Selective Excitation Experiment

One of the assumptions used here is that the $^5D_0$ state has equal population for all sites. This is not true if the method of excitation is selective. Bron and Heller[1] used selective excitation to identify the lines, and their data should be mentioned here. Their results are listed in column 13 of their Table II, and they are in complete agreement with the present authors' identifications here except for the line at 7282.8 Å of the $^7F_2$ group. The authors believe Bron and Heller made a mistake when they identified this line as magnetic dipole transition. The selection rule for magnetic dipole is $\Delta J = 0$ or ± 1. Magnetic dipole transition from $^5D_0$ to $^7F_2$ is expected to be very weak. If this line is an electric dipole transition, it is in agreement with the present work.

VI. CONCLUSIONS

There is insufficient experimental justification in the Zeeman anisotropy patterns for assigning different site symmetry origins for the prominent lines in the $J < 3$ region of KCl: Sm^{2+} fluorescence spectrum, and the evidence presented here is strongly in favor of a C_{2v} site, most likely nearest neighbor, being responsible for these lines. Neither among the prominent lines nor the weaker lines in this region is there any evidence of C_{4v} site origins. Without supporting information such as polarization, intensity, and calculation from a Hamiltonian, the Zeeman anisotropy patterns cannot be used to make reliable identification of the site symmetry.

Other evidence for the strong lines belonging to the same C_{2v} site can be obtained through relative-intensity and relaxation-time measurements. These methods were carried out by the authors and they are in agreement with the present work. The details are reported in Paper III following.

ACKNOWLEDGMENT

The authors wish to express their gratitude to F. K. Fong for making preprints of his publications available.

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Fluorescent Spectra of Sm^{2+} in KCl. III. Site-Symmetry Correlation of Lines by Crystal Comparison and the Thermal Dependence of Intensities and Lifetimes*

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In this final paper in the series on the fluorescent spectra of Sm^{2+} in KCl, the authors present results of studies on the spectra of two different crystals at different temperatures, together with detailed studies in one crystal of the intensity and lifetime behavior versus temperature. Three sets of lines are found corresponding to three different site symmetries for the Sm^{2+}–K^+-vacancy system. All the prominent lines belong to the same set showing that they originate from the same level of the same (site-symmetry) ion. Magnetic studies presented in Papers I and II show that this site symmetry is C_{2v}, in accord with the earlier work of Bron and Heller.

I. INTRODUCTION

The charge compensation of divalent cations doped in alkali halides is generally accomplished by an alkali-ion vacancy, with the divalent ion located at an alkali-ion site of the normal lattice. The imperfect nature of the charge compensation can lead to distortions of the normal lattice, particularly if the vacancy is close to the impurity. The location of the vacancy with respect to the impurity will induce symmetries at the impurity ion site which are lower than the corresponding symmetry of the host lattice.

The case of Sm^{2+} in KCl has been of particular

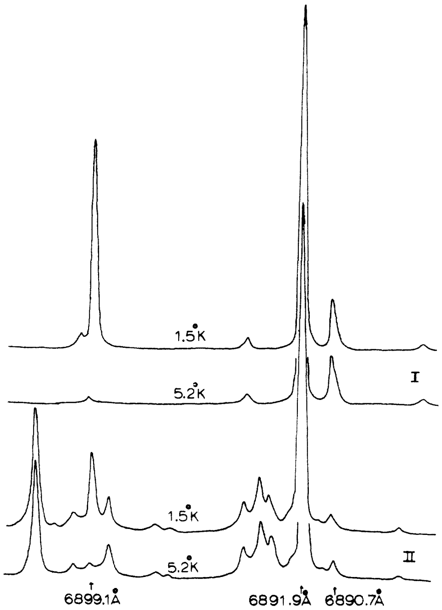


FIG. 1. Spectra of the $^7F_0$ group for type-I and type-II crystals at 4.2 and 1.5 °K.

interest in recent years. Bron and co-workers[1-3] published a series of papers concerning pseudolocalized vibrations of impurity-vacancy complexes in alkali halides, with particular attention to Sm^{2+}. This ion is useful because of its exceptionally strong fluorescence, which is due in turn to the strong $4f^6$-to-$4f^5 5d^1$ absorption bands in the visible region. The energy is transferred to the $^5D_0$ level of the $4f^6$ configuration which fluoresces to different J levels of the ^7F term. On the basis of the vibronic absorption spectrum and the polarized emission under selective excitation, Bron and co-workers came to the conclusion that most of the Sm^{2+} ions are in C_{2v} sites.

Fong and Wong[4] used first-order Zeeman effects to show that a few lines in the $^7F_4$ and $^7F_6$ regions, i.e., corresponding to transitions from $^5D_0$ to $^7F_4$ and $^7F_6$, belong to the C_{4v} site. Later, Fong[5,6] presented a statistical calculation to show that the dominant sites, in the order of relative importance, are $C_{4v}[200]$, $C_s[211]$, $C_{2v}[110]$, and $C_1[321]$, and identified some of the transitions as being from C_s sites. By using a magnetic field of 93.5 kG, Fong and Bellows[7] identified most of the transitions in the $^7F_1$ and $^7F_2$ regions. There is no doubt that Fong and co-workers are working in the direction that the statistical calculation is correct and that there are many different site symmetries of comparable population for the Sm^{2+} ions.

Wong never shared Fong's opinion on this problem, and has come to the conclusion that Bron and co-workers were right, that most of the Sm^{2+} ions are in the nearest-neighbor C_{2v} site. Indeed, four transitions earlier identified as from a C_{4v} site in Wong's laboratory can better be interpreted as from C_{2v} site symmetry with accidental degeneracy, as pointed out later in this paper.

The present authors have carried out further experiments to attack this discrepancy. A paper for the correction of Fong's[8] $^7F_3$ identifications and another paper[9] concerning the high-field second-order Zeeman effects in the $^7F_1$ and $^7F_2$ regions precede this paper. In this paper the problem of different sites is studied through intensity and relaxation-time measurements.

The idea of the intensity measurement is rather simple. Different methods of preparing the crystals give different population distributions among the various sites, but the ratio of the intensity of a given line in one crystal to the intensity of the same line in a different crystal will be identical for all lines originating from ions in the same site.

Additional work on the temperature dependencies of the intensities and relaxation times in one crystal was also carried out with the idea that the different symmetry sites will have different temperature dependency. These measurements confirm the results of the intensity measurements with different crystals.

