available [Elaatmani, Dieguez]. Such ceramics constitute a new family of lead-free relaxor compositions with TKWB structure and can be used in environment - friendly applications. The synthesis of the less studied tantalates is of the special interest.

Whereas these complex TKWB –type oxides are usually the classical ferroelectrics, their oxyfluoride analogs (like $BaNa_2Nb_5O_{14}F$) shows a relaxor-type behaviour with cationic disorder of $Ba_2^+$ and $Na^+$ ions in the same crystallographic site. We plane therefore to elaborate a series of oxyfluoride compositions ( $A_{2-x}B_{1+x}M_5O_{15-x}F_x$: A,B=alkaline, M= transition metals ) to study the appearing of relaxor phase.

Another available compounds are the complex oxides on base of PKN ($Pb_{1.2}Gd_{0.4}K_{1.4}Nb_5O_{15}$ , $PbK_2LiNb_5O_{15}$ ) that were shown to reveal a number of unusual phases of incommensurate / relaxor type just above ferroelectric transition [Mezzane].

**Properties to be studied and methods to be used.**

Several compounds were already characterized. It was found that they reveal the strongly enhanced dialectical constant in the wide temperature region [Mezzane] – the property which is very important for the listed above applications. There was also discovered the existence of unknown phase just above ferroelectric transition in the large temperature interval of presumably relaxor or incommensurate structural type (see Fig.). We believe that the precise identification of this phase will give the fundamental insight on the phase transition theory in the perovskites and complex oxides.



Two-peaks behavior of ε'(T) in $Pb_{1.2}Gd_{0.4}K_{1.4}Nb_5O$ . The lowest peak corresponds to ferroelectric transition and the highest one to the yet unknown phase of relaxor-type [Mezzane]

To understand the properties of these and the newly synthesized compounds, the following methods will be applied:

*Primary characterizing:* compositional analysis (ICP, EDAX, XPS, ATD, ATG, and MEB techniques) and structural analysis (XDR, HRDRX, RBS techniques).

*Raman spectroscopy:* (10K-1750K) will be used to understand the formation, evolution and local structure of the polar nano-clusters that are responsible for the relaxor-type behaviour [Marssi].

*Impedance spectroscopy:* (10K-1750K) will give the information about dynamical dielectric susceptibility. In addition, the temperature evolution of polarization P(T) and of the hysteresis loop P(E) will be done to understand the eligibility of these materials for application.

**Modelling**

Mathematical and numerical modelling will give the complete description of the ferroelectric transition in studied complex oxides, will clarify the structure of the observed intermediate relaxor and incommensurate phases and will provide the systematic way of optimization of the elaborated materials.

*Microscopic approach* in which polar displacement of the central ion in the $MO_6$ octahedron can be modelled by Ising variable will be based on statistical lattice model [Saber]. The appearing of the relaxor or / and incommensurate state is supposed to be modelled either by frustration mechanism like in [Lukyanchuk] or by the random-field impurities model.

*Landau functional approach* will allow to classify the phase transitions in the whole family of the considered ferroelectrics, calculate the principal thermodynamic parameters and incorporate the tunnelling of the small ions through the structural cavity tunnels and elastic degrees of freedom.

**Research facilities:**

| Marrakech | Meknes | Amiens | Madrid |
|---|---|---|---|
| *Resources and facilities* | | | |
| Elaboration<br>Powder and ceramics synthesis by solid state reaction ( $NaNb_2O_3$, $BaNb_2O_6$, $KNbO_3$,$SrNb_2O_6$, PKN( or SRK, BKN….), Films: cathode pulverisation, electro-deposition, sol-gel)<br>Characterisation<br>Powder X-ray diffraction, Dielectric study, ( $10^o$ - $10^8$ Hz ) , ATD, ATG techniques, and MEB, density measurements | Modelling<br>Experience in statistical microscopic models of ferroelectric relaxor phase transitions, Computers, internet | Caractérisation<br>Micro Raman, J.Y. T64000<br>Helium Cryostat (Oxford): 10-400K<br>Linkam hot stages TS1500, TS600 : (1500°C)<br>Impedancmeter Schlumberger 1260 (10Hz-1MHz)<br>Electrometer Keithly 614 (for P(T)); Ferro-tester radian (for P(E)), Piezoelectric measurements, Laser ablation film deposition.<br>Modelling<br>Experience in phenomenological models (Ginsburg-Landau, elasticity etc.)<br>Numerical modelling (MATLAB, C++)<br>Linux cluster | Elaboration<br>High-quality mono-crystals growth: Different types of Czochralski, Bridgman, Vapor growth, epitaxy and film deposition equipments.<br>Cutting and Polishing machines, 3D goniometer, Perkin Elmer DTA 1700, Atomic absorption Perkin Elmer, |

