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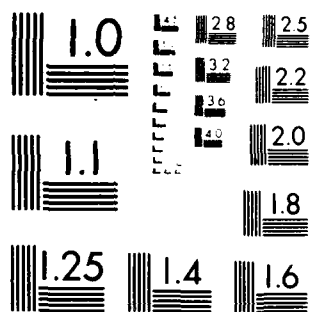
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SPECTROSCOPIC PROPERTIES OF METAL MONOXIDES
AND HYDROXIDES IMPORTANT IN THE IONOSPHERE

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e/f -dependence of B + a intensity factors enables, for the first time, measurement of relative $a^3\Pi$ and $A^1\Pi$ populations from observed B + X fluorescence intensities. A significant excess population (relative to $A^1\Pi$), above the expected 500K thermal distribution is found in the e-parity levels of $a^3\Pi$ ($v_a = 0$ and 1) in a 1-2 Torr Mg + Ar + N₂O Broida-oven flame. Whether these e vs. f and $a^3\Pi$ vs. $A^1\Pi$ population anomalies reflect the nascent product distribution or result from complex, multiple collision processes is under investigation.

The most important use of the MgO B-a system will be proof, via OODR experiments now in progress, that the $\lambda < 320\text{nm}$ continuum arises from the MgO $e^3\Sigma^-$ ($Mg^{-1}S + O^{-3}P$) $\rightarrow a^3\Pi$ transition, thereby providing direct optical spectroscopic bounds to the dissociation energy of the MgO molecules.

Four papers are in preparation describing the results of this research program.

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SPECTROSCOPIC PROPERTIES OF METAL MONOXIDES AND
HYDROXIDES IMPORTANT IN THE IONOSPHERE

A. INTRODUCTION

The monoxides, monoxide ions, and hydroxides of Mg, Ca, and Fe are important species in the ionosphere. Spectroscopic information about these species, needed for atmospheric modelling, is either incompletely available or the subject of controversies. This program of research was originally planned to characterize the lowest lying states of MgO, CaO, and FeO and to provide a direct spectroscopic measure of the dissociation energy of MgO.

Although several projects dealing with the low lying states and electronic structure of CaO^{1,2,3} and CuO⁴ were completed during the period of this contract, the present report will deal exclusively with the MgO molecule.

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B. SUMMARY OF RESULTS

1. MgO $B^1\Sigma^+ - a^3\Pi$ System

The $a^3\Pi$ state is the first excited state of MgO. It was first identified through its perturbations of the $X^1\Sigma^+$ state.⁵ Although the $a^3\Pi$ state is an important "reservoir state" in Mg oxidation reactions and the $e^3\Sigma^- - a^3\Pi$ system has been suggested as responsible for a $\lambda < 320\text{nm}$ continuum (forming $\text{Mg } ^1\text{S} + \text{O } ^3\text{P}$ atoms)⁶, no suitable scheme exists for monitoring populations in specific MgO $a^3\Pi$ rotation-vibration levels.

The main thrust of this program was to record, assign, and deperturb the MgO $B^1\Sigma^+ - a^3\Pi$ intercombination system. Although nominally forbidden, the B-a system borrows oscillator strength from the $B^1\Sigma^+ - A^1\Pi$ and $B^1\Sigma^+ - X^1\Sigma^+$ systems through spin-orbit interactions of the $a^3\Pi$ state with $A^1\Pi$ and $X^1\Sigma^+$. The net result of the A-a mixing is that the $B^1\Sigma^+ - a^3\Pi(\Omega = 1)$ sub-band is computed to have $\sim 1/450$ of the oscillator strength of B-A bands. However, a surprisingly large excess population in the $a^3\Pi$ e-parity levels has made recording parts of the B-a system much easier than initially expected. Unfortunately, the extremely small populations in f-parity levels and the weakness of transitions out of $\Omega = 0$ and 2 components have made characterization of the MgO $a^3\Pi$ state unexpectedly difficult.