II. EXPERIMENTAL METHODS

Two types of crystals were used in this experiment. For all the intensity and relaxation measurements, type-I crystals (grown at UCLA by the Kyropoulos method) were used. In addition, some of the crystals used by Fong and Wong a few years ago were used for intensity measurements only. These crystals give a different spectrum compared with the new crystals grown at UCLA. These old crystals are called type II in the following.

The spectrometer used was a home-made 2-m Czerny-Turner with 4×4-in. 1200-line/mm grating. The first-order spectrum was used and the tested resolving power is about 110 000. The detectors were E. M. I. 955 8B and Dumont 6911 photomultipliers, in a specially built housing for cooling with liquid nitrogen. For intensity measurements, the output of the photomultiplier was fed to a Kin-Tel model 202B dc millivoltmeter. The output of the voltmeter was fed to a Leeds and Northrup type G recorder. The sensitivity of the system was corrected by comparing the output with a type-L-101 standard lamp from Electro Optics Associates. For relaxation-time measurements, the output of the photomultiplier was fed to a Tektronics 565B oscilloscope with Polaroid camera. The excitation source was a 100-W tungsten lamp with a red-cutoff filter. For relaxation-time measurements, a chopper driven by a servo-controlled variable-speed motor was used. Repetitive traces

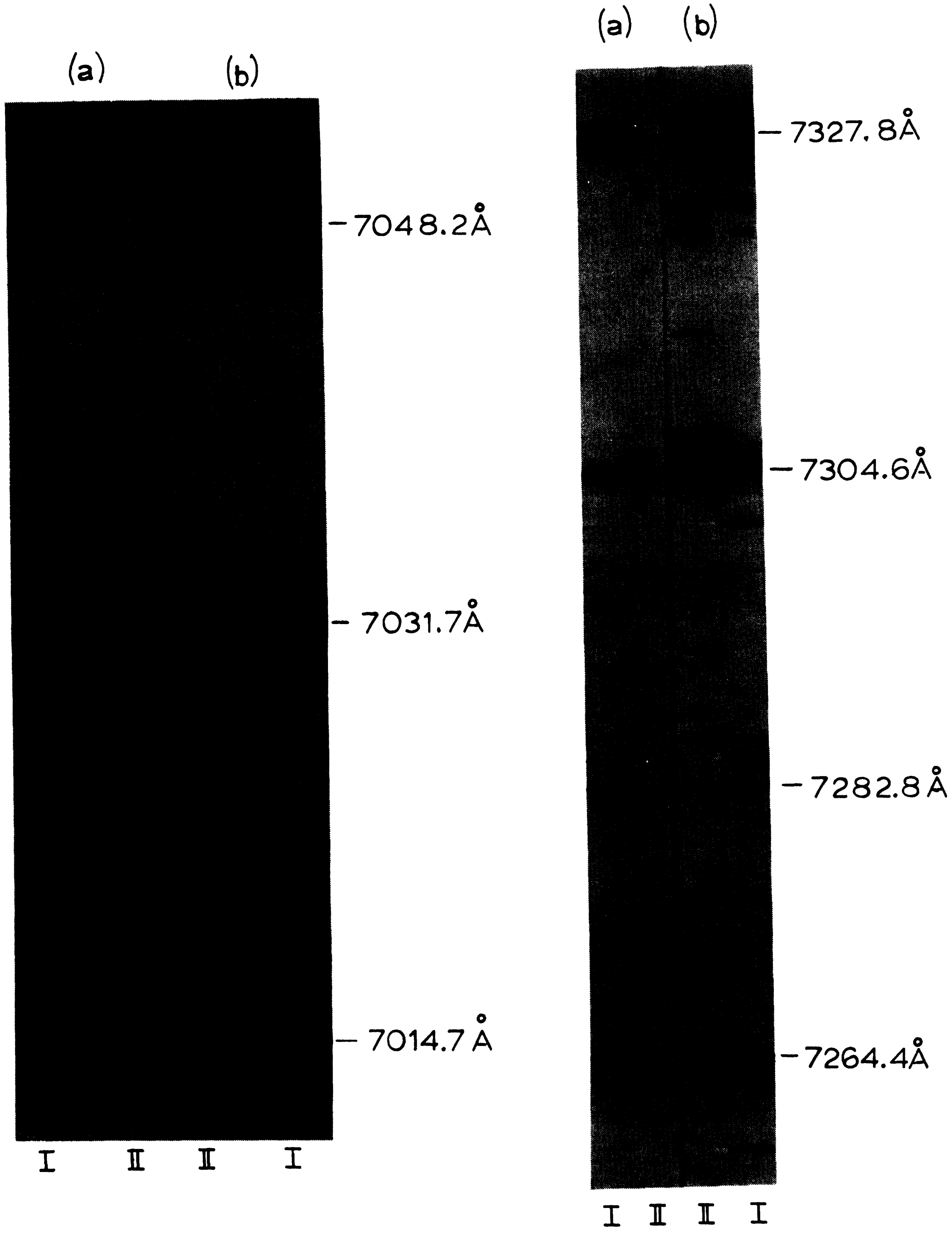


FIG. 2. Spectra of $^7F_1$ and $^7F_2$ group for type-I and type-II crystals at 4.2 °K for (a) weak exposure and (b) strong exposure.

were taken on each photo to provide a photographic average, and each photo was reduced by hand on a semilog plot to determine the decay time.

Both intensity and relaxation-time measurements were made in the temperature range 4.2–40 °K.

