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MASTER

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to the

U. S. Atomic Energy Commission

by

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INTRODUCTION

The primary interest of this research has been directed toward two distinct, but related, types of point defects in ionic crystals. One area has been the study of atomic transport mechanisms for ions. This has involved experimental measurements of ionic conductivity and diffusion in alkali, silver, and thallium halides, and interpretation of the results with the use of theories of defect formation and association and of correlation factors for diffusion. The other area has been the study of electrons trapped at color centers in alkali halides. This has involved experimental measurements of optical absorption, photoconductivity, and electron spin resonance, and development of a detailed theory of electron wavefunctions for F centers. The following report is arranged according to this general outline.

Two appendices provide a summary of the research effort. The first is a list of technical reports arranged in four categories: (A) articles in scientific journals, (B) abstracts of papers presented at scientific meetings, (C) unpublished technical reports, and (D) contract reports. The unnumbered reports in categories (A) and (C) represent anticipated future reports based, at least in part, on effort supported by this contract. References to the reports are given as appropriate at the end of each paragraph. The second appendix is a list of personnel divided into two categories: (A) professional and (B) graduate students. The indicated effort includes both University and Contract Support.

DIFFUSION AND IONIC CONDUCTIVITY

(1) Facilities

During diffusion measurements it is necessary to monitor the sample temperature over long periods of time. This was originally done by taking manual readings of thermocouple e.m.f. with a potentiometer. The procedure has now been completely automated with the output of a digital voltmeter and digital clock being recorded directly on punched cards. The data from these cards is then processed by a computer program to yield the effective time for diffusion. A voltage calibration circuit has also been developed to permit calibration of the digital voltmeter on all ranges against a standard cell.

For the measurements of ionic conductivity it is advantageous to use audio frequencies in the range 0.1 to 100 kHz in order to avoid polarization effects at the electrodes. An improved circuit for connecting decade resistors to minimize errors at high frequency and resistance has been designed and constructed, and recently a more sensitive capacitance bridge has been obtained and installed.

One of the important new experimental techniques has been the measurement of tracer diffusion in an applied electric field. Since the samples have an appreciable ionic conductivity, it is necessary to sustain a dc current through the sample in order to maintain the electric field.

For this purpose a moderately high voltage, high current source (300 v, 300 ma) has been constructed, along with a silver coulometer to measure the integrated current.

For all of the experimental work it has been necessary to use single crystals. Commercial sources have not been very satisfactory, both because nominally pure crystals often contain appreciable amounts of impurities and because doped crystals are usually not readily available. Silver and thallium halides have been grown in evacuated, sealed off quartz tubes, but this method has not been feasible for alkali halides. Hence a crystal growing furnace was constructed to allow growth of alkali halide crystals in a controlled atmosphere. This precaution is often vital to avoid incorporation of OH^- or O^{2-} ions in the crystals. The same motor and gear arrangement can be used either for pulling alkali halide crystals from the melt or lowering the quartz tubes through a temperature gradient for the silver and thallium halides. [COO-1197-19]

(2) Silver Halides

The first effort was devoted to careful measurements of diffusion and ionic conductivity in pure AgCl in order to separate the contributions of collinear and non-collinear interstitialcy motion for the diffusion of Ag . At first it was not possible to obtain a satisfactory interpretation of the results, but the difficulty was removed when newer data on the relative mobility of interstitial and vacancy motion became available. This feature indicated the importance of including Debye-Hückel interactions in the treatment of ionic conductivity data in the silver halides and suggested re-examination of the previous calculations for AgBr . The final publication, therefore, provides the most recent and reliable information on the mobilities (or jump frequencies) for the two types of interstitialcy motion in the silver halides. [COO-1197-1, 12, 22]