Table I shows the $J = 0 - 50$ lines of the complete B - a (0,0) band. Two complementary techniques were used: cw and pulsed dye laser fluorescence ($B^1\Sigma^+ \rightarrow X^1\Sigma^+$, $\Delta v = 0$) excitation spectroscopy (FES). The lower intensity cw laser only weakly saturates the B-A system, thus the relative B-A/B-a intensities sample populations times line strength factors. The high intensity

TABLE 1

MgO B- α -(λ, ν) Observed and Calculated Line Positions (cm $^{-1}$)

J	9- α -10			B- α -11			B- α -12		
	P(J)	Q(J)	R(J)	P(J)	Q(J)	R(J)	P(J)	Q(J)	R(J)
0		17386.356c	17383.553c		17453.457(-7)	17455.782c		17514.822c	17518.302c
1	17381.377c	386.500c	384.858c		453.785(3)	457.257(2)		515.350c	519.990c
2	380.508c	386.788c	386.308c		454.259(1)	458.892(8)		511.470c	521.853c
3	379.789c	387.219c	387.903c		450.769(5)	460.669(-1) ^b		516.053c	523.893c
4	379.206(4) ^b	387.795c	389.631(-12)		450.230c	459.998(6)		511.133c	526.108c
5	378.766(-2)	388.515c	391.528c		449.849(-3)	462.589(-23) ^b		517.989c	528.499c
6	378.485(7) ^b	389.379c	393.569(12) ^b		449.631c	464.712(2)		519.220c	533.805c
7	378.328(-6) ^b	390.386c	395.739(7) ^b		449.573(6)	466.984(-1)		522.627c	536.721c
8	378.328(-7) ^b	391.539c	398.03(-2) ^d		449.658(-1)	469.372(-4)		529.609c	539.813c
9	378.485(3) ^b	392.835c	400.516c		449.910(1)	471.945(2)		532.750c	546.518c
10	378.776(1)	394.276c	403.12(1)		462.035(1)	474.656(-11) ^b		535.382c	553.921c
11	379.206(3) ^b	395.862c	405.880(-1)		463.800(19) ^b	477.548c		536.137(-3)	557.875(-8)
12	379.800(3)	397.592c	408.784(2)		463.800(-3)	480.584(-1)		541.174(3)	562.012(-7)
13	380.521(-6)	399.467c	411.827c		465.683(-3)	483.784(5) ^b		544.334(7)	566.320(-8)
14	381.397(-6)	401.488c	415.016(-3)		467.749(1) ^b	487.121(-1) ^d		547.655(-1)	570.806(-5)
15	382.432(6)	403.655(2)	418.359(3)		469.46(-1) ^b	490.639(2) ^b		551.149(-10)	575.484(-2)
16	383.591(-4)	405.973(8)	421.839(2)		472.348(-7) ^b	494.301(-1)		552.984(4)	580.294c
17	384.905(-6)	408.416(-5)	425.463(-2)		474.900(4)	498.123(1)		556.674(-12) ^b	585.294c
18	386.373c	411.018(-6)	429.235(-4)		477.601(-5)	502.101(1)		558.714(5)	592.466c
19	387.972(10)	413.772(-1)	433.491(-10) ^b		483.479(4)	506.237(1)		562.907(3)	595.809c
20	389.72(-2) ^d	416.664(-4)	437.226(2)		486.652(1)	510.524(1)		565.945c	601.324c
21	391.648(5)	419.718(9)	441.440(4)		489.97(-2) ^d	514.923(5)		568.921(8)	607.011c
22	393.695(1)	422.898c	445.801(6)		490.455(-1) ^b	519.598(3)		569.253c	612.868c
23	395.896(3)	426.227(-6)	450.312(13) ^b		493.486(1)	524.366(18) ^b		575.821(8)	618.397c
24	398.243(3)	429.715c	454.952(1)		497.139(-2)	529.270(-2)		585.411c	625.095c
25	400.735c	433.547(2)	459.750(1)		504.329(-2)	534.357(5)		588.986(2)	631.463c
26	403.381(2)	437.123(1)	464.694c		509.060(-6)	539.594(5)		594.622(5)	638.001c
27	406.171(1)	441.049(1)	469.787(-1) ^d		513.357(-3)	544.984(1) ^b		597.201(-1)	644.709c
28	409.111(3)	445.122c	475.619(-6)		517.613c	550.536(-1)		599.204c	651.586c
29	412.199(-2)	449.346(1)	480.411c		522.420(-7)	556.249(2)		601.324c	658.846c
30	415.441(2)	453.716(-1)	485.94(-1) ^d		527.731(-10)	562.123(6)		607.011c	665.846c
31	418.836(9)	458.240(3)	491.616(-1) ^b		532.134c	568.141c		612.405c	673.230c
32	422.365c	462.91(1) ^d	497.456(-1)		534.300(-2)	574.326c		614.317c	680.780c
33	426.053(1)	467.727c	503.424(-10)		542.489(-3)	581.688c		618.397c	688.490c
34	429.889c	472.692(-4)	509.561(1)		547.499(6)	587.168c		624.398c	696.386c
35	433.876c	477.812(2)	515.837(4)		553.456c	593.726c		629.039c	704.431c
36	438.014c	483.086c	522.254(-2)		559.200c	600.643c		633.033c	712.659c
37	442.302c	488.481(-3) ^d	528.622(-4)		565.94c	606.643c		637.033c	721.045c
38	446.741c	494.079c	535.548(2)		571.148c	614.749c		641.664c	729.596c
39	451.311c	499.303c	542.417(3)		577.362c	622.039c		646.354c	738.317c
40	456.072c	505.681(3)	549.421(-1) ^d		583.737c	629.487c		651.355c	747.290c
41	460.964c	511.699(-6)	556.597c		590.273c	637.093c		656.395c	756.247c
42	466.008c	517.885(1)	563.912c		596.969c	644.955c		661.409c	765.454c
43	471.203c	524.216c	571.374(-2)		603.425c	652.776c		666.466c	774.732c
44	476.549c	530.705(4)	578.990c		610.442c	660.854c		671.500c	784.366c
45	482.048c	537.345(5)	586.753c		618.020c	669.089c		676.550c	794.090c
46	487.699c	544.130c	594.664c		625.358c	677.480c		681.664c	803.817c
47	493.500c	551.072(-2)	602.726c		632.457c	686.029c		687.732c	813.542c
48	499.854c	558.175(3)	610.937c		640.518c	694.732c		693.817c	823.272c
49	505.560c	565.425c	619.297c		648.339c	703.490c		699.926c	833.001c
50	511.813c	572.832c	627.806c		656.122c	712.605c		706.079c	842.732c

