Theoretical approach for collisional depolarization of Rydberg atoms

G. Hermann, B. Kaulakys*, T. Udem

I. Physikalisches Institut der Universität Giessen, Heinrich-Buff-Ring 16, D-35392 Giessen, Germany

Abstract. A theoretical analysis of the collisional depolarization of Rydberg atoms is presented. Using the general formalism of irreducible tensors for collisional depolarization and an approximate expression of the collision matrix for the Rydberg-perturber scattering, simple analytical expressions for the depolarization cross sections of different multipoles are obtained. It is shown that depolarization cross sections may be expressed in terms of the universal cross section for the collisional broadening of Rydberg states. Explicit expressions for cross sections of the one-electron $nP_{3/2}$ and np states are presented.

PACS: 34.10; 34.60.

1. Introduction

The studies of interaction and collisions between the highly excited atoms and the surrounding atomic particles started more than half a century ago [1]. Various theoretical investigations of collisional broadening and shift of the Rydberg levels, inelastic collisions between Rydberg atoms and neutral atomic or molecular targets have been made during the last decade (see [2–10] and references therein). Simple analytical expressions have been obtained for the cross sections of broadening and shift of Rydberg levels by elastic collisions, inelastic $nl \rightarrow n'l'$ transitions, and ionization processes.

On the other hand, collisional depolarizations and m-changing collisions of Rydberg atoms are investigated only incompletely. We can point to the paper [11] and references therein. It is the purpose of this paper to present a consistent approach for the theoretical analysis of the collisional vector and tensor depolarization of the

Rydberg states and to obtain some analytical expressions for the polarization relaxation cross sections. Such depolarization cross sections can be written according to the broadening of Rydberg states taking into account the anisotropy of the Rydberg-perturber potential. Therefore, the experimental investigation of the dependences on the quantum numbers of the Rydberg states and species of the perturbers can give additional information about the Rydberg-neutral interaction potentials and collisional processes.

2. Collisional depolarization processes

The general problem of collisional depolarization of excited atoms has been treated by Dyakonov and Perel [12], Omont [13], and Wang and Tomlinson [14]. Collisional depolarization (relaxation of orientation and of alignment) of Rydberg states is one of the relaxation processes and can be investigated on the basis of the irreducible tensors of the density matrix technique [15, 16]. If one uses a standard representation of the atomic states $|\alpha J M\rangle$, where J and M label the total angular momentum and its projection on the z-axis, and α denotes the other indices necessary to specify the state, then the density matrix ρ can be written

$$\rho = \sum_{\alpha J_\alpha M_\alpha \beta J_\beta M_\beta} \rho_{\alpha J_\alpha M_\alpha, \beta J_\beta M_\beta} |\alpha J_\alpha M_\alpha\rangle \langle \beta J_\beta M_\beta|, \qquad (1)$$

with

$$\rho_{\alpha J_\alpha M_\alpha, \beta J_\beta M_\beta} = \langle \alpha J_\alpha M_\alpha| \rho |\beta J_\beta M_\beta\rangle. \qquad (2)$$

The relations between irreducible $\rho_q^k(\alpha J_\alpha, \beta J_\beta)$ and ordinal (2) components are [16, 17]

$$\rho_{\alpha J_\alpha M_\alpha, \beta J_\beta M_\beta} = (-1)^{J_\beta - M_\beta} \sum_{k, q}$$
$$\cdot \langle J_\alpha J_\beta M_\alpha - M_\beta | k q \rangle \rho_q^k(\alpha J_\alpha, \beta J_\beta), \qquad (3)$$

$$\rho_q^k(\alpha J_\alpha, \beta J_\beta) = \sum_{M_\alpha, M_\beta} (-1)^{J_\beta - M_\beta}$$

$$\cdot \langle J_\alpha J_\beta M_\alpha - M_\beta | k q \rangle \, \rho_{\alpha J_\alpha M_\alpha, \beta J_\beta M_\beta}. \qquad (4)$$

The irreducible tensors formalism provides the simplest and the most physical expressions for the relaxation terms.

Most relaxation processes can be considered to be isotropic, e.g. thermal collisions give a quite isotropic relaxation. The evolution of the system is well described by a master equation taking into account the randomness of the interaction and all the necessary averages.

