PROGRESS REPORT
THEORY OF STRUCTURE AND DYNAMICS IN CONDENSED MATTER
AND
APPLICATIONS IN GEOPHYSICS AND SOLAR ENERGY

1 April 1975 - 31 March 1976

J. A. Krumhansl

ERDA Contract No. E(11-1)-3161

NOTICE
This report was prepared as an account of work
sponsored by the United States Government. Neither
the United States nor the United States Energy
Research and Development Administration, nor any of
their employees, nor any of their contractors,
subcontractors, or their employees, make any
warranty, express or implied, or assume any legal
liability or responsibility for the accuracy, completeness
or usefulness of any information, apparatus, product or
process disclosed, or represents that its use would not
infringe privately owned rights.

Laboratory of Atomic and Solid State Physics
Cornell University
Ithaca, New York 14853

March 1976

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Progress Report
Theory of Structure and Dynamics in Condensed Matter
and
Applications in Geophysics and Solar Energy

SUMMARY

This has been the most successful year in the history of the program supported by this contract. Not only have the basic materials science objectives been met, but also we have tested theory built up over the past years in this program on the properties of rocks, and on the design of composite photothermal conversion surfaces.

The major topics have been:

1. Theory of Structural Phase Transitions. (Krumhansl and Bishop)
2. Theory of Interatomic Interactions, with Application to Defect Structures and Surface Complexes. (Krumhansl and Hathaway)
3. Application of the Theory of Disordered (Heterogeneous) Materials to the Elastic Properties of Rocks, Commercial Graphite, and Composite Solar Energy Absorbing Materials.

DISCUSSION

(1) Structural phase transitions occur in many situations: Ferroelectrics, Spinodal Decomposition, and other Diffusionless transitions, to mention a few. The dynamics of how these transitions take place has been a subject of extensive investigation in materials science, by x-ray, neutron and optical scattering, particularly.

One model is the "soft mode" idea, whereby the oscillation of the lattice in a mode whose displacement would take it to a new structural phase becomes unstable as the temperature is varied, and the system spontaneously changes

structure. It is this idea which we have sought to place on a firmer theoretical basis. The foundation was laid in work done by Krumhansl and J. R. Schrieffer, begun on sabbatic at the University of Pennsylvania.

During the past (contract) year we have moved on in several directions from that work:

- (1) Confirmed by computer simulation that the non-linear nature of the large amplitude displacement fields in structural phase transitions leads to new features in the structure near transitions. That is, "microdomains" of the alternate phase appear; moreover, the statistical mechanics can be constructed in such a way as to relate these to atomistic interactions.
- (2) The basic theory is related to an extensive body of literature on non-linear waves, particularly those called "solitons". Our work has been the first to combine non-linear dynamics and statistical mechanics.
- (3) For the last few years, in studies of the dynamics near phase transitions, a particular feature, the "central peak", has been noted experimentally as a very narrow quasi-elastic peak in neutron and Raman scattering. There are probably several alternative ways in which this may be explained, but the presence of "microdomains" provides a new possibility.
- (4) We have also applied the methods to the unusual quasi one dimensional organic conductors, such as TTF-TCNQ. In this situation the physics is somewhat different because electronic and structural transitions are coupled. We have, in collaboration with M. J. Rice (Xerox), extended our theoretical ideas to discuss the low temperature insulating phase of these materials, and the novelty of the approach has drawn considerable attention.

The above work has attracted general interest. It has been noted in *Physics Today* ("Search and Discovery", February, March 1976), and at the "European Conference in Ferroelectricity", Zurich, September, 1975. Invited papers were given at the "International Conference on Low Lying Excitation and Super-

conductivity", San Juan, December 1975, and the Solid State Sciences Panel, December 1975. A number of seminars and regular papers have been presented, and the following papers have been published by J. A. Krumhansl with co-authors listed:

"Dynamics and Statistical Mechanics of a One-Dimensional Model Hamiltonian for Structural Phase Transitions", Phys. Rev. B 11, 3535 (1975) with J. R. Schrieffer.

"Mean Field and Exact Results for Structural Phase Transitions in 1-D and very Anisotropic 2-D, 3-D Systems:", with A. R. Bishop, Phys. Rev. B 12 2824 (1975).

"Molecular Dynamics Simulation of Model For (One-Dimensional) Structural Phase Transitions", Solid State Communications, Vol. 17, (1975) with T. R. Koehler, A. R. Bishop, and J. R. Schrieffer.

