EFFECTIVE CROSS SECTIONS FOR ELECTRON EXCITATIONS OF TRANSITIONS BETWEEN EXCITED STATES OF THE IONS Ba^+ *AND* Sr^+

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The effective cross sections for inelastic scattering of electrons by excited Ba^+ and Sr^+ ions are obtained from data on the electronic width of the corresponding spectral lines, using the method proposed by Sobel'man[1]. The investigations were performed with the aid of a shock tube, and the electron-concentration values needed for the calculations were determined by two independent methods. The experimentally obtained values for the cross sections are in good agreement with the results of a calculation by the Born-Coulomb approximation with normalization.

1. INTRODUCTION

KNOWLEDGE of the effective cross sections for inelastic collisions of electrons with atoms or ions is essential for the solution of many problems in plasma physics, quantum generators, astrophysics, etc. An experimental determination of the cross sections is practically impossible for many cases of interest. It is therefore very important to verify, using a number of the most accessible cases, the approximate theoretical methods of calculating the cross sections. There has been particularly little study of the transitions between excited states of ions. No direct measurements of such cross sections have been made so far.

One of the few possibilities of indirectly determining the cross sections is to investigate the broadening of the spectral lines. The theoretical foundations of this method were considered by Sobel'man[1]. The width of the spectral line connected with the transition $k \to n$ (Fig. 1) is determined by perturbation of the levels k and n by the nearest neighboring levels. If it is possible to separate one nearest level m such that the dipole interaction of the levels n—m plays the predominant role, then the cross section of this interaction can be determined from the measured values of the electronic line width γ_e, the electron density N_e, and the temperature T. If in addition

$$\beta = \left(\frac{f\,\mathrm{Ry}}{|\Delta E|}\right)^{1/2} \frac{\pi |\Delta E|}{8kT} \ll 1, \qquad (1)$$

then the principal role is played by the inelastic transitions, i.e., we obtain the cross section for the inelastic transition between the nearest excited levels of the atom or ion. In (1), f and ΔE are the oscillator strength and the energy of the $n \to m$ transition, and Ry = 13.6 eV.

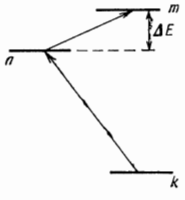

FIG. 1. Level and transition scheme explaining the measurement method of [1].

Of course, under plasma conditions one measures the cross section averaged over the Maxwellian velocity distribution: $\langle v\sigma \rangle$. The inequality (1) limits the possible values of ΔE from above. On the other hand, for a comparison with the theory, greatest interest attaches to the largest possible ΔE, for at very small ΔE the Born approximation is apparently valid. The need for satisfying both requirements greatly limits the choice of levels.

An investigation of the transitions in the atom HeI by the indicated method was carried out in[2,3]. In the present paper we describe the experimental procedure and the results for transitions in the ions Ba^+ and Sr^+. We also calculate the measured cross sections by various approximate methods. The preliminary results of this work were reported in[4,5].

2. FORMULATION OF EXPERIMENT

A feature of our work is an investigation of the width of the lines excited in a shock tube, where it is possible to obtain a plasma with a controllable electron concentration. The values of N_e, and also the temperature, pressure, and other parameters can be calculated on the basis of the gas-dynamic relations using the readily-measured velocity of the incident shock wave. In addition, the relatively good homogeneity of the investigated plasma sample, and the absence of electric and magnetic fields, make it possible to measure N_e reliably by an independent method, (we performed such measurements on the basis of the half-width of the H_β line).

The values $N_e \sim 10^{17}$ cm^{-3}, at which the electronic part of the line width exceeds the other contributions to the width, were attained in the shock tube by heating argon to $T \sim (1-1.5) \times 10^4$°K with a reflected shock wave.

