

On the possibility of studying autoionization states of negative ions in terms of the broadening and displacement of the Rydberg series of neutral atoms

B. P. Kaulakis, L. P. Presnyakov, and P. D. Serapinas
P. N. Lebedev Physics Institute, USSR Academy of Sciences

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It is shown that resonance scattering introduces an important contribution to the broadening and displacement of the Rydberg series in atoms and explains the presence of oscillations. Analysis of the experimental data permits the reconstruction of the binding energy of the quasidecrete level for a negative ion, its autoionization width, orbital angular momentum, and its multiplicity.

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Fermi's concept¹ concerning the relationship between the broadening and displacement in the Rydberg series for atoms in an atmosphere of buffer gas and the characteristics of elastic scattering of slow electrons by atoms, was realized in its most general form by Alekseev and Sobel'man.² It has been shown that the concept of quasi-free electron scattering¹ corresponds to the region of applicability of the impulse approximation within the framework of the shock broadening theory for calculating the line displacement Δ and width γ :

$$\begin{aligned} \Delta(n, l) &= \sum_{L, S_T} (2L + 1) C(S_T) \Delta_{L, S_T}(n, l); \quad \gamma(n, l) \\ &= \sum_{L, S_T} (2L + 1) C(S_T) \gamma_{L, S_T}(n, l) \end{aligned} \quad (1)$$

$$-2\Delta_{L, S_T}(n, l) + i\gamma_{L, S_T}(n, l) = -4\pi N \int_0^{\infty} f_L(q) |G_{n, l}(q)|^2 q^2 dq. \quad (2)$$

Here, N is the buffer gas density, n and l are the principal and orbital quantum numbers of a highly-excited electron, $G_{n, l}$ is the radial part of the wave function in the impulse representation, $C(S_T)$ is the spin statistical weight for a system with spin S_T , and f_L is the amplitude for the elastic scattering of an electron with orbital momentum L . Calculations² taking into account the potential scattering of slow electrons by atoms led to a good description of experimental data in terms of the broadening and displacement of inert gases in the atmosphere,³ and raised a question concerning the study of elastic scattering of slow electrons by spectroscopic techniques.

However, an experiment⁴ carried out in an atmosphere of alkali element gases led to unexpected results: the values for the line width and displacement turned out to be much larger than the best (in a variational sense) theoretical values,² and had an oscillatory component for $10 < n < 35$. Along with quantitative differences (polariza-

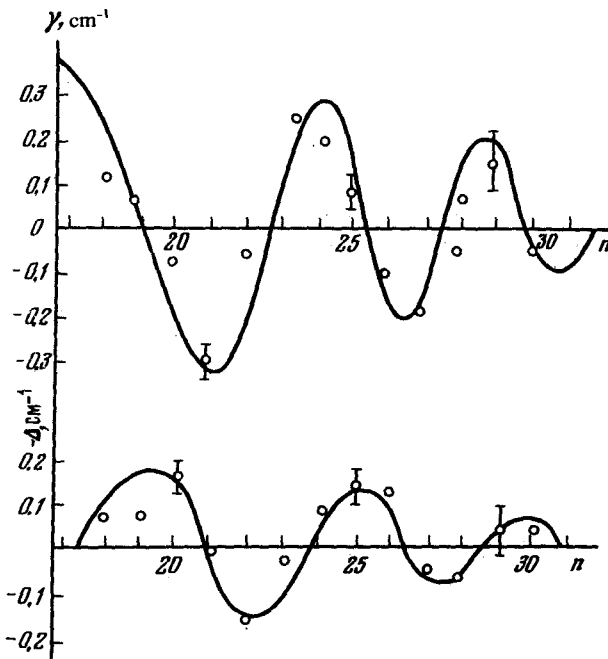


FIG. 1. Oscillatory component of the width γ and displacement Δ as functions of the principal quantum number: circles denote experimental data,⁴ solid lines indicate the results of our work.

tion potentials, etc.), there is also a qualitative difference in the scattering processes by atoms of inert gases and alkali elements, due to the possible formation in the latter of weakly-bound states (of negative ions) during the addition of an electron to the atom in the ground state. This was also indicated earlier⁵; however, taking into account only *s*-scattering by discrete⁵ and quasi-discrete⁶ levels of the negative ion cannot give a complete interpretation of all the experimental data.⁴

In this work, the general approach of Eqs. (1) and (2) is supplemented by taking into account scattering by quasi-discrete levels with $L > 0$. It is shown that this completely explains the oscillatory component of Δ and γ , and introduces an important contribution to their monotonic component. A comparison of the oscillation periods and the amplitudes of the calculated and measured oscillatory components Δ and γ leads to a single-valued reconstruction of the binding energy of the quasi-discrete level, its autoionization width, orbital moment, and multiplicity.