**Planning:** *(see also the planning of visits)*
1th year. General coordination of the project (Marrakech-Amiens). Preliminary selection and characterization of the most perspective compounds (Marrakech, Madrid). More elaborative analysis of these compounds by appropriate techniques in Amiens (Collaboration Marrakech-Amiens). In parallel: Developing the relevant theoretical models (Meknes-Amiens).
2nd year. Continuation of experimental activity in Amiens (Collaboration Marrakech,Madrid-Amiens). Collaboration: experiment-theory (Meknes-Marrakech, Amiens-Amiens). Application of the obtained results for the further optimization of compounds (Marrakech, Madrid). Expert evaluation of the elaborated oxides as candidates for microelectronics applications. (Marrakech, Amiens) and their eligibility for laser ablation technology (Amiens). Preparing the publications and recommendations for the perspective study (Meknes, Marrakech, Amiens, Madrid).

**Expected results:**

We suppose to characterize the available complex oxygen octahedral oxides by the described above experimental techniques and construct the relevant theoretical description of ferroelectric / relaxor states in these compounds. Basing on these results we will:

(i) Understand the fundamental physics of phase transitions in these oxides and microscopic structure of the observed intermediate phases.

(ii) Make the expert evaluation of possibility to elaborate the nano- films and superlattice structures and of the eligibility of theses materials for microelectronic applications.

All the results will be published in the international scientific journals and reported in the international meetings.

**Impact for research and academic infrastructure:**
For Morocco
Consolidation of university-industry relations that will be based on the already installed contacts with GEOPROJET society (Marrakech), exchange of scientific and technological know-how, design of high permittivity compensators, electro-mechanic sensors, infra-red detectors, frequency modulators.
For NATO countries
Reducing of costs of design and research of smart ferroelectric compounds related with their elaboration and primary characterisation that will be done in Morocco.
Perspectives of the cooperation
This project will allow to create the well-equipped and structured long-term NATO - Mediterranean dialogue research network able to resolve the complex problems of material research in area of ferroelectric oxides.
Young researchers training. is planned. The proposed material research network will allow to engage the young research students in the frame of joint PhD program "These cotutelle"

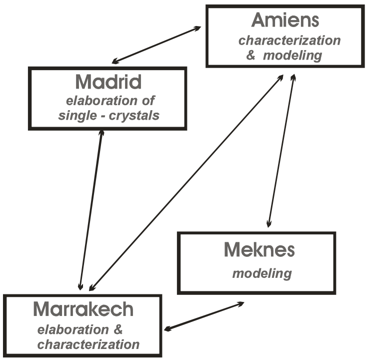
**References**


[Lines] M. E. Lines and A. M. Glass, *Principles and application of ferroelectric materials*; Clardemon Press, Oxford, 1977
[Landolt] Landolt - Bornstein; New Series, Gr. III V.16a *Ferroelectrics and Related Substance*; Springer-Verlag, 1981
[Scott] J.F. Scott. *The Physics of Ferroelectric Ceramic Thin Films for Memory Applications*; Ferroelectrics Review **1**, 1 (1998)
[Lukyanchuk] I. Luk'yanchuk, A. Jorio and M.Pimenta, Phys. Rev. **B57**, 5086 (1998); **B61**, 3147 (2000)
[Saber] N. El Aouad, B. Laaboudi, M. Kerouad and M. Saber; J. Phys.: Condens. Matter **13**, 797-809 (2001)
[Elaatmani] R.Von der Muhll,Asimon, M. Elaatmani and J.Ravez; Materials Letters **55**; 138 (2002)
[Mezzane] Y. Gagou, D. Mezzane, et al., Ferroelectrics, **251**, 131-137 (2001); and Ferroelectrics accepted (2002)
[Marssi] R. Farhi, M. El Marssi, A. Simon, J. Ravez, European Phys. J. **B9**, 599 (1999) **B18**, 605 (2000),
[Dieguez] See web-site :www.uam.es/departamentos/ciencias/fisicamateriales/especifica/grupos/crystal_growth_lab/main~1.htm

