(to be published)].
[7]E. M. Henley and L. Wilets, Phys. Rev. A 14, 1411 (1976).
[8]S. Weinberg, Phys. Rev. Lett. 19, 1264 (1967).
[9]A. Salam, in *Proceedings of the Eighth Nobel Symposium*, edited by N. Svartholm (Almkvist and Wiksell, Stockholm, 1968).
[10]In fact the transition has an E_2 contribution (see Fig. 3). This produces no rotation, but has been taken into account in our calibration, etc.
[11]The details of this and other feedback loops, gating circuitry, etc., will be described elsewhere.
[12]L. L. Lewis *et al.*, preceding Letter [Phys. Rev. Lett. 39, 795 (1977)].

Time-Delayed Li, Be, and B Autoionization Spectra Excited in Low-Energy (200 keV) Single Gas Collisions

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Metastable, autoionizing high-spin states are studied using the ion-beam time-of-flight method in connection with single-collision excitation. High-resolution, time-delayed spectra are obtained from Li, Be, and B excited in 200-keV impact on CH_4. Evidence is found for the $(1s2s2p^2)^5P$ state in B II. Excitation energies for the observed quartet and quintet states are given, and the branching ratio associated with the B II $(1s2s2p^2)^5P$ decay is estimated.

Electron spectroscopy is now established as a powerful technique for the study of ion-atom collision processes.[1,2] However, until now, no time-delayed, electron-emission studies using single gas excitation have been performed. Therefore, excitation mechanisms of metastable, autoionizing high-spin states from fast-moving projectiles have not been investigated in the context of ion-atom collisions. In this Letter, we present for the first time high-resolution, time-delayed Li, Be, and B electron spectra measured under single-collision conditions with ground-state ion beams. It is demonstrated that specific metastable states are strongly populated, and the corresponding autoionizing transitions can be isolated. Therefore, the excitation cross sections and angular dependences of such autoionization lines can be measured. In particular, the nonisotropic population of magnetic substates[3-5] of lithiumlike ^4L and berylliumlike ^5L terms can be studied. We further note that (i) excitation by means of single gas collisions provides high-energy resolution, and (ii) variation of the collisional parameters (e.g., projectile, target, impact energy, observation angle, delay time) makes possible a selective study of specific states such as $(1s2s2p)^4P^\circ$ and $(1s2s2p^2)^5P$. This facilitates a complete line identification of the observed autoionization features. The high resolution provided by this technique permitted analysis of excitation energies, branching ratios, and spectroscopic character of several unusual lines; our results are presented below. Lifetimes and metastable fractions with respect to the single-collision process are topics of the planned extension of this work.

Generally, metastable autoionizing states are accessible for few electron systems from projectiles using foil excitation.[6-13] These beam-foil measurements were primarily limited to studies of electrons ejected at 42.3° to the direction of the incident ion beam. The Oak Ridge group reported delayed electron spectra, using high beam energies and highly stripped heavy ions.[6-8,12] Similar work at low beam energies and for light projectiles (Li, Be, B, and C) were investigated by Bruch and co-workers.[9-11,13] Because of kinematical broadening effects arising from the foil-excitation process, only limited resolution was obtained, especially at low beam energies. Previous direct observations of metastable autoionizing states, produced by single-ion-atom collisions, were performed by Ziem, Bruch, and Stolterfoht[14] and by Stolterfoht *et al.*[15] Ziem, Bruch, and Stolterfoht bombarded Li atoms in the vapor phase with 200-keV He^+ ions, and Stolterfoht and co-workers directed 200-MeV Xe^{31+} onto Ne. In both cases, lithiumlike $(1s2s2p)^4P^\circ$ target states are very likely to be created; however, because the excitation was of the target atom, no time-delayed anisotropic Auger electron emission could be studied.

