PHYSICAL REVIEW A VOLUME 13, NUMBER 1 JANUARY 1976

$K\alpha$ x-ray satellite intensities with proton impact on Al to Fe at a fixed scaled velocity

V. Dutkiewicz, H. Bakhru, and N. Cue
Physics Department, State University of New York at Albany, Albany, New York 12222
(Received 26 June 1975)

The $K\alpha$ x-ray satellite intensities were measured with proton impact at a scaled velocity (relative to that of the L-shell electron) $V_L = 2.1$ for $Z = 13$–20 and $V_L = 1.0$ for $Z = 20$–26. The intensity ratios $I_{K,1L}/I_{K,0L}$ are found to be well characterized by multiple-Coulomb ionization predictions. Taking into account the fluorescence yields, the comparisons of the measured intensity ratio with the corresponding ratio of theoretical vacancy cross sections yield a set of empirical vacany-rearrangement correction factors. Theoretical estimates of these factors based on the atomic transition rates of McGuire are in reasonable over-all agreement with the extracted values.

I. INTRODUCTION

The $K\alpha$ x-ray satellite intensities observed in atomic collisions in general do not directly reflect the primary vacancy productions because the branching ratios of the KL^n vacancy configuration depend on the specific defect. For the convenience of making approximations, this dependence can be expressed in terms of the effect on the fluorescence yield and the probabilities for vacancy rearrangement. The changes in the fluorescence yield with increasing vacancies are well recognized. On the other hand, the consideration of vacancy-rearrangement effects has not been widespread. Their effects have either been assumed[1] to be negligibly small or estimated[2-5] using the assumption that the level widths do not vary with increasing vacancies. In such estimates, the correction factor varies with Z and, depending upon the set of level widths used, can differ by as much as a factor of 2.[4] Although the isolation of these effects is difficult in a single-counter-type measurement, the dependence on Z can nevertheless be examined in situations where the mechanisms for the primary production of vacancies are reasonably understood.

In proton bombardment, only the KL satellite is usually observed. Both the magnitude and velocity dependence of its intensity relative to that of the diagram line, $I_{K,1L}/I_{K,0L}$, have been shown[4,6,7] to be well characterized by the predictions of the multiple-Coulomb ionization theory. Although the rearrangement effects were not considered in many cases, the agreements nevertheless suggest that multiple-Coulomb ionization is the principal mechanism for the primary productions of vacancies in proton impact. Thus a systematic measurement of the KL satellite intensity over a range of Z would enable one to examine the Z dependence of the vacancy-rearrangement effects. The following sections describe the details of our investigation at a scaled velocity $V_L = 2.1$ for $Z = 13$–20 and $V_L = 1.0$ for $Z = 20$–26. Here $V_L = (E/\lambda U)^{1/2}$, with λ being the projectile mass in units of electron rest mass and U_L denoting the weighted average binding energy of target L-shell electrons.

II. EXPERIMENTAL PROCEDURE

Atomic and molecular hydrogen beams from the 4-MV Dynamitron accelerator at Albany were used to bombard thin solid targets. These targets, with a thickness range of 100–200 $\mu g/cm^2$, were evaporated onto either a thick Al or Cu disk. Elemental materials were used except for the cases indicated in Table I. An ARL 4-in. curved crystal spectrometer coupled directly to the target chamber at 90° relative to the beam direction was used to detect the K x rays. The spectrometer was mounted with an ADP crystal for the measurements with $Z \leq 20$ and a LiF crystal for $Z \geq 20$. Published values of the $K\alpha_{1,2}$ energies were used to establish the energy scale of the spectrometer.

The diffracted x rays were detected with a gas-flow proportional counter equipped with a 2-μm-thick Makrofoil window and operated with a P10 gas mixture. Signals from the preamplifier were processed by standard electronics with the single-channel analyzer (SCA) set to accept only photopeak pulses. The x-ray spectra were recorded through a multichannel scaler, each channel corresponding to a specific Bragg angle and an accumulation of the SCA pulses for a preset number of beam particles. Incremental stepping of the spectrometer was automated through a Canberra Model 1712 scanner unit.

In order to avoid a major modification on the accelerator, whose nominal operating range is 1.8–4.2 MV, two sets of measurements were

TABLE I. Relative energies and intensities of the KL satellites and the corresponding calculated ratios of fluorescence yields and primary vacancy cross sections.

