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EMISSION COEFFICIENT FOR A SINGLY IONIZED URANIUM PLASMA: EXPERIMENTAL AND THEORETICAL TREATMENT

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ABSTRACT

Absolute emission coefficient measurements on arc-generated singly ionized uranium (UII) in local thermodynamic equilibrium are described for a wavelength bandwidth of 1050 to 6000 Å. Plasma temperature and uranium partial pressure at the arc centerline were approximately 8000 K and 0.01 atm, respectively. The arc emission data compare favorably with experimental results obtained from UF_6 discharges, once allowance is made for the effects of cold-layer UF_6 photoabsorption on uranium plasma emission.

Observed variation in the emission coefficient is well correlated with a composite of calculated oscillator-strength distribution for selected UII and UIII transition arrays. The theoretical treatment is based on a modified Hartree-Fock method for calculating the appropriate radial wavefunctions. Slater-Condon theory provides a detailed calculation of energy-level structures for both the upper and lower configuration of each transition array computed. However, the number of terms have been truncated, when necessary, to accommodate size limitations of the matricies involved. Using this theoretical approach, we also offer predictions as to the location of strong emission features for a hypothetical plasma dominated by doubly and triply ionized uranium. We suggest that the capability now exists to predict successfully the location of major emission features of uranium and other complex systems for a substantial range of ionization stages.

EMISSION COEFFICIENT FOR A SINGLY IONIZED URANIUM PLASMA: EXPERIMENTAL AND THEORETICAL TREATMENT

I. INTRODUCTION

Technologies that utilize uranium plasmas, including gaseous and plasma core reactors, laser isotope enrichment, and nuclear-pumped lasers require knowledge of the plasma properties. One area of interest is the characterization of uranium plasma emission as a function of temperature, pressure, and wavelength. Historically, attempts to measure emission coefficients of uranium plasmas have been made using shock tubes, induction-heated discharges and arcs, but adequate correlation among the results of these experiments has been lacking. Schneider et al.[1] have addressed this problem and found marginal agreement in magnitude between Marteney's induction-heated UF_6 discharge[2] and Mack's dc uranium arc data,[3] but correlation in shape as a function of wavelength was poor. We ascribe the discrepancies between experiments to UF_6 photoabsorption of the uranium plasma emission. In addition, we have calculated oscillator strength distribution to give indication of the position of major emission features of UI, UII, UIII, and UIV.

To calculate plasma emission coefficients from first principles, a model is necessary which adequately treats the quantum and statistical mechanics for the radiating system under investigation. The spectral line emission coefficient for spontaneous emission is directly proportional to the product of statistical weight and oscillator strength (gf), and to the population density of the energy states involved in spontaneous transitions. The calculation of gf distribution for interesting transition arrays yields prediction of spectral line wavelengths and strengths. Establishing such distributions provides insight into the variation of emission coefficient without having to include the population mechanics.

Uranium radial wavefunctions and the necessary radial integrals were calculated with a computer code developed by R. D. Cowan at the Los Alamos Scientific Laboratory. Using this information within the framework of Slater-Condon theory, the oscillator-strength distribution for singly and doubly ionized uranium has been computed for important transition arrays and compiled into a single curve of oscillator strength versus wavelength. The variation in the measured emission coefficient for 8000 K is well correlated with the calculated oscillator-strength distribution. We also calculated the oscillator-strength distribution for triply ionized uranium as a guide to the wavelengths at which one can expect its major emission features.

II. EXPERIMENT DESCRIPTION

A detailed description of the dc arc and data acquisition has been given elsewhere;[3] the essential features of this system are presented as an overview. A wall-stabilized arc employing a uranium anode and tungsten cathode was used to generate the uranium plasma. The arc was operated with a small flow of helium (at three atmospheres), which was directed the length of the arc column, for added stabilization. The large difference in ionization potentials between uranium and helium suggests that the uranium constituent dominates the arc plasma characteristics. Spectral analysis of the arc plasma emission indicated a majority of

UII lines, some UI lines, and a significant number of lines that did not correspond to UI or UII. It is possible that these are emitted by doubly ionized uranium (UIII). The absence of helium emission lines supports the assumption that a relatively uncontaminated uranium plasma was formed.

The instrumentation, which detected emission to 1050 Å, consisted of a 1/3-m McPherson scanning spectrometer, in conjunction with a sodium salicylate-photomultiplier combination. The intensity data were calibrated with a tungsten ribbon standard and deuterium discharge. Digital signal-averaging was also applied to the raw intensity data to improve the signal-to-noise ratio.