a Number in parentheses is (observed - calc.) in 10 $^{-4}$ cm $^{-1}$.

b Blended lines.

c Calculated lines.

d These lines are obtained by using a pressure-scanned pulsed dye laser with a HWHM of 0.02cm $^{-1}$. Experimental uncertainties for these lines are 0.04cm $^{-1}$.

TABLE II
 Constants for the a, B, and X States of MgO (cm⁻¹)^a

	$a^3\pi$		$B^1\Sigma^+$		$X^1\Sigma^+$	
	$v = 0$	$v = 1^b$	$v = 0$	$v = 0$	$v = 3$	$v = 4$
T_0	2553.755(19)	3195.4625(16)	20003.575(11)	0.0 ^d	2293.67(43)	3038.54(46)
B_v	0.500341(12)	0.495556(7)	0.580058(12)	0.571987(25)	0.5568 ^c	0.55151(19)
$D_v \times 10^6$	1.1979(49)	1.1072(40)	1.1485(47)	1.2173(97)	1.29 ^c	1.31 ^c
A_v	-63.489(14)	-64.1727(73)	-	-	-	-
$A_0 \times 10^4$	-1.214(27)	-2.953(80)	-	-	-	-
A_{01}	17.69(27)	20.889(29)	-	-	-	-
A_{11}	-65.11(11)	-64.85 ^c	-	-	-	-
D_v	-1.046(28)	-0.619 ^c	-	-	-	-
P_v	-0.02001(25)	0.02391(36)	-	-	-	-
$q_v \times 10^4$	-0.96(10)	-1.96 ^c	-	-	-	-
$Q_0 \times 10^4$	-0.741(88)	-	-	-	-	-

^a Number in parentheses is one standard deviation uncertainty.

^b Preliminary values. Constants for $B^1\Sigma^+$ were fixed.

^c Held fixed in the fit.

^d Zero of energy.

pulsed laser saturates both transitions, hence sampling relative populations. Although B-a lines are weaker than B-A in the cw laser spectra, the B-A lines are barely detectable in the pulsed laser spectra! The use of pulsed laser FES will provide the crucial missing $\Omega = 0$ and 2 lines in the B-a (0,1) band.