Target	$E_{sat}-E_{K\alpha_{1,2}}$ (eV)	$I_{K,1L}/I_{K,0L}$ (%) $V_L=1$	$I_{K,1L}/I_{K,0L}$ (%) $V_L=2.1$	$\omega_{K,1L}/\omega_{K,0L}$	$\sigma^V_{K,1L}/\sigma^V_{K,0L}$ (%) $V_L=1$
Al	5.1±0.7 ($K\alpha'$)		27.5[d]	1.097	47.5
	8.3±0.4 ($K\alpha_3$)				
	9.8±0.6 ($K\alpha_4$)				
Si (SiO_2)	7.8±1.0 ($K\alpha'$)		18.2[d]	1.092	35.4
	13.1±0.6 ($K\alpha_3$)				
	15.8±0.8 ($K\alpha_4$)				
P (CoP)	13.7±1.2		16.0	1.086	27.1
S	16.3±1.0		14.1	1.081	21.5
Cl ($CaCl_2$)	17.8±1.3		10.0	1.072	17.6
K (KBr)	23.8±2.1		5.8	1.063	12.2
Ca ($CaCl_2$)	20.5[a]±1.1	9.8	6.0	1.060	10.4
Sc	22.2[a]±1.3	8.3		1.058	9.0
Ti	22.5[a]±1.5	8.0		1.052	7.9
V	23.3[a]±1.8	6.8		1.051[c]	7.0
Cr	26.0[a,b]	5.8		1.049[c]	6.3
Mn	27.4[a,b]	4.4		1.047[c]	5.6
Fe	28.7[a,b]	3.9		1.044	5.1

[a] Relative to the $K\alpha_1$ line.
[b] Held to the weighted average value of the satellite group reported in Ref. 8.
[c] Interpolated values.
[d] Sum of $K\alpha'$, $K\alpha_3$, and $K\alpha_4$ intensities.

made: $V_L=1.0$ for $Z=20-26$ and $V_L=2.1$ for $Z=13-20$. For a given set, H_2^+ and H_3^+ as well as H_1^+ ions were required to cover the proton energy range implied by a given V_L. The resulting K x-ray spectra at the same proton velocity are not expected to depend on the beam species used; indeed, no discernible differences were observed in the few cases where such comparisons were made.

III. DATA ANALYSIS AND RESULTS

As expected, the only satellite excited with any appreciable intensity corresponds to single-K–single-L vacancies (KL), except for the cases of Al and Si where the KL^2 satellite is also seen. Typical spectra are shown in Figs. 1 and 2. Using a double-crystal spectrometer, Parratt[8] had earlier shown that the KL satellite consisted of several transitions. In the present study these transitions are partially resolved only for Al and Si, and unresolved for the higher-Z targets (see Figs. 1 and 2).

Peak areas were extracted by a least-squares fitting of the spectrum with a function consisting of a sum of standard line shapes plus a linear background. For Al and Si, the numerical line

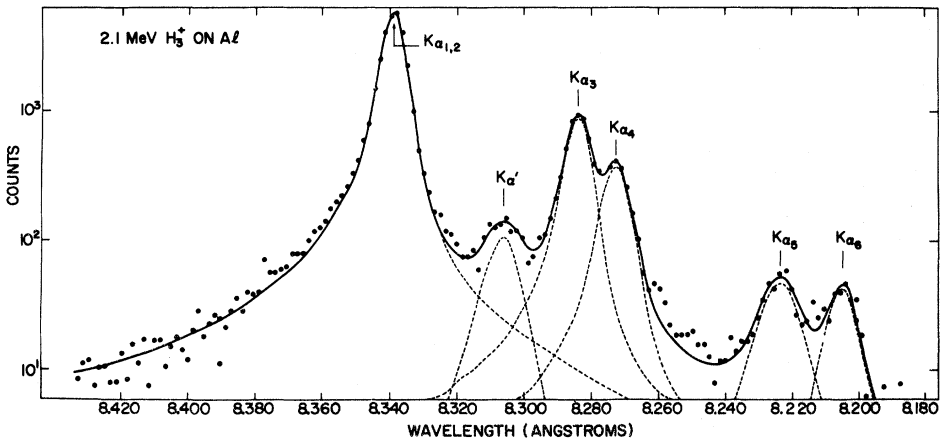

FIG. 1. Al $K\alpha$ x-ray spectrum excited by 2.1-MeV H_3^+ with a linear background subtracted out. Dashed curves, results of a line-shape fit for each peak as described in text; sum of the fitted peaks shown as a solid curve.