III. TEMPERATURE AND DENSITY DIAGNOSTICS

The temperature and partial pressure of the uranium arc plasma at the arc centerline are 8000 K and 0.01 atm respectively. These values are inferred from earlier work[4,5] that employed the same arc system operated under the same current, voltage, and total-pressure conditions. Justifications for the likelihood of obtaining similar temperature-pressure conditions with the present arc are based on a detailed comparison of the spectra emitted by both. The diagnostics assumed local thermodynamic equilibrium and involved the Fowler-Milne, Boltzmann Plot, line-ratio, and absolute-line methods. The temperature diagnostics resulted in an uncertainty of -13% to +20%; the density measurement is an order-of-magnitude estimate.

Saha analysis in the quoted temperature-pressure regime indicates a transition region where the number density of UIII becomes of comparable magnitude to that of UII. Subject to the uncertainties of the factors involved in the Saha equation, one can anticipate radiation from singly and doubly ionized uranium. Near 8000 K most of the emission should be characteristic of UII, and some of the features of UIII radiation may be present, particularly from transitions involving the lower configurations.

IV. EXPERIMENTAL RESULTS

The measured emission coefficient from the uranium arc was compared to similar results obtained by Marteney[2] with an induction-heated UF_6-argon discharge. Both experiments acquired data in the visible, ultraviolet, and vacuum ultraviolet; for this paper we shall concentrate only on the wavelength variation of emission coefficient. After the effects of potential cold-layer UF_6 photoabsorption were unfolded from the Marteney results, favorable agreement resulted to around 1900 Å; below 1900 Å some obvious discrepancies occurred. There is now experimental and theoretical evidence for the existence of two peaks in the vacuum ultraviolet:[3] a peak at 1540 Å detected in the arc experiment, and a peak at 1800 Å detected from the UF_6 discharge. Figure 1 shows a composite uranium emission coefficient as a function of wavelength at a temperature of about 8000 K. The variation is defined by the prominent features exhibited by the arc data and the peak at 1800 Å observed in the UF_6 experiment. The remainder of this report presents a theoretical basis established to predict the major features of the observed emission coefficient variation.

