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Quenching Cross Sections for Alkali-Inert Gas Collisions

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The cross sections for quenching the lowest n^2P states of the alkali atoms Li, Na, K, and Rb by the inert gases He, Ne, Ar, Kr, and Xe are presented for $5\,\text{eV} \leqq E_{\text{c.m.}} \leqq 100\,\text{eV}$. These cross sections are derived from the corresponding cross sections for collisional excitation by applying the principle of microreversibility. Upper estimates for the quenching cross sections at thermal energies are given; in all studied cases the quenching cross sections are $< 8 \cdot 10^{-3}\,\text{Å}^2$. These new upper limits are in most cases much lower than those obtained from other methods previously.

1. Introduction

For many years people have tried unsuccessfully to determine the cross sections for quenching of the resonance fluorescence of the alkali atoms (M) colliding with inert gas atoms (R) at thermal energies [1–3]. Even the most refined experiments were only able to set upper limits for the cross sections of these processes; these are of the order of or lower than $10^{-1}\,\text{Å}^2$ at thermal energies (see Table 1(a)). This failure to detect the cross section for quenching can easily be explained in the molecular description of the collision process [3]: large cross sections for inelastic processes at low collision energies are only expected when the quasimolecular state formed by the colliding atoms in the course of the collision becomes nearly degenerate with other quasimolecular states at some value of the internuclear distance. There the adiabatic description of the collision would break down and quenching processes would become possible by transitions to other quasimolecular states. This effective way to convert electronic into kinetic energy of the colliding atoms is not available for the alkali-inert gas systems since all more recent calculations of the relevant molecular states, namely those correlating with $M(^2P)+R$ and $M(^2S)+R$, have shown no indications for regions with strong non-adiabatic behavior [4, 5]; the potentials of the molecular states resulting from $M(^2S)+R$ and $M(^2P)+R$ are running more or less parallel down to less than 3 atomic units having an energy separation of the order 1 eV. The Massey criterion [33] predicts for such cases that quenching processes at such low energies have to be very improbable.

Evidence for the nature of the possible quenching mechanisms has come from the study of the inverse process, the collisional excitation in alkali-inert gas collisions, at moderate collision energies [6–8]: by studying the threshold behavior of these cross sections one is in principle able to get information on the various mechanisms which can lead to quenching processes.

The aim of this paper is therefore as follows: (i) By applying the principle of microreversibility to measured excitation cross sections in the threshold region, the quenching cross sections as a function of collision energy are obtained for $5 \leqq E_{\text{c.m.}} \leqq 80\,\text{eV}$. Upper limits are derived for the quenching cross sections at thermal energies. (ii) From a comparison of the experimental results with available model calculations for the collisional excitation process we can establish the mechanisms for the quenching processes in the energy range given above.

2. Determination of Quenching Cross Sections

2.1. Relation Between the Cross Sections for Quenching and Collisional Excitation

The starting point for all further considerations is the Eq. (1)