The choice of the investigated ions and lines was dictated primarily by the condition that there be sufficiently intense lines in the visible part of the spectrum. The lower level (k, Fig. 1) should in this case lie sufficiently high so that the cold boundary layer of the gas in the shock tube will make no contribution to the absorption (we investigated the absorption spectrum, see below). Finally, as already noted, there

should exist a perturbing level m, making the main contribution to the width, satisfying the condition (1), and having not too small a ratio $\Delta E/kT$. The lines $\lambda = 4525$ Å $Ba^+(6^2P_{1/2} \rightarrow 7^2S_{1/2})$ and $\lambda = 3465$ Å Sr^+ $(5^2P_{3/2} \rightarrow 5^2D_{5/2})$ satisfy these conditions sufficiently well (Figs. 2 and 3). In these cases the broadening is due mainly to perturbation of the upper level by the levels 7^2P and 6^2P, respectively. Thus, measurement of the line widths makes it possible to estimate the value of $\langle v\sigma \rangle$ for the transitions $7^2S \rightarrow 7^2P$ in Ba^+ $(\Delta E/kT \approx 0.9)$ and $5^2D_{5/2} \rightarrow 6^2D_{3/2}$ in Sr^+ $(\Delta E/kT \approx 0.4)$.

3. EXPERIMENTAL PART

A diagram of the experimental setup is shown in Fig. 4. Here ST is a shock tube with three optical windows near the end.

The vertical windows, the lenses L_1, L_2, L_3, the rotating mirrors M_1 and M_2, and the slotted disc D_1 were used to photograph the absorption spectrum of the gas heated by the reflected wave, using a DFS-13 spectrograph. The light source was a powerful pulsed discharge in a capillary tube (the EV-45 instrument). The horizontal window, the rotating system RS, the lens L_4, the slotted disc D_2, and the spectrograph ISP-51 with a camera having f = 270 mm were used to register the time-resolved emission spectrum of the gas heated by the reflected wave.

Shock Tube

We used a stainless-steel shock tube (inside diameter 90 mm, lengths of high- and low-pressure sections 1 m and 3.2 m respectively). The propelling gas was hydrogen. The shock wave propagated in argon (initial pressure 5.0 mm Hg) with 5% hydrogen and traces of $BaCl_2$ or $SrCl_2$. The latter compounds were deposited on the internal walls of the tube by vacuum evaporation of the aqueous solution. In the plasma of the investigated gas heated by the reflected wave, as shown by the reduction of the obtained absorption spectra, the amount of Ba^+ or Sr^+ impurity was about 0.01%. The Ba- or Sr-compound impurity actually had no influence on the heat contents of the mixture and on the electron concentration N_e.

In order to verify that the Ba^+ (or Sr^+) ions are distributed sufficiently homogeneously inside the tube volume occupied by the gas heated by the reflected wave, additional experiments were performed. First, the usual photographic scanning method with a filter for the Ba^+ lines has shown that when the glow of Ba^+ is observed from the side, it remains practically homogeneous over the entire length of the heated layer of gas and during the entire lifetime of this layer (~ 100 μsec) up to the arrival of the perturbation from the contact region. Second, it was established that the distribution of the glow of Ba^+ along the radius of the shock-tube cross section (averaged over the thickness of the layer of the heated gas) is constant and does not change during the time interval needed by us.

The velocity of the incident shock wave u_s = 3.0–3.5 km/sec was measured with the aid of four ionization pickups located in succession along the shock tube; the pickup signals were recorded with an oscilloscope

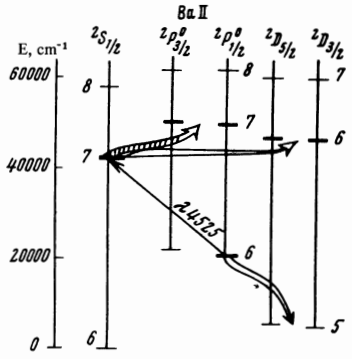

FIG. 2. Level scheme of Ba^+. The arrows denote transitions making a contribution larger than 10% to the broadening of the line $\lambda = 4525$.

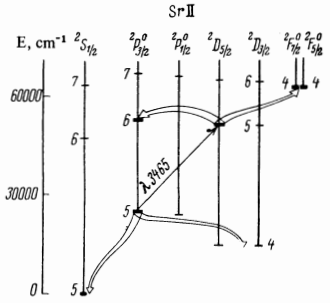

FIG. 3. Level scheme of Sr^+. The arrows denote transitions making a contribution larger than 10% to the broadening of the line $\lambda = 3465$.