Using single gas excitation and small observation angles, we have recently shown that high-

resolution projectile-electron spectra can be obtained even for low beam energies.[16] In this connection, we note that our prompt lithium,[17] beryllium,[16,17] and boron[17,18] autoionization spectra have already indicated strong population of the $(1s2s2p)^4P^\circ$ states in single-collision excitation. To suppress the contribution of the promptly decaying projectile states in the observed spectra, we have performed delayed time-of-flight measurements by using single-collision excitation.

The Li, Be, and B ion beams, which were mass and momentum analyzed, were supplied by the 600-keV heavy-ion accelerator at the University of Aarhus. The electrons emitted from the ion beam at angles between 15° and 150° with respect to the ion-beam axis were energy analyzed with a parallel-plate spectrometer (intrinsic resolution 0.5%), which can be rotated. A preretardation system in front of the spectrometer was used to further improve the resolution. The ion beam was excited in a small, differentially pumped gas cell (length 8 mm), the center of which was displaced 60.5 mm away from the center of the spectrometer viewing region. With beam energies of 200 keV, this gives time delays of 9 ± 3, 11 ± 4, and 12 ± 4 nsec for Li, Be, and B, respectively. For Li, no measurable metastable fractions of heliumlike and lithiumlike states are present in the incoming beams.[19,20] We further note that core-excited Be and B metastable states are very unlikely to be created in the ion source. The pressure in the excitation region was kept sufficiently low (typically 10^{-3} Torr) to ensure single-collision conditions. The escape of target gas in the beam direction leads to a small residual pressure in the viewing region of the electron spectrometer. Hence, small contributions from promptly decaying states were present in the delayed spectra. These lines can be used for absolute energy calibration of the time-delayed spectra and the isotropic $(1s2s^2)^2S_{1/2} \rightarrow (1s^2\epsilon s)^2S_{1/2}$ transition can serve as a reference line (autoionization to an s state in the continuum) when measuring angular dependences. Furthermore, CH_4 was chosen as target gas because of its preferential excitation of quartet and quintet states.[16-18]

In Figs. 1–3 are shown representative autoionization spectra from Li, Be, and B, respectively. Figure 1 exhibits the Li autoionization features taken at a time delay of 9 ± 3 nsec. The prominent peaks in this spectrum originate from initial $1s2snl$ and $1s2pnl$ states in Li I. The strongest peak in Fig. 1 centered at ~52.0 eV is attributed to the Li I $(1s2s2p)^4P^\circ \simeq {}^4P^\circ(1)$ term. This interpretation is consistent with the $^4P^\circ(1)$ term energy given by Ziem, Bruch, and Stolterfoht.[14] We note that the $^4P^\circ(1)$ state is metastable against both Coulomb autoionization and electric-dipole transitions,[21] whereas the energetically higher-lying quartet states are depleted mainly by allowed radiative decays to lower-lying quartet states. The peaks lying at about 51 and 53.5 eV can be assigned solely to the initial $(1s2s^2)^2S$ and $1s(2s2p^3P^\circ)^2P^\circ$ states, respectively. These promptly decaying states are produced due to restgas excitation. The features between 54 and 56 eV are attributed to the $1s(2s2p\,^1P^\circ)^2P^\circ$, $(1s2p^2)^4P$, and $(1s2p^2)^2D$ states. The peak at about 58 eV is composed of several closely spaced autoionization lines. Above the $(1s2s)^3S$ series limit

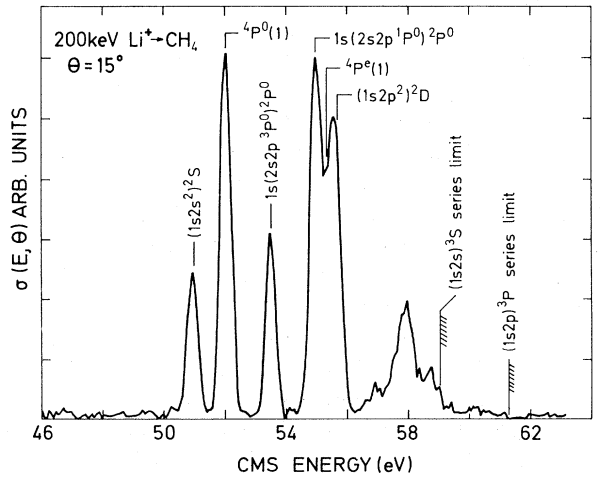

FIG. 1. Li autoionization spectrum measured with delay of 9 ± 3 nsec.

