

BROADENING AND SHIFT OF HIGHLY EXCITED LEVELS OF POTASSIUM AND CESIUM ATOMS

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Submitted August 7, 1970

Zh. Eksp. Teor. Fiz. 60, 541-551 (February, 1971)

Measurements were made of the width and shift of the absorption lines of potassium in potassium vapor and of cesium in cesium vapor near the limit of the series (up to $n \sim 40-50$). Unlike the case of interaction with inert-gas atoms, the line width turned out to be much larger than the shift. The large width can be attributed to the influence of the short-range potential of the alkali-metal atoms, which have a strong electron affinity. A comparison of the experimental values of the shift with the theoretical calculations makes it possible to assess the methods of calculating the phases of elastic scattering of extremely slow electrons (with energy $\sim 10^{-2}$ eV) by atoms of potassium and cesium.

A spectroscopic investigation of the interaction of highly excited atoms with perturbing particles makes it possible to obtain information on the scattering of electrons of extremely low energy by these particles (see the papers of Fermi^[1] and of Alekseev and Sobel'man^[2]).

In the greater part of the experiments performed to date, the absorption spectra of the atoms of alkali metals have been investigated in an atmosphere of inert gases. In particular, in^[3,4] the line widths and shifts of cesium were measured in an atmosphere of argon, and it was shown that the theory of^[1,2] describes correctly both the width and the shift of the lines.

According to Fermi, the qualitative picture of the phenomenon is as follows. The absorption lines near the limit of the series (large values of the principal quantum number $n > 20-30$) correspond to such large electron orbits that at a perturbing-particle density on the order of $10^{17}-10^{18}$ cm⁻³ there are tens and hundreds of these particles inside the orbit. In the field of the atomic residue, the particles become polarized, leading to a shift of the highly excited levels (energy is consumed in the polarization of the particles, and therefore the shift is always red). In addition, a scattering of the (quasifree) optical electron by the perturbing particles takes place, and this also leads to a shift of the levels. In this case the sign of the shift depends on the nature of the scattering center.

Under ordinary experimental conditions, both effects are statistically independent and make additive contributions to the line width and shift. The main contribution to the line width is made by the polarization effect, and that to the shift by the scattering effect.

In^[2] measurements were made of the line shift of cesium in cesium. The theoretically calculated^[5] shift turned out to be very sensitive to the method of calculating the phases of the electron-atom scattering. A comparison of the experimental and the calculated quantities has made it possible to assess the different methods of scattering-phase calculations in the electron-energy region which is the most complicated both for the theory and for the direct experiment, namely 0-0.2 eV.

In the present paper we report measurements of the widths and shifts of the absorption lines of cesium in cesium vapor and of potassium in potassium vapor.

DESCRIPTION OF EXPERIMENT AND RESULTS

The measurements were performed on the absorption lines of the principal series, corresponding to transitions to levels with principal quantum numbers n ; studies were made of the line widths γ at $n = 20-40$ and of the line shifts Δ at $n = 15-50$.

The procedure for measuring the width and the shift did not differ, in essence, from that described earlier.^[3,4]

1. The spectra were registered photographically with the aid of a DFS-3 spectrograph. Photographs for the measurement of the Cs line widths were obtained in second order of diffraction with a 1200-line/mm grating, and all the remaining spectra were obtained in first order of diffraction (the reciprocal dispersions were 1 and 2 Å/mm, respectively). The apparatus function made a contribution of $\lesssim 3\%$ to the Cs line widths and was disregarded. For the K lines, the correction for the apparatus width was introduced and did not exceed 7%.

The lines of the principal series of the alkali-metal atoms have a hyperfine structure which, in the main, is determined by the splitting of the level $n_0^2S_{1/2}$ into two components. The magnitude of the splitting of the potassium lines is $\Delta\nu^K \sim 0.01$ cm⁻¹,^[6] making practically no contribution to the width (the same pertains to the isotopic shift of the lines of the isotopes K³⁹ and K⁴¹). For the cesium lines, $\Delta\nu^{Cs} = 0.31$ cm⁻¹,^[6] and its lines must be regarded as consisting of two components. In measuring the shift it was sufficient to determine the displacement of the center of gravity of the summary contour, and in measuring the width the separation of the contours was carried out graphically.^[4] The ratio of the intensities of the hfs components, in accordance with the statistical weights, is 7:9, which agrees within a few percent with the experimental value obtained on photographs with separated components.