Introducing a relaxation matrix G we can write the corresponding master equation for the density matrix

$$(\mathrm{d}\rho/\mathrm{d}t)_{\mathrm{rel}} = -G\rho, \qquad (5)$$

where the relaxation matrix G describes a linear transformation of the Liouville space L of the system. In the semi-classical theory of collisions with one state perturbers one gets for the relaxation matrix [16]

$$\left(\frac{\mathrm{d}\rho}{\mathrm{d}t}\right)_{\mathrm{coll.}} = 2\pi N_p \left\langle v \int_0^\infty b\,\mathrm{d}b\,[S(b,v)\,\rho S^+(b,v) - \rho] \right\rangle, \qquad (6)$$

where N_p is the density of perturbers, the average is over the velocities v and the direction of the impact parameter $\mathbf{b}$. S is the collision matrix which relates the states after the collision Ψ' with states before collision Ψ by

$$\Psi' = S\Psi. \qquad (7)$$

In the special case of isotropic collisions and relaxation within a single Zeeman multiplet $|\alpha J\rangle$ the relaxation matrix in the irreducible representation is diagonal, i.e.,

$$\mathrm{d}\rho_q^k/\mathrm{d}t = -g_k\,\rho_q^k, \qquad (8)$$

where g_k are the collisional relaxation rates of the different multipoles of the Zeeman multiplet. They are real and positive except $g_0 = 0$ and may be expressed as

$$g_k = N_p \langle v \sigma_k(v) \rangle, \qquad (9)$$

$$\sigma_k = 2\pi \int_0^\infty \Pi_k(b,v)\,b\,\mathrm{d}b. \qquad (10)$$

Here σ_k are the depolarization cross sections of the multipoles rank k and Π_k are the irreducible components of an intermediate relaxation matrix averaged over the Euler angles of collision axes

$$\Pi_k = 1 - \sum_{MNM'N'q} (-1)^{2J-N-N'}$$

$$\cdot \begin{pmatrix} J & J & k \\ M & -N & q \end{pmatrix}\begin{pmatrix} J & J & k \\ M' & -N' & q \end{pmatrix} S_{MM'}\,S_{NN'}^*. \qquad (11)$$

Since (11) is rotation-invariant, the matrix elements of S may be expressed in any frame [16].

It follows from (11) the relationship

$$\sum_k (2k+1)\,\Pi_k = (2J+1)^2 - \sum_{MN} S_{MM}\,S_{NN}^*, \qquad (12)$$

which relates the summed intermediate relaxation rate with the diagonal matrix elements of the collison matrix and may be used for checking the accuracy of calculations.

3. Depolarization cross sections

Thus, the main problem in the theory of collisional relaxation of the excited states is the calculation of the collision S matrix for the Rydberg-perturber scattering. A theoretical analysis of the Rydberg-neutral collision process taking into account the anisotropy of the Fermi potential [18] and rotation of the diatomic axis [19] results to approximate expressions for the S matrix of $|nl\rangle$ and $|nj\rangle$ one electron states. In the convenient collision frame of reference with the z-axis directed opposite to the collision velocity $\mathbf{v}$ and y-axis opposite to the impact parameter $\mathbf{b}$ (see [10]) the most significant matrix elements of the collision matrix are $S_{m,m}$, $S_{m,-m}$ and S_{m_j,m_j}. Therefore, if in some frame of reference the S matrix may be written as

$$S_{M'M} = \delta_{M',M}\,S_{MM} + \delta_{-M',M}\,S_{-M,M}, \qquad (13)$$

then, according to (11)

$$\Pi_k = 1 - \sum_{MN_q} \begin{pmatrix} J & J & k \\ M & -N & q \end{pmatrix}^2 S_{MM}\,S_{NN}^*$$

$$- (-1)^k\, 2 \sum_{M>0} \begin{pmatrix} J & J & k \\ M & -M & 0 \end{pmatrix}^2 |S_{-M,M}|^2. \qquad (14)$$

In the following we mainly restrict our consideration to the collisional depolarization of $nP_{3/2}$ and np states.

3.1 Depolarization of $nP_{3/2}$ states

In the paper [10] the approximate expressions for the matrix elements of the S matrix for the Rydberg-neutral scattering described by the superposition of the polarization potentials and the Fermi potential have been derived. For the scattering in $nP_{3/2}$ states the non-zero matrix elements of the approximate S matrix may be written as

$$S_{1/2,1/2} = S_{-1/2,-1/2} = \exp(-i\eta_{1/2}),$$

$$S_{3/2,3/2} = S_{-3/2,-3/2} = \exp(-i\eta_{3/2}), \qquad (15)$$

where $\eta_{1/2} = \frac{1}{2}\eta_e$, $\eta_{3/2} = \frac{3}{2}\eta_e$ and

$$\eta_e(b) = -L/(2vn^{*3}\sqrt{b}) \qquad (16)$$

is the semiclassical phase-shift due to the isotropic part or the Fermi potential, with n^* and L being the effective principal quantum number of the Rydberg state and the electron-perturber scattering length, respectively [3].