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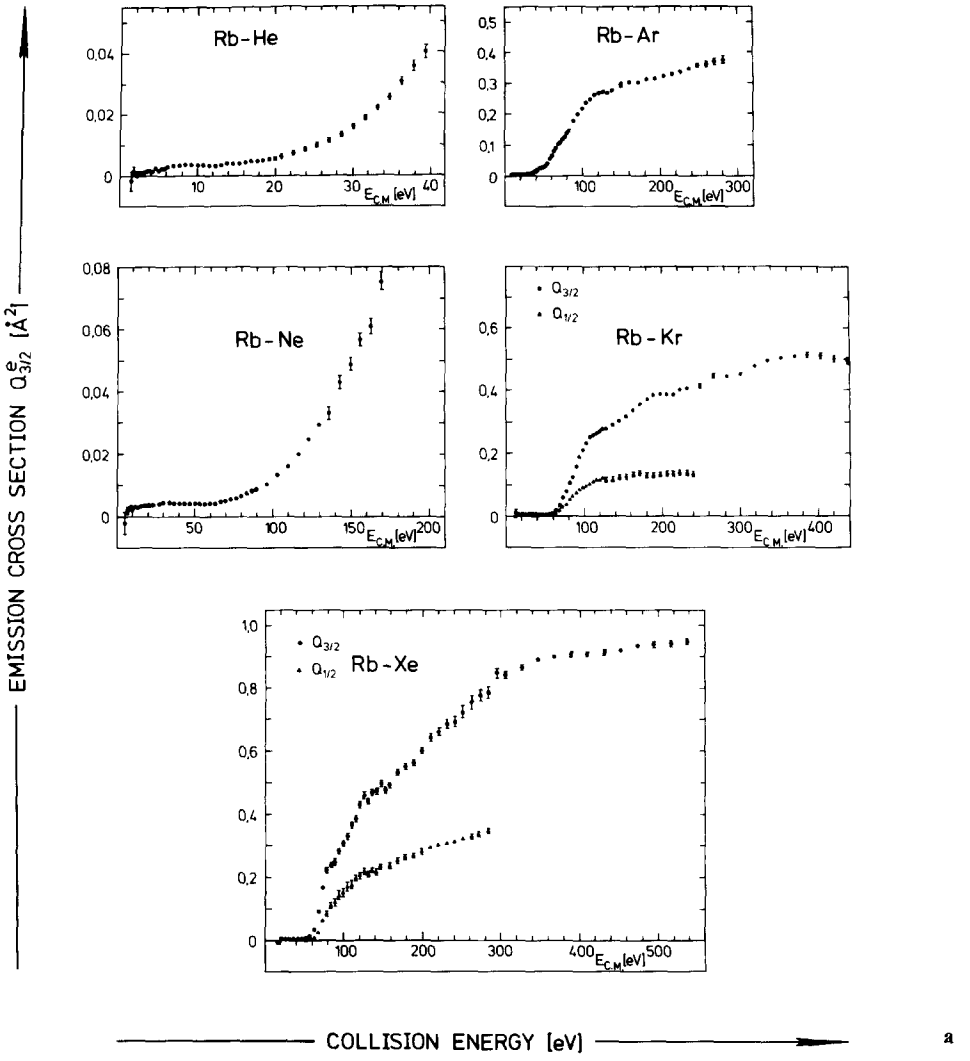

Fig. 1a–c. Excitation of Rb($5\,^2P$) in collisions with the inert gases: **a** $Q^e_{3/2}$ versus $E_{c.m.}$. **b** $Q^e_{3/2}/Q^e_{1/2}$ versus $E_{c.m.}$. **c** Polarisation of the $^2P_{3/2} \rightarrow {}^2S_{1/2}$ transition versus $E_{c.m.}$

$$E \cdot (2J_1+1)\, Q_{1\to 2}(E) = (E-E_0)(2J_2+1)\, Q_{2\to 1}(E-E_0) \tag{1}$$

which relates the cross section for the process $1 \to 2$ to that for the inverse process $2 \to 1$ [9, 10]. Both the initial and final state are characterized by definite values J_1, J_2 of the angular momentum. Both cross sections are summed over the contributions from all magnetic substates M_1, M_2 of the states J_1, J_2. Eq. (1) relates $Q_{1\to 2}$ at an energy E to $Q_{2\to 1}$ at an energy $E-E_0$; E_0 is the endoergicity of the process $1 \to 2$.

When the experiment resolves both fine structure components $^2P_{3/2} \to {}^2S_{1/2}$ and $^2P_{1/2} \to {}^2S_{1/2}$, we present the cross section $Q^q_{3/2}$ for quenching $^2P_{3/2}$ to the ground state $^2S_{1/2}$ and obtain from (1)

$$Q^q_{3/2}(E-E_0) = \frac{E}{(E-E_0)} \cdot \tfrac{1}{2} Q^e_{3/2}(E) \tag{2}$$

$Q^e_{3/2}$ is the cross section for exciting $^2P_{3/2}$. Because we have measured the ratio of the cross sections for exciting the two fine structure components, $Q^e_{3/2}/Q^e_{1/2}$, over a wide energy range, we can also obtain

$$Q^q_{1/2}(E-E_0) = \frac{E}{E-E_0} \cdot \frac{Q^e_{1/2}(E)}{Q^e_{3/2}(E)} \cdot Q^e_{3/2}(E). \tag{3}$$