The line asymmetry, a measure of which was taken

to be the ratio of the red to blue half-widths, did not exceed 1.1. The contribution of the statistical wing to the line width thus did not exceed 5% and was disregarded.

At large n , the contours of neighboring absorption lines overlap and distort each other. Graphical construction of the overlapping contours makes it possible to estimate the error and to introduce the corresponding correction to the line width. The measurements were carried out up to values of n where the correction did not exceed 10% ($\delta\lambda_{n, n+1} \leq 2\gamma$).

2. The wavelengths of the principal series of K and Cs at a vapor density $N \sim 10^{15} - 10^{16} \text{ cm}^{-3}$ were measured by Kratz.^[7] Our measurements were carried out at densities larger by 1.5–2 orders of magnitude, and therefore the wavelengths measured by Kratz could be regarded as the initial zeroth wavelengths in the measurement of the line shifts.

In the present study we determined the absolute values of the wavelengths. The references were the Ar and Fe lines of the spectrum of a discharge in a hollow cathode (the values of the wavelengths were taken from the tables^[8]). Monitoring of the monotonicity of the dispersion with respect to the wavelength, and also the determination of the wavelengths of certain reference lines relative to the others, has shown that the accuracy of these wavelengths was not worse than $\sim 10^{-3} \text{ \AA}$.

3. In measurement of the width and shift of the lines, no account was taken of the presence of a fine structure. For the Cs spectrum this is perfectly justified, since the intensity of the weak component is at least one order of magnitude lower than that of the strong one, starting with $n \sim 10$.^[9, 10] For the K spectrum there are only theoretical data,^[10] according to which the ratio of the intensities of the fine-structure components of K at $n = 15 - 30$ is 3–3.5. The error in the determination of the width and shift of the lines of K at $n = 15$ does not exceed 0.1 cm^{-1} and decreases rapidly with increasing n , becoming negligible at $n \gtrsim 25$.

The error in the determination of the line width and shift is determined mainly by the scatter of the individual measurements and amounts to $\sim 10\%$.

4. Absorption tubes were made of stainless steel, and sapphire windows were fused to their ends. In order for the density of the atoms to correspond to the saturated vapor pressure, liquid metal was used to coat the greater part of the tube length. To prevent the metal from settling on the windows, their temperature was maintained several degrees higher. The distribution of the temperature along the tube was monitored by five thermocouples, and the temperature was measured accurate to $\pm 1^\circ$. The density of the atoms was calculated from the following formulas (p —vapor pressures in millimeters of mercury, T —in degrees K):

for K^[11]

$$\lg p = 7.1830 - 4430 / T;$$

for Cs^[9]

$$\lg p = -3529 / T + 3.6572.$$

The accuracy of these formulas is apparently not higher than 5% for potassium or 10% for cesium, and this governs the accuracy with which the density of the atoms is measured.