Another set of measurements on diffusion in $\text{AgCl}:\text{Cd}$ provided an opportunity for an accurate check of the experimental procedures used for comparing tracer diffusion to ionic conductivity. This was desirable because of the uncertainty mentioned above and because of small but tantalizing indications from measurements on NaCl and TlCl (discussed below). The theoretical value of $f_v = 0.78146$ was well-known for diffusion by single vacancies on a fcc lattice; the difficulty was obtaining an experimental system in which it was known unambiguously that all of the diffusion and ionic conductivity was occurring by motion of single vacancies. The most suitable system seemed to be $\text{AgCl}:\text{Cd}$ at moderate temperatures, in which nearly all of the interstitial silver ions are suppressed by the divalent cadmium, leaving only silver ion vacancies. After small corrections for concentration gradients and for residual interstitial ions, the experimental results did agree with the theoretical value within 1 per cent. Thus not only was the theory of correlation factors confirmed (of which there was not much doubt), but more importantly the absolute accuracy of the comparison of ionic conductivity and diffusion was established as a basis for interpretation of other experiments. [COO-1197-18, 21, 24]

An intriguing problem for many years has been a large, anomalous increase in the ionic conductivity of both AgCl and AgBr in the temperature region of about 100°C below the melting points. It is believed that this is due to a lowering of the formation energy for Frenkel defects through Debye-Hückel type interactions because of the rather large concentration of defects (0.1 and 1 per cent at the melting point for AgCl and AgBr, respectively). An accurate theory for this effect has been developed by K. L. Kliewer and R. Sevenich at Iowa State University, and they have expressed an interest in obtaining experimental data to test the theory. Hence an experimental program has been under way for the past year and will continue this year to obtain accurate, closely-spaced measurements of ionic conductivity vs. temperature for the high temperature region of AgCl. The principal experimental problems seem to be obtaining good electrode contact (attempts with evaporated silver and colloidal graphite), accurate measurement of the conductivity (new capacitance bridge), calibration of the thermocouple at fixed temperature points (in progress), and calibration of the digital voltmeter (voltage calibration circuit). These results, in conjunction with the theory, will provide an important extension of the understanding of the formation of defects in ionic crystals. [unnumbered reports by Aboagye and by Aboagye, Valge, Holmberg, and Friauf]

(3) Alkali Halides

At one time it was thought that single cation vacancies were the predominant defect for intrinsic ionic conductivity and cation diffusion. If this were true, it should be possible to obtain a direct comparison of the two phenomena through the Einstein relation along with the correlation factor f_v . Since this had never been done with any real accuracy, we initiated diffusion measurements of Na in NaCl for this purpose. After correction for anion diffusion the first results showed a 10 to 15 per cent excess diffusion that could not be accounted for from the ionic conductivity. This suggested a contribution to diffusion of Na by neutral vacancy pairs, but experimental problems with conductivity measurements made a quantitative analysis infeasible at the time. These results, however, along with other indications of vacancy pair contributions from anion diffusion in doped crystals, provided the impetus for the more extended experiments below. [COO-1197-6, 9, 33]

There are two major problems for a more general interpretation of diffusion processes in the alkali halides: one is separating the single vacancy and vacancy pair contributions for both cations and anions, and the other is separating the cation and anion contributions to the conductivity. Usually only three types of data have been available: diffusion of cation tracers, diffusion of anion tracers, and measurement of total ionic conductivity. Since the traditional transport number measurements of Tubandt appear to be inherently untrustworthy, an additional kind of measurement is needed. This is why we have measured tracer diffusion with an applied electric field: the charged single vacancies should drift along the electric field and produce a corresponding shift of the diffusion profile, whereas the diffusion of the neutral vacancy pairs should not be influenced by the field. In order to make the technique feasible, it has been necessary to show how to obtain a solution of the diffusion equation even when the field varies with time; this is accomplished by relating the time integral of the field to the total charge passing through the sample. In addition it has

been helpful for analyzing diffusion profiles to obtain an analytical solution of the diffusion equation with a barrier at the interface between two samples. The experimental problems of ensuring a good contact at the interface have been only partially overcome, and some difficulties remain with deterioration of the samples near the electrodes, sometimes extending into the diffusion region, and with possible variation of the electric field with position. Despite these difficulties a direct experimental check at two temperatures shows that the tracer drift mobility measurements with this technique are reliable to better than 15 per cent. [COO-1197-26, 27]