For Li we can only determine $Q^e_{1/2,\,3/2}$ because the fine structure components cannot be separated. From Eqs. (1) and (3) we obtain

$$Q^q_{1/2,\,3/2}(E-E_0) = \frac{2}{3} \frac{E}{E-E_0} Q^e_{1/2,\,3/2}(E). \tag{4}$$

Here we have assumed that $Q^e_{3/2} = 2Q^e_{1/2}$, equal to the statistical ratio, which is a reasonable approximation for all studied combinations at most energies.

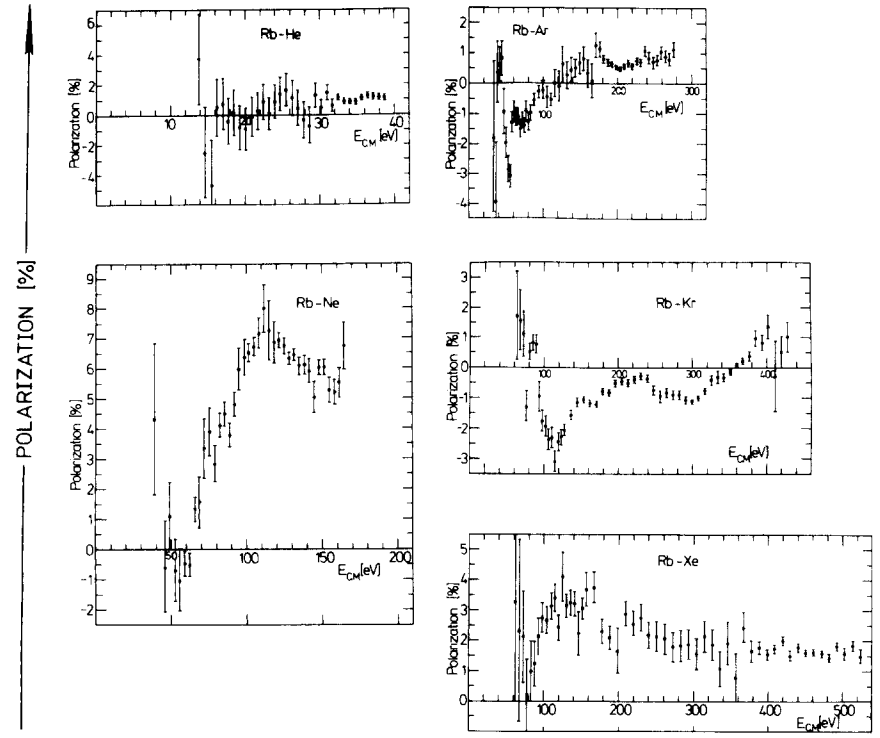
POLARIZATION [%]
Rb - He
Rb - Ar
Rb - Ne
Rb - Kr
Rb - Xe
Polarization [%]
E_C.M. [eV]
COLLISION ENERGY [eV]