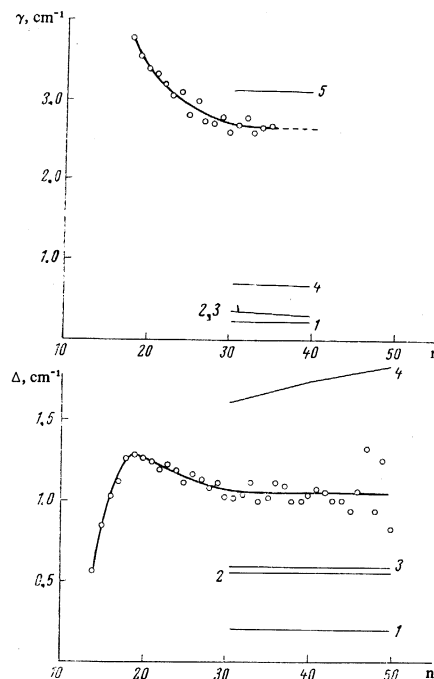


FIG. 1. Width γ and shift Δ of the potassium lines ($N = 2.6 \times 10^{17} \text{ cm}^{-3}$, $T = 481^\circ \text{C}$) as functions of principal quantum number. \circ —experimental results; 1—polarization part of the width γ_p and shift γ_p ; 2, 3, 4—sum $\gamma_c + \gamma_p$ and $\Delta_c + \Delta_p$, where γ_c and Δ_c are calculated; 2, 3—with allowance for exchange and the contribution made to the polarization potential by the first one and two excited states, respectively, 4—with allowance for one p level, without allowance for exchange in the polarization potential, 5—width calculated with allowance for the short-range potential of the atom.

5. The measurements were made at an atom density $N^K = 2 - 4.8 \times 10^{17} \text{ cm}^{-3}$ ($T = 466 - 520^\circ \text{C}$) for potassium and $N^{\text{Cs}} = (1.2 - 2.5) \times 10^{17} \text{ cm}^{-3}$ ($T = 360 - 400^\circ \text{C}$) for cesium. Figures 1 and 2 show the characteristic plots of the widths and shifts of the lines against the principal quantum number, obtained at $N^K = 2.6 \times 10^{17} \text{ cm}^{-3}$ ($T = 481^\circ \text{C}$) and $N^{\text{Cs}} = 1.3 \times 10^{17}$ ($T = 360^\circ \text{C}$).

In the region $n > 20$ in the spectrum of K and $n > 30$ in the spectrum of Cs, the line width and shift are proportional to the density of the perturbing particles. Table 1 gives, by way of example, data on the line shift of K at different densities. At $n \gtrsim 30$ the width and shift change very little with changing n and are given respectively by:

for K

$$\begin{aligned} \gamma / N &= (1.02 \pm 0.15) \cdot 10^{-17} \text{ cm}^{-1} / \text{cm}^{-3}, \\ \Delta / N &= (0.40 \pm 0.06) \cdot 10^{-17} \text{ cm}^{-1} / \text{cm}^{-3}; \end{aligned}$$

for Cs

$$\begin{aligned} \gamma / N &= (1.1 \pm 0.2) \cdot 10^{-17} \text{ cm}^{-1} / \text{cm}^{-3}, \\ \Delta / N &= (0.61 \pm 0.12) \cdot 10^{-17} \text{ cm}^{-1} / \text{cm}^{-3}. \end{aligned}$$

The results obtained on the line shift of Cs agree qualitatively with the results of^[3], except that the shift is approximately 25% larger. The difference is apparently due to the fact that the saturated-vapor density was not reached in^[3] (cesium reacts strongly with glass, and in^[3] we used a glass tube with a long stub).

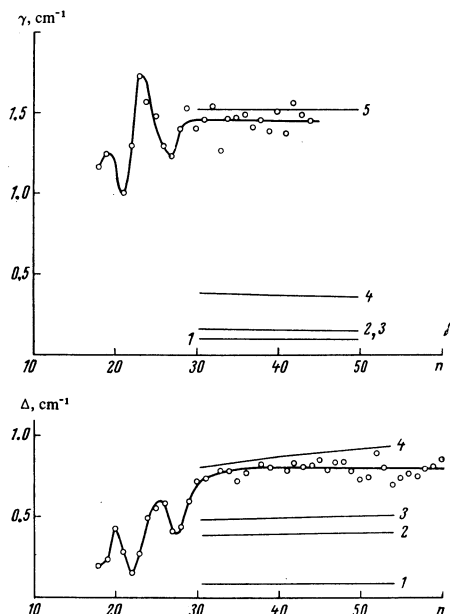


FIG. 2. Width γ and shift Δ of the cesium lines ($N = 1.3 \times 10^{17} \text{ cm}^{-3}$, $T = 366^\circ\text{C}$) as functions of the principal quantum number. \circ — experimental results; the designations of the curves 1–5 are the same as in Fig. 1.

