PHYSICAL REVIEW A VOLUME 14, NUMBER 6 DECEMBER 1976

United-atom limits for molecular-orbital continua and the characteristics of broadening beyond the limit

George H. Pepper* and Louis Brown
Department of Terrestrial Magnetism, Carnegie Institution of Washington, Washington, D. C. 20015
(Received 12 July 1976)

In collisions of 0.2–3.0-MeV Na^+ with thick targets of Ti through Zn, relatively flat molecular-orbital continua are observed that extend from the characteristic separated-atom lines to the united-atom L-shell limits. The broadening of these continua beyond their respective united-atom limits is essentially target independent and can, to a good approximation, be fitted with half-Gaussians characterized by widths proportional to $E^{1/4}$, where E is the energy of the incident ion. Similar continua are observed for Na^+-target combinations that have as their limit the united-atom M shell.

I. INTRODUCTION

In collisions of heavy ions wherein both target and projectile have inner electronic shells that interpenetrate, numerous phenomena have been explained through the proposal[1] that during the collision atomic orbitals of the separated atoms transform through molecular orbitals into atomic orbitals of a united atom. The occurrence of continuum radiation extending to energies well beyond characteristic lines, and not explainable as bremsstrahlung, is generally attributed to the filling of vacancies in these molecular orbitals.[2] In this paper we are concerned with two features of this molecular-orbital continuum: (1) the existence and location of a united-atom limit and (2) the characteristics of the spectral broadening beyond this limit. First we discuss previous work on continua originating from molecular orbitals extending to the united-atom K shell (K-band continua), L shell (L band), and M shell (M band).

The initial expectation[3] that K-band spectra would show distinct limits given by $K\alpha, \beta$ transition energies of the united atoms has not yet been realized. This is not to imply that K-band continua are not at least partially of molecular origin.[4] Part of the evidence for this is to be found in theoretical predictions for the shapes of the K-band continua.[5,6,7] However, although these predictions tend to fit experimental spectra reasonably well, there have been papers recently presented on coincidence spectra[8] and on primary bremsstrahlung processes,[9] which in certain cases have raised doubts as to the conclusiveness of this agreement between theory and experiment.

Just as with K-band spectra, L-band spectra previously reported[10-13] have not exhibited distinct limits. Although these limits may well have been present, they have been obscured by rather severe attenuation or efficiency problems,[10] by the presence of characteristic lines from projectile, target, or impurities,[11] and by the possibility of a contribution from primary bremsstrahlung in high-energy, heavy-ion bombardment.[12,13]

Only for M-band continua have united-atom limits definitely been observed.[13-15] These continua, resulting from bombardment of high-Z targets (Sn, Yb, Ta, Au, Pb, Bi, Th, U) with equally heavy projectives (Xe, I, Au), show distinct peaking at energies corresponding to $M\alpha, \beta$ transitions in the united atoms. However, this peaking, which is a distinctive feature of the united-atom limit in these spectra, disappears with increasing ion energy until near 0.3 MeV/amu these M-band continua have the same exponential shape as K-band continua. Coincidence experiments in this region[16] show the M-band continuum to have the same shape in the coincidence as in the singles spectrum, a condition not consistent with theoretical concepts developed for K-band continua.[5,6] A further complication arises at ion energies above the threshold for production of characteristic L x rays from the heavy collision partner, as these lines obscure the shape of the continuum.

Investigations of the characteristics of the spectral broadening beyond the united-atom limit have not been carried out for the L-band, and only limited characteristics can be inferred from M-band spectra previously reported.[13-15] Specifically, the broadening increases with increasing projectile velocity[14] and appears insensitive to changes in target. The velocity dependence is similar to observations of K-band continua, but the target dependence is considerably less.

Theoretical predictions of the shape of this broadening beyond the limit have only been made for K-band continua.[5,6] These calculations attribute the shape entirely to the rapid energy variation of the molecular orbitals during the collision and result in an exponential shape for the broadened spectra. A simple formula, derived from these theoretical calculations, gives the velocity and Z dependence of the half-width of this exponential as[17,18]

$$H = \frac{1}{3}\left(\hbar v \frac{I_U - I_H}{R_H + R_L}\right)^{1/2}, \qquad (1)$$