## 7. INTERNATIONAL COOPERATION

**Elaboration**

The group from Marrakech will produce the powder and ceramic oxides of the tetragonal tungsten bronze (TKWB) type derived from $Ba_2NaNb_5O_{15}$ composition: $Sr_3TiNb_4O_{15}$, $Ba_2BiTi_2Nb_3O_{15}$, $Pb_2KNb_5O_{15}$, $BaNa_2Nb_5O_{14}F$, etc. and will attempt to elaborate the films of the same stochiometry by cathode pulverisation and / or sol gel deposition. The optimisation of the synthesis and composition of materials as a result of feed-back from experimental and theoretical characterisation is planned.




Network structure

The Madrid team will elaborate several high-quality single-crystals using Czochralski method to compare their structural anisotropic properties (dielectric constant, Raman spectroscopy etc) with averaged characteristics of powder and ceramic samples.

**Characterisation**

The group from Marrakech will be specialized in the structural and electrical characterisation of materials.
*Powder X-ray diffraction* will be used to determine the crystallographic structure and the limits of the solid solution region.
*The dielectric study* of the complex dielectric permittivity above room temperature in frequency region ( $10^o$-$10^8$ Hz ) will be done on the ceramic disks with deposited gold or platinum electrodes. The characterization by *ATD, ATG techniques*, and MEB, *density* measurements is planned
The Amiens group has considerable expertise in study of the electrical and spectroscopic features of ferroelectric-relaxors [Marssi].
*The micro-Raman spectroscopy* will be performed in the temperature range of 10K-1750K (cryostat + hot stage), to study the evolution of polar nano-clusters.
*The impedance spectroscopy* of the dielectric susceptibility will complete the measurements of the Marrakech group to the wide temperature region (10K-1750K). In addition, the temperature evolution of polarization P(T) and of the hysteresis loop P(E) will be done to understand the eligibility of the materials for application.
Expert evaluation. The group of laser ablation in Amiens will evaluate the eligibility of these compounds for preparation substrate-deposited nano-films and sandwiched superlattice structures.

**Modelling**

The group from Meknes will apply the microscopic modelling on base of statistical Ising model with frustrations to understand the origin and structure of intermediate non-uniform phases (incommensurate phase, relaxor etc.)
The group from Amiens will implement the modelling on the base of alternative phenomenological Landau functional and numerical modelling to understand the macroscopic properties of the materials.
Results of both groups will be used for further optimisation of ferroelectric oxides.

**Visits**

| | *Name* | *Destinations* | *Duration* | Justification |
|---|---|---|---|---|
| **1th year** | I. Luk'yanchuk | Amiens–Marrakech – Meknes | 14 days | Coordination, "start-up" the modeling |
| | M. El Marssi | Amiens – Marrakech | 7 days | Coordination "start-up" and participation in experiment, |
| | V.Bermudez | Madrid – Marrakech | 7 days | Coordination of elaboration of ceramics |
| | D. Mezzane | Marrakech – Amiens | 7 days | Participation in experiments (Raman) |
| | M. Elaatmani | Marrakech – Madrid | 7 days | Elaboration of single-crystals, materials exchange |
| | M. Saber | Meknes – Amiens | 7 days | Discussion of results of modelling, |
| **2nd year** | I. Luk'yanchuk | Amiens – Meknes | 7 days | Coordination in modeling, recommendations to optimization |
| | D. Mezzane | Marrakech – Amiens | 14 days | Participation in experiments (Raman, Impedance), Congress |
| | J. L. Dellis | Amiens – Marrakech | 7 days | Joint experiments and coordination of dielectric measurements |
| | E.Diéguez | Madrid – Amiens | 7 days | Raman characterization on the elaborated crystals |
| | A. Ainane | Meknes – Amiens | 7 days | Final results in modelling, preparing the publication |
| | M. El Marssi | Amiens – Marrakech | 7 days | Final results in experiment, publication, Congress |
| | M. Elaatmani | Marrakech – Amiens | 7 days | Results in elaboration, closing the project. Final report. |