Fig. 1 b

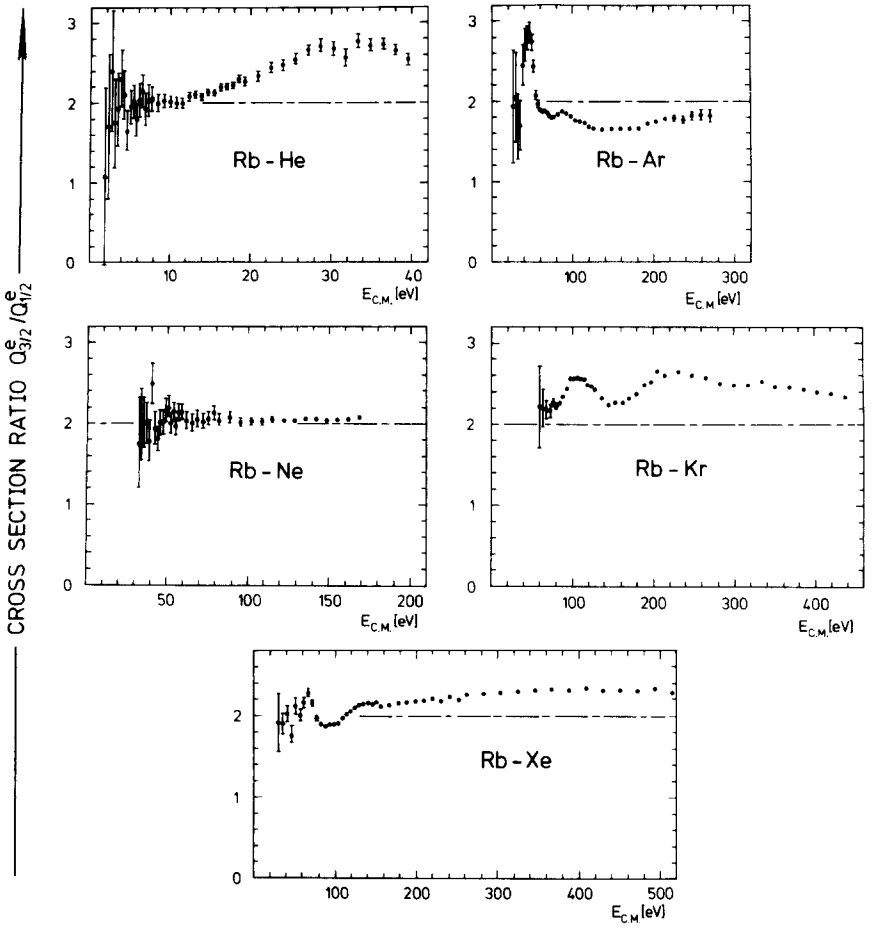
CROSS SECTION RATIO Q3/2e / Q1/2e
Rb - He
Rb - Ar
Rb - Ne
Rb - Kr
Rb - Xe
E_C.M. [eV]
COLLISION ENERGY [eV]