The interpretation of the diffusion experiments in a field has required clarification of the concept of tracer drift mobility. With this interpretation the results indicate that vacancy pairs do make an appreciable contribution to cation diffusion (nearly 40 per cent at the melting point). The comparison with the vacancy pair contribution to anion diffusion is consistent with the general theory of vacancy pair diffusion, but comparison to previously reported high frequency dielectric loss measurements in NaCl leads to an unresolvable discrepancy (recent results show that the dielectric loss measurements are probably not ascribable to vacancy pairs in any case). The activation energies for cation diffusion by both single vacancies and vacancy pairs are noticeably larger than previous values and indicate in both cases that modifications in the theoretical calculations of these energies may be necessary. It was also necessary to examine the possible contributions of cation Frenkel defects and of vacancy triplets in order to show that the experimental check of the tracer drift mobility experiments is independent of the type of defect mechanism. In summary the pioneering use of this new experimental technique has contributed to a much more complete and sophisticated understanding of ionic transport processes in NaCl. [COO-1197-30, 32]

(4) Thallium Halides

Extensive experimental measurements have been made of ionic conductivity and diffusion in TlCl. The measured conductivity is appreciably larger than that reported in the older literature, but a possible electronic contribution seems to be ruled out by consideration of polarization effects, general temperature dependence, and reproducibility of results. Nevertheless the sum of cation and anion diffusion coefficients gives an excess diffusion of 10 to 15 per cent more than can be accounted for from the ionic conductivity. [COO-1197-9]

There appear to be two possible explanations for the excess diffusion. One is contributions from vacancy pairs, as for NaCl. The other is that there can be jumps from next nearest neighbors as well as nearest neighbors into a single vacancy because of the more open s.c. structure of TlCl. Theoretical correlation factors have been calculated for both possibilities, and either one could provide a suitable explanation. There is perhaps a slight preference for the second explanation, but additional diffusion measurements on doped samples and with an electric field will be required to provide a complete understanding. [COO-1197-35 and unnumbered report by Friauf]

(5) Theory of Diffusion and Correlation Factors

The experimental test of the correlation factor for single vacancies required diffusion of Ag in AgCl containing divalent Cd. In order to ensure that there was no contribution to diffusion of Ag by vacancies complexed with Cd ions, it was necessary to understand the theory of diffusion of divalent ions in ionic crystals. A review of the literature showed that there is a complete, formal theory by R. Howard and A. B. Lidiard, but also revealed that there have been many incorrect applications of this theory. Hence a paper was written to present the general results of the theory from the experimental viewpoint and to provide critical comments about the application of the theory to a number of experimental arrangements for the alkali halides. The analysis of the results for AgCl provided the needed information for the test of the correlation factor and suggested an explanation for an apparent change of diffusion mechanism at intermediate temperatures. The analysis of the results for AgBr seems to remove the necessity for making a very peculiar assumption about an increase in mobility of silver ion vacancies with an increase in divalent ion concentration. Thus this work has not only provided the desired information for interpreting diffusion in AgCl:Cd but has also clarified a number of other problems. [COO-1197-25, 36]

The experimental results for both NaCl and TlCl have suggested contributions to diffusion by vacancy pairs. As long as the vacancy pairs remain bound, the relative contributions to cation and to anion diffusion are determined, in a rather complicated way, by the relative jump frequencies of cations and of anions into the vacancy pair. In order to make comparisons to experimental results, therefore, it has been necessary to examine the theory of correlation factors for diffusion by vacancy pairs. Although much of the basic theory has been done by other persons, it has still been advantageous to reformulate the results in several ways that are particularly appropriate for making experimental comparisons. It has also been necessary to work out the atomic theory of dielectric loss measurements for vacancy pairs in order to try to compare these measurements to the diffusion results. All of these aspects have been particularly important for interpreting the results in NaCl and are summarized in the paper describing those results. [COO-1197-7, 32]