III. RESULTS ON INTENSITY MEASUREMENTS FROM TWO TYPES OF CRYSTAL

The spectra of type-I and type-II crystals are given in Figs. 1–3. Figure 1 shows the spectra of the $^7F_0$ group. At 5 °K there are three lines at

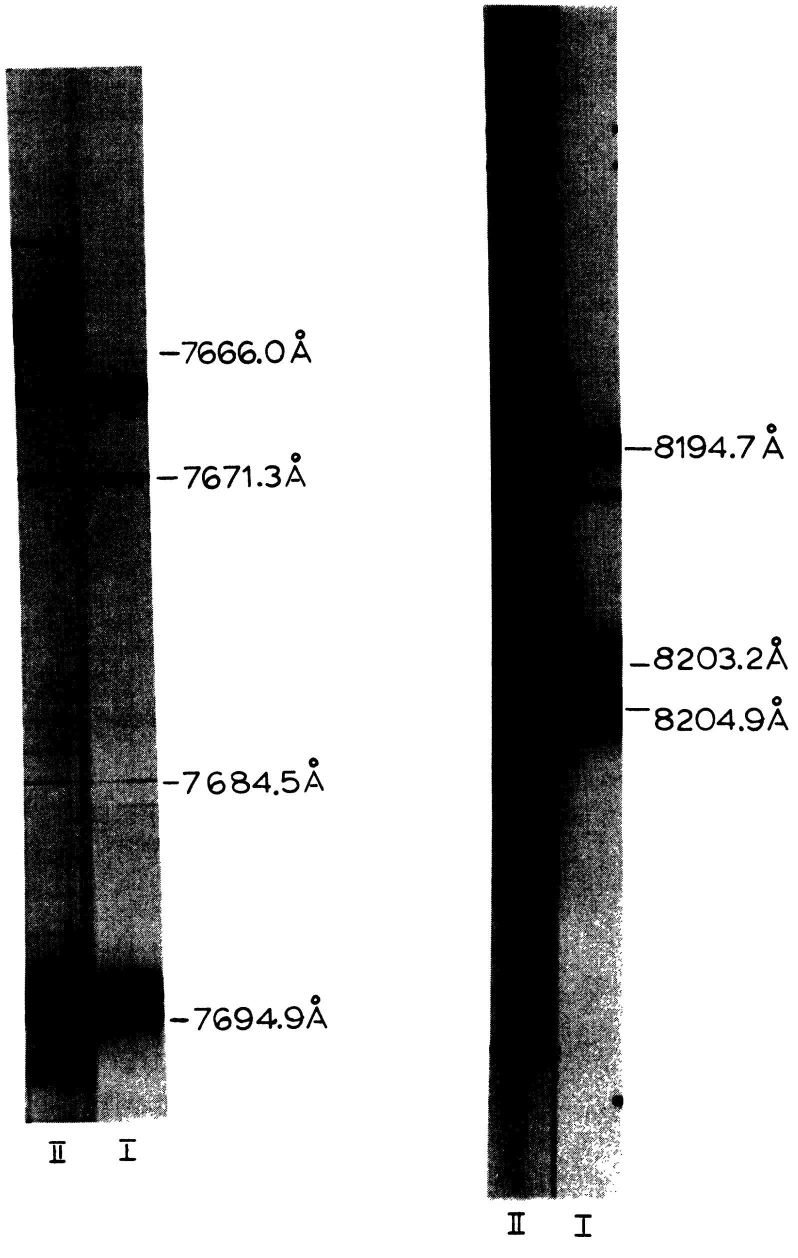


FIG. 3. Spectra of $^7F_3$ and $^7F_4$ group for type-I and type-II crystals at 4.2 °K.

6890.7, 6891.9, and 6899.1 Å which are in common for both crystals. At 1.5 °K the line at 6899.1 Å increased in intensity and it is quite different from the rest of the spectrum. Since only one line is expected for the $^5D_0$ to $^7F_0$ transition, each line must correspond to a different symmetry site. The possibility of vibronic transitions is unlikely because of the sharpness of the lines. The vibronic transitions associated with this group were observed by a fast scan and higher gain, and they are given in Fig. 4. These vibronic transitions have been studied by Bron,[10] and the theoretical calculations were reported by Buchanan and Woll.[11] The spectrum shown in Fig. 4 agrees well with the calculations.

Figures 2 and 3 give the spectra for both crystals at 4 °K taken with a 2.5-m Ebert spectrometer in the third and fourth order, with a Hartman diaphragm. The high resolving power (about 200 000) of the spectrometer and the use of the Hartman diaphragm gives a more reliable choice of common lines between the types of crystal.

The intensity ratios between type-I and type-II crystals are given in Table I. At 1.5 °K, the ratios are 3.3, 1.0, and 2.7 for the three common lines in the $^7F_0$ group at 6890.7, 6891.9, and

6899.1 Å, and the corresponding sites are called γ, α, and β, respectively. The intensity ratios vary with the experimental conditions and are normalized to 1.0 for the strong line at 6891.9 Å. All through the spectrum, if a line appears in both crystals and has an intensity ratio of about 1, it will be identified as site α. If the ratio of intensity is about 3, it could be either site γ or β because these lines are weak, and the weak lines do not give intensity ratios accurate enough to discriminate between 3.3 and 2.7. However, site β shows a great increase of intensity when cooled from 4.2 to 1.5 °K, and this unique temperature dependency can be used to identify site β.

In Table I, the $^7F_1$ group has three strong lines with intensity ratios of 1.0, and these three lines are identified as magnetic dipole transitions in the previous paper of the authors and by Bron and Heller.[1] This is in agreement with the site-α identification except for a discrepancy in the intensity measurements. For magnetic dipole transitions, the intensities of all three transitions belonging to $^7F_1$ must be equal. The difference of intensity between the two lines at 7014.7 and 7031.7 Å is about 10%, and this is within the accuracy of the intensity measurement; but the line at 7048.2 Å has an intensity about half that of the other two lines, which is beyond the error of the measurement. This may be due to the mixing with the vibronic state. The broad linewidth more or less supports this idea.

The $^7F_2$ group shows four transitions with inten-

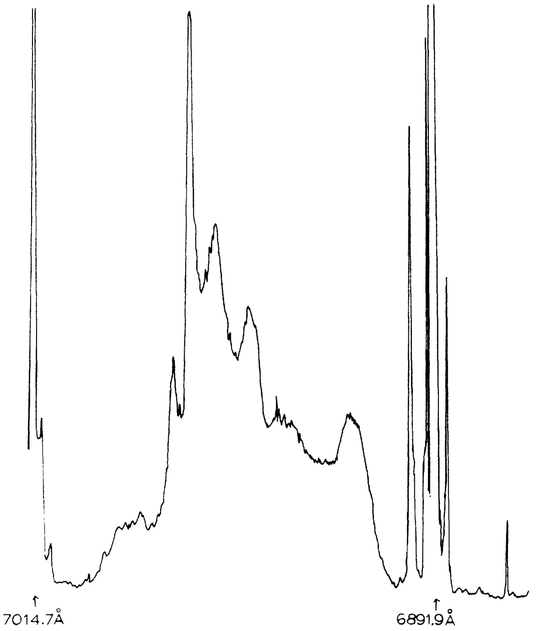


FIG. 4. Vibronic spectrum of $^7F_0$ group at 4.2 °K.