Fig. 1 c

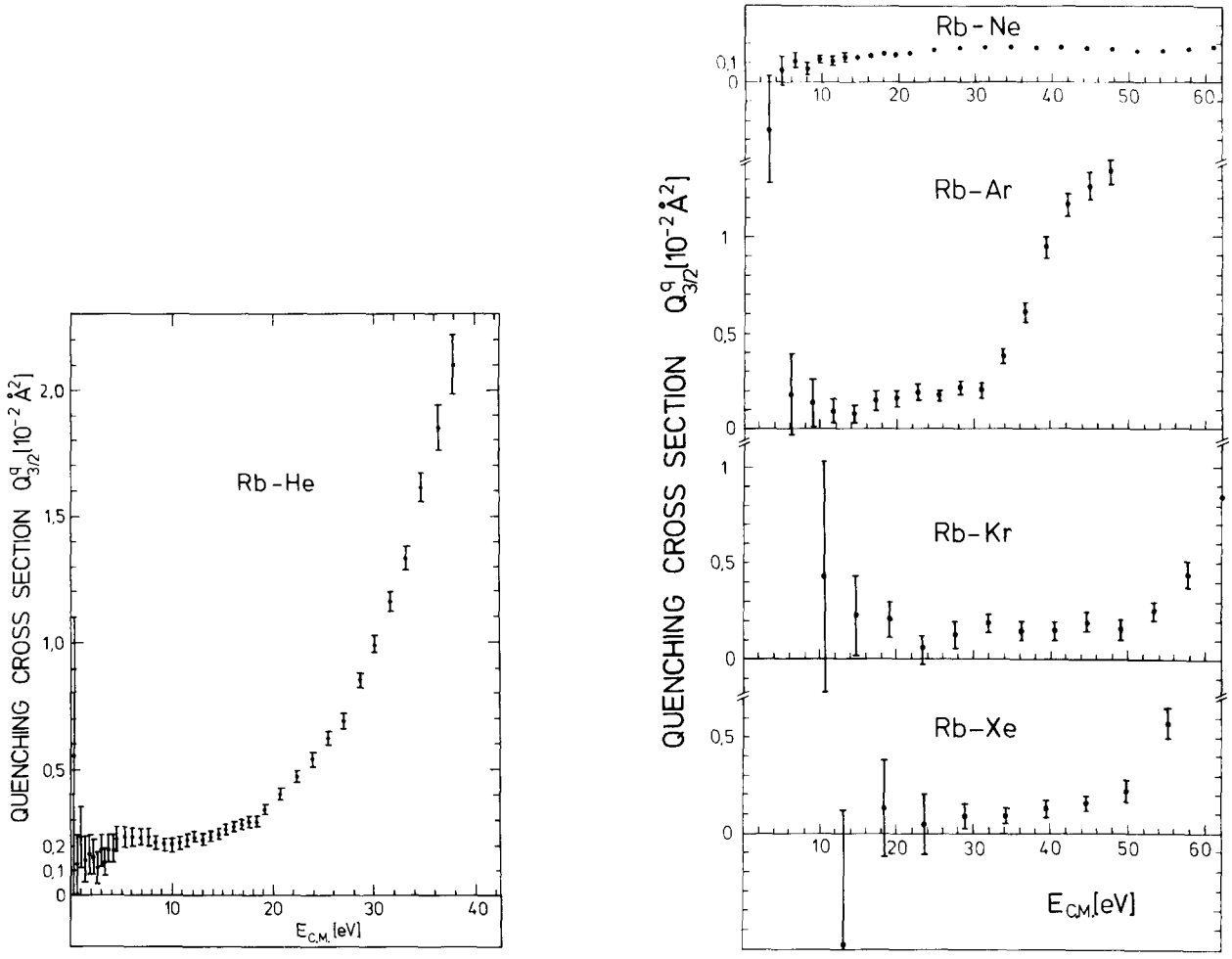

Fig. 2. Quenching cross section $Q^q_{3/2}$ versus $E_{c.m.}$ for the Rb-inert gases

The Eqs. (2), (3), and (4) have been applied to derive the quenching cross sections Q^q from the measured excitation cross sections Q^e.

2.2. *Apparatus*

Detailed descriptions of the apparatus employed to measure the cross sections for collisional excitation required for Eqs. (2) through (4) do exist [11, 12] and are available on request; a short version can be found in [7] together with the results for the sodium-inert gases. In the following a comprehensive description of the apparatus is presented in order to demonstrate the capability to measure absolute values of the integral excitation cross sections.

The alkali beams were produced by neutralising alkali ions obtained by surface ionisation. The energy spread of the alkali atoms and the nominal beam energy were determined by a time-of-flight-analysis. The energy spread was of the order of 1 eV at collision energies below about 100 eV.

The alkali resonance lines were isolated by appropriate interference filters and the light was viewed by a cooled photomultiplier perpendicular to the beam direction. The absolute sensitivity of the optical detection system was determined using a tungsten radiation standard.

Collisionally excited alkali atoms were produced in a scattering chamber. Photons escape perpendicular to the beam axis into the optical system through a quartz window in one of the scattering chamber walls. The gas pressure in the chamber was measured with a capacitance manometer; it was kept well within the range where the photon count rate showed linear pressure dependence and was typically $3 \cdot 10^{-3}$ Torr.

For Na, K, and Rb the beam intensity was measured with a surface ionisation detector. The coefficient for reflection of the impinging atoms as ions is rather well known for K as a function of the beam energy [13]. The same energy dependence of the reflection coefficient was assumed to hold for Na and Rb.