The experimental results for TlCl have suggested two new kinds of diffusion mechanisms, vacancy pairs on a s.c. lattice and mixed nearest and next nearest neighbor jumps into single vacancies. In developing a general procedure for calculating the needed correlation factors for these mechanisms, a method has been found for obtaining an exact expression for the sum of the infinite series arising from correlation between non-consecutive jumps of the tracer. The method is somewhat similar to a treatment published independently by R. Howard, but some differences also remain. Computer calculations are then used for obtaining the correlation factor for the first return of the diffusing entity to the tracer. Some results have already been obtained for the mixed vacancy jumps in TlCl, and calculations are continuing for the vacancy pair diffusion. [COO-1197-28, 35 and unnumbered report by Friauf]

Summary

Diffusion of Ag in AgCl has yielded quantitative data on collinear and non-collinear interstitialcy motion, and has also provided an absolute test of the comparison of diffusion and ionic conductivity measurements as a basis for interpreting other experiments of this type. Diffusion of Na in NaCl has first suggested a contribution from vacancy pairs, and application of the new experimental technique of tracer diffusion in an electric field has given a better understanding of all ionic transport processes, in addition to quantitative results for cation diffusion by single vacancies and by vacancy pairs. Diffusion of Tl and Cl in TlCl has confirmed the basic picture of diffusion by single vacancies with some additional diffusion by either vacancy pairs or next nearest neighbor jumps. The necessary theory for the formulation and interpretation of these experiments has been developed, considerable clarification has been provided for the application of the theory of diffusion by divalent ions, and a general method has been devised for calculating correlation coefficients for the proposed new diffusion mechanisms in TlCl.

COLOR CENTERS

(1) Facilities

The original observation of color centers was by optical absorption, and this method is still important for identifying centers and determining the relative amounts of different kinds of centers in the crystal. For this purpose a Perkin-Elmer spectrophotometer was obtained and installed, and several small dewars were fabricated to permit measurements at liquid nitrogen temperature. A number of polarizing prisms and depolarizers were obtained and mounted for polarized bleaching and absorption experiments.

For photoconductivity experiments a stainless steel helium dewar was purchased and mounted on tracks to provide ready access for sample mounting and insertion into the spectrophotometer. A dynamic capacitor electrometer was installed to measure the very small currents, with particular attention paid to electrostatic shielding, leakage resistance, and minimization of capacitance for the input to the pre-amplifier unit. A stereo camera shutter was installed in the spectrophotometer to permit short light pulses for photoconductivity measurements.

Originally an X-band electron spin resonance (ESR) spectrometer was assembled, with all electronics constructed locally. Since attempts to modify this spectrometer for ENDOR measurements were unsuccessful, a commercial ESR spectrometer and magnet system were obtained (with other funds), and the new system is now giving excellent sensitivity for both ESR and ENDOR.

The crystal growing furnace described earlier is also used to grow alkali halide crystals, both pure and doped, for color center experiments. An Inconel bomb has been constructed for additive coloring, and with the help of an oil manometer the density of color centers produced can be conveniently controlled. A small monochromator provides light of selected wavelength for causing bleaching and optical transformations of color centers.

(2) Optical Absorption

It has been known for many years that colored Li F crystals often contain an absorption band at 222 nm in addition to the prominent F band at 250 nm. We were able to produce and study the 222 nm band by coloring with neutron irradiation at 300°C. By correlating the height of the band with the amount of Mg doping, we were able to show unambiguously that the 222 nm band is associated with the presence of magnesium, and it was proposed that this may be similar to the Z bands in KCl:Sr, for instance. Broad bands were also observed at 320 and 350 nm, and it was proposed that these are F₂-bands that are stabilized in some way by proximity to magnesium. [COO-1197-5]

Several experiments were attempted to produce polarized bleaching of Z₁ and Z₂ bands in KCl:Sr, KCl:Ca and NaCl:Ca. Unfortunately no significant effects were obtained, in agreement with the experience of other workers in this field. Some additional broad absorption was observed near the F band in NaCl:Ca, but it was not possible to identify separate Z bands. An indication of L bands was obtained during preliminary experiments in NaCl, but the results were not confirmed because of changes in personnel. [COO-1197-10]