where v is the incident-ion velocity, I_U (I_H) is the binding energy of the more tightly bound level of the x-ray transition in the united atom (heavy collision partner), and R_H (R_L) is the hydrogenic radius of the heavy (light) collision partner. Considering the assumptions in this formula it is remarkable that it predicts within 25% the half-widths of most available K-band continua and is only off by 35% in the few other cases[17] (cf. Table I). These data represent half-widths ranging from 50 eV to over 6 keV, from C+C to I+I. Variations of this formula have met with similar success over a more limited range of data.[5,18] Were it not for some troubling assumptions in the derivation and the lack of consideration for primary bremsstrahlung processes,[9] the success of these simple formulas would seem to uphold the validity of the underlying assumptions of the theoretical treatments from which they are derived.[5,6] An analysis of these assumptions is not within the scope of this paper, but we shall present data that will indicate that another approach may be in order.

II. EXPERIMENT

We accelerated beams of Na^+ from the Carnegie Van de Graaff onto thick targets using techniques described elsewhere.[11] Changes in the original apparatus were a Si(Li) detector with a 0.008-mm beryllium window and an amplifier with pileup rejection and baseline restoration. The only x-ray attenuation was by the Be window and a 7.6-μg/cm^2 gold contact layer; attenuation by the Si dead layer was insignificant.

TABLE I. Exponential half-widths for K-band continua read primarily from published experimental spectra (H_{expt}) and half-widths calculated using Eq. (1) in the text (H). The projectile (target) is Z_1(Z_2) and E_1 is the projectile energy.

Z_1	Z_2	E_1 (MeV)	H_{expt} (keV)	H (keV)	Ref.	Z_1	Z_2	E_1 (MeV)	H_{expt} (keV)	H (keV)	Ref.
53	53	82	10	7.3	a	13	13	5	0.50	0.54	
36	40	200	5.6	5.6	b			2.5	0.38	0.46	
35	40	60	5.8	4.1	a, c	16	10	48	0.84	0.82	i
35	35	60	4.6	3.7	a, c	10	14	1.9	0.55	0.38	j
32	32	81	4.6	3.6	d			0.9	0.43	0.31	
35	28	57	3.1	3.0	c			0.7	0.38	0.29	
35	26	30	2.6	2.4	a			0.5	0.35	0.27	
28	28	95	2.7	3.2	e			0.3	0.29	0.24	
		70	2.5	2.9		9	12	40	0.74	0.68	k
		40	2.1	2.5				20	0.55	0.57	
		20	1.6	2.1				10	0.42	0.48	
35	22	60	3.0	2.4	a			6	0.40	0.42	
16	13	95	1.1	1.2	f			4	0.33	0.38	
		90	1.1	1.2				3	0.33	0.35	
		55	0.96	1.1				2	0.28	0.32	
		32	0.74	0.95		8	13	8	0.43	0.44	h
		16	0.71	0.80		16	6	32	0.50	0.46	f
		7	0.55	0.65				16	0.45	0.38	
14	14	2	0.66	0.48	g	6	6	0.22	0.099	0.089	l
13	13	20	0.82	0.77	h			0.050	0.073	0.062	
		10	0.68	0.65				0.019	0.050	0.048	

[a] Reference 7.
[b] W. E. Meyerhof, T. K. Saylor, and R. Anholt, Phys. Rev. A 12, 2641 (1975).
[c] C. K. Davis and J. S. Greenberg, Phys. Rev. Lett. 32, 1215 (1974).
[d] P. Gippner, K.-H. Kaun, F. Stary, W. Schulze, and Yu. P. Tretyakov, Nucl. Phys. A 230, 509 (1974).
[e] P. Vincent, J. S. Greenberg, and H. P. Trautretter, Bull. Am. Phys. Soc. 21, 32 (1976).
[f] Reference 18.
[g] Ch. Heitz, J. Phys. B 8, L271 (1975).
[h] R. Laubert, H. H. Hazelton, J. R. Mowat, R. S. Peterson, and I. A. Sellin, Phys. Rev. A 11, 1468 (1975).
[i] F. Bell, H.-D. Betz, H. Panke, E. Spindler, W. Stehling, and M. Kleber, Phys. Rev. Lett. 35, 841 (1975).
[j] B. Knaf and G. Presser, Phys. Lett 49A, 89 (1974).
[k] G. H. Pepper and T. J. Gray (unpublished).
[l] J. R. Macdonald, *et al.*, in Ref. 2.