Table I. Line shift of K at different atom densities (in arbitrary units).

$T, ^\circ\text{C}$	$10^{17} N, \text{ cm}^{-3}$	Δ/N			
		$n = 20$	$n = 25$	$n = 30$	$n = 35$
466	2.0	0.49	0.47 ₅	0.44	0.41
473	2.3	0.48	0.48	0.44	0.40 ₅
481	2.6	0.48	0.46	0.41	0.40
520	4.8	0.50 ₅	0.47	0.44	0.42

DISCUSSION OF RESULTS

The condition for statistical independence of the effects of polarization and scattering is given by [2]

$$\rho_i = \left(\frac{\pi \alpha e^2}{4\hbar V} \right)^{1/2} \ll a_0 n^2. \quad (1)$$

Here α is the polarizability and V is the relative velocity of the perturbing particles.

Assuming the polarizabilities of the potassium and cesium atoms to be respectively $\alpha^{\text{K}} = 41 \text{ \AA}^3$ and $\alpha^{\text{Cs}} = 53 \text{ \AA}^3$, [12] we find that condition (1) is satisfied for both atoms, starting with $n \gtrsim 30$. At a particle density $N \sim 10^{17} \text{ cm}^{-3}$, the impact approximation is valid, and the polarization width and shift are equal to [2]

$$\gamma_p = 11.4 \left(\frac{\alpha e^2}{2\hbar} \right)^{1/2} V^{1/2} N; \quad \Delta_p = -\frac{\sqrt{3}}{2} \gamma_p. \quad (2)$$

For the K lines we obtain at $T = 481^\circ\text{C}$

$$\gamma_p/N = 0.078 \cdot 10^{-17} \text{ cm}^{-1}/\text{cm}^{-3}, \quad \Delta_p/N = 0.067 \cdot 10^{-17} \text{ cm}^{-1}/\text{cm}^{-3},$$

For the Cs lines at $T = 360^\circ\text{C}$

$$\gamma_p/N = 0.074 \cdot 10^{-17} \text{ cm}^{-1}/\text{cm}^{-3}, \quad \Delta_p/N = 0.063 \cdot 10^{-17} \text{ cm}^{-1}/\text{cm}^{-3}.$$

In Figs. 1 and 2, the quantities γ_p and Δ_p calculated from formulas (2) are represented by the lines—(γ_p and Δ_p) are independent of n).

It follows from the figures that in the case of line broadening by alkali atoms the polarization contribution to both the shift and to the width is quite negligible, whereas in the interaction with an inert gas the polarization contribution to the width was the main one. [4]

The contribution made to the width and to the shift by the effect of scattering of an optical electron is expressed in terms of the scattering phase shifts δ_l or the cross section σ : [2]

$$\begin{aligned} \gamma_p &= N \frac{\hbar}{m} \sum_{S_T} C(S_T) \int \left[\frac{4\pi}{q} \sum_l (2l+1) \sin^2 \delta_l \right] W(q) dq \\ &= N \frac{\hbar}{m} \int q \sigma(q) W(q) dq; \end{aligned} \quad (3)$$

$$\Delta_p = N \frac{\hbar}{m} \sum_{S_T} C(S_T) \int \left[\frac{\pi}{q} \sum_l (2l+1) \sin 2\delta_l \right] W(q) dq. \quad (4)$$

Here $C(S_T) = (2S_T + 1)/2(2s + 1)$, where s is the spin of the perturbing atom, and the values of the total spin of the system $S_T = |s \pm 1/2|$ correspond to the antisymmetrical and symmetrical scatterings; $W(q)$ is the distribution function with respect to the momenta $\hbar q$ of an electron in the state n .