In view of the additivity of potential and resonance scattering, we shall discuss the contribution of the latter in Eqs. (1) and (2) separately. Using a general equation for the amplitude of *L*-scattering by the quasi-discrete level⁷ with binding energy $\epsilon_r > 0$ and for the autoionization width $\Gamma_L \ll \epsilon_r$, and also the analytic properties of the functions $G_{n,l}(q)$, we may obtain the resonance scattering contribution in the form

$$\frac{\Delta_{L,S_T}^{res}}{4\pi N(2L+1)C(S_T)} = - \left[\frac{3\nu^{-2} + n^2}{2(\nu^{-2} + n^2)^2} \Gamma_L \nu^{-3} n^2 + (-1)^{l+1} A_L \sin(4n^2\nu) \right], \quad (3)$$

$$\frac{\gamma_{L, S_T}^{e, s}}{8\pi N(2L+1)C(S_T)} = \left[\frac{\Gamma_L \nu^{-6} n}{(\nu^{-2} + n^2)^2} + (-1)^{l+1} A_L \cos(4n^2\nu) \right], \quad (4)$$

$$A_L = \frac{\Gamma_L \nu^{-6} n}{(\nu^{-2} + n^2)^2} \left(\frac{\nu^{-2} + n^2 - \Gamma_L \nu^{-3} n}{\nu^{-2} + n^2 + \Gamma_L \nu^{-3} n} \right)^n, \quad \nu = \sqrt{2\epsilon_r} \ll n^{-1},$$

$$\Gamma_L \ll 1/2n^2. \quad (5)$$

Here the amplitude of the oscillatory component is bounded by the condition $A_L \ll \nu^{-3} [2en(\nu^{-2} + n^2)]^{-1}$. The period of the oscillatory component is uniquely related to the energy ϵ_r , which is slowly reduced during the period of the experimental oscillations. We shall apply this result to interpreting an experiment⁴ in the broadening and displacement of the np -levels of Cs in Cs doublets (the concentration is $N = 1.3 \times 10^{17} \text{ cm}^{-3}$). From Eqs. (3)–(5) it follows that the experimental values for the oscillatory components of γ and Δ cannot be obtained for $L = 0$. Coincidence of the amplitude maxima (in Fig. 1) occurs for $L = 1$ and $S_T = 1$. It can be shown that the values $L > 1$ lead to a discrepancy with the experiment in terms of the decrease in the damping of the oscillations. Thus, we must assume the existence of an autoionization 3P level in the Cs^- ion with a binding energy $\epsilon_r = 7.5 \times 10^{-4} \text{ eV}$ and a width $\Gamma \approx 3.10 \times 10^{-4} \text{ eV}$. We note that resonance scattering also makes a contribution to the monotonic part γ^{res} and Δ^{res} which is 1.33 and 0.22 cm^{-1} , respectively. Potential scattering² gives $\gamma^{\text{pot}} = 0.40 \text{ cm}^{-1}$, $\Delta^{\text{pot}} = 0.5 \text{ cm}^{-1}$, and the sum of the potential and resonance parts describes the data satisfactorily⁴: $\gamma_{\text{exp}} = 1.5 \text{ cm}^{-1}$, $\Delta_{\text{exp}} = 0.75 \text{ cm}^{-1}$.

The existence of the autoionization 3P level in Cs^- is not unexpected. Direct variational calculations⁸ have shown the existence of similar levels for ions of the lighter alkali metals Li^- , Na^- , and K^- with respective binding energies 6.0×10^{-2} , 8.3×10^{-2} , and $2.4 \times 10^{-3} \text{ eV}$ and widths 5.7×10^{-2} , 8.5×10^{-2} , and $5.8 \times 10^{-4} \text{ eV}$.

The results given here reveal new possibilities for studying autoionization states by spectroscopic techniques.

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