"Weakly-Pinned Fröhlich-Charge-Density-Wave Condensates: A New, Nonlinear, Current-Carrying Elementary Excitation", Physical Rev. Letters 36, 432 (1976) with M. J. Rice, A. R. Bishop, and S. E. Trullinger.

The following are in preprint or preparation:

"Non-Linear Excitations in Anisotropic X-Y Models", submitted to Physics Letters, A. R. Bishop and J. F. Currie.

"Classical Particle-Like Behavior of Sine-Gordon Solitons in Scattering Potentials and Applied Fields", submitted to Phys. Rev. Letters, M. B. Fogel, S. E. Trullinger, A. R. Bishop.

"Microscopic Derivation of Domain Walls", submitted to the Journal on Ferroelectrics, E. Domany, A. R. Bishop.

It is likely that in the solid state community there will be considerable general activity in this topic during the next year, and we hope to continue to be among the main contributors. During part of the contract year Visiting Professor R. A. Guyer has consulted with this program.

(2) Ms. K. B. Hathaway and J. A. Krumhansl have essentially completed the work modifying "Extended Hückel Theory" for solid state applications, particularly surface physics and defect structures. We have possibly accomplished a more systematic comparison of the methods used with first principle quantum chemical

computations, and thereby suggested improvements. These have been tested successfully for ionic molecules, and noble gas interactions. In addition Hathaway, in collaboration with D. Welch, of Brookhaven, has set up the programs for looking at defects and at the cohesive energy of MgO; results to date are promising. Hathaway's thesis is now completed.

Publications: (J.A. Krumhansl with co-authors)

"Extended Hückel Calculation of Repulsive Interactions in Noble Gases", Journal of Chemical Physics, Vol. 63, No. 10 (1975), Kristl B. Hathaway.

"Potential Energy and Force Constants of a Potassium Chloride Molecule from Extended Hückel Calculations", J. Chem. Phys. Vol. 63, No. 10, (1975) with Kristl B. Hathaway.

(3) For some years the principal investigator worked on the theory of random or heterogeneous systems, particularly on the quantum mechanics and lattice vibrations of atomically disordered systems (c.f. R. J. Elliott, J. A. Krumhansl, P. L. Leath, Rev. Mod. Phys. 46, 465 (1974)).

With some apology for "advertising", it is nonetheless noteworthy that theoretical work has a certain multipurpose value not always self evident from its immediate appearances. Here we make the point that the methodology developed for those basic studies has now been transcribed to:

(a) "The Elasticity of Polycrystals and Rocks", E. Domany, J. E. Gubernatis, J. A. Krumhansl; Journal of Geophysical Research, 80, 4851 (1975).

(b) "Macroscopic Engineering Properties of Polycrystalline Materials" Elastic Properties", Journal of Applied Physics, 46, 1875 (1975).

(c) "Theory and Computation of Effective Electrical and Optical Properties of Composite (Photothermal) Materials", J. A. Krumhansl, Tucson (Nov. 1975)

Symposium on Optical Properties of Solids Relevant to Solar Energy Conversion. (Invited)

- (d) "Self Consistent Calculations of Elastic Constants of Polycrystalline Graphite", R. E. Smith, G. B. Spence, J. E. Gubernatis, and J. A. Krumhansl, Submitted to "Carbon".

We believe that these applications to geophysics, solar energy materials, and reactor structural materials can, with further development, contribute to better materials design methods.

GENERAL

Numerous seminars, or contributed papers, on our work have been given by A. R. Bishop, J. A. Krumhansl, and by colleagues supported by other sources (M. Fogel, S. E. Trullinger, J. E. Gubernatis, and E. Domany). These are not listed in detail, since the above listed publications are more to the point as an official record.

Personnel associated with J. A. Krumhansl during 1975-76, and receiving some ERDA support were:

Dr. A. R. Bishop

Visiting Professor R. A. Guyer

Ms. K. Hathaway

Mr. P. Muzikar

Mr. D. Wood

PUBLICATIONS

COO-3161-24

MACROSCOPIC ENGINEERING PROPERTIES OF POLYCRYSTALLINE MATERIALS, I: GENERAL METHODS

J. Appl. Phys. 46 1875 (1975)

J. E. Gubernatis and J. A. Krumhansl

COO-3161-25

STOCHASTIC THEORY OF SPIN-PHONON RELAXATION

Phys. Rev. B. 11 4470 (1975)