Radiation chemists have studied trapped electrons in glass-like alkaline ices formed by rapid freezing of aqueous solutions of NaOH, for instance. First reports showed an optical absorption band at 585 nm, and the presumably F-like nature of this band was confirmed by ESR measurements. On prolonged gamma irradiation, however, it was observed that both the optical absorption and ESR signals disappeared, and we therefore suspected the formation of dielectron centers, with two trapped electrons in a singlet state, similar to F' centers in alkali halides. We observed a very broad optical absorption in the near infrared with a maximum near 1000 nm and were able to show reasonable correlation between the increase of this band and the decrease of the F band by either prolonged irradiation or optical bleaching. [COO-1197-29]

Optical absorption measurements have also been an integral part, of course, of the photoconductivity and ESR experiments described below.

(3) Photoconductivity

Measurements have recently been completed for Z₁, Z₂, and Z₃ bands in KCl:Sr. This represents an extension of much earlier work of similar nature in this laboratory; experimental techniques have been improved considerably both in obtaining a much more nearly complete conversion of F to Z₁ and Z₂ centers and in the improved electrometer arrangements. It is known from many experiments that all of the Z bands are definitely associated with the presence of the divalent ion, but no definite models have been established. Early models proposed that Z₃ centers were formed by adding an electron to Z₂ centers, whereas later models propose that Z₃ centers are formed by removing an electron from Z₂ centers.

The photoconductivity results for Z₂ show a behavior qualitatively similar to F centers: low quantum efficiency at -180°C rising to unity near -100°C (compared to -130°C for F centers); these results give an activation energy of about 0.1 eV from the relaxed excited state to the

conduction band. The Z_3 centers show no photoconductivity at all (quantum efficiency zero) throughout the temperature range (-180° to -100°C) where they are stable; this behavior indicates a much more tightly bound relaxed excited state than for Z_2 centers and confirms the newer proposal for the relationship between Z_2 and Z_3 centers. The raw data for Z_1 centers give a small quantum efficiency at room temperature (approximately one-third in agreement with earlier results) and suggest a fairly rapid decrease at lower temperatures. [Unnumbered reports by Renneke and by Renneke and Friauf]

Throughout these experiments control measurements have been made on pure KCl crystals containing F centers, and there continue to be tantalizing indications of non-uniform quantum efficiency for different wavelengths within the general range of F absorption. These results are probably associated with underlying absorption to higher excited states of M and perhaps R centers, and accurate photoconductivity measurements would give interesting information about the relaxed excited states of these centers. There are also intriguing questions about the behavior of photoconductivity for the higher energy regions of the K band. It is contemplated that work will continue in these areas.

(4) ESR and ENDOR

The first ESR results were obtained for Z_1 centers in KCl:Sr. These showed a broad, isotropic ESR absorption with no resolved hyperfine structure, with the same g-value as F centers but with a slightly larger width. Although no definite correlation could be made with an atomic model, simple perturbation calculations showed that the results were not inconsistent with proposed models. A somewhat different ESR band was obtained after thermal ageing at room temperature, with detectable changes in both g-value and width. This was tentatively ascribed to Z_2 centers, but more recent results of other workers have cast some doubt on this assignment; the matter appears to be still an open question. [COO-1197-2, 3]

Subsequent attempts to obtain ENDOR spectra were unsuccessful for some time, but a new experimental system is now yielding good results. With the improved conversion of F to Z_1 and Z_2 centers it will be possible in the near future to examine the question of Z_2 center resonance; the more positive identification of ENDOR spectra will eliminate possible confusion or oversight in comparison to a small signal from residual F centers. Other work is in progress in cooperation with the University of Missouri and Oak Ridge National Laboratory to investigate ENDOR spectra of F centers in BaO and BaS.