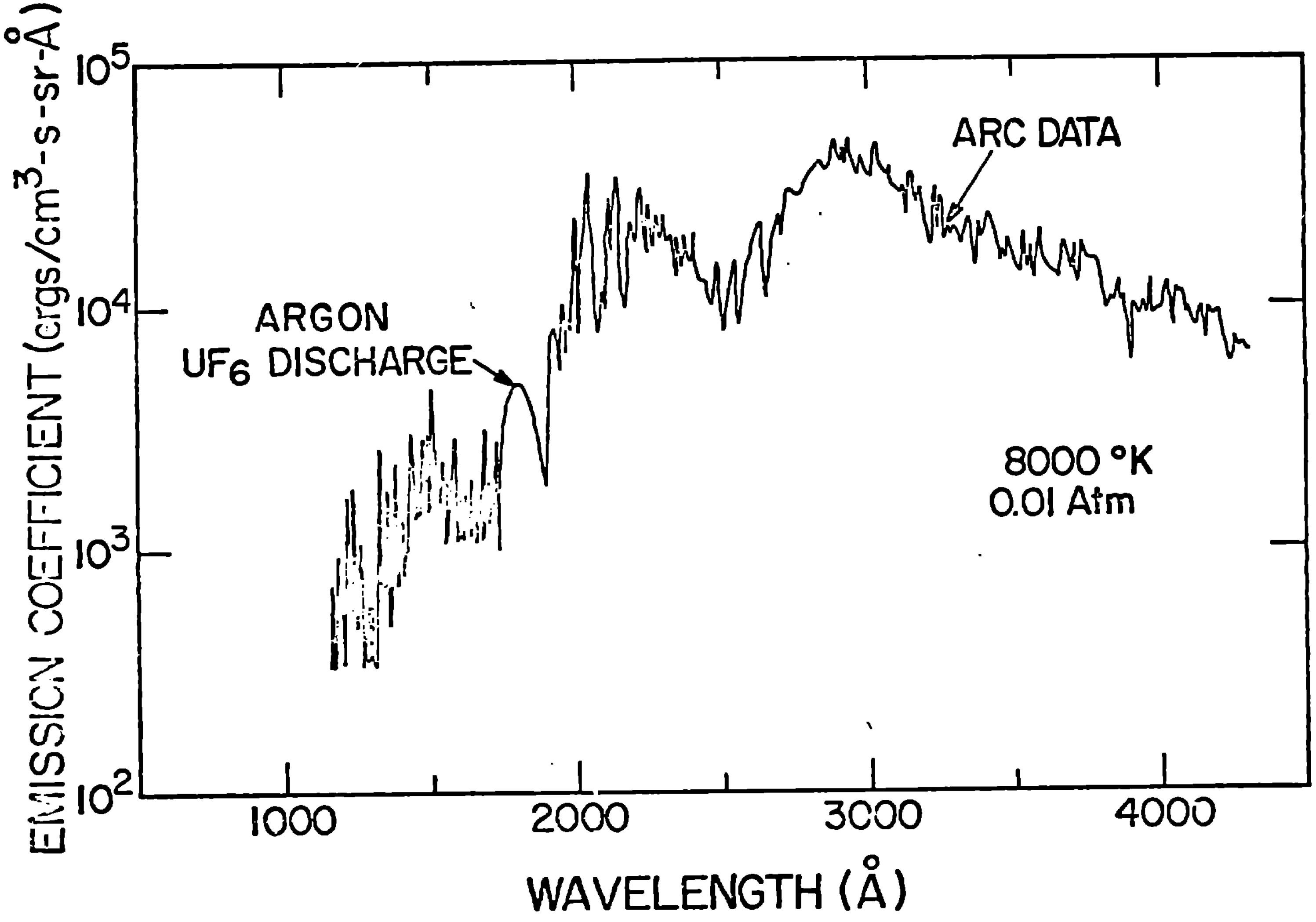

Fig. 1.

Observed uranium emission coefficient.

V. THEORETICAL CALCULATIONS AND RESULTS

In order to substantiate the observed uranium plasma emission and make predictions of emission from uranium plasmas at higher temperatures, we have calculated energy levels and oscillator strengths for many transition arrays in UI through UIV. The computations are based on the familiar Slater-Condon theory and involve two steps:

1) One-electron radial wavefunctions are computed by solving an approximate form of the Hartree-Fock equations.[6,7] These provide an absolute energy for the center of gravity of the electronic configuration (including correction for relativistic effects), numerical values for the radial integrals which describe the electrostatic and spin-orbit interactions among the electrons, and the reduced dipole matrix elements connecting configurations of different parity.

2) The center of gravity and radial integrals are used as input for a computer program which calculates the detailed energy level structure for each configuration, the eigenvector compositions for the levels, and the oscillator strengths when configurations of opposite parity are computed together.[8,9]

Figure 2, taken from Ref.10 (First Conference on Uranium Plasmas in Gainsville, 1970), depicts schematically the results for the energy level structures of the ground state configurations of UI through UVI. We were more interested in the distribution of oscillator strength, and after demonstrating that we could reproduce the level positions from basic theory for UI and UII, we proceeded to calculate level structures and oscillator strengths for many arrays of UI through UIV. Configuration interaction was neglected in these computations and might tend to broaden the distribution