Spectra of thick-target absolute yields for 1-MeV Na^+ on Ti, Fe, and Zn (L band) and on Pb and Sn (M band) are shown in Fig. 1. Both groups of spectra have been normalized to the yields at their respective united-atom limits. The L-band spectra are shown with the united-atom limits indicated by vertical arrows. The Z dependence is clear, as the positions of the shoulders of the continua follow the united-atom limits precisely. This is in contrast to K-band continua, which show only an exponential decrease in intensity (cf. Ref. 18), and is similar to the Z dependence

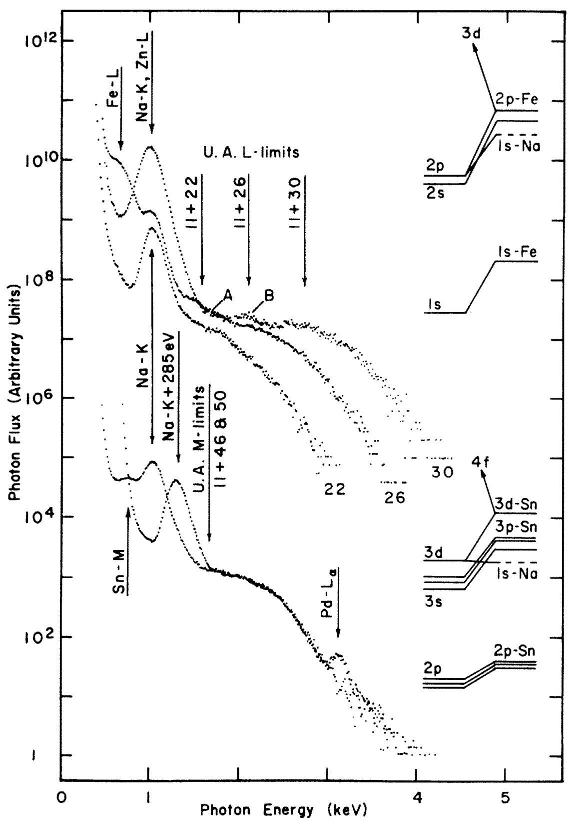

FIG. 1. Target Z dependence of the molecular-orbital continuum. All spectra are efficiency-corrected thick-target yields produced by 1-MeV Na^+ and observed with a Si(Li) detector having a 0.008-mm beryllium window. The upper three spectra are from Ti, Fe, and Zn. United-atom L-shell limits ($2s$ united-atom binding energies) are indicated by vertical arrows and the three spectra are normalized to the yields at these photon energies. The lower two are from Pd and Sn; the united-atom M-shell limits ($3s$ united-atom binding energies) are shown as a vertical arrow with the Pd spectrum shifted 285 eV so that the two limits coincide. Normalization at this point shows the two spectra are identical beyond their limits except for their characteristic separated-atom lines. Counting rates were held below 100/sec, but pileup is still observed for Fe at A and Zn at B, owing to the high intensity of their L lines.

observed for M-band continua[13] although without the peaking near the limit that characterizes these continua at the lowest projectile energies.[15]

The M-band spectra in Fig. 1 are presented with the Pd spectrum shifted 285 eV so that the united-atom limits of the two continua coincide. This method of presentation is used in order to illustrate the Z dependence of the broadening beyond the limit. It is observed that for a given projectile energy the shapes of the two continua beyond their united-atom limits are identical. This same procedure yields identical results when applied to the L-band continua in Fig. 1. One must then conclude that, within the statistics of the data, there is no Z dependence of the broadening for this series of projectile-target combinations. This is the same dependence observed for superheavy M-band continua[13,15] and is in opposition to both theory and experiment for K-band continua.

It is appropriate at this point to discuss the meaning of the term united-atom limit as used in Fig. 1. The exact location of this limit is uncertain because of three circumstances: (1) There are several L x-ray lines that span a range of energies. (2) This range may be extended owing to the presence of multiple vacancies. (3) The possible changes in populations and methods of depopulating levels due to molecular effects can shift the centroid of the L x-ray distribution. We have determined experimentally that the position we choose for the limit does not affect the results of this experiment as long as it is chosen between the $L\alpha(3d\text{-}2p)$ transition energy and the $2s$ binding energy in the united atom. The $2s$ binding energies are chosen for convenience as upper limits for the united-atom limits in Fig. 1. Likewise, the $3s$ binding energies are used as limits for the M-band data in Fig. 1.