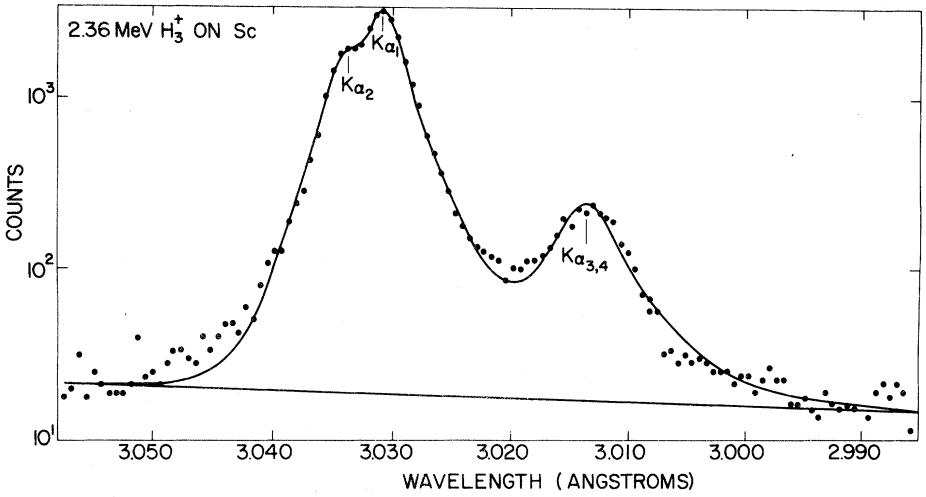

FIG. 2. Sc $K\alpha$ x-ray spectrum induced by 2.36-MeV H_3^+. The solid curve represents the results of the least-squares fit as described in the text.

shape from the 11-keV electron-excited Al $K\alpha_{1,2}$ line was used with the position, width, and height as adjustable parameters. The remainder were fitted with an analytical line shape consisting of a Lorentzian function attached with asymmetric exponential tails. As can be seen from Figs. 1 and 2, the fits are good. Entered in Table I are the relative energies of the KL satellites extracted from the fitting procedure. The yield ratios $I_{K,1L}/I_{K,0L}$ formed from the extracted areas are also tabulated in Table I and displayed in Fig. 3. Since the background in all cases is relatively small, the errors are dominated by the peak shape uncertainties. Based on repeated data runs, the errors in the intensity ratios are estimated to be ~6%. No corrections for the various absorptions and crystal reflectivity were made since the corresponding energy difference is small and such corrections would essentially cancel out in the ratio. In Fig. 3, the intensity ratios at $V_L = 2.1$ have also been scaled up to the corresponding ratios at $V_L = 1$, based on the reasonable assumption[6] that the V_L dependence of these ratios is the same for all Z, the scaling factor being determined from the two measurements on Ca.

IV. DISCUSSION

Our main interest is in the intensity ratios, and therefore no detailed consideration was given to the observed satellite energies except to note that these relative energies are in reasonable over-all agreement with the observed trend.[9] Although chemical bonding effects in the $K\alpha$ x-ray satellites excited by light ions have been reported[10] for Al and Si, they are manifested mainly in terms of the energy shifts. The observed effects on the intensity ratio $I_{K,1L}/I_{K,0L}$ are minimal, perhaps reflecting their near cancellation in the ratio. Further, the effects are expected to diminish with increasing Z. Thus the ratios measured here with compounds are expected to reflect the corresponding elemental cases.