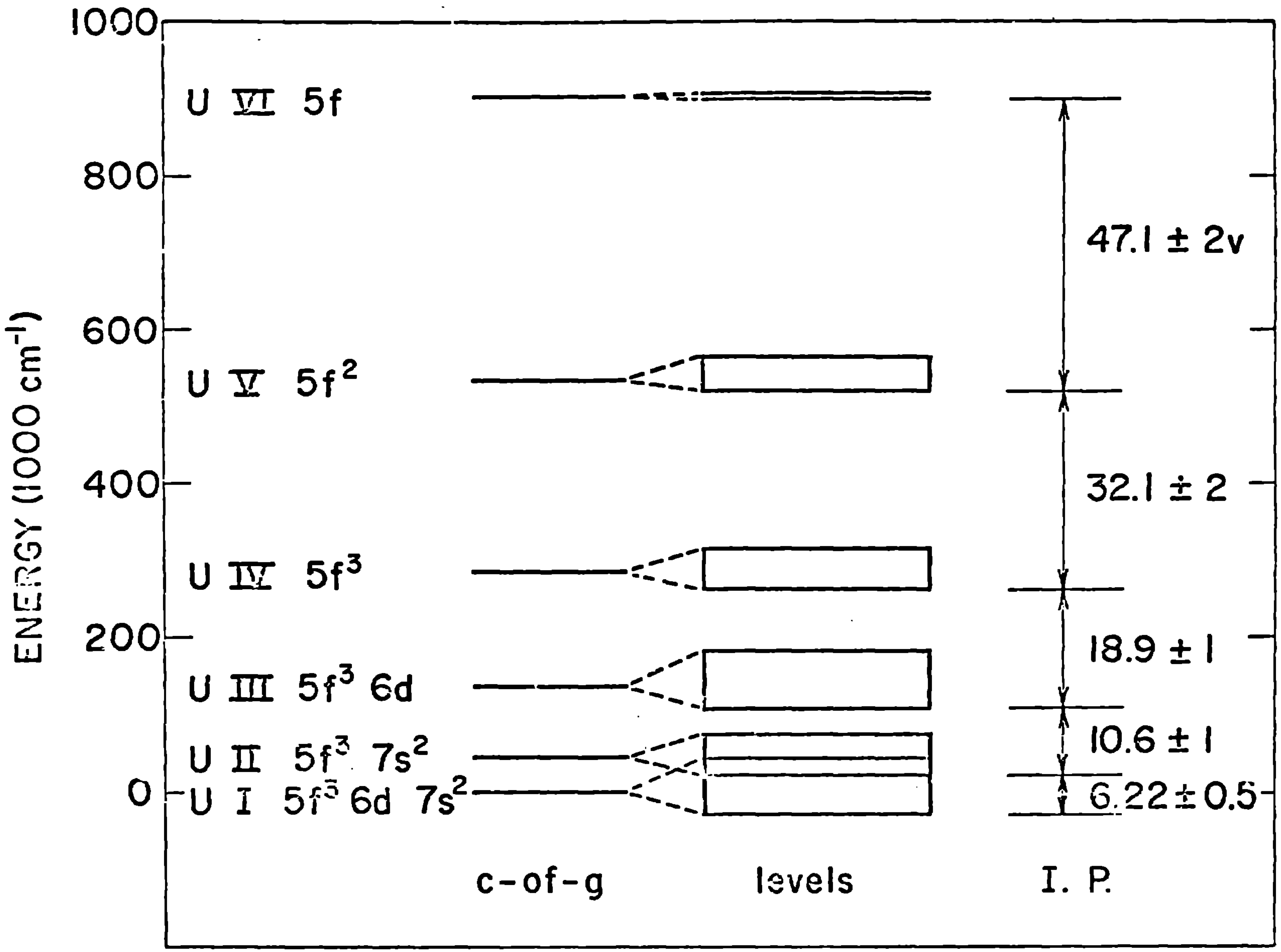

Fig. 2.

Energy-level range for ground-state configurations of UI through UVI and ionization potentials of UI through UV.

of oscillator strength to some extent. In some cases the number of parent terms was necessarily truncated to reduce the size of the matrices.

Tables I through IV show the transition arrays calculated for the first four stages of ionization of uranium. Arrays with very little total oscillator strength have been omitted from the tables. In each table the first column indicates the optical electrons of the two configurations involved in the transitions; the second column contains the total gf for that array and is a useful guide to the most important types of transitions (usually 6d-7p and 7s-7p). Columns 3 and 4 contain the wavelength and wave-number means of the oscillator strength distribution; column 5 gives the computed center of gravity energies for the configurations involved in the transition, which may be useful in estimating which configurations may be well populated at various temperatures.

Figures 3 through 6 contain composites of the oscillator strength distributions for UI, UII, UIII and UIV, formed by summing the contributions from the individual transition arrays for a given ionization stage. (The UI curve consists of an experimentally determined emission coefficient[3] correlated to the largest oscillator-strength distribution found for neutral uranium.) The main peak shifts slowly to the ultraviolet with increasing ionization, while the smaller peaks at lower wavelength shift toward the vacuum ultraviolet more rapidly. Significant emission from UIII should occur in the vacuum ultraviolet, whereas UIV emission should occur below 1000 Å. Figure 7 shows the same trend for the transition array of largest total oscillator strength in each ion stage, those being $f^n 6d7s$ - $f^n 6d7p$, save for the case of UI.

TABLE I

TRANSITION ARRAY COMPILATION FOR NEUTRAL URANIUM (UI)

Transition Array	Total gf	Mean Wavenumber [1000 cm^{-1}]	Mean Wavelength [Å]	Configuration E_{av} [cm^{-1}]
$5f^36d7s^2$ - $5f^36d7s7p$	10896	27	3704	0 - 17992
$5f^47s^2$ - $5f^47s7p$	2930	24	4167	15652 - 31325

TABLE II

TRANSITION ARRAY COMPILATION FOR SINGLY IONIZED URANIUM (UII)

Transition Array	Total gf	Mean Wavenumber [1000 cm^{-1}]	Mean Wavelength [Å]	Configuration E_{av} [cm^{-1}]
$5f^36d7s$ - $5f^36d7p$	9680	29	3448	4600 - 33400
$5f^37s7p$ - $5f^37s7d$	4505	34	2941	25800 - 58600
$5f^47s$ - $5f^47p$	2610	27	3704	16600 - 43160
$5f^46d$ - $5f^47p$	1688	16	6250	29700 - 43160
$5f^36d7s$ - $5f^37s7p$	1350	23	4348	4600 - 25800
$5f^37s^2$ - $5f^37s7p$	1206	37	2703	0 - 25800
$5f^37s7p$ - $5f^37s8d$	704	50	2000	25800 - 74300
$5f^36d7s$ - $5f^36d8p$	211	61	1639	4600 - 65600
$5f^36d7s$ - $5f^37s8p$	52	59	1695	4600 - 62400

