PRODUCTION OF HIGHLY EXCITED HYDROGEN MOLECULES AND ATOMS BY FAST H_2^+ AND H_3^+ IONS PASSING THROUGH H_2, Ne, AND Mg AND Na VAPOR

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The formation of highly excited H_2^* molecules and H^* atoms (principal quantum numbers $n > 8$) by passage of fast 20–180 keV H_{2+} and H_3^* ions through Ne, H_2, and Na and Mg vapor is investigated. The cross sections for formation of highly excited hydrogen molecules ($\sigma_{H_2^*}$) and hydrogen atoms ($\sigma^n_{H^*}$) are measured. The yield of highly excited molecules and atoms relative to the primary ion beam as a function of target thickness (H_2 or Mg vapor) is also measured. The yields of highly excited H^* atoms for various fast ions (H^+, H_2^+, H_3^+) and various targets are compared. The most effective target for highly excited hydrogen atom production is Mg vapor. In this case the greatest yield of highly excited atoms with energies below 40 keV is obtained as a result of fast proton charge exchange. At higher energies, dissociation of fast H_3^+ ions is the more effective process.

INTRODUCTION

THE present paper is a continuation of a cycle of investigations aimed at obtaining highly excited hydrogen atoms (principal quantum numbers $n > 8$) in atomic collisions [1-3]. The purpose of these investigations was to find inelastic processes that are the most effective in this respect. The earlier investigations [1-3] were devoted to obtaining highly excited atoms by charge exchange of fast protons with different atoms and molecules. It was observed that at proton energies 10–50 keV the best charge-exchange targets are vapors of alkali and alkali-earth metals. At higher energies, the charge exchange cross section is relatively small, and gas targets are comparatively more convenient (H_2, N_2, inert gases).

Since, as follows from [4,5], the dissociation of fast H_2^+ ions gives at energies on the order of 50 keV/proton a larger yield of hydrogen atoms than charge exchange of fast protons, it can be expected that at these energies the dissociation is also a more convenient means for obtaining highly excited atoms. In this connection, we investigated in this study the formation of highly excited hydrogen atoms by collision of fast H_2^+ and H_3^+ ions with different target particles. The targets were chosen to be typical representatives of different groups: molecular gas (H_2), inert gas (Ne), alkali metal vapor (Na), and alkali earth metal vapor (Mg). This choice of target makes it possible to draw more general conclusions on the qualitative features of the production of highly excited H^* atoms connected with the structure of the target particles.

In general form, the processes of dissociation and charge exchange in collisions of fast H_2^+ ions with atoms or target molecules M can be written in the form of the following reactions:

$$H_2^+ + M \to H + H^+ + M, \quad (1)$$

$$H_2^+ + M \to 2H^+ + e + M, \quad (2)$$

$$H_2^+ + M \to 2H + M^+, \quad (3)$$

$$H_2^+ + M \to H_2 + M^+. \quad (4)$$

The cross sections of these reactions are denoted respectively σ_1, σ_2, σ_3, and σ_4. Then the cross section σ_{H^+} for proton production and the cross section σ_H for the production of hydrogen atoms are expressed in terms of the cross sections of these reactions in the form

$$\sigma_{H^+} = \sigma_1 + 2\sigma_2, \quad (5)$$

$$\sigma_H = \sigma_1 + 2\sigma_3. \quad (6)$$

The cross section of reaction (4) is the cross section for the production of neutral hydrogen molecules, i.e., $\sigma_4 \equiv \sigma_{H_2}$. The total neutralization cross section is defined as

$$\sigma_0 = \sigma_1 + 2\sigma_3 + 2\sigma_4 = \sigma_H + 2\sigma_{H_2}. \quad (7)$$

The highly excited hydrogen atoms H^* can be produced as a result of reactions (1) and (3). We shall denote the corresponding cross section for the production of H^* by $\sigma^n_{H^*}$.

Barnett and Ray [6] and the present authors [2] have observed that the charge exchange of fast molecules of hydrogen ions without dissociation (reaction (4)) can lead to the formation of highly excited molecules H_2^* (electron excitation). We shall denote the cross section for the production of H_2^* by $\sigma^n_{H_2^*}$. The excited atoms H^* and molecules H_2^* can also be produced as a result of dissociative processes occurring with fast H_3^+ ions.