TABLE I. Fluorescent lines of KCl : Sm^{2+} in the 6880–8780-Å region. I is the integrated intensity (arbitrary units) for the type-I crystal and η is the intensity ratio of type I to type II.

J region	λ(Å)	I	η	Site
0	6887.6	12	a	δ
	6890.7	164	3.3	γ
	6891.9	1000	1.0	α
	6893.8	31	a	ϵ
	6899.1	635	2.7	β
1	7013.5	~0	a	
	7014.7	830	1.0	α
	7015.3	10	a	
	7018.0	54	3.4	γ
	7018.8	10	a	
	7022.1	5	a	
	7022.7	67	b	γ
	7031.3	76	b	β
	7031.7	718	1.0	α
	7039.4	82	2.1	β
	7043.3	101	3.2	β
	7048.2	393	1.0	α
2	7258.0	31	a	
	7263.2	28	5.0	γ
	7264.4	437	1.0	α
	7278.0	138	3.2	β
	7279.1	5	b	β
	7279.5	16	5.3	β
	7280.1	5	a	
	7282.8	99	1.0	α
	7284.8	29	4.0	γ
	7285.9	5	a	
	7300.9	39	3.7	γ
	7304.6	261	1.0	α
	7320.8	84	b	γ
	7322.7	24	a	
	7327.8	402	1.0	α
3	7666.0	2	1.0	α
	7667.6	17	3.1	
	7671.3	12	1.1	α
	7673.2	3	•••	β
	7684.5	4	1.0	α
	7685.6	3	a	
	7687.5	5	a	β
	7689.4	3	a	
	7690.1	2	a	β
	7693.2	3	a	
	7694.0[c]	300	0.9	α
	7694.9[c]	137	1.0	α
	7701.9	3	a	β
	7704.8	35	a	β
4	8132.6	17	1.2	α
	8136.8	2	a	
	8194.7	94	1.3	α
	8196.6	10	a	
	8202.4	31	6.5	
	8203.2	30	1.2	α
	8204.9	199	1.2	α
5	8742.8	0.2	a	
	8746.4	0.6	1.2	α
	8747.3	1.8	1.1	α
	8778.4	4.4	1.1	α

[a] Lines not observed in type-II crystal.

[b] No reliable intensity can be obtained due to overlap with other lines.

[c] In Papers I and II, these two lines designated as 7693.5 and 7694.5 Å.

sity ratios of 1.0, which are identified as from site α. For electric dipole transitions between $^5D_0$ and $^7F_2$, four lines are expected under C_{2v} site symmetry, which fact agrees with the present results.

The $^7F_3$ group has five transitions with intensity ratios within 10% of 1.0; five lines are expected for this group under C_{2v} symmetry. Two of these lines at 7694.0 and 7694.9 Å have been identified as from a C_{2v} site by low-field Zeeman effects.[8]

The ratios of intensity closest to 1.0 in the $^7F_4$ group are 1.2 and 1.3. If the associated lines are identified as from site α with C_{2v} symmetry, there will be a serious discrepancy with the identification of C_{4v} symmetry through first-order Zeeman effects.[4]

The three lines at 8194.6, 8203.2, and 8204.9 Å show first-order Zeeman effects; the anisotropy patterns agree with the calculated values for $S=1$ and $g=5.04$, 5.34, and 5.50 under C_{4v} symmetry. Furthermore, when the magnetic field was along the [111] direction, all three lines showed two components, in agreement with C_{4v} symmetry. The polarization data of these three lines, not reported by Fong and Wong in detail, are also in agreement with C_{4v} symmetry.

The decomposition of a $J=4$ level contains two Γ_5 states under C_{4v} symmetry that would give first-order Zeeman effects; the sum of the g factors should obey the sum rule. (Note: For spin of $\frac{1}{2}$, the sum of the g factors should be 12, but since Fong and Wong assumed a spin of 1, the sum should be 6.) Since no two of the experimental g values can be chosen to give the proper sum, it was assumed that these three lines originated from three different C_{4v} sites.

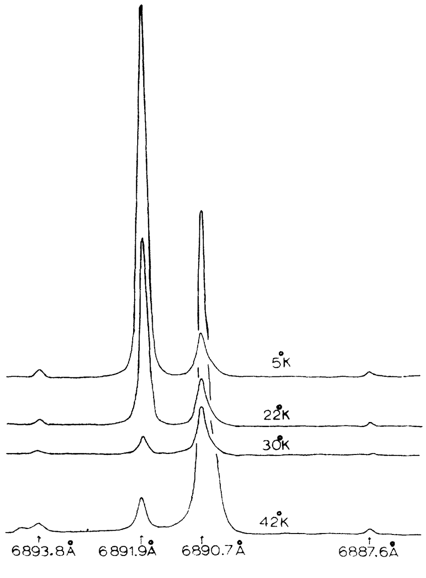


FIG. 5. Temperature dependence of $^7F_0$ group for type-I crystal. (The 4.2 °K spectrum has a gain increase of 10 over the others.)

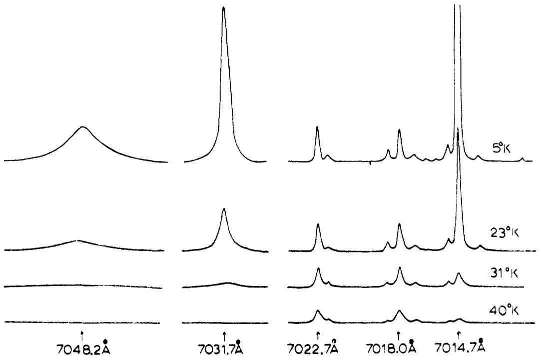


FIG. 6. Temperature dependence of $^7F_1$ group for type-I crystal.

There are some points that were not really understood by Fong and Wong. For example, no explanation was given to explain why the dotted line in Fig. 7 of Fong and Wong was missing, or why the line at 8204.9 Å has only a half-pattern.

Since these lines have the same intensity ratio in Table I, they must belong to the same site, and the Zeeman effect will be very complex since the separations of these lines are of the same order as the Zeeman effect. This point was not taken into account by Fong and Wong. If these three lines belong to the same site, then the sum rule will disagree with the C_{4v} identification.