TABLE III

TRANSITION ARRAY COMPILATION FOR DOUBLY IONIZED URANIUM (UIII)

Transition Array	Total gf	Mean Wavenumber [1000 cm^{-1}]	Mean Wavelength [Å]	Configuration E_{av} [cm^{-1}]
$5f^2 6d7s$ - $5f^2 6d7p$	2635	41	2439	15900 - 55300
$5f^2 6d^2$ - $5f^2 6d7p$	2042	43	2326	14600 - 55300
$5f^2 7s7p$-$5f^2 7s7d$	1382	49	2041	60400 - 107500
$5f^3 7s$ - $5f^3 7p$	1046	38	2632	0 - 37136
$5f^3 6d$ - $5f^3 7p$	880	38	2632	1800 - 37136
$5f^2 5d7s$-$5f^2 7s7p$	438	46	2174	15900 - 60400
$5f^2 7s^2$-$5f^2 7s7p$	318	49	2041	24900 - 60400
$5f^3 6d$ - $5f^2 6d^2$	317	16	6250	1800 - 14600
$5f^2 7s7p$-$5f^2 7s8d$	145	78	1282	60400 - 137200
$5f^3 7s$ - $5f^2 6d7s$	98	22	4545	0 - 15900
$5f^2 6d^2$-$5f^2 6d8p$	90	94	1064	14600 - 107800
$5f^3 6d$-$5f^3 8p$	46	86	1163	1800 - 87200
$5f^2 6d7s$-$5f^2 6d8p$	31	92	1087	15900 - 107800

TABLE IV

TRANSITION ARRAY COMPILATION FOR TRIPLY IONIZED URANIUM (UIV)

Transition Array	Total gf	Mean Wavenumber [1000 cm^{-1}]	Mean Wavelength [Å]	Configuration E_{av} [cm^{-1}]
$5f6d7s$ - $5f6d7p$	427	51	1961	81600 - 131000
$5f^2 7s$ - $5f^2 7p$	277	49	2041	34200 - 81400
$5f^2 6d$ - $5f^2 7p$	246	61	1639	21800 - 81400
$5f^2 6d$ - $5f6d^2$	124	43	2326	21800 - 65100
$5f^3$ - $5f^2 6d$	61	28	3571	0 - 21800
$5f^2 6d$ - $5f^2 8p$	17	132	758	21800 - 153200

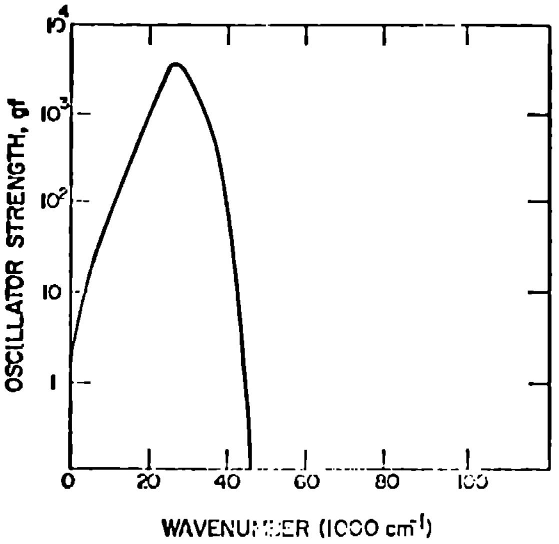

Fig. 3.
Experimental emission coefficient curve normalized to the peak of that calculated UI transition array which produced the greatest total oscillator strength, $5f^36d7s^2$-$5f^36d7s7p$.

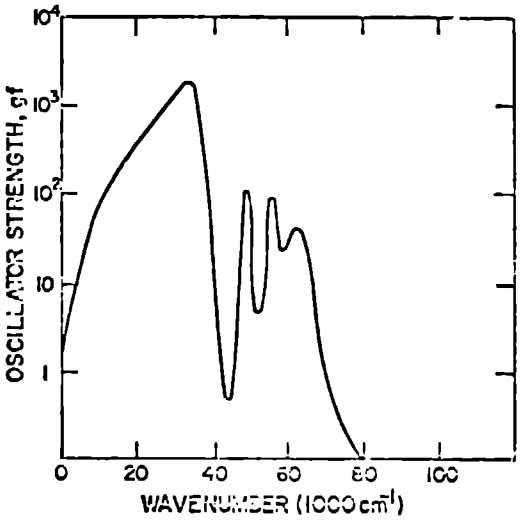

Fig. 4.
Calculated UII oscillator-strength distribution.