(5) Theory of F Center Wavefunctions

The original impetus was provided by a desire to understand and explain the hyperfine interaction constants obtained for F and Z_1 centers from ESR and ENDOR experiments. For this purpose a rather detailed treatment of the ground state of the F center is required, and it was considered to be paramount to treat the atomic environment with some realism. For this purpose the pseudopotential approach of Cohen and Heine was introduced for the neighboring ions, marking the first use of this method in color center calculations. Two other new features of this work were the expansion of

the crystal field in Kubic harmonics and a somewhat novel use of perturbation theory to determine the first order correction to the wavefunction. The results pointed out obvious shortcomings of the point ion potential, particularly when nonspherical terms in the crystal potential are considered, and provided an improved estimate of the location of the ground state of the F center with respect to the conduction band.

[COO-1197-11, 13, 15, 16]

This work was continued in order to incorporate additional features. Some of these were inclusion of exchange with the Slater approximation, consideration of relaxation of the nearest neighbor ions, and phenomenological treatment of interaction with lattice vibrations. The problems of calculating spin-orbit splittings, oscillator strengths, and hyperfine interactions for wavefunctions of cubic rather than spherical symmetry were formulated exactly, with full use of group theory, and an improved method of calculating overlap integrals was introduced. The numerical results show some improvement in agreement with experimental values. Even more significant, however, is the indication that lattice distortion may make major changes in the F center wavefunction and that even small nonspherical parts of the wavefunction (5-10 per cent) lead to large changes (50 to 100 per cent) in computed values of oscillator strength, spin-orbit splitting, etc. [COO-1197-20, 31, 34 and unnumbered reports by Taylor and Friauf]

There are still many problems that remain to be considered from this point of view. Among the physical problems are better treatment of lattice relaxations, improvement of free ion wavefunctions for use in crystals, and incorporation of dynamic electronic polarization. Other interesting centers that may be considered are F_A centers, and U_2 centers. Treatment of centers with appreciably lower symmetry, such as M and R centers, will require appreciable modification of the expansion techniques.

Summary

The optical absorption experiments have identified bands associated with Mg in Li F and with trapped dielectrons in alkaline ice, and have also been a basic part of other experiments. The photoconductivity measurements have helped to substantiate an important feature of the recently proposed models for Z_2 and Z_3 centers in KCl:Sr and promise to give useful information about higher excited states from M centers and in the K band region. The early ESR measurements suggested some features for Z_1 and Z_2 centers in KCl:Sr, and the present ENDOR apparatus should permit further study of Z_1 , Z_2 , and Z_3 centers as well as investigation of F centers in BaO and BaS. The theoretical work has introduced the use of pseudopotentials and nonspherical crystal fields into color center calculations, has thereby pointed out the inherent deficiencies of the point ion potential, and promises to provide a fruitful way of treating many aspects of the interaction between the trapped electron and the surrounding ion cores in some detail.