Actually the Zeeman effect pattern also agrees with the C_{2v} identification if there is accidental degeneracy. For example, if a Γ_1 is very close to a Γ_2 state (the character table is given in Paper II),[9] it will give exactly the same first-order Zeeman effect pattern as the C_{4v} symmetry; the polarization data with the magnetic field along the [100] direction are also identical. There is no requirement that the Γ_1 have the same intensity as the Γ_2 state, so if one of them is very weak, the missing dotted line can be accounted for. The sum rule will not apply here. Since the C_{2v} identification agrees better with the experimental data, the identification of site α with C_{2v} symmetry for the three lines at 8132.6, 8194.7, and 8203.2 Å is most likely though not definite. However, the site-α origin of the line at 8204.9 Å will be established by the lifetime measurements in Sec. IV.

(Fong and Bellows have recently analyzed the lines in this region.[12] In double checking that experiment, the authors find that the polarizations are exactly the opposite from that given by Fong and Bellows. Since those authors also assert incorrectly that the polarization labels given in Fig. 11 of Fong and Wong were reversed,[4] we sug-

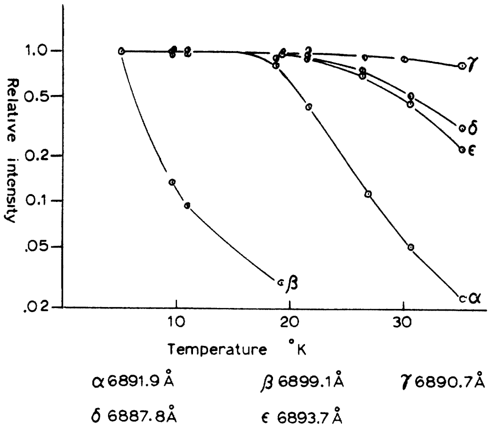


FIG. 7. Relative intensity vs temperature for $^7F_0$ group of type-I crystal.

gest that the workers at Purdue check their analyzer for mislabeling.)

There is no problem with the weak lines in the $^7F_5$ group. The intensity ratios indicate the three lines at 8746.4, 8747.3, and 8778.4 Å belong to site α; one of them has been identified as from the C_{2v} site symmetry. The $^7F_6$ group was not studied because it was outside the range of the spectrometer used.

IV. TEMPERATURE-DEPENDENT INTENSITY

The intensities in the fluorescent spectrum have a very strong temperature dependence as shown in Figs. 5 and 6. Since different sites may have different thermal dependencies of the intensity and lifetime, it is possible to study the problem of different sites through the temperature-dependent intensity of different transitions. As reported by Alam and Di Bartolo[13] for the case of Sm^{2+} doped in CaF_2 and SrF_2, the intensity remains constant at low temperature and falls off at higher temperature. Figure 7 gives the intensity of the $^7F_0$ group at temperatures up to 40 °K. Site β has not reached a low enough temperature to show the limiting-case lifetime, but measurement at pumped helium temperature does indicate the leveling off of the intensity. Different sites do not seem to draw intensity from each other, which is expected if they are isolated.

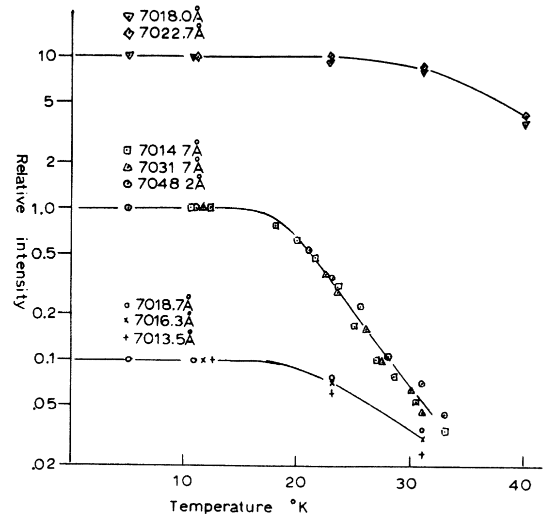


FIG. 8. Relative intensity vs temperature for $^7F_1$ group of type-I crystal. (For convenience the data on the upper curve have been raised by a factor of 10 and the data on the lower curve have been lowered by a factor of 10.)

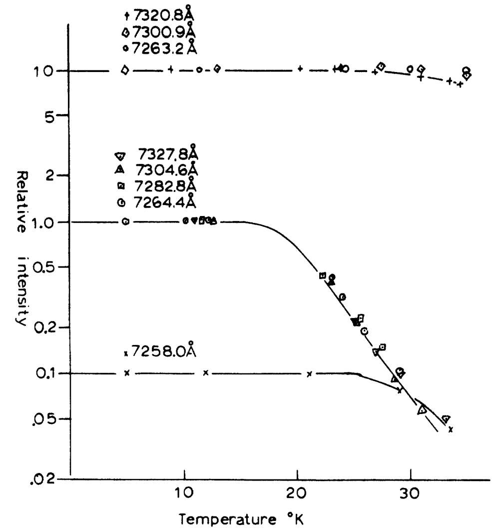


FIG. 9. Relative intensity vs temperature for $^7F_2$ group of type-I crystal. (For convenience the data on the upper curve have been raised by a factor of 10 and the data on the lower curve have been lowered by a factor of 10.)

Figure 7 indicates the existence of five different sites in the type-I crystal. Two of the sites, δ and ε, have almost the same temperature dependence and leave doubt as to their identification. The other three sites, α, β, and γ, differ significantly in the thermal dependence and seem securely identified.

The three lines in the $^7F_1$ region belonging to site *B* were too weak to be measured at higher temperature, but the other eight lines were measured and the results are shown in Fig. 8. It is clear that the three strong lines at 7014.7, 7031.7, and 7048.2 Å have the same thermal dependence as the 6891.9-Å line in the $^7F_0$ group, and all three lines are identified as from site α. This is in good agreement with the identification presented previously. The two lines at 7018.0 and 7022.7 Å must belong to site γ because they have the same

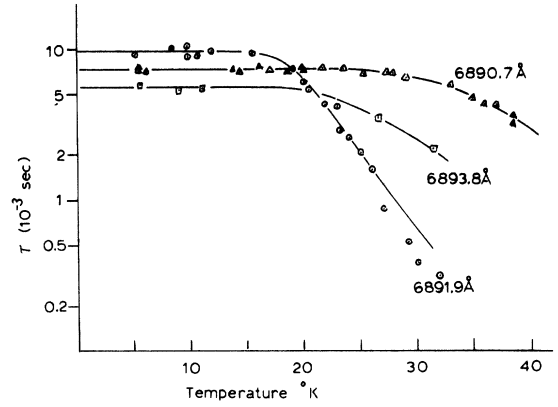


FIG. 10. Relaxation time vs temperature of $^7F_0$ group for type-I crystal.

temperature dependence as the line at 6890. 7 Å in the $^7F_0$ group. The other three lines at 7013. 5, 7016. 3, and 7018. 8 Å are either site δ or site ϵ.