In the present paper we investigate more systematically the features of the production of such highly excited particles by collision between fast molecular hydrogen ions in gases (H_2, Ne) and in metal vapors (Mg, Na).

EXPERIMENTAL SETUP AND MEASUREMENT PROCEDURE

The experimental setup was described earlier in [1,2] and consists essentially of the following: A monoenergetic beam of fast molecular hydrogen ions passes through a collision chamber filled with metal vapor or gas. After passing through this chamber, the beam of fast neutral particles, from which the charged fraction

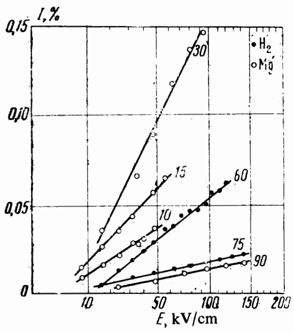


FIG. 1. Dependence of the relative yield I of the highly excited molecules H_2^*, produced upon collision of fast ions H_2^+ with molecular hydrogen H_2 and with Mg vapor, on the ionizing electric field E. The number on the curves denote the energy in keV/proton.

was eliminated by a weak transverse electric field, enters a region with a strong longitudinal electric field $E \le 160$ kV/cm, where the highly excited atoms and molecules experience Lorentz ionization. The H^+ and H_2^+ ions produced in this case are diverted with the aid of a magnetic analyzer into a Faraday receiver, and the neutral-particle beam is recorded via secondary electron emission. The ratio of the current of protons produced by Lorentz ionization (i_*) to the total flux of neutral particles (i_0) gives the relative yield of the highly excited hydrogen atoms

$$I = i_* / i_0. \tag{8}$$

It was shown earlier [1-3] that if the population of the highly excited states of the hydrogen atoms with principal quantum number n is equal to a/n^3, then

$$I(E) = 6{,}4 \cdot 10^{-4}\, a\sqrt{E}, \tag{9}$$

where a is a quantity that depends on the energy of the fast ions and on the type of targets; E is the ionizing electric field in kV/cm.

It was established experimentally that the relation $I \sim \sqrt{E}$ is satisfied for both highly excited atoms (see [2]) and for highly excited hydrogen molecules (see Fig. 1). This is evidence that for H_2^* the law of population of the highly excited state $\sim n^{-3}$ is close to the true one.

In the case of highly excited H_2^* molecules, in addition, it is assumed that the dependence of the ionizing field E on n is the same as for the highly excited H^* atoms. This was recently confirmed by Barnett and Ray [6]. Therefore the relative yield of the highly excited molecules I is described by the same expression (9).

It is also shown in [1,2] that if the total neutralization cross section σ_0 and the relative fraction of the neutral particles in the beam Φ_0 are determined in an independent experiment, then the cross section for the production of the highly excited atoms $\sigma_{H^*}^n$ and their fraction Φ^n can be calculated from the formulas

$$n^3 \sigma_{H^*}^n = a_0 \sigma_0, \tag{10}$$

$$n^3 \Phi^n = a \Phi_0. \tag{11}$$

In formula (11) the value of a is usually chosen for a "thick" target, and in (10) it corresponds to the case of "thin" target, i.e., to the condition of single collisions, and is denoted by a_0.

The cross section for the production of the highly excited hydrogen molecules $\sigma_{H^*}^n$ and their fraction Φ^n are determined from similar formulas.

The error in the measurement of the values of the cross sections and of the value of a is approximately ±20%. The energies of the fast H_2^+ and H_3^+ ions ranged from 20 to 180 keV.

RESULTS AND THEIR DISCUSSION

Total Neutralization Cross Section σ_0. Proton Production Cross Section σ_{H^+}

To determine the cross sections for the production of highly excited atoms and molecules it is necessary, according to (10), to known the total neutralization cross section σ_0. It was measured for all the investigated targets. At the same time, we measured also the proton-production cross section σ_{H^+}. Although measurement of σ_{H^+} is not the main task of our investigation, information concerning this cross section is necessary for the interpretation of the data on the production of highly excited atoms and molecules of hydrogen.

