ORNL/FTR--3783

DE91 001526

OAK RIDGE NATIONAL LABORATORY

OPERATED BY MARTIN MARIETTA ENERGY SYSTEMS, INC.
POST OFFICE BOX 2008, OAK RIDGE, TENNESSEE 37831-6285

ORNL

FOREIGN TRIP REPORT

ORNL/FTR-3783

Date: October 15, 1990

Subject: Report of Foreign Research Assignment of M. M. Abraham,
Senior Research Staff Member, Ceramics and Interfaces Section,
Solid State Division, ORNL

To: Alvin W. Trivelpiece

From: M. M. Abraham

Purpose: To perform collaborative research involving optical
characterization techniques applied to high-temperature ceramic
oxides at Clarendon Laboratory, Oxford, United Kingdom.

Sites Visited: 10/3/89-9/25/90 Clarendon Laboratory, M. J. M. Leask
Oxford University, J. M. Baker
Oxford, U. K. B. Bleaney
M. Wells
R. Ward

Abstract: The traveler collaborated with M. J. M. Leask, J. M. Baker,
B. Bleaney, and others at the Clarendon Laboratory, Oxford
University, Oxford, U. K., to study tetragonal rare-earth phosphates
and vanadates by optical and magnetic spectroscopy. This work is
related to similar studies that have been performed at ORNL by the
Synthesis and Properties of Novel Materials Group in the Solid
State Division. Some of the work was reported at an International
Meeting in Stuttgart, Federal Republic of Germany, September 1990,
and several papers are in preparation.

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.

CLARENDON LABORATORY, OXFORD UNIVERSITY, OXFORD, U. K.

The purpose of this foreign research assignment was to initiate collaborative research on the tetragonal rare-earth phosphates and vanadates with colleagues at the Clarendon Laboratory of Oxford University, Oxford, U. K. Single crystals of these materials have been grown and subsequently characterized at the Clarendon Laboratory by employing measurements of the magnetic susceptibility and specific heat.

Discussions were carried out with M. Wells and M. J. M. Leask regarding the history of such measurements at the Clarendon Laboratory. It was discovered that while many of these rare-earth phosphates and vanadates were ferromagnetic at room temperature, they pass through a phase transition at lower temperatures and become either antiferromagnetic or ferrimagnetic. The particular temperature at which the transition takes place is called the Neél temperature, and for these materials, the Neél temperature lies in the temperature region only attainable using liquid helium. Below this Neél temperature, the magnetic moments are not randomly oriented, but may be antiparallel in pairs (for antiferromagnetic) resulting in two nonzero magnetic fields that are equal but opposite in direction. Ferrimagnetic materials have two nonzero magnetic fields that are unequal but opposite in direction.

Research on similar structured tetragonal silicates, phosphates, and vanadates have been performed at ORNL by the Synthesis and Properties of Novel Materials Group in the Solid State Division. Our magnetic studies have revealed the presence not only of substitutional sites for various impurities, but also of specific orthorhombic impurity sites which were symmetrically oriented in the tetragonal host. Subsequently, at the Clarendon Laboratory, the electronic energy levels of the rare-earth phosphates and vanadates have been measured by magnetic and optical spectroscopy using state-of-the-art equipment, including superconducting-solenoidal magnets and dye lasers of different frequencies.

Specifically, it was found that a normal strictly forbidden optical transition for Eu^{3+} in EuVO_4 [$^5\text{D}_0 \leftrightarrow ^7\text{F}_0$] had satellite lines in the spectrum indicating breakdowns in the local tetragonal symmetry. By burning holes in the

satellite lines, new information regarding the different defects could be obtained by utilizing the quadrupole interaction with the local crystal field of the two naturally occurring Eu isotopes, Eu^{151} and Eu^{153} . To reduce the complexity of the experiment, special LuPO_4 crystals doped with only one of the Eu isotopes were grown at ORNL. The traveler took them to the Clarendon Laboratory, but unfortunately they were not of sufficient optical quality to perform the proposed experiment. Therefore, attention was directed toward other rare-earth systems. Optical spectroscopy on holmium energy levels in HoF_3 and BaHo_2F_8 was performed, and these materials were studied at temperatures below the Néel temperature where they are antiferromagnetic. Either a magnetic field or uniaxial stress was applied during the course of the measurements in order to create a transition to a ferromagnetic state from the initial antiferromagnetic state. Additional magnetic resonance experiments were performed on impurities in the crystals DyVO_4 , GdVO_4 , and DyPO_4 , while they were in the antiferromagnetic state. We were able to observe the transition to the ferromagnetic state (the so-called "spin-flip" or "spin-flop" transition) directly by magnetic resonance and correlate the results with previous work on these same substances. Resonance signals could be observed from both domains, and measurements were performed at different microwave frequencies which enabled a direct calculation of the internal fields in the antiferromagnetic state.