Table II gives the constants for the $a^3\Pi v = 0$ and 1 levels obtained from the measured lines of the B-a (0,0) and (0,1) bands. Reduction to a set of mechanically and magnetically meaningful constants required deperturbation of $a^3\Pi v_a$ with respect to interactions with $X^1\Sigma^+ v_X = v_a + 3$ and $A^1\Pi v_A = v_a$. In turn this deperturbation model yields precise basis function mixing coefficients which enable calculation of v, J, Ω , and e/f dependent B-a intensity factors.

2. Intensities in the MgO $B^1\Sigma^+ - a^3\Pi_i$ System

In order to make quantitative measurements of the relative populations of $a^3\Pi_{\Omega}$ e vs. f, $a^3\Pi_{\Omega}$ vs $a^3\Pi_{\Omega\pm 1}$, or $a^3\Pi_{\Omega}$ vs $A^1\Pi$ levels, it is necessary to know the relative linestrength factors. Such factors are never trivially available for a nominally forbidden band system. Fortunately, the B + a (v_B, v_a) band of the FES spectrum monitored by B $\rightarrow$ X ($v_B, v_X = v_B$) fluorescence, borrows more than 99% of its intensity from the B-A ($v_B, v_A = v_a$) band. This means that knowledge of the $A^1\Pi$ character in each nominal $a^3\Pi$ v_a, J, Ω , e/f level is sufficient to calculate the intensity of any B-a line relative to any B-A or other B-a line. The $A^1\Pi$ fractional character varies over several orders of magnitude, and the mixing fractions obtained from the deperturbation model are accurate to better than 1% of their calculated magnitudes. The deperturbation model yields mixing fractions which should be considerably more accurate than state-of-the-art absolute intensity measurements.

Although unimportant for most purposes, quantum mechanical interferences between B-A and B-X transition amplitudes can cause certain B-a lines to behave anomalously. P and R lines from a common J'' level would normally have comparable linestrengths. However, for B + a bands detected by B $\rightarrow$ X $\Delta v \neq 0$ fluorescence, the P/R intensity ratio will deviate significantly from 1. Whether it is greater or less than 1 determines the relative signs of the B - X and B - A transition moments.

For the first time, populations in the most important "reservoir state" of MgO, the MgO $a^3\Pi$ state, can be monitored systematically and without assignment ambiguity. The monitoring transition is in the convenient Rhodamine 6-G region and had the simplest possible branch structure. Although

the d-a system has been used for monitoring $a^3\Pi$, its complex structure has, to date, frustrated analysis. The availability of $a^3\Pi$ combination differences should facilitate assignment of the $d^3\Delta$ - $a^3\Pi$ system and resolution of several contradictory features of speculative bandhead assignments.^{5,7}

3. Relative $a^3\Pi/a^3\Pi f$ and $a^3\Pi/A^1\Pi$ Populations.

Armed with the calculated linestrength factors from the $a^3\Pi \sim A^1\Pi$, $X^1\Sigma^+$ deperturbation, it becomes possible to measure population ratios and to compare these ratios to those expected from a thermal ($T \sim 500K$) distribution. At 1-2 Torr total pressure (99% Ar, 1% N_2O , 0.1% MgO and Mg), it is reasonable to expect the metastable X, A , and a levels to be equilibrated. However, the $a^3\Pi v = 0, \Omega = 1$ levels have more than a factor of 10 higher population relative to $A^1\Pi v = 0$ than the 950 cm^{-1} separation of these levels would imply at 500K. Similarly, the $a^3\Pi \Omega = 1$ e-levels have more than a factor of 2 greater population than the near degenerate ($\Delta E < 0.1\text{ cm}^{-1}$) f-levels. It is difficult to imagine how such large departures from equilibrium could exist at such high pressure.

Single collision $Mg + N_2O$ experiments, in collaboration with Professor Paul Dagdigan at Johns Hopkins University, are in progress. The purpose of these experiments is to determine whether the 1-2 Torr population anomalies reflect nascent product distributions or complex multiple-collision population funnelling and removal processes.