Figure 9 shows four strong lines at 7264. 4, 7252. 8, 7304. 6, and 7327. 6 Å having the same thermal dependence as site α, in agreement with the previous identification.

The temperature-dependent intensity was not measured for the higher-J groups due to the low sensitivity of the S-1 photomultiplier. The results from the temperature-dependent intensity work should be the same as those from the work on the thermal dependence of the lifetime. The relaxation-time measurements were carried out to verify the intensity-measurement results. The results, given in Sec. V, are in very good agreement with the work discussed earlier.

V. TEMPERATURE-DEPENDENT RELAXATION TIME

Relaxation times at various temperatures were measured for all lines of sufficient intensity. Since fast circuitry was necessary, the number of lines studied was much less than in the intensity study, where integration over time could be used. On the other hand, for broad lines that were not strong enough to give reliable data in the temperature-dependent intensity scan, a wide slit width could be used to obtain lifetime measurements. Particular care was taken in the case of the $^7F_3$ lines at 7694. 0 and 7694. 9 Å (referred to in Papers I and II as 7693. 5 and 7694. 5 Å). A 100-μ slit width was used to assure resolution of these two lines. The results for the $^7F_0$ group are given in Fig. 10 and those for all the other strong lines, in Figs. 11(a)–11(c). These curves are in good agreement with the previous identification. The solid curves drawn through the experimental data of the temperature-dependent intensity, as well as of the lifetime, are calculated from the equation

$$\frac{1}{\tau} = \frac{1}{\tau_0} + \frac{1}{\tau_q} e^{-\Delta E/kT},$$

with $\tau_0 = 9.8\times10^{-3}$ sec, $\tau_q = 9.3\times10^{-7}$ sec, and $\Delta E = 137$ cm^{-1} for site α. This thermal behavior, encountered in a variety of circumstances in crystals,[14–16] arises when there is a level or band with fast relaxation time lying above the fluorescent state by an energy gap ΔE. The derivation is straightforward and assumes the two levels are in thermal equilibrium with each other. For the present no speculation of the nature of the quenching level will be given.

VI. CONCLUSIONS

Bron and co-workers' assertion that most of the Sm^{2+} ions are in the C_{2v} site symmetry is now supported by the following arguments:

(a) polarized excitation/emission,[1]

(b) vibronic absorption spectrum,[12]

(c) anisotropy pattern of first-order Zeeman effect,[4]

(d) anisotropy pattern of second-order Zeeman effect,[8,9]

(e) polarization of different components in the Zeeman effect,[4,8,9]

(f) relative intensity of different components in the Zeeman effect,[9]

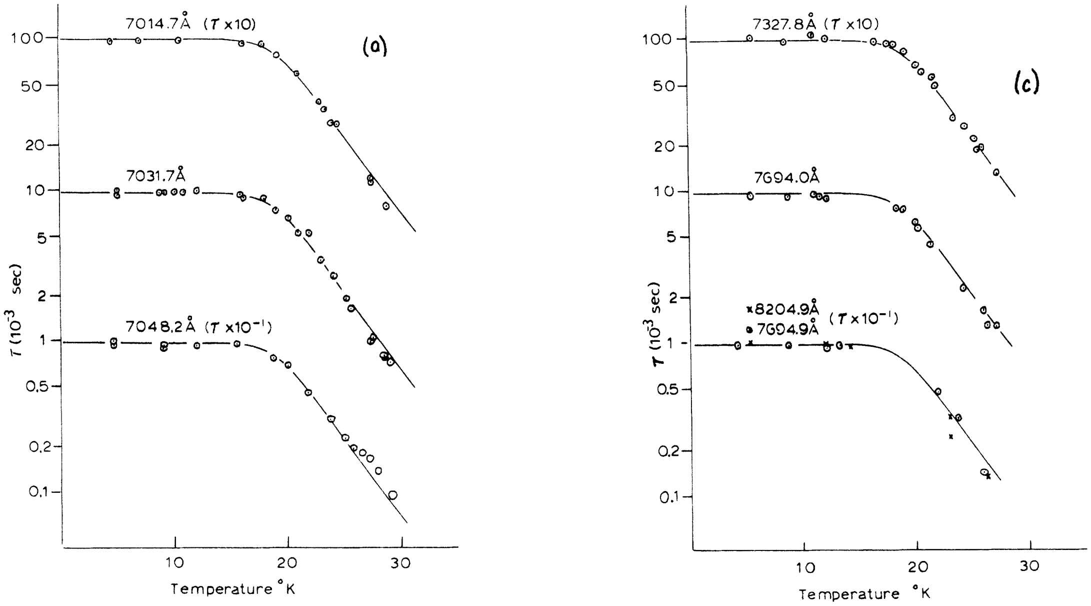
(g) comparison of intensities in different crystals (present work),

(h) thermal dependence of intensities (present work),

(i) thermal dependence of lifetimes (present work).

Items (g)–(i) show that all the prominent lines come from the same site, and items (c)–(f) show that the only consistent symmetry assignment for this site is C_{2v}. At 1. 8 °K, this site α accounts for 73% of the (line) radiation in the region 6800–9000 Å. At 4. 2 °K this figure climbs to 90% due to the quenching of site β. Although this quenching, by "nonradiative" processes not understood at present, prevents using the spectral line intensities to accurately measure the population distribution of different site symmetries, the indication is clear that a majority of ions are in site α. Another estimate, made from the intensity of the magnetic dipole transition of the $^7F_1$ group, gives the relative population of Sm^{2+} ions in site α as 0. 8 ± 0. 1. Site α with C_{2v} symmetry will thus account for 70–80% of the Sm^{2+} ions in the type-I crystal, and site β with lower symmetry will account for 10–20%, the remainder being distributed among other sites.