## 8. EXPECTED DURATION OF THE COLLABORATION

ONE YEAR [ ] TWO YEAR [X]

**9. INVESTIGATORS**

**(a) Provide below the names of the <u>other</u> Investigators participating in the project who will benefit from NATO funding under this CLG**

| ***<u>Name</u>*** | ***<u>Discipline</u>*** | ***Highest Degree*** | ***Affiliation •*** | *<u>% of time</u>* |
|---|---|---|---|---|
| | | | | |
| **N. ACHARGUI** | Solid State Physics | Ph. D. | Marrakech | **30 %** |
| **A.ZEGZOUTI** | Solid State Chemistry | Ph. D. | Marrakech | **20 %** |
| **D. MEZZANE** | Solid State Chemistry | Ph. D. | Marrakech | **70 %** |
| | | | | |
| **A. AINANE** | Solid State Physics, | Prof. | Meknes | **40 %** |
| | | | | |
| **M. EL MARSSI** | Solid State Physics, | Ph. D. | Amiens | **70 %** |
| **J.- L. DELLIS** | Solid State Physics, | Ph.D. | Amiens | **30 %** |
| **M. KARKUT** | Solid State Physics, | Prof. | Amiens | **5 %** |
| | | | | |
| **V.BERMUDEZ** | Solid State Chemistry | Ph. D | Madrid | **20%** |

(•)

- Team “**Marrakech”**: Lab.Min.Solid St.Chem.,& Fac. Sci.. Techn. Univ. Cadi Ayyad, Marrakech, MOROCCO
- Team “**Meknes”**: Dept. of Phys., Univ. Meknes, MOROCCO
- Team “**Amiens”**: Lab. Phys. Cond. Mat., Univ. Picardie, Amiens, FRANCE
- Team **“Madrid”**: Lab. Cryst. Growth., Autonomia Univ. Madrid, SPAIN

**(b) Have the Project Coordinators or any of the other Principal Investigators or Investigators been supported in the past by a NATO grant?**

**<u>NO</u>**

**10. REFEREES**

*Suggest three referees from NATO member countries other than those of Investigators, and provide their <u>complete</u> addresses. Referees should not be former research directors or associates, or postgraduate students of participants.*

**(a)** **Prof. Jan Petzel**, Inst. of Physics AS CR, Na Slovance 2, 182 00 Praha 8 Czech Republic ;
phone: (++420) - (2) - 66 05 11 11 fax: **- 868 90 527 E-mail: petzelt@fzu.cz.

**(b) Priv. Doz. Dr. Peter Lemmens**, Max-Planck-Institut für Festkörperforschung, Heisenbergstr. 1, D - 70569 Stuttgart, Germany Tel.: 0711-689-1754; Fax: **16 32; E-mail: lemmens@fkf-mpg.de

**(c) Prof. Paolo Mazzoldi** University of Padova, Department of Physics, IT-Padova Italy Tel: 39 49 8277002 Fax: **003
E-mail: Mazzoldi@padova.infm.it.

**11. COSTS**

*Give an <u>estimate</u>, in US$, of the total yearly cost for the research (excluding salaries and costs covered by NATO grant), and indicate sources of support (excluding NATO support).*

| ***<u>Team</u>*** | ***<u>Source</u>*** | ***<u>Cost / year</u>*** |
|---|---|---|
| **Marrakech** | Ministry and Industry financial support ( PROTARS II PROJECT) | **6000 $$** |
| **Meknes** | University support | **small** |
| **Amiens** | Regional grant “Multi-functionality of materials”+ Educational Ministry Support | **40.000 $$** |
| **Madrid** | Ministry and Regional projects support | **25000 $$** |

**Date: 25.02.2003** ***Please ensure that both Project Coordinators sign on Page 1***