The quantities γ_c and Δ_c for cesium lines perturbed by cesium atoms were calculated by Alekseev and Vainshtein [5] under certain approximations. Analogous calculations were carried out for potassium. [1] In Figs. 1 and 2, curves 2–4 correspond to the results of the calculations of γ_c and Δ_c in different approximations (the figures show the quantities $\gamma = \gamma_c + \gamma_p$ and $\Delta = \Delta_c + \Delta_p$). These results will be discussed in greater detail below. At present we wish to call attention to the fact that the theoretical values of the shift are close to the experimental ones, but the calculated widths are much smaller than the measured ones.

From the general expression (3) it follows that $\gamma_c \rightarrow 0$ as $q = 1/na_0 \rightarrow 0$. As is seen from the figure, the width, if it does decrease with increasing n , does so very slowly. In addition, the line width in the case of perturbation by alkali atoms is several times larger than the shift, whereas in the case of broadening by argon and other inert gases [4, 13] there was always observed $\gamma < \Delta$. As follows from (3) and (4), at large scattering phase shifts the ratio γ_c/Δ_c can, generally speaking, be larger than unity. However, the scattering phase shifts that follow from the experimental data (from the simplified expressions for γ_c , Δ_c , and γ_c/Δ_c as $q \rightarrow 0$) differ from one another quite appreciably.

Thus, the theory of broadening of lines with large n upon perturbation by alkali atoms does not describe the line width, although no serious contradictions arise in the description of the shift. It is apparently necessary to take into consideration some new phenomena which are not taken into account by the Fermi mechanism. To this end it is necessary to consider a number of interaction processes capable of making an additional contribution to the line width and shift.

1. At temperatures 300 – 500°C , at which the measurements were performed, there are several percent of molecules in the alkali-metal vapors. [14] It is difficult to estimate the contribution of the molecules to the line

¹⁾The authors are most grateful to V. A. Alekseev for performing these calculations.

broadening. At the same temperatures, however, when the atom density changes by a factor of 2, the density of the molecules changes by an approximate factor of 3. The proportionality of the observed width and shift to the density of the atoms demonstrates that the contribution of the molecules to the line broadening is quite small.

2. When an excited atom interacts with a like unexcited atom, resonant broadening takes place. Its magnitude is proportional to the oscillator strength of the transition.^[15] With increasing n , the oscillator strengths of the transitions in the principal series of alkali-metal atoms decrease very rapidly, and the resonant broadening turns out to be significant only for the resonant transition.

3. When the core of a strongly excited atom collides with perturbing atoms, symmetrical charge exchange can occur: $A^+ + A = A + A^+$. Such a process shortens the lifetime of the excited atom and increases the line width. The cross section of such a process is about 4×10^{-14} cm²,^[16] and the width due to it turns out to be smaller than the observed one by approximately two orders of magnitude.

4. It is known that in the presence of excited cesium atoms there is observed associative ionization: $Cs^* + Cs \rightarrow Cs_2^* \rightarrow Cs_2^+ + e$.^[17] The process is due to interatomic forces, and it can be assumed that it is significant only on a relatively slight excitation of the cesium atoms, namely at such values of n where the interaction of the atoms can be described at least approximately by Van der Waals forces.

Indeed, the process of associative ionization is not observed in potassium,^[18] and the behavior of the lines at $n > 30$ in potassium and cesium is approximately the same. Associated with the indicated process may be the nonresonant behavior of the line width and shift of cesium at values near $n \sim 20-30$, which is not observed in potassium.

5. Associative ionization is an example of a process that is capable, under the conditions of our experiment, of producing charged particles. Owing to the relatively low temperature ($T \lesssim 800^\circ\text{K}$), the generation of charged particles can proceed only via excited states or via direct photoionization. The density of such particles should depend on the power of the radiation entering the tube. Special control measurements were made by placing filters that attenuate the light intensity by a factor of 5 in front and behind the absorption tube. In both cases identical line widths were observed, and consequently the contribution of the charged particles to the line broadening is negligible.

6. Presnyakov^[19] proposed the following explanation of the phenomenon. The interaction of a strongly excited atom A^* with an unexcited atom B can lead to a strong line broadening, if the atom B is capable of producing a negative ion. (As is well known, the alkali metal atoms, unlike inert-gas atoms, have a large electron affinity.) The presence of a short-range potential for such atoms leads to a splitting of the initially degenerate levels of the atom A^* and to transitions between these sublevels.