A paper entitled "Magnetic Resonance of Probe Yb^{3+} and Er^{3+} Ions in the Antiferromagnetic Phase of DyPO_4 " by M. M. Abraham, J. M. Baker, B. Bleaney, A. A. Jenkins, P. M. Martineau, and J. Z. Pfeiffer was presented (by J. M. Baker) at the International Meeting on Magnetism, 25th Colloque Ampère, in Stuttgart, Federal Republic of Germany, September 9-14, 1990, and will be submitted for publication. Other papers based on the traveler's research at Oxford are in preparation.

The Clarendon Laboratory has a specific research group whose primary function is to grow single crystals for the use of all the other research groups. The group is headed by R. Ward, and the traveler was able to see the inventory of crystals that were grown in the past by various members of the group. The techniques employed in growing the vanadates and phosphates were of particular interest. They were grown by a molten-flux

procedure that is similar to our method here at ORNL. However, unlike ORNL where we have settled on one particular flux and one geometry, the Clarendon group has experimented with many different fluxes and many geometries with variable success. It was gratifying to find that the ORNL method compared favorably with the best of the Clarendon methods for the growth of these materials.

The traveler gained considerable new insight into the complexity of these systems by performing optical experiments. These experiments naturally required a large, experienced support system, and the Clarendon Laboratory at Oxford certainly fulfilled that requirement. For example, there was a constant production of over 1000 liters of liquid helium every week for these and other low-temperature experiments. All helium gas was recovered with a return line in every individual laboratory and reliquified to meet the demand.

The Clarendon Laboratory is commencing a new venture on molecular beam epitaxy growth, and a large investment has been made to purchase capital equipment for the proposed work. The group will be headed by R. Ward, M. Wells, and J. F. Gregg and will most probably provide an addition to Clarendon's existing research on high- T_c superconductors. Additionally, metalorganic vapor phase epitaxy has been used to produce very thin layers of semiconductors (a few atomic layers thick) with the goal of creating a new microchip.

A facility which produces superconducting solenoids for high-magnetic fields is also located at Clarendon. A solenoid manufactured by this group was used in the traveler's research and was capable of a maximum magnetic field of 9 T. It was used routinely in the laboratory at fields of approximately 6 T.

Within the Clarendon Laboratory, many groups are involved with lasers, either developing new types or using them as a tool to carry out research into optical effects in solids or gases. These lasers were of many forms and operated in the ultraviolet, visible, and infrared regions of the spectrum.

Low-temperature nuclear orientation experiments were performed on radioactive samples at temperatures as low as 0.5 mK. Gamma rays are detected from these oriented nuclei and various aspects of nuclear and solid state physics are studied.

Another area of research at the Clarendon Laboratory that has received much attention in recent years is the study of turbulence or chaos. This is a universal phenomenon which occurs in diverse situations, from water flow out of a faucet to air circulation in the upper atmosphere. Chaos theory is the current rage in scientific circles and has been applied to the problem of weather forecasting, among other things. During the period of this report, an introductory series of lectures were given at the Clarendon Laboratory on this new theory of chaos with numerous experimental examples. A visitor, B. Mandelbrot, the originator of the closely related field of fractals, delivered a ceremonial seminar on the same subject. The traveler felt fortunate to be able to attend these talks.

In conclusion, the traveler spent a productive year performing research in a first-rate foreign laboratory collaborating with experienced colleagues. Several scientific publications will be forthcoming, and plans have been made for the collaboration to continue. The experience gained at the Clarendon Laboratory in crystal growth of these complex sample materials will directly benefit research projects here in the Solid State Division of ORNL. The traveler has returned from the U. K. with a fresh understanding of rare-earth optical spectroscopy, which will assist him in his ORNL research duties on high-temperature ceramic oxides.

APPENDICES

- A. ITINERARY
- B. LITERATURE ACQUIRED DURING FOREIGN TRAVEL

APPENDIX A

ITINERARY

Dates	Location	Individuals Contacted	Subject of Discussion
10/1-3/89	Travel from Knoxville, TN, to Oxford, U. K. via plane		
10/3/89- 9/25/90 9/25/90	Assignment at the Clarendon Laboratory, Oxford University, Oxford, U. K.	M. J. M. Leask J. M. Baker B. Bleaney M. Wells	Research performed on optical spectroscopy, magnetic resonance, and magnetic susceptibility.
9/26-28/90	Travel from Oxford, U. K., to Knoxville, TN, via plane		

APPENDIX B

LITERATURE ACQUIRED DURING FOREIGN TRAVEL

Thesis, M. G. Robinson, "High-Resolution Optical Studies of Some Magnetic Compounds," Lincoln College, Oxford University, Oxford, U. K. (1986).

Thesis, P. C. Hansen, "Magneto-optical Spectroscopy of Rare-Earth Insulating Compounds," Wadham College, Oxford University, Oxford, U. K. (1990).

Thesis, M. E. Newton, "Magnetic Resonance Measurements in Diamond," Merton College, Oxford University, Oxford, U. K. (1989).

END

DATE FILMED

11 / 15 / 90