The continuum observed from $Na^+ + Ni$ has the largest cross section in the range of this experiment and it has no complicating structure from characteristic lines near its shoulder. For these reasons it was chosen for a detailed study of the velocity dependence of the broadening beyond the limit. Figure 2 shows spectra from Na^+ incident on Ni at 0.5, 1.5, and 2.5 MeV. Since it is quite obvious from Figs. 1 and 2 that the shape of the spectra beyond the united-atom limit is not exponential, as proposed in theoretical predictions for K-band continua,[5,6] we have chosen to fit this broadened part of the continuum with half of a Gaussian. This choice is somewhat arbitrary, but a consideration of the broadening from the point of view of the uncertainty principle makes Gaussian broadening at least plausible, if not mandatory, particularly as opposed to the use of exponential broadening. To take into account the

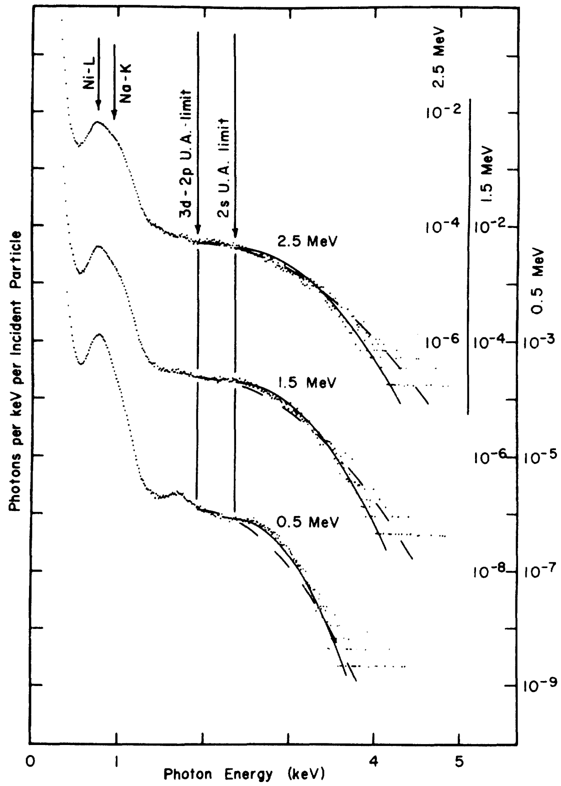

FIG. 2. Velocity dependence of the broadening beyond the united-atom limit for 0.5-, 1.5-, and 2.5-MeV Na^+ incident on thick Ni targets. The dashed and solid curves were calculated from Eq. (2) in the text using yield values given in Fig. 3 and by curves B and C in Fig. 4.

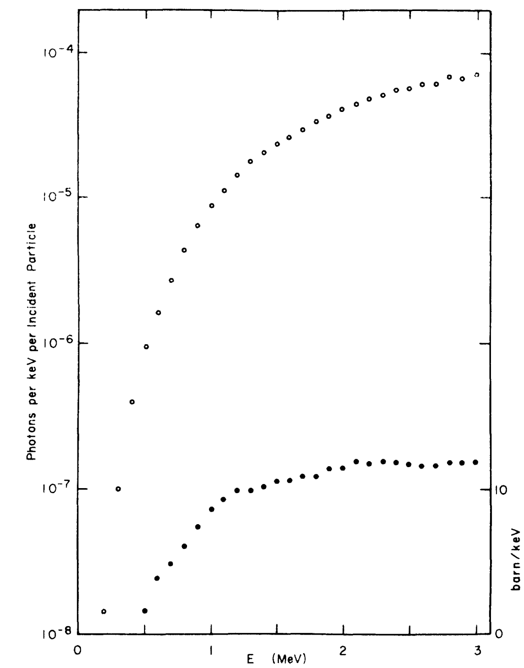

FIG. 3. Thick-target yields of the continuum at the united-atom limit for Na^+ incident on Ni, and the cross sections derived from these yields, plotted as functions of the projectile energy.