The treatment of vacancy-rearrangement effects on the x-ray yields corresponding to KL^n vacancies can be simplified by considering only the L_2 and

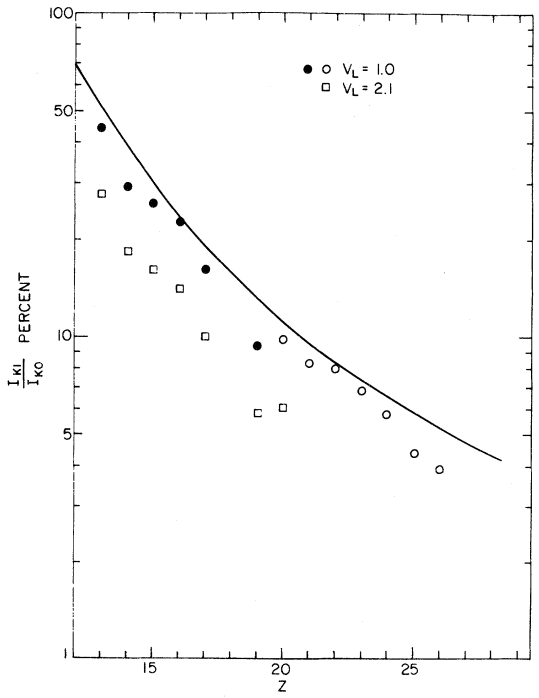

FIG. 3. $K\alpha$ satellite intensity ratio vs Z. The relative error for each V_L is estimated to be ~6%. Solid circles, data at $V_L = 2.1$ which have been scaled up to correspond to $V_L = 1$ (see text). Solid curve, predictions of Coulomb ionization without rearrangement effects.

L_3 subshells and not distinguishing these, and assuming that the vacancies are in dynamical equilibrium. Vacancies in the L_1 subshell can be incorporated but will only add complications to the equations with no improvement on the clarity. Using ω_n, σ_n^X, and σ_n^V to denote, respectively, the fluorescence yield, K x-ray, and primary vacancy-production cross sections corresponding to the KL^n configuration, the relation between the cross sections can be written as

$$\sigma_n^X = \omega_n (\sigma_n^V + \sigma_n^T)(1 - T_{n-1,n}). \quad (1)$$

Here T_{mn} $(0 \leq m < n)$ expresses the probability that the KL^n configuration is transformed into the KL^m configuration prior to a K x-ray transition. Only $m = n - 1$ is expected to be significant, since the probability of simultaneously filling two or more L holes is small. The term σ_n^T arises from the rearrangement of the KL^{n+1} configuration, and is given by

$$\sigma_n^T = T_{n,n+1}(\sigma_{n+1}^V + \sigma_{n+1}^T). \quad (2)$$

For the case of a single K vacancy $(n = 0)$, there is no vacancy rearrangement and the term inside the right-most parentheses in Eq. (1) should therefore be set to unity. We see from Eqs. (1) and (2) that the x-ray satellite intensity is not proportional to the primary vacancy cross section. Indeed, in heavy ion bombardment, where the K x-ray intensities are observed[11, 12] to peak at about the KL^2 or KL^3 satellite, the cascading contributions in the σ_n^T term can introduce substantial distortion if the T_{mn} are not negligibly small. Thus the interpretation of x-ray satellite intensities in terms of the primary vacancy cross sections must be exercised with care.

For proton impact, the largest number of vacancies is seen here to correspond at most to the KL^2 defect configuration, with an intensity of $\lesssim 0.1$ times $I_{K,1L}$. For the purpose of comparison with the present experimental results, we write the x-ray cross section for KL vacancies relative to that for a single K vacancy as

$$\frac{\sigma_1^X}{\sigma_0^X} = \frac{1}{A_{KL}} \frac{\omega_1}{\omega_0} \frac{\sigma_1^V}{\sigma_0^V}, \quad (3)$$

where the A_{KL} will be termed the "rearrangement correction factor." This factor has the explicit form

$$A_{KL} = \frac{1 + T_{01}\sigma_1^V/\sigma_0^V + T_{01}T_{12}\sigma_2^V/\sigma_0^V}{(1 - T_{01})(1 + T_{12}\sigma_2^V/\sigma_1^V)}. \quad (4)$$

In view of the over-all successes of Coulomb ionization theories in accounting for the systematics of proton-induced single-vacancy-production cross sections[11] and the fact that the satellite intensities are relatively small, as observed here and elsewhere[6, 7], multiple-Coulomb ionization is expected to be the principal mechanism for primary vacancy productions. Since the ω_1/ω_0 ratios are close to unity and, moreover, can be estimated with reasonable accuracy, comparisons of the observed satellite intensity ratios with the theoretical σ_1^V/σ_0^V would enable us to extract the A_{KL} factors. Although the magnitudes of the extracted A_{KL} will depend on the theory used to calculate σ_1^V/σ_0^V, the experimental A_{KL} are expected to reflect the actual Z dependence. The reason for this follows from the observation that the probability of double ionization is relatively small, and this results[13] in an expression for the σ_1^V/σ_0^V which is nearly proportional to the cross section σ_L for single-L ionization, and all simple Coulomb ionization theories predict similar Z dependence for σ_L at a fixed scaled velocity.