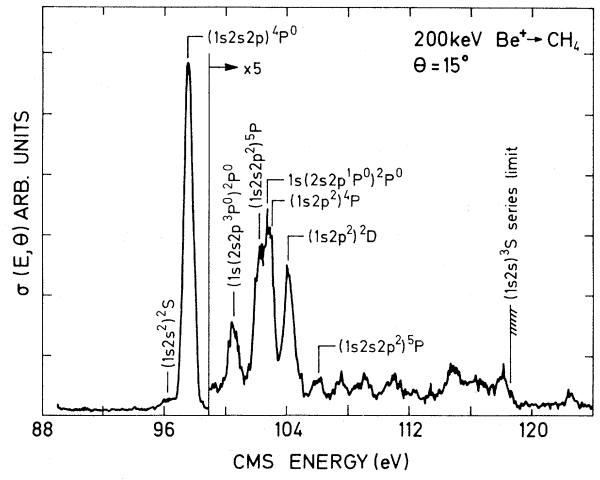

FIG. 2. Be autoionization spectrum measured with delay of 11 ± 4 nsec.

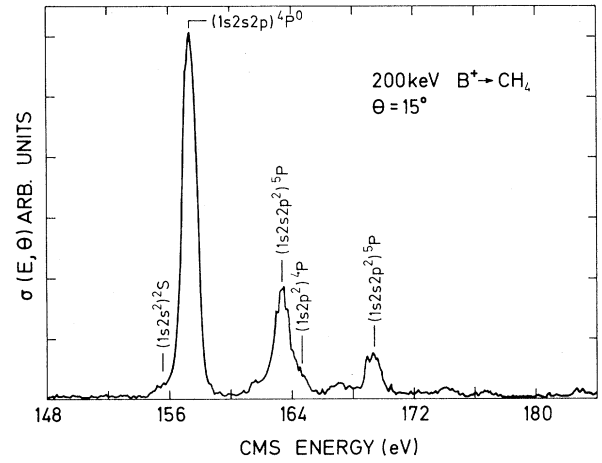

FIG. 3. B autoionization spectrum measured with delay of 12 ± 4 nsec.

(see Fig. 1), no structure was observed.

Results for 200-keV $Be^+ \rightarrow CH_4$ are plotted in Fig. 2. It is seen that the most prominent line is due to the $(1s2s2p)^4P^\circ$ decay in Be II. The feature at about 102.5 eV may contain contributions for $(1s2s2p^2)^5P$, $1s(2s2p^3P^\circ)^2P^\circ$, and $(1s2p^2)^4P$ decays. This is supported by the agreement between experimental line energies and calculated transition energies (see Table I). The small hump at about 106.5 eV is probably due to the initial $(1s2s2p^2)^5P$ term in Be I. Above 106.5 eV, the identification of individual lines is difficult.

Fortunately, the 200-keV $B^+ \rightarrow CH_4$ spectrum, as displayed in Fig. 3, shows well-separated spectral structures so that a complete analysis of the three prominent lines is possible. In this case, the contribution arising from promptly decaying states is small. The most intense line arising at about 157 eV is due to the initial B III $(1s2s2p)^4P^\circ$ states. The spectral feature at about 163 eV is a composite of two lines associated with the decay of B II $(1s2s2p^2)^5P$ and B III $(1s2p^2)^4P$.

The remaining peak in Fig. 3 near 169 eV is attributed to the B II $(1s2s2p^2)^5P$ state decaying to the $(1s^22s)^2S$ final ionic state. We note that the centroid energy of 169.0 ± 0.3 eV agrees well with a recent calculation of Schwarz and co-workers.[22,23] The line identification of the peaks lying between 162 and 170 eV is consistent with the $1s^22p$-$1s^22s$ relative spacing in B III, which is 6.0 eV. From a peak deconvolution of the spectrum, the branching ratio corresponding to the $(1s2s2p^2)^5P$ decay was estimated to be (2.9 ± 0.7):1.