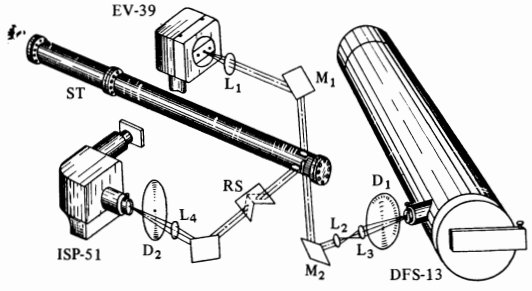

FIG. 4. Diagram of experimental setup.

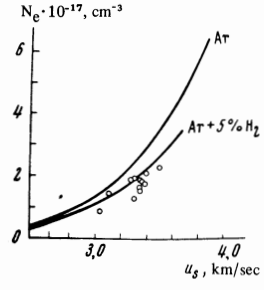

FIG. 5. Dependence of the electron concentration N_e behind the reflected wave on the velocity of the incident shock wave.

with a spiral sweep. The value of u_S increased smoothly and reached near the end an almost constant value, which was measured accurate to about 0.5%.

Measurement of the Electron Concentration N_e

The electron concentration N_e is one of the most important parameters. This parameter was determined by a gas-dynamic computer calculation using the program described in[6]. Figure 5 shows plots of $N_e(u_S)$ for Ar + 5% H_2 and pure Ar. As is well known, the gas parameters actually observed in a shock tube, especially behind the reflected wave, deviate appreciably from the calculated ones. The values of these

parameters can vary significantly with increasing distance from the end of the tube, i.e., in those sections where the existing one-dimensional theory predicts constant values for them (see, for example,[7]).

We have therefore made independent measurements of N_e by determining the width of the hydrogen line H_β (in emission). The measurement setup, which is also shown in Fig. 4, is analogous to that used in[8] and was supplemented by a rotating system (RS). It made it possible to focus the entire optical-window slit located along the tube axis onto the input slit of the spectrograph. Therefore the radial slots of the disc, which moved transversely to the spectrograph slit, provided both temporal resolution (~ 30 μsec/mm) and spatial resolution (~ 5 mm) of the obtained spectra. N_e was measured at the same distance from the end of the tube (~ 10–15 mm) and at the same instants of time as the Ba^+ or Sr^+ absorption line width. Since the half-width of H_β was 60–120 Å, a correction was made for the variability of the continuous spectrum under the line. A special check has shown that H_β is not registered in absorption, i.e., the optical thickness for H_β is sufficiently small.

The value of N_e was determined from the measured width of H_β in accordance with[9]. The results represented by the circles in Fig. 5 confirm the course of the function $N_e(u_s)$, but give on the average values of N_e that are lower by 15%. The scatter of the experimental points turns out to be much larger than the corresponding scatter in the widths of the Ba^+ and Sr^+ lines. We therefore used the theoretical values of N_e reduced by 15% in the succeeding calculations.

Determination of the Electronic Widths γ_e of the Ba^+ and Sr^+ Lines

The widths of the chosen lines were determined by measuring the absorption spectra obtained by passing light through the investigated gas mixture, heated by the reflected shock wave (Fig. 4). The absorption method has made it possible to use a spectrograph with low transmission but with large dispersion (2 Å/mm) in conjunction with a time resolution of 10–20 μsec. The latter is essential, since the "pure conditions" of heating by the reflected shock wave existed for 50–100 μsec. We used a powerful pulsed light source EV-45, which gave single rectangular light pulses of duration from 150 to 300 μsec. Its radiation is close to that of an absolutely black body with a brightness temperature 39,000°K. The EV-45 source was synchronized with the aid of a ionization pickup. The time resolution was produced, as in the registration of H_β, by using a rotating disc with slots (0.5 mm wide each) mounted at a distance ~2 mm from the input slit of the spectrograph.