LIST OF REPORTS

(A) Articles in Scientific Journals

COO-1197- 1
June, 1962	R.J. Friauf	"Ionic Transport Processes in the Silver Halides," J. Phys. Chem. <u>66</u> , 2380-2383 (1962).
COO-1197- 3
Dec., 1962	G.E. Conklin
and R.J. Friauf	"Electron Spin Resonance of the Z_1 Band in KCl with $SrCl_2$," Phys. Rev. <u>132</u> , 189-194 (1963).
COO-1197- 5
Apr., 1963	F. Pilcher,
D. Schinke,
and R.J. Friauf	"Optical Absorption of Color Centers in Lithium Fluoride Containing Magnesium Additive," J. Phys. Chem. Solids <u>24</u> , 1687-1691 (1963).
COO-1197-15
May, 1965	J. Kuebler
and R.J. Friauf	"Perturbation Calculations of F-Center Wavefunctions with Point Ion and Pseudopotentials," Phys. Rev. <u>140</u> , A1742-A1753 (1965).
COO-1197-22
Feb., 1967	M.D. Weber
and R.J. Friauf	"Interstitialcy Motion in the Silver Halides," J. Phys. Chem. Solids <u>29</u> , XXXX (1968).
COO-1197-24
May, 1968	J. Gracey
and R.J. Friauf	"Experimental Verification of the Correlation Factor for Vacancy Diffusion in Silver Chloride," J. Phys. Chem. Solids <u>29</u> , YYYY (1968).
COO-1197-25
May, 1968	R.J. Friauf	"Comments on Diffusion by Divalent Ions in Alkali and Silver Halides," J. Phys. Chem. Solids <u>29</u> , ZZZZ (1968).
COO-1197-29
Jan., 1968	L. Kevan,
D.R. Renneke,
and R.J. Friauf	"Optical Absorption Spectrum of Trapped Dielectrons in Alkaline Ice," Solid State Communications <u>6</u> , 469-471 (1968).
COO-1197-32
Aug., 1968	V.C. Nelson
and R.J. Friauf	"Diffusion of Vacancies and Vacancy Pairs in NaCl," submitted to J. Phys. Chem. Solids.
COO-1197-33
Aug., 1968	H.L. Downing
and R.J. Friauf	"Self-Diffusion of Na in NaCl," submitted to J. Phys. Chem. Solids.
Feb., 1969	L. Taylor
and R.J. Friauf	"Effect of Crystal Fields, Slater Exchange, and Lattice Distortion on F Center Wave Functions"
Feb., 1969	L. Taylor
and R.J. Friauf	"Calculation of Spin-Orbit, Hyperfine, and Electric Quadrupole Splittings for F Centers in KCl, CsF, and MgO"
Feb., 1969	L. Taylor
and R.J. Friauf	"Interaction of Lattice Vibrations with F Centers in KCl, CsF, and MgO"

Mar., 1969	R.J. Friauf	"Diffusion Processes in TlCl"
Mar., 1969	R.J. Friauf	"Calculation of Correlation Factors for Diffusion in Cubic Crystals"
Apr., 1969	D.R. Renneke and R.J. Friauf	"Photoconductivity of Z_1 , Z_2 , and Z_3 Centers in KCl:Sr"
May, 1969	J. Aboagye, L. Valge, G. Holmberg, and R.J. Friauf	"High Temperature Ionic Conductivity of AgCl"

(B) Abstracts of Papers Presented at Scientific Meetings

COO-1197- 2 Aug., 1962	R.J. Friauf, G.E. Conklin, and D.L. Foster	"Hyperfine Interactions of Z_1 Centers in KCl:Sr," International Symposium on Color Centers in Alkali Halides, Stuttgart, Germany (1962).
COO-1197- 6 Apr., 1963	R.J. Friauf	"Ionic Transport Processes in Ionic Crystals," J. Electrochem. Soc. <u>110</u> , 66C (1963).
COO-1197- 7 Aug., 1963	R.J. Friauf	"Diffusion and Correlation Effects in TlCl and AgCl," Gordon Research Conference, Meriden, New Hampshire (1963).
COO-1197-13 Jan., 1965	R.J. Friauf and J. Kuebler	"Calculation of F-Center Wavefunctions with Point-Ion and Pseudopotentials," Bull. Am. Phys. Soc. II, <u>10</u> , 306 (1965).
COO-1197-16 May, 1965	J. Kuebler, L. Taylor, and R.J. Friauf	"Pseudopotential Calculations for F Centers," International Symposium for Color Centers Alkali Halides, Urbana, Illinois (1965).
COO-1197-20 Jan., 1967	L. Taylor and R.J. Friauf	"Crystal Field Effects on the 1S and 2P States of a Localized Center in a Cubic Potential," Bull. Am. Phys. Soc. II, <u>12</u> , 279 (1967).
COO-1197-21 Jan., 1967	R.J. Friauf and J. Gracey	"Experimental Verification of the Correlation Factor for Vacancy Diffusion," Bull. Am. Phys. Soc. II, <u>12</u> , 326 (1967).
COO-1197-27 Oct., 1967	R.J. Friauf and V.C. Nelson	"Diffusion of Sodium in Sodium Chloride in an Applied Electric Field," in <u>Mass Transport in Oxides</u> , edited by J. B. Wachtman, Jr. and A. D. Franklin, National Bureau of Standards Special Publication 296, Washington, D.C. (1968), p. 9.
COO-1197-28 Jan., 1968	R.J. Friauf	"Correlation Factor for Self-Diffusion by Mixed Vacancy Jumps," Bull. Am. Phys. Soc. II., <u>13</u> , 489 (1968).