Finally, experimental transition energies and excitation energies associated with the $(1s2s2p)^4P^\circ$ states and the $(1s2s2p^2)^5P$ states for $Z \leq 5$ are collected in Table I together with theoretical values.

TABLE I. Spectroscopic assignment, transition energies, and excitation energies associated with the observed line structures in the delayed Li, Be, and B spectra. The excitation energies are measured from the corresponding atomic or ionic ground state.

Atom	Initial state	Final state	Transition energy (eV)	Excitation energy
Li I	$(1s2s2p)\,^4P^\circ$	$1s^2$	52.04 ±0.05[a]	57.43 ±0.05[a]
			51.993±0.01[b]	57.385±0.010[b]
			52.030±0.004[c]	57.422±0.004[c]
			52.08[d]	57.47[d]
Be II	$(1s2s2p)^4P^\circ$	$1s^2$	97.6 ±0.1[a]	115.8 ± 0.1[a]
			97.79[d]	116.00[d]
B III	$(1s2s2p)^4P^\circ$	$1s^2$	157.0 ±0.3[a]	194.9 ± 0.3[a]
			157.07[d]	195.00[d]
Be I	$(1s2s2p^2)^5P$	$1s^22p$	102.2 ±0.3[a]	
			102.22[e]	
			102.4 ±0.5[f]	
			103.2[g]	
	$(1s2s2p^2)^5P$	$1s^22s$	106.1 ±0.3[a]	115.4 ±0.3[a]
			106.18[e]	115.50[e]
			106.4 ±0.5	115.7 ±0.5[f]
			107.9[g]	116.5[g]
B II	$(1s2s2p^2)^5P$	$1s^22p$	163.0 ±0.3[a]	
			162.96[e]	
			163.2 ±0.5[f]	
			163.7[g]	
	$(1s2s2p^2)^5P$	$1s^22s$	169.0 ±0.3[a]	194.1±0.3[a]
			168.96[e]	194.10[e]
			169.2 ±05[f]	194.3±0.5[f]
			169.7[g]	194.8[g]

[a] This work.
[b] Ziem *et al.*, Ref. 14.
[c] Berry, Refs. 21 and 25.
[d] Holøien and Geltman, Ref. 26.
[e] Schwarz *et al.*, Refs. 22 and 23.
[f] Bruch, Ref. 13.
[g] Safranova *et al.*, Ref. 27.

The principal feature of this work is the observation and identification of metastable, autoionizing lithiumlike quartet and berylliumlike quintet states under single ion-atom conditions. The existence of such long-lived states of high spin and high excitation is of principal importance since as yet, unidentified lines in the optical beam-foil and beam-gas spectra[13,21,24,25] may arise from transitions among quartet, quintet,

and even sextet states.

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[1]M. E. Rudd and J. H. Macek, in *Case Studies in Atomic Physics*, edited by M. R. C. McDowell and E. W. McDaniel (North-Holland, Amsterdam, 1973), Vol. 3, p. 47.

[2]Q. C. Kessel and B. Fastrup, in *Case Studies in Atomic Physics*, edited by M. R. C. McDowell and E. W. McDaniel (North-Holland, Amsterdam, 1973), Vol. 3, p. 137.

[3]B. Cleff and W. Mehlhorn, J. Phys. B 7, 593 (1974).

[4]U. Fano and J. H. Macek, Rev. Mod. Phys. 45, 553 (1973).

[5]J. H. Macek in *Beam-Foil Spectroscopy*, edited by I. A. Sellin and D. J. Pegg (Plenum, New York, 1976), Vol. 2, p. 781.

[6]B. Donally, W. W. Smith, D. J. Pegg, M. Brown, and I. A. Sellin, Phys. Rev. A 4, 122 (1971).

