HAL
open science

# LASER SPECTROSCOPY OF HIGH RYDBERG STATES OF LIGHT ALKALINE-EARTH ELEMENTS : Be AND Mg

R. Beigang, D. Schmidt, P. West

▶ **To cite this version:**

R. Beigang, D. Schmidt, P. West. LASER SPECTROSCOPY OF HIGH RYDBERG STATES OF LIGHT ALKALINE-EARTH ELEMENTS : Be AND Mg. Journal de Physique Colloques, 1983, 44 (C7), pp.C7-229-C7-237. 10.1051/jphyscol:1983719 . jpa-00223276

**HAL Id: jpa-00223276**

**https://hal.science/jpa-00223276**

Submitted on 4 Feb 2008

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

JOURNAL DE PHYSIQUE
Colloque C7, supplément au n°11, Tome 44, novembre 1983 

# LASER SPECTROSCOPY OF HIGH RYDBERG STATES OF LIGHT ALKALINE-EARTH ELEMENTS: Be AND Mg

R. Beigang, D. Schmidt and P.J. West

*Inst. f. Atom- und Festkörperphysik, FU Berlin, 1000 Berlin 33, F.R.G.*

Résumé - Les séries de Rydberg des configurations électroniques msns et msnd ont été étudiées entre les nombres quantiques principaux $10 \leq n \leq 70$ pour Be (m=2) et Mg (m=3). Les états de Rydberg ont été peuplés à l'aide d'une excitation en échelon ou d'une transition directe à deux photons avec des lasers à colorant pulsés doublés en fréquence (235 nm $\leq \lambda <$ 330 nm). Les atomes de Rydberg excités ont été détectés avec une diode thermo-ionique fonctionnant en régime de charge d'espace. L'analyse à l'aide de la théorie du défaut quantique à plusieurs voies des séries de Rydberg dans Be et Mg a conduit à la détermination des mélanges des états $mp^2\ ^1D_2$ avec ceux de la série msnd $^1D_2$.

Abstract - Rydberg series of msns and msnd configurations of Be(m=2) and Mg (m=3) were investigated between principal quantum numbers $10 \leq n \leq 70$. The Rydberg states were populated using stepwise excitation or direct two-photon transitions with frequency doubled light (235 nm $\leq \lambda <$ 330 nm) from pulsed dye lasers. The excited Rydberg atoms were detected with a space charge limited thermionic diode. An analysis of the Rydberg series both in Be and Mg by means of multi-channel quantum-defect theory (MQDT) results in the determination of $mp^2\ ^1D_2$ admixtures in msnd $^1D_2$ states.

## I. INTRODUCTION

Extensive investigations of high Rydberg states of the alkaline-earth elements Ca, Sr and Ba have been carried out over the past years using various spectroscopic methods (see e.g. /1-5/). The analysis of even and odd parity Rydberg series by means of an extended Rydberg-Ritz formula or a more sophisticated multi-channel quantum-defect theory (MQDT) resulted in the determination of configuration interactions, singlet-triplet mixing and accurate ionization limits. In the case of the light alkaline-earth elements Mg and Be the knowledge of high Rydberg states is incomplete. For Mg only the 3s3p $^1P_1$ series is known up to high principal quantum numbers /6/ whereas all other series like 3sns, 3snd and 3snf are not tabulated for quantum numbers higher than 25. In Be no 2snl Rydberg state was ever measured above n=12. The knowledge of these states, however, is desirable e.g. for astrophysicists and atomic theorists because Be and Mg are the lightest alkaline-earth elements and ab initio calculations of energy levels and wave functions seem feasible.

Article published online by EDP Sciences and available at http://dx.doi.org/10.1051/jphyscol:1983719

The determination of complete Rydberg series also allows for the investigation of configuration interactions between different series. Although the strength of perturbations decreases with decreasing atomic weight, Be and Mg exhibit marked deviations from the regular behavior of Rydberg series. In contrast to Ca, Sr, and Ba where the Rydberg series are mainly perturbed by doubly excited states originating from the open d-shell, the dominating perturber in Be and Mg is the $mp^2$ configuration. In particular the msnd $^1D_2$ series is strongly perturbed by the $mp^2$ $^1D_2$ level. A MQDT analysis will be presented which determines the admixtures of this state into the $^1D_2$ series.