COO-1197-30
June, 1968	V.C. Nelson
and R.J. Friauf	"Diffusion of Vacancies and Vacancy Pairs in NaCl," International Symposium on Color Centers in Alkali Halides, Rome, Italy (1968).
COO-1197-31
June, 1968	L.H. Taylor
and R.J. Friauf	"Crystal Field Effects on F Center Wavefunctions in KCl, CsF, and MgO," International Symposium on Color Centers in Alkali Halides, Rome, Italy (1968).
COO-1197-35
Aug., 1968	R.J. Friauf	"Diffusion in TlCl and Calculation of Correlation Factors," International Conference on Science and Technology of Non-metallic Crystals, New Delhi, India (1969).
COO-1197-36
Aug., 1968	R.J. Friauf	"Comments on Diffusion of Divalent Ions in Alkali and Silver Halides, "International Conference on Science and Technology of Non-metallic Crystals, New Delhi, India (1969).

(C) Technical Reports, Unpublished

COO-1197- 9
June, 1964	H. Downing	"Self-Diffusion of the Sodium Ion in Pure NaCl"
COO-1197-10
Aug., 1964	D. Lauer	"Polarized Absorption Measurements of NaCl with Ca Impurity"
COO-1197-11
Dec., 1964	J. Kuebler	"Wave Function Calculations for F Centers in NaCl, KCl, and NaF"
COO-1197-12
Dec., 1964	M. Weber	"Self-Diffusion of the Silver Ion in Pure AgCl"
COO-1197-18
Jan., 1967	J. Gracey	"Experimental Verification of the Correlation Factor for Vacancy Diffusion in Silver Chloride"
COO-1197-19
Jan., 1967	R. Mellott	"A Controlled Atmosphere Crystal Grower"
COO-1197-26
Aug., 1967	V.C. Nelson	"Diffusion of Sodium in Sodium Chloride in an Applied Electric Field"
COO-1197-34
Aug., 1967	L.H. Taylor	"Crystal Field Effects on the Wavefunction and Physical Properties of the F Center in Solids with the NaCl Structure"
Feb., 1969	D.R. Renneke	"Photoconductivity of Z_1 , Z_2 , and Z_3 Centers in KCl: Sr"
May, 1969	J. Aboagye	"High Temperature Ionic Conductivity of AgCl"

(D) Contract Reports

COO-1197- 4 Mar., 1963	R.J. Friauf	First Annual Progress Report
COO-1197- 8 Mar., 1964	R.J. Friauf	Second Annual Progress Report
COO-1197-14 Mar., 1965	R.J. Friauf	Third Annual Progress Report
COO-1197-17 Mar., 1966	R.J. Friauf and G.G. Wiseman	Fourth Annual Progress Report
COO-1197-23 Mar., 1967	R.J. Friauf	Fifth Annual Progress Report
COO-1197-37 Sep., 1968	R.J. Friauf	Final Report

LIST OF PERSONNEL

(A) ProfessionalMan-Months

Prof. Robert J. Friauf	Principal Investigator	33
Prof. Gordon G. Wiseman	Senior Investigator	7.2
Dr. Paolo Podini	Research Associate	<u>21</u>
Total		61.2

(B) Graduate Research AssistantsMan-Months

Harold Breedlove	5.5
Harry Downing	2.0
Donald Foster	19.5
Joseph Gracey	2.5
Gerald Holmberg	5.5
Shih-Pei Hu	1.0
Soo-Ung Kim	10.0
Juergen Kuebler	7.8
Dennis Lauer	4.0
Robert Mellott	25.0
Vaughn Nelson	27.5
Frederick Pilcher	0.5
David Renneke	18.0
Lyle Taylor	16.0
Lawrence Walsh	5.5
Max Weber	<u>1.0</u>
Total	151.3