[7]I. A. Sellin, Nucl. Instrum. Methods 110, 477 (1973).

[8]D. J. Pegg, I. A. Sellin, R. Paterson, J. R. Mowat, W. W. Smith, M. D. Brown, and J. R. MacDonald, Phys. Rev. A 8, 1350 (1973).

[9]R. Bruch, G. Paul, J. Andrä, and L. Lipsky, Phys. Rev. A 12, 1808 (1975).

[10]R. Bruch, G. Paul, and J. Andrä, J. Phys. B 8, L253 (1975).

[11]R. Bruch, J. Andrä, and G. Paul, in *Beam-Foil Spectroscopy*, edited by I. A. Sellin and D. J. Pegg (Plenum, New York, 1976), Vol. 1, p. 781.

[12]J. P. Forester, R. S. Peterson, P. M. Griffin, D. J. Pegg, H. H. Haselton, K. H. Liao, I. A. Sellin, R. J. Mowat, and R. S. Thoe, in *Beam-Foil Spectroscopy*, edited by I. A. Sellin and D. J. Pegg (Plenum, New York, 1976), Vol. 1, p. 451.

[13]R. Bruch, thesis, Freie Universität Berlin, West Germany, 1976 (unpublished).

[14]P. Ziem, R. Bruch, and N. Stolterfoht, J. Phys. B 8, L480 (1975).

[15]N. Stolterfoht *et al.*, in *Proceedings of the Fourth Conference on the Scientific and Industrial Applications of Small Accelerators, Denton, Texas, 1976*, edited by G. L. Duggan and I. L. Morgan (IEEE, New York, 1076), and references therein.

[16]M. Rødbro, R. Bruch, and P. Bisgaard, J. Phys. B 10, L275 (1977).

[17]M. Rødbro, R. Bruch, P. Bisgaard, P. Dahl, and B. Fastrup, to be published.

[18]M. Rødbro, R. Bruch, P. Bisgaard, P. Dahl, and B. Fastrup, to be published.

[19]P. Hvelplund, J. Phys. B 9, 1555 (1976).

[20]P. Hvelplund, Dissertation, Institute of Physics, University of Aarhus, Denmark, 1977 (unpublished).

[21]H. G. Berry, Phys. Scr. 12, 5 (1972).

[22]W. H. E. Schwarz, private communication.

[23]R. Bruch, D. Schneider, and W. H. E. Schwarz, to be published.

[24]D. R. Beck and C. A. Nicolaides, Phys. Lett. 61A, 227 (1977).

[25]H. G. Berry, Rep. Progr. Phys. 40, 155 (1977).

[26]E. Holøien and S. Geltman, Phys. Rev. 153, 81 (1967).

[27]U. I. Safranova and V. N. Kharitonova, Opt. Spectrosc. 27, 300 (1967).

Coupling of Spatial and Temporal Coherence in Depolarized Double Scattering

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Measurements of strongly density-dependent Rayleigh linewidth of depolarized doubly scattered light are presented. A new theory based on the coupling between spatial and temporal parts of coherence is proposed to explain our data. In addition, it is shown experimentally that successive application of Van Hove space-time correlation formulation to double scattering is not valid in general.

In this Letter, we report measurements and interpretation of a strongly density-dependent Rayleigh linewidth (Γ_{HV}) of depolarized light scattered in succession by two statistically independent Rayleigh-Gans spheres in Brownian motion. Our experimental data of Γ_{HV} is in direct contrast with the previously reported results[1] and furthermore is not compatible with available existing theories.[1-3] Γ_{HV} has so far been interpreted by using the Van Hove space-time density correlation formulation applied successively to two scattering events. This kind of interpretation is based on two important assumptions: (a) The total linewidth is contributed by *both the first and second scatterers in random motion;* (b) the complex degree of coherence of incoherently doubly scattered light can be factorized into spatial and temporal parts. We present an explicit experimental demonstration showing that assumption (a) is *not*, in general, valid. We also present a new theory for Γ_{HV} using a so-far unexplored mechanism, viz., the *coupling* be-