From (10), (14)–(16) we have the depolarization cross sections of the different multipoles (rank k)

$$\sigma_0 = 0, \qquad \sigma_1 = \sigma_3 = \tfrac{2}{5}\sigma_e'(L), \qquad \sigma_2 = \tfrac{4}{5}\sigma_e'(L), \qquad (17)$$
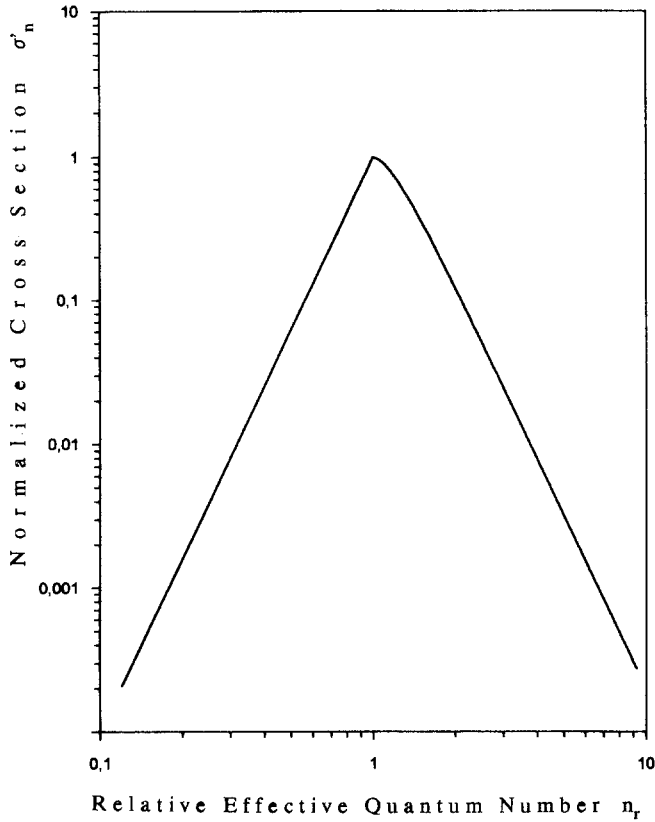
Fig. 1. Dependence of the normalised universal cross section $\sigma'_n = \sigma'_e(L)/4\pi n_1^{*4}$ on the relative effective principal quantum number $n_r = n^*/n_1^*$ according to (18)

where $\sigma'_e(L) = \sigma'(0, L)$ is the broadening cross section of the Rydberg state with the effective principal quantum number n^* due to the Fermi potential, without taking into account the polarization attraction of the perturber with the core of the Rydberg atom, i.e. with $\alpha = 0$ (see [10]). $\sigma'_e(L)$ may be calculated according to (26) from the paper [3], i.e.

$$\sigma'_e(L) = \begin{cases} 4\pi n^{*4} & n^* < n_1^* = (|L|/4v)^{1/4}, \\ \dfrac{8\pi n_1^{*8}}{n^{*4}}\left(1 - \dfrac{n_1^{*8}}{2n^{*8}}\right), & n^* > n_1^*. \end{cases} \quad (18)$$

A representative plot of normalized cross section against the relative effective principal quantum number is shown in Fig. 1.

3.2 Depolarization of np states

For np states the approximate non-zero matrix elements of the S matrix may be written as [10]

$$S_{00} = \exp(-i\eta_c), \quad S_{11} = S_{-1,-1} = \tfrac{1}{2}[\exp(-i\eta_1^+) + 1],$$
$$S_{-1,1} = S_{1,-1} = \tfrac{1}{2}[\exp(-i\eta_1^+) - 1], \quad (19)$$

where $\eta_c = \pi\alpha/4vb^3$ is the scattering phase-shift due to the polarization attraction between the perturber and the core of the Rydberg atom while $\eta_1^+ = 3\eta_e$. Analysis

of (10), (14), and (19) shows that the influence of the phase η_c is negligible and as a result we have

$$\sigma_0 = 0, \quad \sigma_1 = \tfrac{2}{3}\sigma'_e(3L), \quad \sigma_2 = \tfrac{2}{5}\sigma'_e(3L). \quad (20)$$

Here $\sigma'_e(3L)$ is the broadening cross section calculated according to (18) with replacing L by $3L$. We see that the depolarization of np states is more effective, than of $nP_{3/2}$ states.