The obtained absorption spectra described a distinct picture of the phenomena occurring at the end of the shock tube, namely, the arrival of the incident wave, then the arrival of the reflected wave, and the subsequent arrival of the perturbations distorting the "pure" conditions. The investigated lines of Ba^+ (λ = 4525 Å) and Sr^+ (λ = 3465 Å) could not be seen behind the incident wave. Near the front of the reflected wave (~ 30 μsec), the line width changed somewhat, after which it remained constant up to the arrival of the perturbations from the contact surface, following which the indicated lines vanished. The measurements were made photometrically using sections of constant line width; the measurement results are summarized in Table I.

Table I. Summary of experimental data (line width in Å, $\delta\lambda = \lambda^2\gamma/2\pi c$)

u_s km/sec	T, 10^3 °K	N_e 10^{17} cm^{-3}	$\delta\lambda$, Å	$\delta\lambda_e$, Å	γ_e/N_e 10^6 cm^3/sec
		Ba^+			
3.11	12.0	1.16	0.60	0.36	2.93
3.37	12.5	1.74	0.77	0.51	2.74
3.39	12.6	1.80	0.85	0.58	3.02
3.47	12.8	2.06	1.20	0.90	3.99
3.51	12.9	2.21	1.21	0.91	3.99
					Av. 3.33
		Sr^+			
3.27	12.3	1.48	0.57	0.40	4.25
3.28	12.3	1.50	0.57	0.40	4.35
3.35	12.5	1.68	0.70	0.51	4.79
3.37	12.5	1.74	0.67	0.49	4.41
3.40	12.6	1.83	0,60	0.42	3.67
					Av. 4.29

All the processed line contours have a more or less pronounced asymmetry connected with the static broadening by the ions. According to[10], the shock width γ is approximately equal to double the value of the "blue" half width of the line. In Table I, the experimental values of γ in sec^{-1} correspond to the values of $\delta\lambda$ expressed in Å.

We then subtracted the apparatus, Doppler, and Van der Waals half-widths from the obtained value of γ. The Van der Waals constant for the interaction with neutral Ar atoms was calculated in accordance with[11], and was also determined for Ba^+ from the broadening under the conditions of low ionization (T ~ 8,000°). The measured value (1.0×10^{-29} cm^6/sec) is approximately double the calculated value, but such a difference has little effect on the results.

The half-width obtained after the subtraction is due to the shock broadening by electrons and ions. To obtain γ_e, the ratio γ_i/γ_e was estimated in accordance with the quasiclassical theory[12] and amounted to ~ 0.1. The final values of γ_e correspond to the values of $\delta\lambda_e$ given in Table I. We see that the difference between $\delta\lambda_e$ and $\delta\lambda$ and accordingly between γ_e and γ is not very large, so that the requirements with respect to the accuracy with which the effects listed above are estimated are quite moderate.

Comparison with Theoretical Calculations

The electronic width γ_e is due, generally speaking, to the influence of a number of perturbing levels acting on both the upper and lower states of the optical transition. Estimates show that although it is possible for the two observed lines to separate the principal perturbing level, which contributes approximately 50% of the entire broadening effect, one cannot neglect the remaining levels.

The contribution of each perturbing level is connected a) with inelastic transitions to this level, and b) with elastic scattering by the corresponding part of the polarization potential.

The cross sections for the inelastic collisions were calculated by us in the Born-Coulomb approximation

with normalization by the R-matrix method. For an alkaline-earth ion, the cross section is represented by

$$\sigma = \sum_{l_0 l_1} \frac{\sigma^{BC}(l_0 l_1)}{(1+R^2)^2}, \quad R^2 = \frac{k_0^2}{4\pi(2l_0+1)}\sigma^{BC}(l_0 l_1), \qquad (2)$$

where $\sigma^{BC}(l_0 l_1)$ is the partial cross section for the electrons with initial angular momentum l_0 and wave number k_0. It is calculated in the first order of perturbation theory with allowance for the distortion of the wave function by the Coulomb field of the ion. The calculation method was described in greater detail in[13].

The values of $\langle v\sigma_{nm} \rangle$ obtained in this manner for transitions to different perturbing levels are given in Table II for $T = 12.5 \times 10^3$°K. Figure 6 shows the dependence of the total frequencies of the elastic collisions $\langle v\sigma^{in} \rangle$ on the temperature.