The ω_1/ω_0 ratios used in the present analysis were estimated following the prescription of Larkins[14] and using the transition rates of McGuire.[15] As can be seen in Table I, they are close to unity. Possible simultaneous outer-shell vacancies, whose number is expected not to differ for KL and K vacancies, do not alter significantly these calculated ratios and further may be filled prior to the K x-ray emission, since solid targets were used. For Al and Ar where Hartree-Fock-Slater (HFS) calculations[12, 16] are available for comparisons, the HFS ratios for both $1s^{-1}2p^{-1}$ and $1s^{-1}2s^{-1}$ configurations differ from the Larkin's values by no more than 10%.

Multiple-Coulomb ionization theories based on impact-parameter formulation have been described by several authors.[13, 17, 18] Since the probability for double ionization is relatively small for the cases considered here, Eqs. (34) and (35) in McGuire and Richard[13] would provide adequate estimates of the primary vacancy cross-section ratios for our purpose. The calculated σ_1^V/σ_0^V using weighted-averaged L-shell binding energies are tabulated in Table I. The products $(\sigma_1^V/\sigma_0^V)(\omega_1/\omega_0)$ are shown as the solid curve in Fig. 3 and are seen to simulate the Z dependence of the observed intensity ratios. In conjunction with Eq. (3) and the over-all data at $V_L = 1$, a set of empirical rearrangement factors A_{KL} can be extracted, and these are shown in Fig. 4. As indicated earlier, not much weight should be attached to the absolute magnitudes, but the over-all Z dependence is expected to be significant.

For the purpose of comparison, A_{KL} has been calculated from Eq. (4) using the assumption that the level widths are independent of the number of

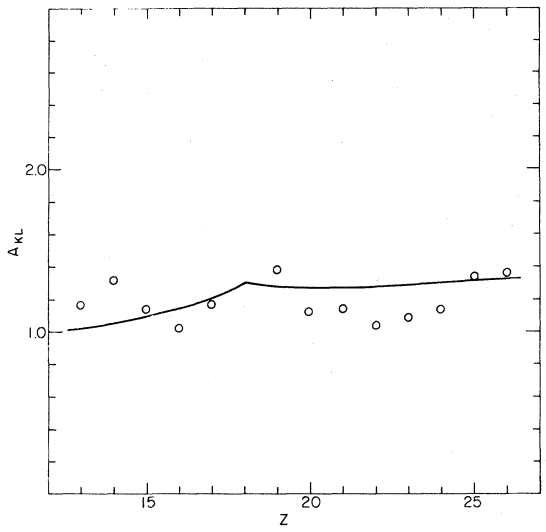

FIG. 4. Empirical rearrangement correction factor displayed as a function of Z. The relative errors are estimated to be ~10% for $Z \leq 19$ and ~6% for $Z \geq 20$. The solid curve represents the values obtained from Eq. (5).

L vacancies, i.e., $T_{12} = T_{01} = \Gamma_L/(\Gamma_L + \Gamma_K)$, where the Γ_L and Γ_K are the corresponding level widths of the atom. Γ_L is the average L-shell width and may be taken to be Γ_{L_3}, since both Γ_{L_1} and Γ_{L_2} include Coster-Kronig contributions to the L_3 subshell where such transitions are energetically possible. Using the transition rates of McGuire,[15] the T_{01} value ranges from 0.015 ($Z = 13$) to 0.242 ($Z = 26$), while the corresponding σ_1^V/σ_0^V ratio ranges from 0.48 to 0.05. Since the simple ionization theory[13] gives $\sigma_2^V/\sigma_1^V = \frac{7}{16}(\sigma_1^V/\sigma_0^V)$ and $\sigma_2^V/\sigma_0^V \simeq \frac{1}{2}(\sigma_1^V/\sigma_0^V)^2$, Eq. (4) is approximately

$$A_{KL} \simeq \frac{1}{1-T_{01}} = \frac{\Gamma_K + \Gamma_L}{\Gamma_K} . \qquad (5)$$