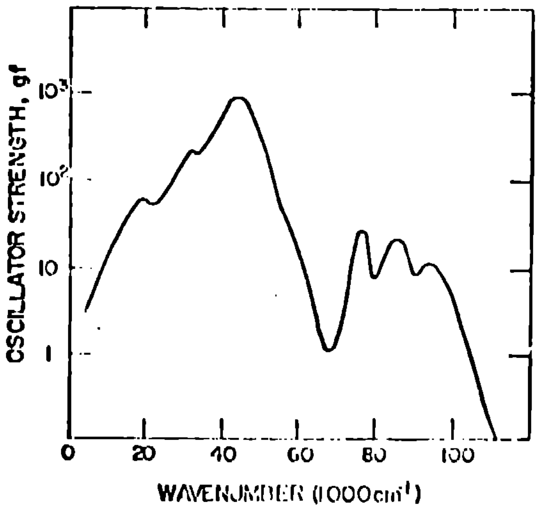

Fig. 5.
Calculated UIII oscillator-strength distribution.

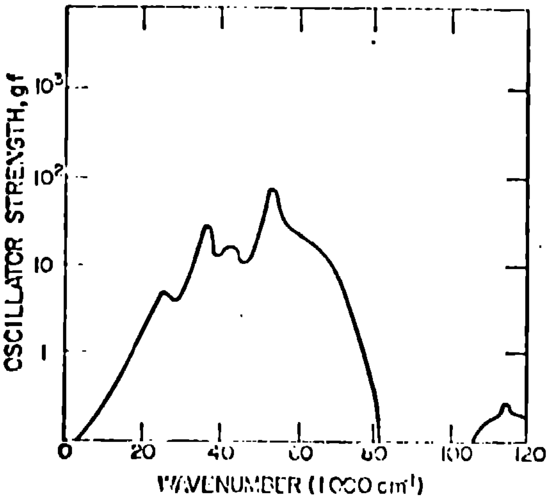

Fig. 6.
Calculated UIV oscillator-strength distribution.