## II. EXPERIMENTAL

From the $ms^2$ $^1S_0$ ground state an energy gap of 75 000 $cm^{-1}$ and 61 000 $cm^{-1}$ has to be bridged to excite high lying Rydberg levels for Be and Mg, respectively. The relevant part of the energy level diagrams is shown schematically in Figs. 1a and b. In the case of Mg a direct two-photon excitation was chosen to populate even parity msns and msnd Rydberg states. For Be the vapor density achieved in these experiments was too low to allow for the detection of two-photon transitions. Therefore the Rydberg states were excited with stepwise transitions via the 2s2p $^1P_1$ intermediate level. In both cases the required uv radiation was provided by frequency doubled light of pulsed dye lasers. The dye lasers were pumped with the second or third harmonic of a Nd:YAG laser producing peak powers up to several 10 kW at a linewidth of about

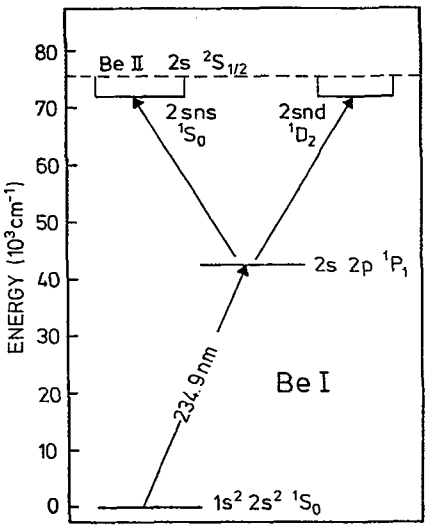



Fig. 1a Part of the energy level diagram of Be illustrating stepwise excitation of 2sns and 2snd Rydberg states.

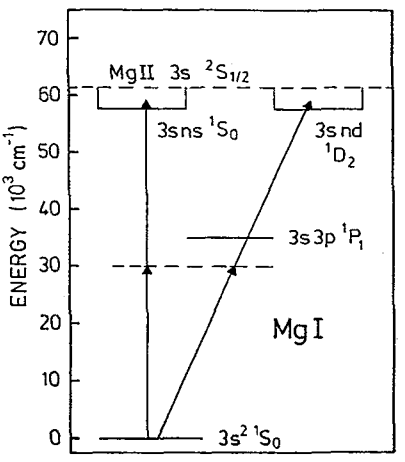



Fig. 1b Part of the energy level diagram of Mg with direct two-photon transitions to 3sns and 3snd Rydberg states.

0.1 $cm^{-1}$ between 450 and 660 nm. With a single dye (Rhodamine 6G) and an angle tuned KDP crystal (efficiency ∿ 10 %) the wavelength range from 322 to 330 nm was covered, corresponding to the energy range from 60 600 $cm^{-1}$ up to the ionization limit of Mg using two-photon transitions. For Be a second dye laser was necessary for the first step to the 2s2p $^1P_1$ intermediate level: A stilbene 3 dye laser was frequency doubled at a fixed frequency in a KB5 crystal producing 234.9 nm radiation with a peak power of several kW. A second frequency doubled Rhodamine 6G laser was then tuned to promote the exited Be atoms to high Rydberg states. A schematic diagram of the experimental set up is shown in Fig. 2. The metal vapor was produced in a stainless steel pipe which was heated to temperatures between 500 °C and 1200 °C. The typical vapor pressure used for the experiments was 50 to 150 mTorr in the case of Mg. A buffer gas of Ne at 2-4 Torr protected the cell windows. At the maximum temperature obtainable with the simple hot pipe oven the Be vapor pressure was limited to values below 1 mTorr. Therefore a very sensitive detection method for the excited Rydberg atoms was required. We have used thermionic detection with a space-charge limited diode. The diode was formed by a stainless steel wire inserted in the center of the pipe and negatively biased (-0.5...- 2 V) through a load resistor (10 ... 10 kΩ). The sensitivity of the thermionic diode increases with decreasing energy difference to the ionization limit, thus partly compensating for the decreasing transition probability. The sensitivity may be illustrated by the detection of a spin forbidden transition in Be. From the $2s^2\ ^1S_0$ ground state the 2s2p $^3P_1$ level was excited and detected with the thermionic diode although this state is more than 50 000 $cm^{-1}$ below the ionization limit. With this transition as the first step it should be possible to populate and investigate also the corresponding triplet series in Be.