The depolarization cross sections of other states may be calculated in the similar way. The main problem, however, is the evaluation of the collision S matrix for the Rydberg states.

It should be noted that the relaxation rates of the hyperfine Zeeman multiplets may be expressed in terms of the relaxation rates for the electronic multipoles (8)–(10) (see, e.g. [16]).

4. Conclusions

Theoretical analysis of the collisional process between a Rydberg atom and a neutral atomic particle taking into account the anisotropy of the Rydberg-perturber potential results in simple analytical expressions for the cross sections of collisional polarization relaxation of the Rydberg states due to the Fermi potential. In addition, the dependences of the collisional depolarization cross sections on the effective principal quantum number of the Rydberg state and on the electron-perturber scattering length may be expressed in terms of the universal cross section (18). However, the strength of the cross section's dependence on the scattering length varies with the electronic states (compare, e.g. (18) with (20)).

While the broadening of optical lines is essentially determined by the isotropic part of the potential, the depolarization is entirely caused by its anisotropic part. So, both types of observations are required to get information about the whole potential. Comparison of the theoretical cross sections with experimental results would give additional information about the Rydberg-neutral interaction potentials and collisional processes. On the other hand, the broadening of the Rydberg levels is caused by the superposition of the polarization potential and the perturber-Rydberg-electron short range interaction. In addition, the broadening of the high Rydberg levels is determined by the interaction of the perturber with the core of the Rydberg atom [10], while the depolarization is essentially caused by the interaction of the perturber with the Rydberg electron. Therefore, the experimental investigations of dependences of the collisional depolarization of the Rydberg atoms on the spectral terms, fine structure, effective principal quantum number, and species of the perturbing particles are desirable.

The calculations of the cross sections for the collisional polarization relaxation of the Rydberg states in the free electron model is not a straightforward generalization of the method [7, 9] but a task for the future.

122

References

1. Fermi, E.: Nuovo Cimento **11**, 157 (1934)
2. Stebbings, R.F., Dunning, F.B.: Rydberg states of atoms and molecules. Cambridge: Cambridge University Press 1983
3. Kaulakys, B.: J. Phys. B **17**, 4485 (1984)
4. Gounand, F., Petitjean, L.: Phys. Rev. A **30**, 61 (1984)
5. Petitjean, L., Gounand, F.: Phys. Rev. A **30**, 2946 (1984)
6. Kaulakys, B.: J. Phys. B **18**, L167 (1985)
7. Kaulakys, B.: Zh. Eksp. Teor. Fiz. **91**, 391 (1986); Sov. Phys. – JETP **64**, 229 (1986)
8. Lebedev, V.S., Marchenko, V.S.: J. Phys. B **20**, 6041 (1987)
9. Kaulakys, B.: J. Phys. B **24**, L127 (1991)
10. Hermann, G., Kaulakys, B., Lasnitschka, G., Mahr, G., Scharmann, A.: J. Phys. B **25**, L407 (1992)
11. Yoshizawa, T., Matsuzawa, M.: J. Phys. Soc. Jpn. **54**, 918 (1985)
12. Dyakonov, M.I., Perel, V.I.: Zh. Eksp. Teor. Fiz. **48**, 345 (1965); Sov. Phys. – JETP **21**, 227 (1965)
13. Omont, A.: J. Phys. Rad. **26**, 26 (1965)
14. Wang, C.H., Tomlinson, W.J.: Phys. Rev. **181**, 115 (1969)
15. Omont, A.: J. Phys. (Paris) **34**, 179 (1973)
16. Omont, A.: Prog. Quant. Electron. **5**, 69 (1977)
17. Sobelmann, I.I., Vainshtein, L.A., Yukov, E.A.: Exitation of atoms and broadening of spectral lines. Berlin, Heidelberg, New York: Springer 1981
18. Hermann, G.: Phys. Lett. **133**A, 225 (1988)
19. Kaulakys, B.: Liet. Fiz. Rinkinys **19**(1), 55 (1979); Sov. Phys.-Collect: Lit. Fiz. Sbornik (Allerton) **19**(1), 38 (1979)