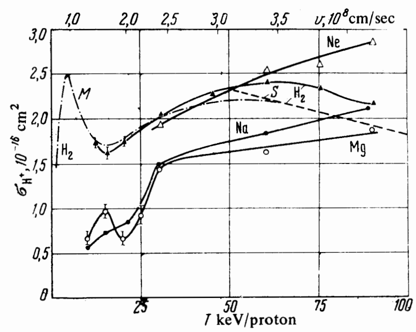


FIG. 2. Dependence of the cross section σ_{H^+} for the production of protons on the velocity and energy of the fast H_2^+ ions in H_2, Ne, and Na and Mg vapor. M–data of McClure[5], S–data of Sweetman[4].

The results of the measurements of the cross sections σ_{H^+} are shown in Fig. 2. The dependence of σ_{H^+} on the energy T for fast H_2^+ ions is nonmonotonic. At low energy, a sharp maximum due to the dissociation of the H_2^+ ion into an atom and a photon is observed on the $\sigma_{H^+}(T)$ curve of H_2.[5,7,8] It is probable that the influence of this process causes the nonmontonicity of the indicated dependences for other targets (Mg, Na), too. The maximum observed in H_2 at higher energy is due to dissociation with production of two protons. The difference between our data and those of McClure [5] and Sweetman [4] for the cross section of the production of protons in H_2 from fast H_2^+ ions lies within the range of the random experimental errors. The cross sections σ_{H^+} for fast H_3^+ ions in H_2 and in Mg vapor are listed in Table I. This cross section increases in the investigated energy range in Mg vapor and decreases in H_2.

The results of the measurements of the total neutralization cross section σ_0 for fast H_2^+ ions are shown in Fig. 3. Similar results for fast H_3^+ ions are given in Table I.

Table I. Proton production cross section σ_{H^+} and total neutralization cross section σ_0 for fast H_3^+ ions (in 10^{-16} cm^2)

Target	Energy of H_3^+ ions, keV/proton					
	20		40		60	
	H+	σ_0	I	σ_0	σ_{H^+}	σ_0
H_2	1,55	13,7	1,45	6,3	1,1	4,2
Mg	1,8	22	3,3	8,1	3,3	5,1

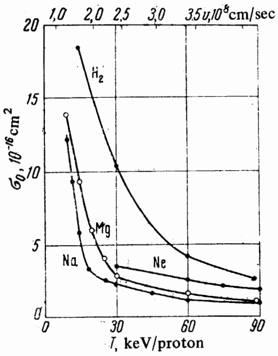


FIG. 3. Dependence of the total neutralization cross section σ_0 on the velocity and energy of the fast $H_2{}^+$ ions in H_2, Ne, and Na and Mg vapor.

It is seen from Fig. 3 that the cross section σ_0, which is the summary cross section of several processes (see (7)), decreases with increasing velocity in the entire investigated velocity interval. At low velocities ($v \leq 2.5 \times 10^8$ cm/sec) the value of σ_0 is determined essentially by the capture process (4) and by the dissociative capture process (3), and at higher velocities apparently a larger contribution is made by the dissociation process (1) besides the reaction (3). It is also seen that for H_2^+ ions the cross section σ_0, as well as the cross section σ_{H^+}, is smaller in magnitude for metal-vapor targets than for gas targets.

The neutral-particle beam produced in the course of charge exchange and dissociation of fast H_2 ions should consist of atoms and molecules of hydrogen, and their ratio, obtained in different cases, should be of interest in itself. To determine the composition of the neutral beam produced in Mg vapor and in H_2, a separate experiment was performed. A neutral beam obtained by passing a beam of fast H_2 ions through a chamber, where it collided with Mg vapor or with H_2, was caused to pass through an additional chamber filled with helium. The cross sections for the loss of an electron by the hydrogen atoms (σ_l) and by H_2 molecules ($\sigma_{H_2^+}$) in collisions with helium atoms were measured by us earlier [9,10]. The cross section for the production of protons from H_2 molecules is negligibly small compared with the indicated cross sections [10]. The ratio of the proton current (i_{H^+}) and the H_2^+ ion current ($i_{H_2^+}$) produced in the beam after passing through the chamber with helium is connected with the ratio of the neutral fluxes produced in the first collision chamber as follows:

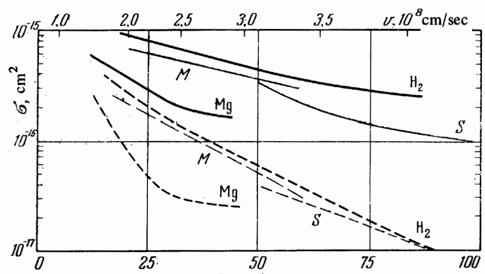


FIG. 4. Cross section σ_H for the production of hydrogen atoms (solid curves) and cross section σ_{H_2} for the production of hydrogen molecules (dashed curves) vs. velocity and energy of fast ions $H_2{}^+$ in H_2, Ne, and in Na and Mg vapor. M—data of McClure [5], S—data of Sweetman [4].

$$\frac{i_H}{i_{H_2}} = \frac{i_{H^+}}{i_{H_2^+}} \frac{\sigma_{H_2^+}}{\sigma_l}. \tag{12}$$

On the basis of (12), knowing the experimentally measured cross section σ_0, we determined the cross section σ_H for the production of the atoms and the cross section σ_{H_2} for the production of the hydrogen molecules. The determined cross sections σ_H and σ_{H_2} are shown in Fig. 4. It is seen from the figure that the cross sections σ_H and σ_{H_2} decrease with increasing energy. The cross section σ_{H_2}, which is the cross section of the capture process only, decreases with the energy more rapidly than the cross section σ_H, which is determined both by the dissociation process (1) and by the dissociative capture process (3). Obviously, the fraction of the neutral molecules H_2 in the beam increases rapidly with decreasing energy. Figure 4 shows for comparison the values of the cross sections σ_H and σ_{H_2} measured by Sweetman [4] and McClure [5] in molecular hydrogen.

Population of Highly Excited States

The quantities a_0, which characterize the population of the highly excited states of both the atoms H^* and the hydrogen molecules H_2^* for a thin target, are shown in Table II. It is interesting to note that for H_2^* molecules obtained from fast H_2 ions (i.e., for the capture process (4)) the $a_0(T)$ dependence has a maximum (with the exception of Ne) whose height decreases with increasing ionization potential of the target, while the maximum itself shifts towards the higher energies. Similar features of $a_0(T)$ were observed by us earlier also in the case of the capture of electrons by fast protons [2,3].

Cross Section $n^3\sigma^n_{H_2^*}$ for the Production of Highly Excited Hydrogen Molecules from fast H_2^+ and H_3^+ Ions

Plots of $n^3\sigma^n_{H_2^*}(T)$ for fast H_2 ions are shown in Fig. 5. It is seen from the figure that in the low-energy region (below 30 keV/proton), the most effective production of highly excited hydrogen molecules is observed in metal vapors (Mg and Na). At high energies (above 40–50 keV/proton), gas targets (H_2, Ne) are more convenient.

We note the following feature. Just as in the case of the capture of electrons by fast protons with formation of highly excited atoms [3], the maximum of the cross section $n^3\sigma^n_{H_2^*}$, which is also the capture cross sec-

Table II. Values of α_0 for highly excited H_2^* molecules and Highly excited H^* atoms produced from fast H_2^+ and H_3^+ ions

	Fast ion	Target	Fast-ion energy, keV/proton												
			10	12.5	15	18.75	20	22.5	25	30	40	45	60	75	90
H^*	H_2^+	H_2	—	—	—	—	—	—	—	0.12	—	—	0.40	0.53	0.67
		Ne	—	—	—	—	—	—	—	0.25	—	—	0.48	0.55	0.77
		Na	—	0.145	0.305	0.46	—	0.90	—	0.73	—	0.41	0.57	—	0.58
		Mg	—	—	0.13	—	0.25	—	0.32	0.40	—	—	0.46	—	0.48
	H_3^+	H_2	—	—	—	—	0.12	—	—	—	0.18	—	0.30	—	—
		Mg	—	—	—	—	0.15	—	—	—	0.35	—	0.57	—	—
H_2^*	H_2^+	H_2	—	—	0.019	—	—	—	—	0.057	—	—	0.12	0.09	0.05
		Ne	—	—	—	—	—	—	—	0.064	—	—	0.065	0.05	0.08
		Na	—	0.25	0.31	0.33	—	0.26	—	0.07	—	0.015	—	—	—
		Mg	0.095	—	0.17	—	0.26	—	0.31	0.27	—	—	0.08	—	0.03
	H_3^+	Mg	—	—	—	—	0.008	—	—	—	0.025	—	0.02	—	—