All spectra were calibrated by means of Rb and Cs reference pipes.

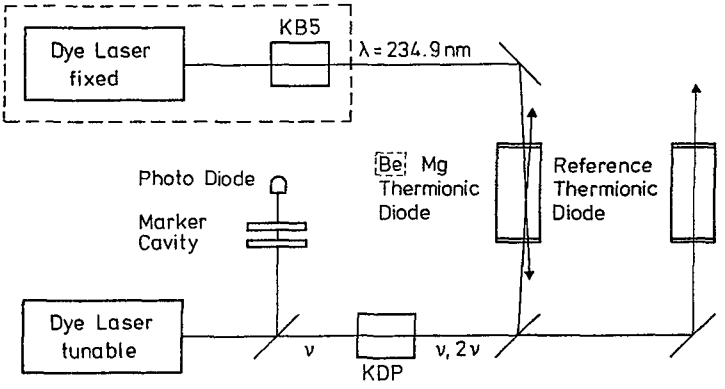



Fig. 2 Experimental set up. The fixed dye laser is only used for the stepwise excitation of Be Rydberg states.

Using known transition wavelengths the level energies in Be and Mg were determined with an accuracy of about $3 \cdot 10^{-6}$. To interpolate between adjacent calibration lines we used the transmission peaks of a Fabry-Perot interfermometer with a free spectral range of 0.5 $cm^{-1}$ in the visible.

## III. RESULTS AND DISCUSSION

### Beryllium

A typical excitation spectrum of 2snd $^1D_2$ Rydberg states close to the ionization limit is shown in Fig. 3. The $^1D_2$ series was resolved up to principal quantum numbers n ∿ 70. Due to the lower transition probability to 2sns $^1S_0$ states this series was only recorded up to n ∿ 40. From the experimentally determined level energies $E_n$ the quantum defect $\delta$ was determined using the Rydberg-Ritz formula

$$E_n = E_{ion} - \frac{R_{Be}}{(n-\delta)^2} \qquad (1)$$

where $E_{ion}$ is the ionization limit and R the Rydberg constant for Be. The result is shown in Fig. 4. It turns out that the 2sns $^1S_0$ series is

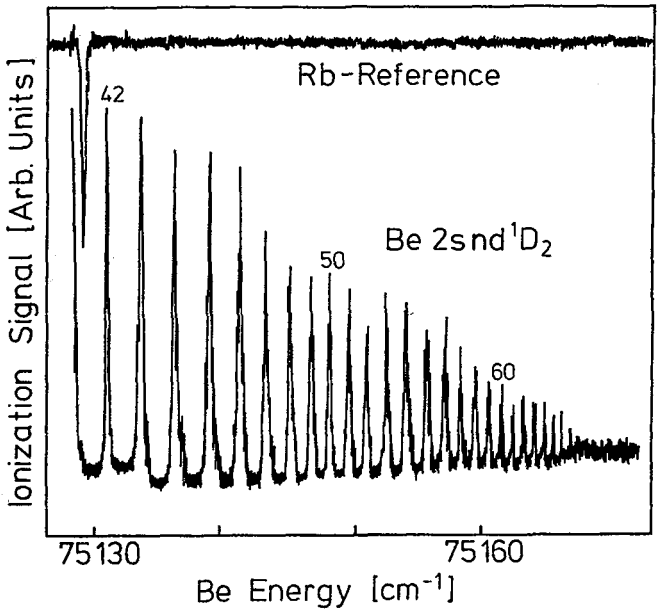



Fig. 3 Stepwise excitation spectrum of 2snd $^1D_2$ Rydberg states of Be. The upper trace displays the reference signal obtained from a Rb pipe.