complications of the detector resolution, we assume that for photons of energy ϵ greater than the united-atom limit, ϵ_0, the width of the Gaussian spectrum at a given projectile energy is $W^2 = W_d^2 + W_{ua}^2$, where W_d is the detector width (FWHM $= 2.355W_d$) and W_{ua}^2 is the Gaussian width of the broadening beyond the limit. The detector width for our system is $W_d = 66.4[1.0 + 0.120\epsilon(\text{keV})]$. The shape of the thick-target spectrum beyond the limit ϵ_0 for a projectile of energy E_N is assumed approximated by

$$\mathcal{Y}(\epsilon, E_N) = \frac{1}{Y(E_N)} \sum_{i=1}^{N} [Y(E_i) - Y(E_{i-1})] \times \exp\left(\frac{-(\epsilon - \epsilon_0)^2}{2W^2(E_i)}\right), \qquad (2)$$

where $Y(E_i)$ is the measured thick-target yield for a small portion of the continuum at photon energies just below ϵ_0. We take $Y(E_0) = 0$, where E_0 (< 200 keV) is the threshold energy for the continuum. Measurements of $Y(E_i)$ in 0.1-MeV steps are shown in Fig. 3 along with the cross sections deduced from these yields.

Having decided on the functional form for fitting the broadening beyond the limit, we must choose the exact position of the united-atom limit. The dots in Fig. 4 are two plots of $W(E_N)$ obtained from the data, each for an assumed value of ϵ_0. The values of $W(E_N)$ were adjusted until the calculated values of $\mathcal{Y}(\epsilon, E_N)$ gave the best fit by eye to the data. Width parameters for energies below 0.5 MeV were erratic owing to the nature of the analysis (Eq. 2), poor statistics, and the proximity of threshold and are not plotted in Fig. 4. In Fig. 2 the $2s$ energy of the united atom $Z_1 + Z_2$ has been indicated, and it is this limit to which the lower curve of points in Fig. 4 corresponds. Transitions $3d$-$2p$ ($L\alpha$) are the most probable for a normal atom, and one can fit half-Gaussians to the data nearly as well using this limit; the upper curve of points in Fig. 4 corresponds to this $3d$-$2p$ limit, which is also noted in Fig. 2. Curves B and C in Fig. 4 correspond to parametrizations of the dotted curves by functions proportional to $E^{1/4}$, chosen when an $E^{1/2}$ dependence clearly failed, as shown by curve A in Fig. 4. This is not to suggest that other parametrizations would not fit as well (e.g., a form such as $a + bE^{1/2}$) but only that $E^{1/4}$ was the simplest form and one suggested by the success of the $E^{1/4}$ dependence of

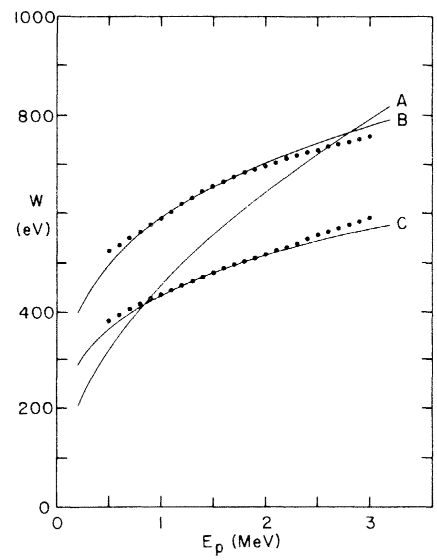

FIG. 4. Gaussian widths of the broadening beyond the united-atom limits for $Na^+ + Ni$ as functions of projectile energy. The dotted curves result from best fits by eye to the continua obtained by varying the width parameters in Eq. 1 in the text. The solid curves B and C, which are proportional to $E^{1/4}$, were adjusted to give reasonably close fits to the dotted curves. Curve A is representative of fits proportional to $E^{1/2}$.

K-band continua.

The widths W given by curves B and C in Fig. 4 are used in Eq. 1 to obtain the dashed and solid curves in Fig. 2. The half-Gaussians using the $2s$ limit (solid curves) fit better at lower projectile energies and those using the $3d$-$2p$ limit (dashed curves) fit better at higher energies. The worst fits are at low energies using the $3d$-$2p$ limit. These results show that considerable improvement in fit would result if a limit were assumed that tended toward lower photon energies with increasing projectile energy.

III. CONCLUSIONS

We have observed continuum radiation that we believe to be the clearest evidence yet reported for being characteristic of the formation of the united atom itself. We have shown that the united-atom limit, though ill defined, does exist and is roughly bounded by the $3d$-$2p$ transition energy on the low-energy side and the $2s$ binding energy on the high side. We have demonstrated that the broadening beyond the united-atom limit is clearly not exponential and can be represented by half of a Gaussian having a width proportional to $E^{1/4}$. We have also determined, within the projectile-target limits of this experiment, that the broadening beyond the united-atom limit is not a function of Z.