Komarov^[20] investigated the splitting of the Coulomb terms of an atom A^* under the influence of the short-range potential of an atom B . From the magnitude of

the splitting of the energy levels ΔE , as is well known^[21, 15] it is possible to calculate the phase shift

$$\eta(\rho) = \frac{1}{V} \int_0^\infty \Delta E_n(\rho) \frac{RdR}{\sqrt{R^2 - \rho^2}}$$

where ρ is the impact parameter, and the cross sections for the width σ' and for the shift σ'' :

$$\sigma' = 2\pi \int_0^\infty \rho d\rho \sin^2 \eta(\rho), \quad (5)$$

$$\sigma'' = \pi \int_0^\infty \rho d\rho \sin 2\eta(\rho). \quad (6)$$

It turns out that $\eta(\rho) \gg 1$ up to $\rho \sim 2n^2$. Then in the effective integration region of (5) and (6) we have $\sin^2 \eta(\rho) = 1/2$ and the line-broadening cross section due to this interaction is of the order of the dimensions of the excited atom, while the shift cross section is $\sigma'' \sim \overline{\sin^2 \eta(\rho)} = 0$ (there is no line shift).

The numerical values of the width, calculated in^[19] for K and Cs (without allowance for the deviation from the hydrogen-like behavior) are shown in Figs. 1 and 2 (curve 5). (We present the calculated curves of^[19] at electron affinities 0.35 eV for K and 0.19 eV for Cs.) A comparison with the experimental data shows that the considered effect of inelastic scattering by the short-range potential makes the main contribution to the line width.

Thus, if the perturbing particles are atoms of alkali metals, then the main part of the line width is not determined by the particle polarization in the field of the atomic core, as is the case with interactions with inert-gas atoms. Scattering by the short-range potential of atoms capable of forming negative ions makes the main contribution to the line width, leaving the shift unchanged.

The line shift is completely described by the Fermi mechanism. The main contribution to the shift is made by elastic scattering of the electrons by the perturbing particles. Consequently, the experimental data on the line shifts can give information on the elastic scattering of electrons by the perturbing atoms also when the latter are those of alkali metals.

The experimentally measured line shift is not directly connected with the cross section for the elastic scattering of the electrons by the atoms. A determination of even the limiting cross section $\sigma(0)$ from the value of the shift $\Delta_C(0)$ by the Fermi formula^[11] is possible only if the scattering at zero is described by one wave. In the case of alkali-metal atoms at $q = 0$ (see formula (4)) there always remain two waves corresponding to symmetrical and antisymmetrical scatterings. The scattering amplitudes can make contributions of opposite signs to the line shift, and the squares of the amplitudes are summed in the scattering cross section. The error resulting from the use of the Fermi formula for atoms with nonzero spin will depend on the relative contribution of the scattering amplitudes at a total spin $s + 1/2$ and $s - 1/2$. According to the available calculations, this ratio can vary in a very wide range, thereby causing a large uncertainty in the value of $\sigma(0)$.

We shall compare the results of the theory with experiment by using the magnitude of the line shift (see formula (4)).

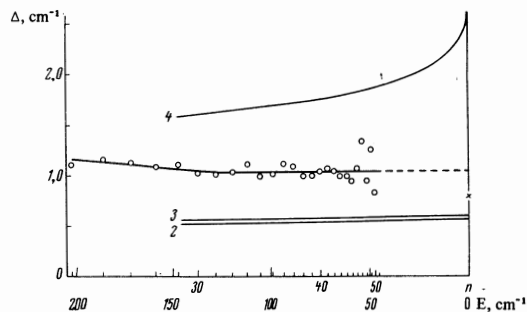


FIG. 3. Shift of potassium lines as a function of the energy E of the upper level with principal quantum number n . \circ —experimental results, \times —calculation [22] by the strong-coupling method (states n_0s-n_0p); the designations of curves 2–4 are the same as in Fig. 1.