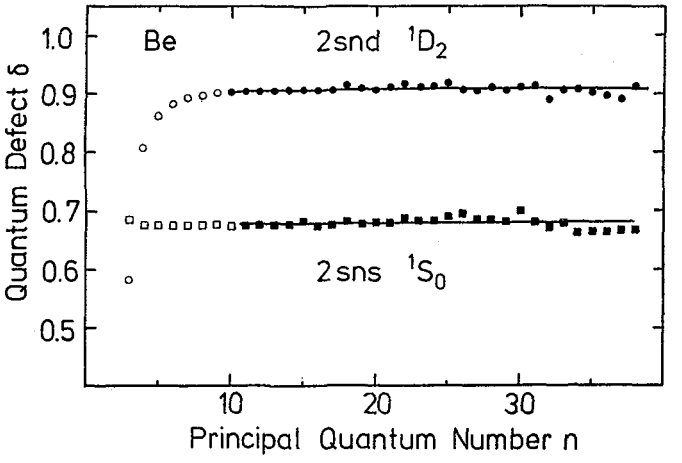



Fig. 4 Plot of the quantum defect δ versus principal quantum number n. Closed circles and rectangles are own measurements. Energy levels for n ≤ 10 are taken from Ref. /7/.

not perturbed by other series resulting in an almost constant quantum defect. The same holds for the $^1D_2$ series for high principal quantum numbers. Using only energy levels with n ≥ 15 the ionization limit was determined to $E_{ion}$ = 75 192.64(6) $cm^{-1}$. In the region of low principal quantum numbers n the 2snd $^1D_2$ series is heavily perturbed due to an interaction with the $2p^2$ $^1D_2$ level. In order to determine the amount of $2p^2$ admixture in the 2snd $^1D_2$ series we have performed a two-channel MQDT analysis with the 2pnp series as second channel. Using MQDT parameter as listed in Table I the 2snd $^1D_2$ series was fitted with this simple two-channel model. The admixture as a function of principal

Table I

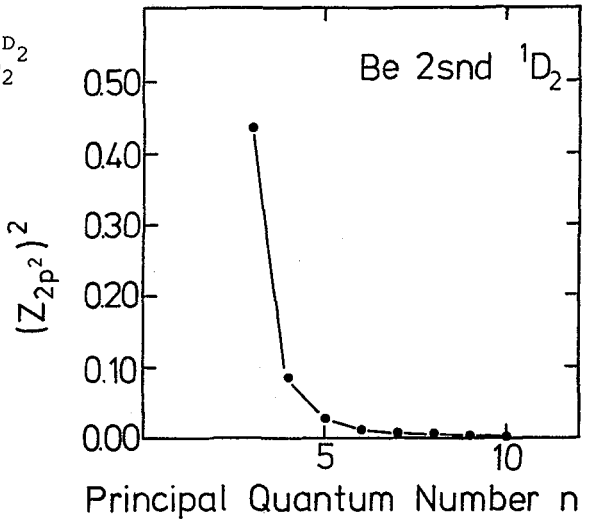
MQDT-Parameter Be, $^1D_2$ bound states.

| 2snd | 2pnp |
|---|---|
| $I_s$ = 75192.64 $cm^{-1}$ | $I_p$ = 107125.22 $cm^{-1}$ |
| $\mu_1$ = 0.96471 | $\mu_2$ = 0.54006 |
| $\frac{d\mu_1}{dE}$=-0.14744 | |
| $U_{11}$= 0.89169 | $U_{12}$= -0.45265 |

The MQDT parameters in Tables I and II are defined in Ref./9/ e.g.

quantum number n is shown in Fig. 5. The lowest bound $^1D_2$ state contains almost equal amounts of 2s3d (57 %) and $2p^2$ (43 %) character. This state was classified before as $2p^2\ ^1D_2$ /7/. Although a clear classification with a single configuration is questionable, as already claimed by Lu /10/, the label 2s3d $^1D_2$ seems more appropriate for this state according to the MQDT analysis.