What is needed now is a correlation of the characteristics of the various continua under one cogent treatment of quasimolecules formed during heavy-ion collisions. Failing this, the next step would seem at least a resolution of the contradiction between exponential broadening for K-band continua and Gaussian broadening for L- and M-band continua.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the value of our stimulating discussions with J. Macek, J. Rafelski, and T. K. Saylor.

---

*Present address: Department of Physics and Astronomy, University of Kentucky, Lexington, Ky. 40506.

[1] U. Fano and W. Lichten, Phys. Rev. Lett. 14, 627 (1965); W. Lichten, Phys. Rev. 164, 131 (1967); M. Barat and W. Lichten, Phys. Rev. A 6, 211 (1972).

[2] F. W. Saris, W. F. van der Weg, H. Tawara, and R. Laubert, Phys. Rev. Lett. 28, 717 (1972); P. H. Mokler, H. J. Stein, and P. Armbruster, Phys. Rev. Lett. 29, 827 (1972); J. R. Macdonald, M. D. Brown, and T. Chiao, Phys. Rev. Lett. 30, 471 (1973).

[3] W. E. Meyerhof, T. K. Saylor, S. M. Lazarus, W. A. Little, B. B. Triplett, and L. F. Chase, Jr., Phys. Rev. Lett. 30, 1279 (1973).

[4] W. E. Meyerhof, in *The Physics of Electronic and Atomic Collisions IX*, edited by J. S. Risley and R. R. Geballe (University of Washington Press, Seattle, 1975), p. 470.

[5] B. Müller, in Ref. 4, p. 481.

[6] J. H. Macek and J. S. Briggs, J. Phys. B 7, 1312 (1974).

[7] W. E. Meyerhof, T. K. Saylor, S. M. Lazarus, A. Little, B. B. Triplett, L. F. Chase, Jr., and R. Anholt, Phys. Rev. Lett. 32, 1279 (1974).

[8] I. Tserruya, H. Schmidt-Böcking, R. Schulê, K. Bethge, R. Schuch, and H. J. Specht, Phys. Rev. Lett. 36, 1451 (1976).

[9] R. Anholt and T. K. Saylor, Phys. Lett. 56A, 455 (1976).

[10] F. W. Saris *et al.*, in Ref. 2; F. W. Saris, C. Foster, A. Langenberg, and J. van Eck, J. Phys. B 7, 1494 (1974); G. Bissinger and L. C. Feldman, Phys. Rev. Lett. 32, 1 (1974); A. Lurio, J. A. Cairns, J. F. Ziegler, and J. Macek, Phys. Rev. A 12, 498 (1975).

[11] L. Brown, G. E. Assousa, N. Thonnard, H. A. van Rinsvelt, and C. K. Kumar, Phys. Rev. A 12, 425 (1975).

[12] W. Wölfli, Ch. Stoller, G. Bonani, M. Suter, and M. Stöckli, Lett. Nuovo Cimento 14, 577 (1975); W. R. Stott and J. C. Waddington, Phys. Lett. 56A, 258 (1976).

[13]P. H. Mokler, S. Hagmann, P. Armbruster, G. Kraft, H. J. Stein, K. Rashid, and B. Fricke, in *Atomic Physics 4*, edited by G. zu Putlitz (Heidelberg U.P., Heidelberg, 1974), p. 301.
[14]P. Armbruster, G. Kraft, P. Mokler, B. Fricke, H. J. Stein, Phys. Scr. 10A, 175 (1974).
[15]F. C. Jundt, H. Kubo, and H. E. Gove, Phys. Rev. A 10, 1053 (1974); H. O. Lutz, W. R. McMurray, R. Pretorius, I. J. van Heerden, R. J. van Reenen, and B. Fricke, J. Phys. B 9, 1157 (1976).
[16]F. Jundt, G. Guillaume, P. Fintz, and K. W. Jones, Phys. Rev. A 13, 563 (1976).
[17]G. H. Pepper and L. Brown, Bull. Am. Phys. Soc. 21, 649 (1976).
[18]H.-D. Betz, F. Bell, H. Panke, W. Stehling, E. Spindler, and M. Kleber, Phys. Rev. Lett. 34, 1256 (1975).