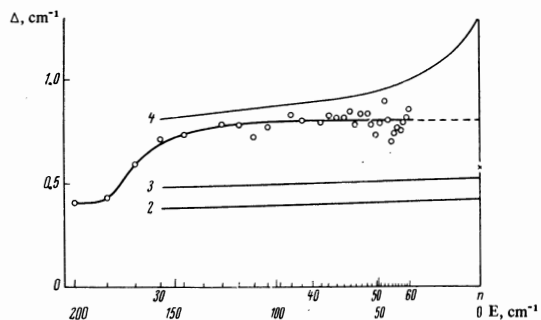


FIG. 4. Shift of cesium lines as a function of the energy E of the upper level with principal quantum number n . \circ —experimental results, \times —calculation [22] by the strong-coupling method (states n_0s-n_0p); the designations of curves 2–4 are the same as in Fig. 1.

To this end, it is more convenient to represent the experimental line shift as a function of the energy of the level with the corresponding quantum number n (Figs. 3 and 4).

The accuracy of the experimental results becomes worse with decreasing energy (at large n the absorption decreases, and the neighboring lines begin to overlap) and it is generally impossible to measure the line shifts when $n > 60$. Extrapolation of the experimental data to zero energy yields the quantity $\Delta(0)$, which is directly connected with the scattering length. Extrapolation cannot claim high accuracy, but it is useful for a comparison with the theory.

The theoretical curves (see footnote 1¹) of $\Delta(E)$ were obtained using the scattering phase shifts calculated in the non-adiabatic approximation with allowance for exchange and for the contribution made to the polarization potential by the first (curve 2) and by the first two (curve 3) p states. Curve 4 corresponds to allowance for one p level, but without allowance for exchange in the polarization potential. In the latter case, the shift increases very rapidly with decreasing energy. On the boundary of the series it is considerably overestimated (the values $\Delta_c(0)$ are listed in Table II). When a more complete account is taken of the exchange (curves 2 and 3), the correct plot of the shift against the energy is obtained, but the absolute values are too low. Values of $\Delta_c(0)$ that are somewhat too low are also obtained from the scattering data calculated by Karule^[22] (calculation

Table II. Theoretical and experimental values of the line shift on the boundary of the series ($n = \infty$).

Source	$\Delta_p(0)/N, 10^{17} \text{ cm}^{-1}/\text{cm}^{-3}$	
	K	Cs
Our results*	0.33	0.55
Calculation by the strong-coupling method (states n_0s-n_0p) [22]	0.23	0.37
Calculation [5]		
a) with allowance for exchange and the contribution of the state n_0p to the polarization	0.15	0.26
b) without allowance for exchange in the polarization potential	0.92	0.92
c) with allowance for exchange and the contribution of the states n_0p and $(n_0 + 1)p$ to the polarization	0.16	0.34

$$*\Delta_c = \Delta_{\text{meas}} - \Delta_p.$$

by the strong-coupling method for the states n_0s and n_0p).

Thus, the existing approximations for the theoretical calculation of the scattering phase shifts lead to results that differ from the experimental data by not more than a factor of 2. It is important to note that the results of rougher approximations, for example the adiabatic approximation,^[5] are in qualitative contradiction to the presented experimental data.

The experimental method employed in the present work makes it possible to assess the quality of the approximate methods of calculating the phase shifts of elastic scattering of electrons at extremely low energies, where the sensitivity to the approximations employed in the calculation, at least for alkali-metal atoms, is exceedingly high.

At high energies ($E \gtrsim 0.1$ eV), where there are experimental methods for measuring directly the cross sections for elastic scattering of electrons by atoms, the differences in the values of the cross sections obtained by different calculation methods are much smaller and actually lie within the limits of experimental error (see [5]).

In conclusion, the authors are deeply grateful to V. A. Alekseev, L. A. Vaĩnshteĩn, S. L. Mandel'shtam, L. P. Presnyakov, and I. I. Sobel'man for interest in the work and for a discussion of the results.

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Translated by J. G. Adashko