Fig. 5 Admixture of $2p^2\ ^1D_2$ character into the 2snd $^1D_2$ series of Be.




Magnesium

In Mg the 3snd $^1D_2$ series was resolved up to principal quantum numbers n = 45. The other even parity Rydberg series which could be excited via two photons from the ground state, the 3sns $^1S_0$ series, has a transition probability of one order of magnitude smaller than the $^1D_2$ series. Therefore only states with maximum principal quantum number n=20 were recorded. The difference in transition probability is illustrated in Fig. 6. The plot of the quantum defect $\delta$ as a function of n again clearly shows that both $^1S_0$ and $^1D_2$ series are not perturbed above $n \gtrsim 15$ (Fig. 7) comparable to Be. Below $n \sim 10$ the 3snd $^1D_2$ series interacts with the $3p^2\ ^1D_2$ level. The level energies can be fitted with a two-channel MQDT analysis using the 3pnp $^1D_2$ series as second channel. The MQDT parameter are listed in Table II. The dominant admixtures occur in the low 3snd $^1D_2$ states with a maximum of 12 % in the 3s3d $^1D_2$ state whereas for higher $^1D_2$ states the admixture decreases rapidly as shown in Fig. 8. The results are in qualititative agreement with MCHF calculations of Froese-Fischer /8/ with a calculated maximum admixture of 21.7 % for the 3s3d $^1D_2$ state, however.

The behavior of Be and Mg Rydberg series is very similar with respect to the perturbations by the $mp^2$ configuration. The strength of interaction decreases when going from Be to Mg. This trend continues in the heavier alkaline-earth elements where configuration interactions with $mp^2$ term cause only localized perturbations.

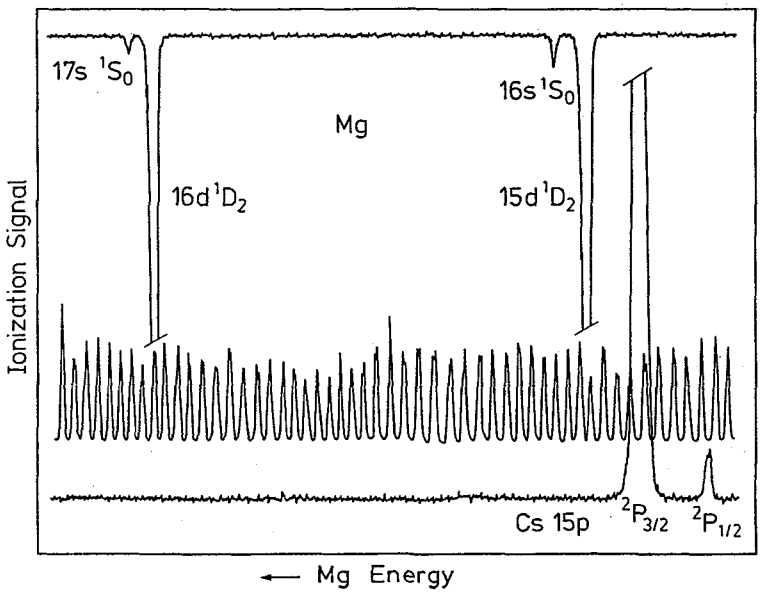



Fig. 6 Two-photon excitation spectrum of Mg. The middle trace shows the transmission fringes of the Fabry-Perot interferometer.

Fig. 7 Plot of the quantum defect δ versus principal quantum number n.