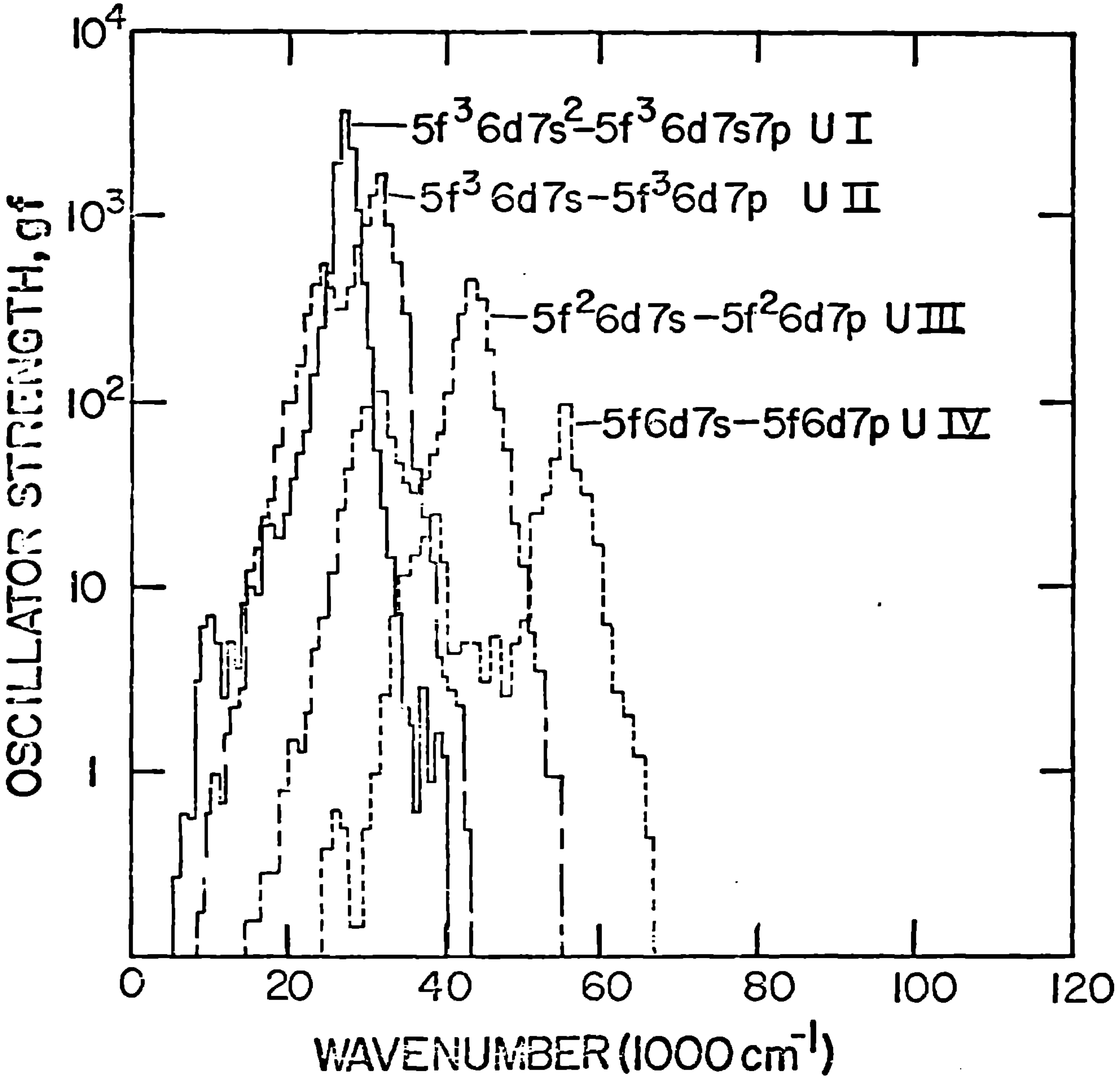

Fig. 7.
Illustration of blue shift for transition array of largest total oscillator strength in each ionization stage.

Upon comparing the variation of the experimental emission data (Fig. 1) to the calculated oscillator-strength distribution for UII, one finds reasonably good agreement except for gaps in the theoretical curve at 45 and 53 kilokaysers. We have attempted to locate UII transition arrays that would produce oscillator strength at the gap locations, but were unsuccessful. However, a composite of UII and UIII theoretical curves, shown in Fig. 8, compares very favorably with the experimental data, which is also sketched in Fig. 8. UIII is observed to contribute substantially to the emission from the 8000 K arc used by Mack;[3] this behavior is consistent with Saha theory and the earlier statement that many of the spectral lines emitted are likely to be of UIII origin. This deduction constitutes a verification of the present status of experimental and theoretical approaches.

VI. RECOMMENDATIONS

Clearly there are two directions to follow in expanding and verifying these results. First, strong shape correlation between measured uranium emission coefficient data and calculated oscillator strength distribution demonstrates the importance on incorporating a rigorous energy-level model into *ab initio* calculations of uranium plasma optical properties. The next phase in such calculations is the inclusion of a statistical-mechanical treatment to predict the state populations as a function of temperature and pressure. The computer size necessary to allow an adequate quantum and statistical mechanical treatment has recently become available, so the potential for improvement over earlier calculations[11] is established. Second, of comparable importance is