On the basis of the data presented by Fong and Wong, the lines in the $^7F_6$ region at 9439. 16 and 9439. 95 Å are clearly of C_{2v} origin. The anisotropy



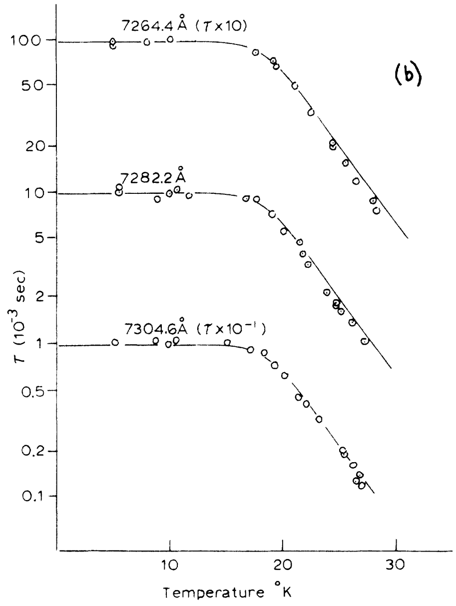


FIG. 11. Relaxation time vs temperature of the strong lines in the $^7F_1$, $^7F_2$, $^7F_3$, and $^7F_4$ groups for type-I crystal. (For convenience the data on the upper curve have been raised by a factor of 10 and the data on the lower curve have been lowered by a factor of 10.)

pattern is an ideal example of a two-level system (Γ_1, Γ_2) amenable to exact diagonalization.[9] The polarization data given by Fong and Wong show that the lines are of electric dipole nature from the $^5D_0$ (Γ_1) level of a C_{2v} [110] site. (It is puzzling why Fong and Wong persist in calling these lines C_{4v} when they invoke a C_{2v} spin Hamiltonian to account for the separation of the lines at zero field. As already mentioned, Fong and Bellows's assertion that the π and σ labels were "inadvertently reversed" is wrong.) Thus these lines might also be from site α, leaving no evidence in the spectrum of C_{4v} sites.

Fong's calculation[5] is a good exercise but is unsupported by the experimental facts, which give C_{2v} as the dominant site and not C_{4v}. Since the two different crystals grown by different methods display different relative populations of sites α, β, and γ, it appears that the Sm^{2+}-K^+-vacancy system does not reach thermodynamic equilibrium at room temperature, at least in a couple of years. Even the presence of Sm^{2+} aggregates, which may account for many of the additional lines in the type-II crystal, would not affect the binding energy of a dispersed Sm^{2+}-K^+-vacancy pair. If such were the case, it would certainly be reflected by a wavelength shift. The different relative populations therefore indicate different effective temperatures for the Boltzmann factor. According to Fong's theory, however, C_{4v} sites should dominate C_{2v} sites at all temperatures up to the melting point of the crystal.[5] Either the binding energies used in the calculation are wrong or the populations are uncorrelated with the spectrum. Regardless, any attempt to utilize the fluorescent spectrum in determining the site distributions must start with the correct identification of the lines.

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Knight-Shift Calculations in Liquid Magnesium

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The Knight shift K_s and nuclear spin-relaxation time T_1 have been calculated in liquid Mg using the pseudopotential theory. The results of this calculation show that both K_s and T_1T are independent of temperature T within the error in our calculation. The calculated value of K_s at the melting temperature (651 °C) is $(0.10 \pm 0.005)\%$ which compares well with the recent experimental measurement $(0.112 \pm 0.004)\%$ made between -196 and 230 °C. From this evidence, it can be concluded that there is no significant change of K_s upon melting, contrary to what has been observed in Cd. The theoretical value of the Korringa ratio, $(K_s^2 T_1 T)_{\rm theor} / (K_s^2 T_1 T)_{\rm ideal}$, departs from its ideal value of unity by about 17%. This departure is attributed to the different role of exchange enhancement on the uniform and nonuniform spin paramagnetic susceptibilities.

I. INTRODUCTION

In this paper, we have investigated the temperature T dependence of the Knight shift K_s and nuclear spin-relaxation time T_1 in liquid Mg. Mg is a divalent metal and has hexagonal-close-packed (hcp) structure similar to Be and Cd. In general, these metals exhibit very interesting band structure and hyperfine properties.[1] The nuclear-magnetic-resonance properties of this divalent group have been extensively studied both experimentally[2–4] and theoretically[5,6] in the solid state. It is interesting to note that Cd shows a strong temperature dependence[4] of K_s, $K_{\rm an}$ (anisotropic Knight shift), and T_1T in the solid state. On the other hand, K_s in Mg[3] and presumably Be[2] is independent of temperature. Furthermore, K_s and T_1T in liquid Cd[4] exhibit no temperature dependence. The source of the temperature dependence has been explained in the solid[5] to be mainly due to the role of electron-phonon interactions and in the liquid[7] through the temperature dependence of the observed ionic interference functions. However, no such investigations have been made either in liquid Be or Mg. We will extend the theory developed earlier[8] for liquid Cd, to study the nuclear-magnetic-resonance properties of liquid Mg and attempt to understand the liquid-state properties as compared to those in the solid state.

The purpose of the present work is to calculate K_s and T_1 using pseudopotentials[9] and interference functions[10–12] in liquid Mg. The role of the Korringa constant and the effects involving the exchange interactions among conduction electrons in