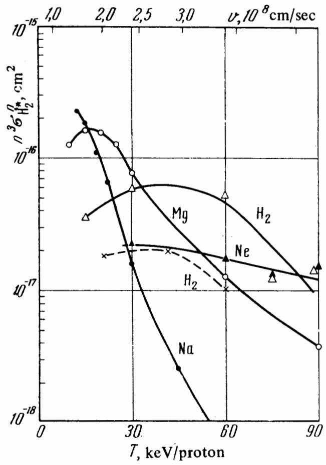


FIG. 5. Dependence of the cross section $n^3\sigma^n_{H_2^*}$ for the production of highly excited hydrogen molecules on the velocity and energy of fast ions H_2^+ (solid curves) and H_3^+ (dashed curve) in H_2, Ne, and in Na and Mg vapor.

tion, shifts towards larger energies with increasing ionization potential of the target, although no rigorous quantitative relations were observed in this case.

Figure 5 shows also the plot for the production of highly excited H_2^* molecules in the dissociation of fast H_3^+ ions in Mg vapor. In our energy interval the corresponding cross section is of the order of 10^{-17} cm^2. In H_2 we did not observe highly excited H_2^* molecules produced from H_3 ions (i.e., $n^3\sigma^n_{H^*} < 1\times10^{-19}$ cm^2).

Cross Section $n^3\sigma^n_{H^*}$ for the Production of Highly Excited Hydrogen Atoms from Fast Hydrogen Ions.

The cross sections $n^3\sigma^n_{H^*}$ for the production of highly excited hydrogen atoms following dissociation of fast H_3^+ and H_2^+ ions are shown in Fig. 6. For comparison we give also the analogous cross sections in the charge exchange of fast protons H^+ and H_2 and in Mg vapor from our earlier papers [2,3].

Let us consider first the $n^3\sigma^n_{H^*}(T)$ plots for fast H_2^+ ions. They have a somewhat different character for targets of metal vapors and of gases. For gases (H_2, Ne) these quantities increase slowly with in-

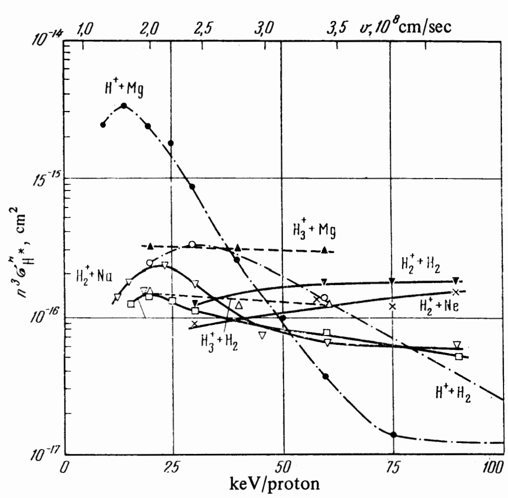
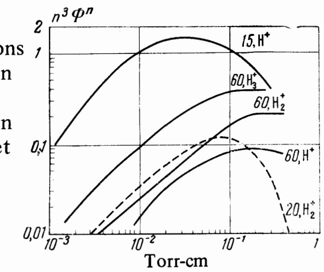


FIG. 6. Cross section $n^3\sigma^n_{H^*}$ for the production of highly excited hydrogen atoms, vs. velocity and energy of fast H^+, H_2^+, and H_3^+ ions in Mg and Na vapor and in H_2 and Ne (points □—for H_2 + Mg).

FIG. 7. Dependence of the fractions $n^3\Phi_{H^*}$ of the highly excited hydrogen atoms (solid curve) and the fractions $n^3\Phi^n_{H_2^*}$ of the highly excited hydrogen molecules (dashed curve) on the target thickness pl (Mg vapor). The energy (in keV/proton) and the type of fast ions are marked on the curves.