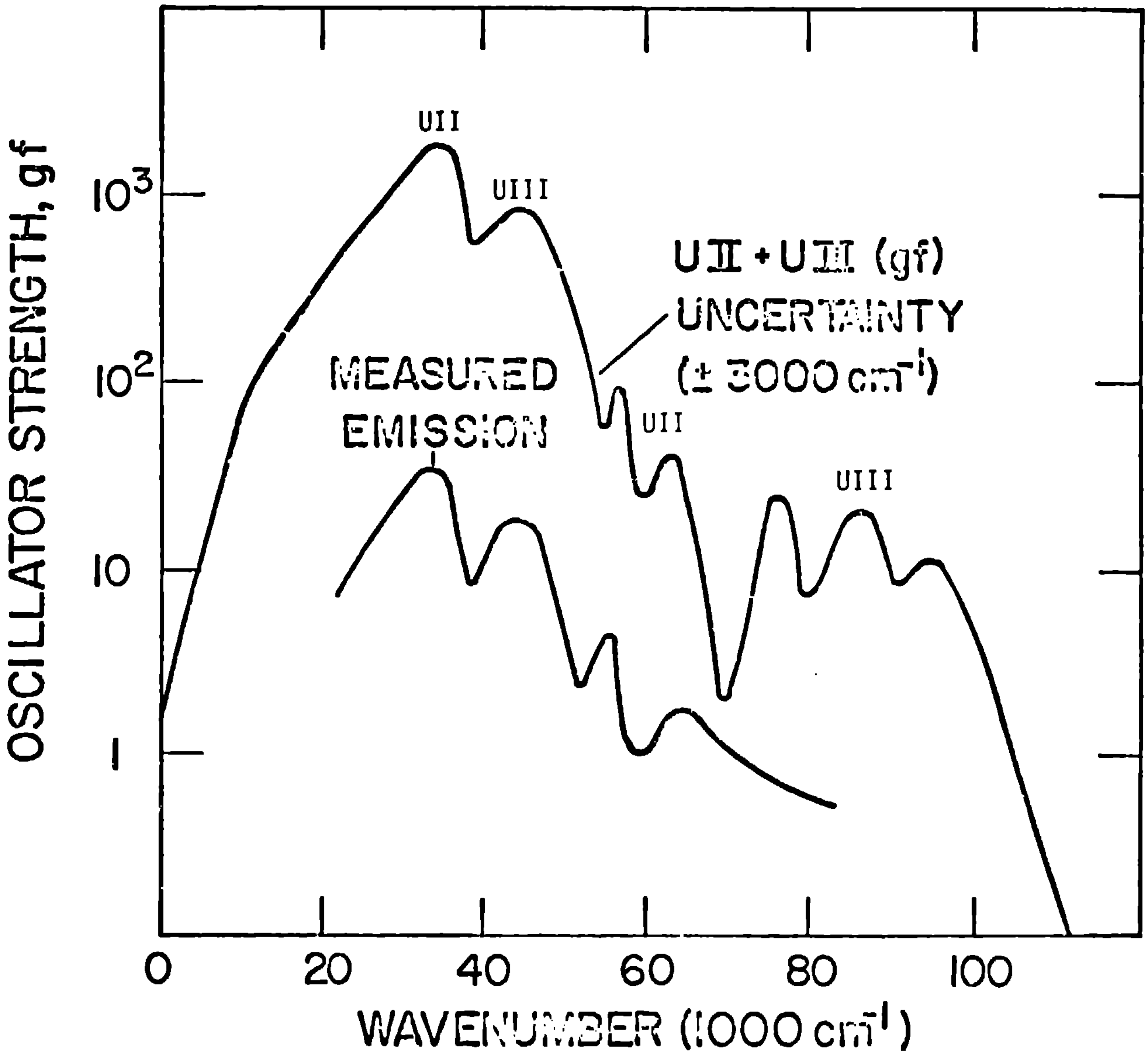

Fig. 8.
Comparison of major features for the observed uranium emission coefficient and calculated UII and UIII oscillator strength distribution.

experimental verification of the general shape of the oscillator strength distribution curves by measuring the emission coefficient for uranium plasmas likely to be dominated by the presence of higher ionization stages.

Each of these two directions is a major program in itself, and the viability of concepts using uranium plasmas will probably dictate whether or not either or both are pursued.

References

1. R. T. Schneider, H. D. Campbell, and J. M. Mack, "On the Emission Coefficient of Uranium Plasmas," Nuclear Technology, 20 p. 15-26, (1973).
2. P. Marteney, A. E. Mensing, and N. L. Krascella, "Experimental Investigation of Spectral Emission Characteristics of Argon-Tungsten and Argon-Uranium Induction Heated Plasmas," United Aircraft Corporation NASA report CR-1314 (1969).
3. J. M. Mack, Jr., "Investigation of Uranium Plasma Emission from 1050 to 6000 Å," Ph. D. Dissertation, University of Florida, Gainesville (December 1977).
4. A. G. Randol III, "A Determination of High Pressure, High Temperature Uranium Plasma Properties," Ph. D. Dissertation, University of Florida, Gainesville, Florida (August 1969).
5. J. M. Mack, Jr., "Temperature Profile Determination of a Uranium Plasma in a Helium Atmosphere," M. S. Thesis, University of Florida, Gainesville, Florida (June 1969).
6. R. D. Cowan, "Atomic Self-Consistent-Field Calculations Using Statistical Approximations for Exchange and Correlations," Phys. Rev. 163, 54 (1967).
7. R. D. Cowan, D. C. Griffin, "Approximate Relativistic Corrections to Atomic Radial Wavefunctions," J. Opt. Soc. Am. 66, 1010 (1976).
8. R. D. Cowan, "Theoretical Calculation of Atomic Spectra Using Digital Computers," J. Opt. Soc. Am. 58, 808 (1968).
9. R. D. Cowan, "The Theory of Rare Earth Energy Levels and Spectra," Nucl. Instr. and Methods, 110, 173 (1973).

10. D. W. Steinhaus, L. J. Radziemski, and R. D. Cowan, "Current Status of the Analysis of the Optical Spectra of Uranium," NASA SP 236 (1971).

11. D. E. Parks, G. Lane, J. C. Stewart, S. Peyton, "Optical Constants of a Uranium Plasma," Gulf General Atomic Incorporated, Report GA-8244 (NASA-CR-72348), (1967).