4. Dissociation Energy of MgO

Ground state Mg and O atoms correlate adiabatically with the MgO $a^1\Pi$ and $e^3\Sigma^-$ states. The e-a transition is predicted to be strong because the $e^3\Sigma^-$ state belongs to the same electronic configuration as $d^3\Delta$, $D^3\Delta$, and $C^3\Sigma^-$. Evans and Mackie⁶ have observed a continuum in absorption from shock-heated MgO at $\lambda < 320$ nm which they have assigned as $e^3\Sigma^- \leftarrow a^1\Pi$. If this assignment can be verified and the long- λ limit of the continuum can be located with greater sensitivity, it should be possible to obtain a direct spectroscopic upper bound to D_0° (MgO).

Our initial purpose for investigating the MgO B-a system was to develop an unambiguous $a^3\Pi$ population monitor for use in an Optical-Optical Double Resonance (OODR) scheme for assigning and extending the Evans-Mackie continuum. Laser 1 destroys MgO $a^3\Pi$ by pumping in the continuum. Laser 2 detects MgO $a^3\Pi$ and shows that the population destruction which occurs when Laser 1 pumps in the continuum is specific to $a^3\Pi$ $v = 0$. If the Evans-Mackie assignment is correct, then population loss in $A^1\Pi$ (monitored by Laser 2 in the B-A system) will occur at a longer delay after the Laser 1 pulse than population loss in $a^3\Pi$.

The pulsed laser FES component of this experiment has been demonstrated. The full OODR experiment will be performed in March, 1983.

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Ph.D., June 1983 (expected)

Jeffrey Norman (pulsed MgO B-a)

David E. Reisner

Ph.D., March 1983 (expected)

PUBLICATIONS

The following papers are in preparation:

1. Precila C.F. Ip, Robert W. Field, and Keith J. Cross, "The $B^1\Sigma^+ - a^3\Pi_i$ Intercombination System of the MgO Molecule," for J. Mol. Spectrosc.
2. Keith J. Cross and Precila C.F. Ip, "The MgO $B^1\Sigma^+ - A^1\Pi$ System. Laser Spectroscopy and Deperturbation," for J. Mol. Spectrosc.
3. Precila C.F. Ip and Robert W. Field, "Intensity Factors for the MgO $B^1\Sigma^+ - a^3\Pi_i$ System Based on $X^1\Sigma^+ \sim a^3\Pi_i \sim A^1\Pi$ Deperturbation Model," for Chem. Phys. Lett.
4. Precila C.F. Ip, Keith J. Cross, and R.W. Field, "Population Anomalies in an Mg+N₂O Flame. Excess $a^3\Pi_i/A^1\Pi$ and $a^3\Pi_e/a^3\Pi_f$ Populations," for J. Phys. Chem.

INTERACTIONS AND PRESENTATIONS

6-80 Molecular Spectroscopy Symposium, Columbus, Ohio, P.F. Bernath, P. Ip, and R.W. Field, "Optical-Optical Double Resonance Spectroscopy of BaF" (contributed talk MF5).

3-81 Yale University, Symposium on Lasers in Chemistry, R.W. Field, "Molecular Spectroscopy Beyond Molecular Constants" (invited).

4-81 Discussion of Faraday Society, No. 71, Bristol, England, R.W. Field, "Tunable Laser Electronic Spectroscopy" (invited).

5-81 International Colloquium on Molecular Spectroscopy, Stockholm, Sweden, R.W. Field, "Perturbations: Sublime and Ridiculous" (invited).

6-81 Molecular Spectroscopy Symposium, Columbus, Ohio, K.J. Cross, P. Ip, and R.W. Field, "Rotational Analysis of the $B^1\Sigma^+ - a^3\Pi_1$ Band System of MgO (contributed talk MG7).

3-82 Bad Honnef, Germany, conference of the Deutsche Bunsengesellschaft für Physikalische Chemie on Small Molecules in the Gas Phase, R.W. Field, "Electronic Structure of Diatomic Molecules Beyond Simple Molecular Constants" (invited).

8-82 Gordon Conference on Molecular Electronic Spectroscopy, R.W. Field, "The Electronic Structure of Ionic Diatomic Molecules" (invited).

Collaborations with Dr. Guy Taieb and Dr. Bernard Bourguignon at Université de Paris-Sud, Orsay and Professors Yarkony and Dagdigan at Johns Hopkins University.