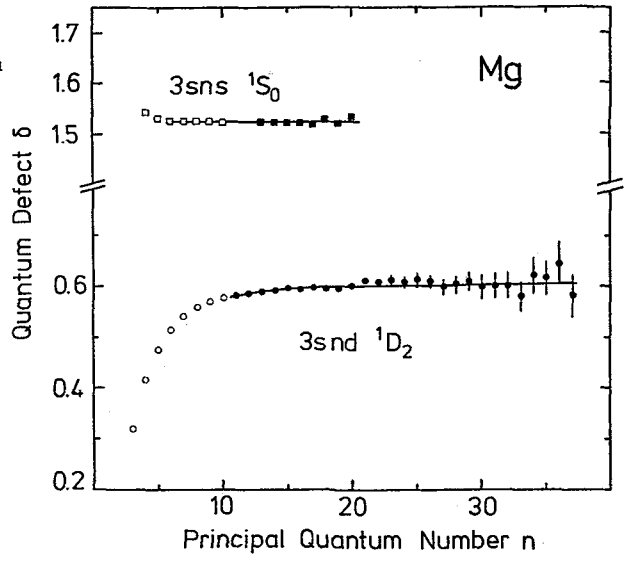


Table II

MQDT-Parameter Mg, $^1D_2$ bound states.

| 3snd | 3pnp |
|---|---|
| $I_s = 61671.02\ cm^{-1}$ | $I_p = 97401.37\ cm^{-1}$ |
| $\mu_1 = 0.96171$ | $\mu_2 = 0.42001$ |
| $\frac{d\mu_1}{dE} = 0.0369$ | |
| $U_{11} = 0.70357$ | $U_{12} = -0.71062$ |

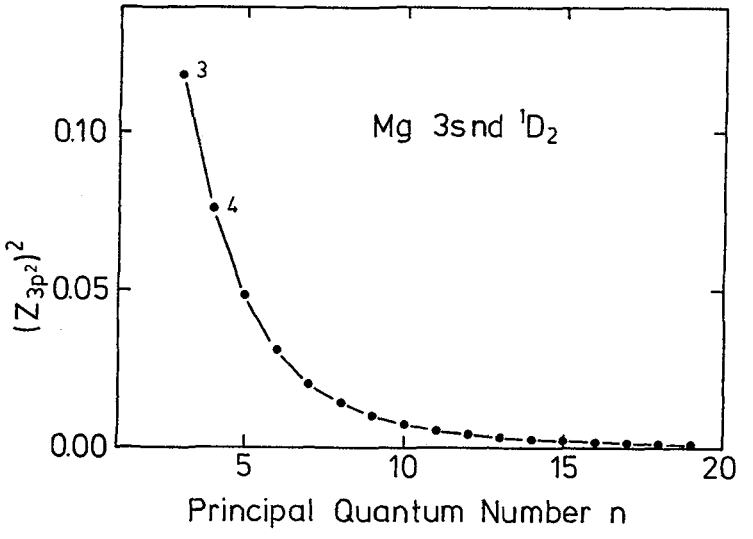



Fig. 8 Admixture of $3p^2\ ^1D_2$ character in the 3snd $^1D_2$ series of Mg.

This work was supported by the Deutsche Forschungsgemeinschaft, Sfb 161.

References

/1/ J.J. Wynne, J.A. Armstrong, and P. Esherick, Phys. Rev. Lett. 39, 1520 (1977)
/2/ S.A. Borgström and J.R. Rubbmark, J. Phys. B: At. Mol. Phys. 10, 3607 (1977)
/3/ M. Aymar, P. Camus, M. Dieulin, and C. Morillon, Phys. Rev. A18, 2173 (1978)
/4/ M. Aymar, P. Grafström, C. Levinson, H. Lundberg, and S. Svanber, J. Phys. B: At. Mol. Phys. 15, 877 (1982)
/5/ T.F. Gallagher, W. Sandner, K.A. Safinya, Phys. Rev. A23, 2969 (1981)
/6/ C.M. Brown, R.H. Naber, S.G. Tilford, and M.L. Ginter, Appl. Opt. 12, 1858 (1973)
/7/ L. Johansson, Physica Scripta 10, 236 (1974)
/8/ C. Froese-Fischer, Can. J. Phys. 53, 184 (1975)
/9/ R. Beigang and D. Schmidt, Physica Scripta 27, 172 (1983)
/10/ K.T. Lu, J. Opt. Soc. Am. 64, 706 (1974)