It should be noted that in the calculation of R^2 in (2) it is actually necessary to sum over all the levels, so that the normalization effect is somewhat undervalued in our calculations. A rough estimate of this effect gives the values listed in Table III, which also gives the values of $\langle v\sigma^{in} \rangle$ calculated without allowance for the renormalization.

In comparing the calculation results with the experimental data (Table III) it must be recognized that the parameter β, determined in formula (1) for the main perturbing level, differs significantly in the cases of Ba^+ and Sr^+ (see Table II). In the case of Ba^+ we have β (7S – 7P) = 1.74. This means that the interaction is close to stationary, the elastic scattering should prevail over the inelastic one, and the elastic and inelastic scatterings are not additive, so that it is difficult to estimate the role of the elastic scattering. The quantity $\langle v\sigma^{in} \rangle$ should then give only the lower limit of γ_e. Consequently, as seen from Table III, the quantum-mechanical value of $\langle v\sigma^{in} \rangle$ is overestimated by at least 50%. It is interesting to note that a calculation by the quasiclassical method in accordance with[12] yields $\gamma_e/N_e = 3.4 \times 10^{-6}$ cm^3/sec, which is in good agreement with the experimental data.

In the case of Sr^+ we have β (5D – 6P) = 0.13, the interaction is nonstationary, $\sigma^e < \sigma^{in}$, and it can be assumed that σ^e and σ^{in} are additive. To estimate the total value of $\langle v\sigma^t \rangle$ we used the inequality

$$\langle v\sigma^t \rangle \leqslant \langle v\sigma^{in} \rangle \left[1 + \frac{\langle v\sigma^e \rangle}{\langle v\sigma^{in} \rangle}\right], \qquad (3)$$

where the term in the square bracket was calculated by the quasiclassical method of[12] without allowance for the possible cancellation of the contributions of different levels to σ^e (this explains the inequality sign in (3)). The results obtained in this manner are in good agreement with the experimental data.

Thus, the Born-Coulomb approximation with allowance for the normalization at $E \sim \Delta E$ results in an error of $\lesssim 50\%$. Without allowance for the normalization (see Table III), the cross section is highly overvalued. An estimate shows that neglect of the effect of the Coulomb field leads to approximately the same error, but such as to decrease the cross section.

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Table II. Calculated rate of inelastic transitions to the perturbing levels m

Level n	Level m	β_{nm}	$\langle v\sigma_{nm} \rangle$ 10^6 cm^3/sec
	Ba^+		
7S	7P	1.74	2.5
	6D	—	1.3
	6P	1.80	0.1
	8P	0.10	< 0.1
6P	5D	1.00	0.6
	6S	1.46	0.6
	7S	1.06	< 0.1
	Sr^+		
5D	6P	0.13	1.7
	4F	0.16	0.7
	5P	0.45	0.3
	6S		0.1
	7P	0,02	< 0.1
	5D		< 0.1
5P	4D	0.44	0.7
	5S	0.30	0.5
	5D	0.51	< 0.1
	6S	0.70	< 0.1

Table III. Comparison of the calculated value of $\langle v\sigma \rangle$ (in units of 10^{-6} cm^3/sec) with the experimental data

$\langle v\sigma^{in} \rangle$ normalization in accord with (2)	$\langle v\sigma^{in} \rangle$ total normalization (estimate)	$\langle v\sigma^{in} \rangle$ without normalization	$\langle v\sigma^t \rangle$ normalization in accord with (2)	$\langle v\sigma \rangle$ experiment
		Ba^+ ($\lambda = 4525$ Å)		
5.3	4.6	8.0	—	3.3
		Sr^+ ($\lambda = 3465$ Å)		
4.1	3.8	8.0	< 5.5	4.3

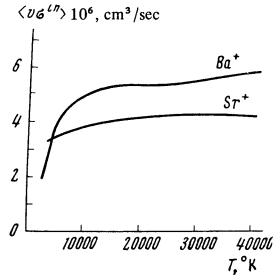

FIG. 6. Temperature dependence of $\langle v\sigma^{in} \rangle$ for the ions Ba^+ and Sr^+.

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