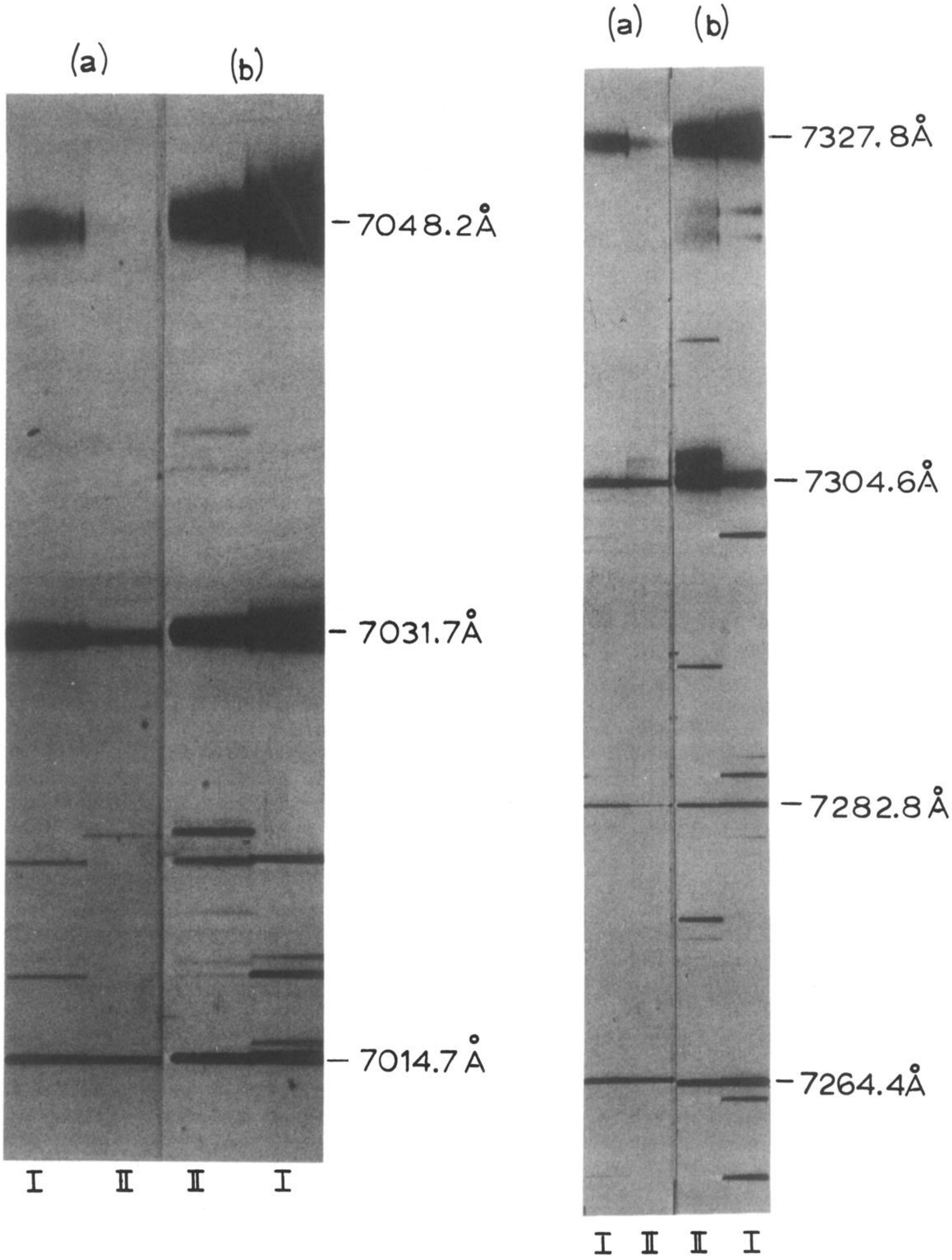

FIG. 2. Spectra of $^7F_1$ and $^7F_2$ group for type-I and type-II crystals at 4.2 °K for (a) weak exposure and (b) strong exposure.

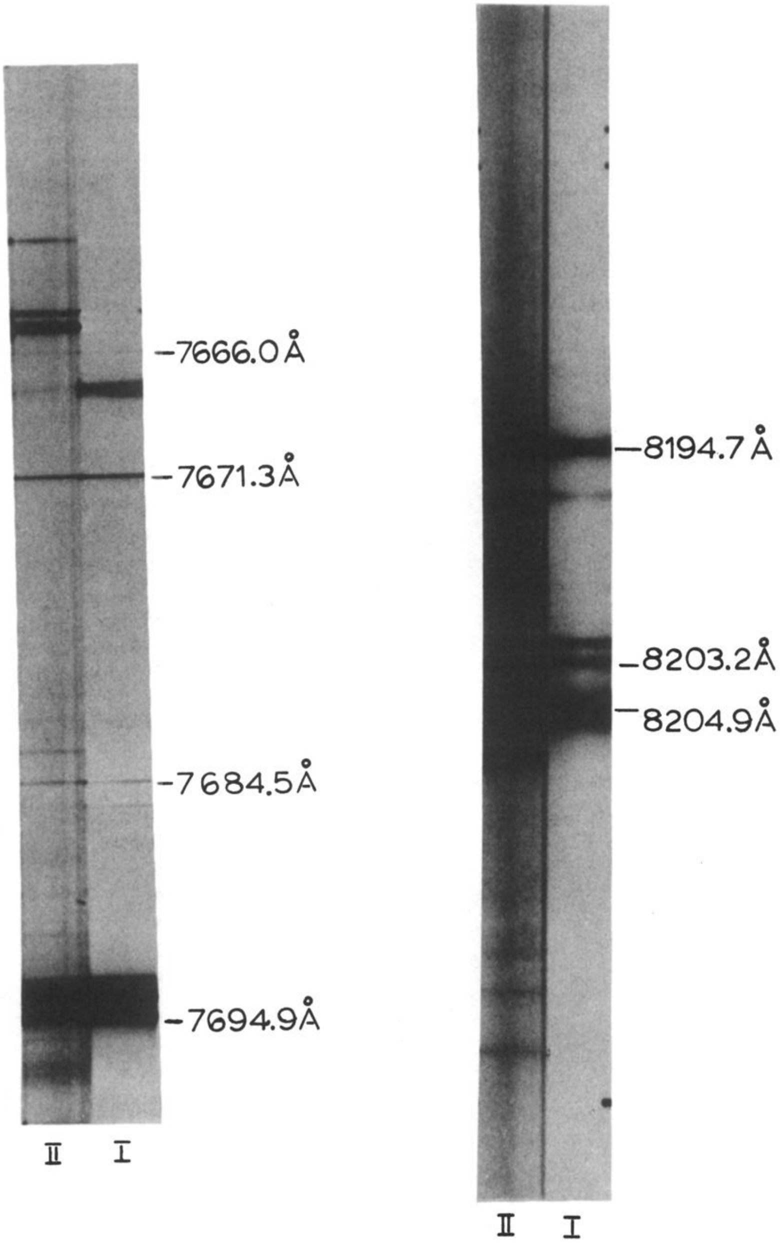

FIG. 3. Spectra of $^7F_3$ and $^7F_4$ group for type-I and type-II crystals at 4.2°K.