R. Pirc and J. A. Krumhansl

COO-3161-26

TWO POINT CORRELATION FUNCTION FOR GENERAL FIELDS AND TEMPERATURES IN THE CRITICAL REGION

Phys. Rev. B 11, 4661 (1975)

M. Combescot, M. Droz, and J. M. Kosterlitz

COO-3161-27

DYNAMICS AND STATISTICAL MECHANICS OF A ONE-DIMENSIONAL MODEL HAMILTONIAN FOR STRUCTURAL PHASE TRANSITIONS

Phys. Rev. B 11, 3535 (1975)

J. A. Krumhansl and J. R. Schrieffer

COO-3161-28

ON THE EQUIVALENCE OF CONFIGURATIONAL AVERAGING FORMALISMS IN THE CPA: TWO-PARTICLE PROPERTIES

J. of Phys. C.: Solid State Physics 8, 3317 (1975)

A. R. Bishop

COO-3161-29

POTENTIAL ENERGY AND FORCE CONSTANTS OF A POTASSIUM CHLORIDE MOLECULE FROM EXTENDED HÜCKEL CALCULATIONS

J. Chem. Phys. vol. 63, no. 10 4313 (1975)

Kristl B. Hathaway and James A. Krumhansl

COO-3161-30

EXTENDED HÜCKEL CALCULATION OF REPULSIVE INTERACTIONS IN NOBLE GASES

J. Chem. Phys. vol. 63, no. 10 4308 (1975)

Kristl B. Hathaway and J. A. Krumhansl

COO-3161-31

THE ELASTICITY OF POLYCRYSTALS AND ROCKS

J. Geophysical Research Vol. 80, No. 35 4851 (1975)

E. Domany, J. E. Gubernatis, and J. A. Krumhansl

COO-3161-32

COMMENTS ON DILUTE ISING SYSTEMS: CLUSTERING AND TRICRITICAL BEHAVIOUR

Submitted to the Journal of Magnetism

A. R. Bishop and E. Domany

COO-3161-33

MEAN FIELD AND EXACT RESULTS FOR STRUCTURAL PHASE TRANSITIONS IN 1-D AND
VERY ANISOTROPIC 2-D, 3-D SYSTEMS

Phys. Rev. B 12, 2824 (1975)

A. R. Bishop and J. A. Krumhansl

COO-3161-34

MOLECULAR DYNAMICS SIMULATION OF A MODEL FOR (ONE-DIMENSIONAL) STRUCTURAL PHASE
TRANSITIONS

Solid State Communications, Vol. 17, 1515 (1975)

T. R. Koehler, A. R. Bishop, J. A. Krumhansl, and J. R. Schrieffer

COO-3161-35

ABSENCE OF LOCALIZATION IN THE COHERENT POTENTIAL APPROXIMATION: THE ONE-
DIMENSIONAL LIMIT

Solid State Communications, Vol. 17, 1405 (1975)

A. R. Bishop

COO-3161-36

MICROSCOPIC DERIVATION OF DOMAIN WALLS

Submitted to the Journal on Ferroelectrics and presented at the Puerto Rico Conference

E. Domany, A. R. Bishop and J. A. Krumhansl

COO-3161-37

WEAKLY-PINNED FROHLICH-CHARGE-DENSITY-WAVE CONDENSATES: A NEW, NONLINEAR,
CURRENT-CARRYING ELEMENTARY EXCITATION

Physical Rev. Letters 36, 432 (1976)

M. J. Rice, A. R. Bishop, J. A. Krumhansl and S. E. Trullinger

COO-3161-38

NON-LINEAR EXCITATIONS IN ANISOTROPIC X-Y MODELS

Submitted to Physics Letters

A. R. Bishop and J. F. Currie

COO-3161-39

CLASSICAL PARTICLE-LIKE BEHAVIOR OF SINE-GORDON SOLITONS IN SCATTERING
POTENTIALS AND APPLIED FIELDS

Submitted to Phys. Rev. Letters

M. B. Fogel, S. E. Trullinger, A. R. Bishop, and J. A. Krumhansl

PERSONNEL ACTIVITIES

The principal investigator, J. A. Krumhansl, devoted 25% of his time at Cornell to work on this project during the academic term 1975-76. Dr. A. Bishop will continue until September 1976, and a new postdoctoral associate will succeed him. Ms. K. Hathaway will complete her Ph.D. in June 1976. Two new research assistants Mr. P. Muzikar and Mr. D. Wood have received partial support for exploratory work on photothermal materials. Visiting Professor R. A. Guyer (University of Massachusetts) is consulting with us on the structural phase transition work,