This same expression has been given by Li *et al.*,[4] and they showed that the value calculated for a particular Z can differ by as much as a factor of 2 depending on the set of level widths used. In Fig. 4, the solid curve represents the estimated A_{KL} using the widths of McGuire.[15] Based on the errors in the intensity ratios, the relative errors in the extracted A_{KL} are ~10% for $Z \leq 19$ and ~6% for $Z \geq 20$. Although the calculated value differs from the extracted value in some cases by as much as 21%, the over-all agreement is reasonable, particularly in view of the fact that average parameters have been used for the calculations of σ_1^V/σ_0^V and A_{KL} with no adjustment on the parameters.

V. SUMMARY

Accurate estimates of the vacancy-rearrangement effects would require a major undertaking of the detailed calculations of all possible transition rates corresponding to a given defect configuration. In many instances of interest, the exact configuration may not even be known. For simple defect configurations, atomic level widths have been used to estimate[2-5] the rearrangement corrections in x-ray measurements. These corrections have been shown[4] to depend on the particular set of level widths used. Based on the premise that multiple-Coulomb ionization is the primary vacancy-producing mechanism, we have obtained a set of empirical rearrangement factors from the measurements of $I_{K,1L}/I_{K,01}$ ratios with proton impact at a fixed V_L. These empirical factors are found to be in close agreement with the simple estimates based on the use of atomic level widths from McGuire.[15] For larger defect configurations, the use of more realistic level widths may be necessary for the estimates of rearrangement effects.

ACKNOWLEDGMENTS

We thank M. Haas and E. Wolin for their assistance in the data reductions. One of us (N.C.) would like to acknowledge the support provided by the Research Corporation whose Cottrell Grants made possible the acquisition of the spectrometer used in this study.

[1] P. Richard, R. L. Kauffman, J. H. McGuire, C. F. Moore, and D. Olsen, Phys. Rev. A 8, 1369 (1973).
[2] V. P. Sachenko and V. F. Demekhin, Zh. Eksp. Teor. Fiz 49, 765 (1965) [Sov. Phys.—JETP 22, 532 (1966)].
[3] T. Aberg, Ann. Acad. Sci. Fenn. A 6, 308 (1969).
[4] T. K. Li, R. L. Watson, and J. S. Hansen, Phys. Rev. A 8, 1258 (1973).
[5] R. L. Watson, F. E. Jenson, and T. Chiao, Phys. Rev. A 10, 1230 (1974).
[6] N. Cue, V. Dutkiewicz, P. Sen, and H. Bakhru, Phys. Lett. 46A, 151 (1973).
[7] R. Kauffman, J. H. McGuire, P. Richard, and C. F. Moore, Phys. Rev. A 8, 1233 (1973).
[8] L. Parratt, Phys. Rev. 50, 1 (1936).
[9] See, e.g., R. L. Kauffman and P. Richard, in *Methods of Experimental Physics: Spectroscopy*, edited by D. Williams (Academic, New York, 1975).
[10] P. G. Burkhalter, A. R. Knudson, D. J. Nagel, and F. L. Dunning, Phys. Rev. A 6, 2093 (1972); J. McWherter, D. F. Olsen, H. H. Wolter, and C. F. Moore, Phys. Rev. A 10, 200 (1974).
[11] J. D. Garcia, R. J. Fortner, and T. M. Kavanagh,

Rev. Mod. Phys. 45, 111 (1973).
[12]F. Hopkins, D. O. Elliott, C. P. Bhalla, and P. Richard, Phys. Rev. A 8, 2952 (1973).
[13]J. H. McGuire and P. Richard, Phys. Rev. A 8, 1374 (1973).
[14]F. P. Larkins, J. Phys. B 4, L29 (1971).
[15]E. J. McGuire, Phys. Rev. A 2, 273 (1970); 3, 587 (1971).
[16]C. P. Bhalla, Phys. Rev. A 8, 2877 (1973).
[17]J. Bang and J. M. Hansteen, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 31, 13 (1959); J. M. Hansteen and O. P. Mosebekk, Z. Phys. 234, 281 (1970).
[18]J. S. Hansen, Phys. Rev. A 8, 822 (1973).