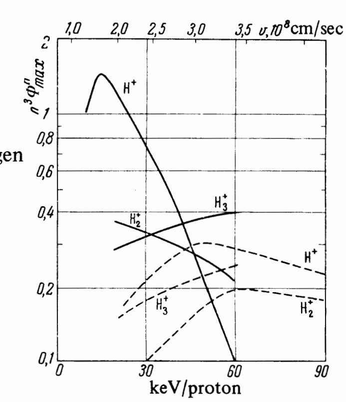
creasing energy, and for vapor (Mg, Ne) they have a maximum in the region of 20 keV/proton, after which they decrease slowly. The slow variation of the cross section $n^3\sigma_{H^*}$ at energies higher than 30 keV/proton is probably connected with the increasing contribution made to the production of H* by dissociation into a proton and an atom, $H_2^+ \to H^+ + H^*$. In the region of lower energies (near 20 keV/proton and below), where measurement data are available only for Mg and Na, the cross sections are close in magnitude to the cross sections $n^3\sigma^n_{H_2^*}$ (cf. Fig. 5). This allows us to assume that at such energies a large contribution to the production of H* is made by the dissociative capture $H_2^+ \to H + H^*$.

In the case of dissociation of fast H_3 ions, the cross sections for the production of highly excited atoms were measured only for two targets, H_2 and Mg vapor. Comparing all the cross sections for the production of highly excited hydrogen atoms in the charge exchange of H^+ protons and ion dissociation H_2^+ and H_3^+, we can see that the largest cross section $n^3\sigma^n_{H^*}$ is observed in Mg vapor, for fast protons up to 40 keV/proton, and for fast H_3 ions at higher energies.

Production of Highly Excited Molecules and Hydrogen Atoms in a "Thick" Target

All the cross sections were measured under single-collision conditions ("thin" target). In practical problems requiring the production of neutral beams enriched with highly excited hydrogen particles, it is advantageous to use targets in which multiple collisions take place ("thick" targets). Figure 7 shows the dependence of the fractions $n^3\Phi^n(pl)$ plots for highly excited hydrogen atoms H* obtained from fast H_1^+, H_2^+, and H_3^+ ions in Mg vapor. The same figure shows a plot of $n^3\Phi^n(pl)$ for highly excited hydrogen molecules H_2^* obtained via capture of electrons by fast H_2^+ ions.

FIG. 8. Maximum fractions $n^3\Phi^n_{max}$ of highly excited hydrogen atoms vs. velocity and energy of fast H^+, H_2^+ and H_3^+ ions in Mg vapor (solid curves) and in H_2 (dashed curves).



It is seen from the figure that whereas in the case of fast molecular ions H_2^+ and H_3^+ the $n^3\Phi^n(pl)$ for highly excited atoms H* increase monotonically and reach a plateau (corresponding to attainment of equilibrium), in the case of fast protons H^+ the curves have a maximum. This maximum is due to the presence of two competing processes: production of highly excited atoms and their stripping, the latter process preceeding more rapidly[1)].

The maximum on the curve (Fig. 7) for the highly excited molecules H_2^* is due to complete dissociation of the molecules as $pl \to \infty$, therefore the equilibrium fraction of the highly excited molecules is $n^3\Phi_\infty^n = 0$.

The dependence of the measured maximum fractions $n^3\Phi_{max}^n$ of the highly excited hydrogen atoms on the energy of the fast ions H^+, H_2^+ and H_3^+ in molecular hydrogen and in magnesium vapor is shown in Fig. 8. In some cases this fraction coincides with the equilibrium fraction.

The main conclusion that can be drawn from the figure is as follows: The most convenient target for the production of highly excited hydrogen atoms is magnesium vapor, to obtain highly excited atoms with energy larger than 40 keV it is preferable to use charge exchange of fast protons, and for still higher atom energies it is preferable to use dissociation of fast H_3^+ ions.

[1)] A special analysis shows that if transitions between close values of n are neglected, the dependence of the fraction Φ^n of the highly excited atoms on the target thickness pl in the case of charge exchange of protons is

$$\Phi^n(pl) = Ae^{-\alpha pl} - Be^{-\beta pl} + C,$$

where A, B, and C are positive constant coefficients that depend only on the cross sections: $\alpha = \sigma_c + \sigma_l$; $\beta = \sigma_l^n$ (σ_c – total cross section for the capture of electrons by fast protons, σ_l – cross section for the stripping of fast atoms, averaged over all the states of the atom, and σ_l^n – cross section for the stripping of highly excited atoms). The exponential with argument α characterizes the beam neutralization process, and the exponential with argument β characterizes the stripping of the highly excited atoms. The condition for the presence of the maximum is $\sigma_l^n > \sigma_c + \sigma_l$.

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221