The excitation cross sections for Na – He, Ne obtained in this manner [7] agree within 10% in their absolute

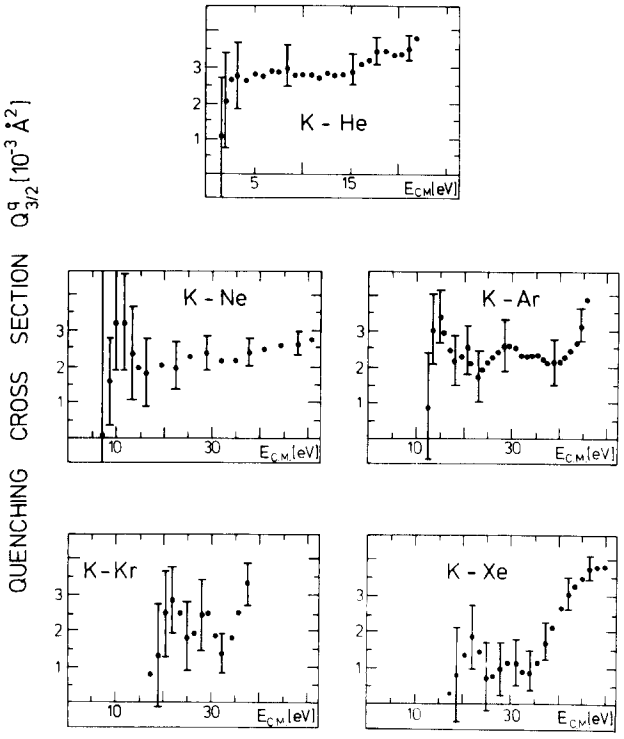

Fig. 3. Quenching cross section $Q^q_{3/2}$ versus $E_{c.m.}$ for the K-inert gases

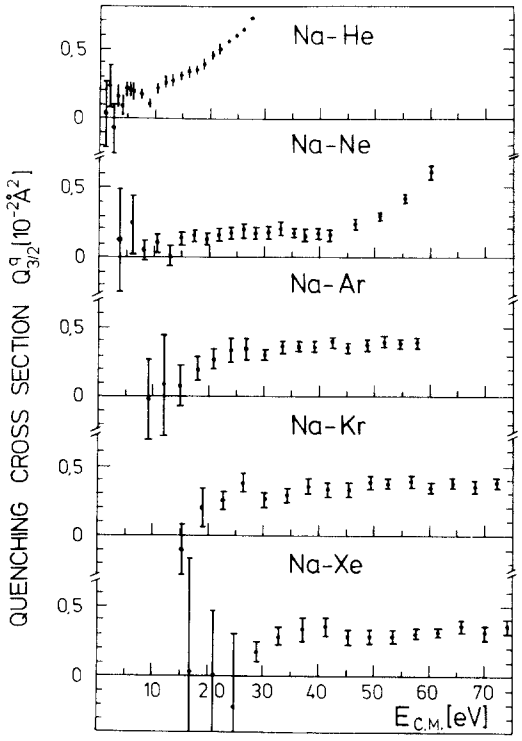

Fig. 4. Quenching cross section $Q^q_{3/2}$ versus $E_{c.m.}$ for the Na-inert gases

values with the results of [8] in the region of mutual overlap. In [8] the beam intensity was determined with an absolute detector.

For unknown reasons the absolute cross sections for K-collisions obtained by the same group [14] are about a factor three lower than ours in the region of overlap [6, 11]. We judge their absolute values as more accurate since their determination of the neutral beam flux is more reliable; the values employed in Eq. (2) have been normalized to their results.

For Rb no high energy results do exist; we estimate that the absolute values should be of the same quality as the Na results [12].

For Li the beam intensity at energies above 100 eV was determined both with a surface ionization detector and by using an absolute bolometer detector [15]. For Li–He, Ne the absolute cross sections at high energies agree within 30% with those in [8] in the region of overlap. In the region 10^2 to 10^3 eV the reflection coefficient for Li was found to be very weakly depending on the energy. It was assumed that the same reflection coefficient (0.1) also applies to lower energies where only the surface ionization detector could be used.

The excitation cross sections have been calculated in the same manner as in [7] for the Na-inert gases. Cascade contributions and polarization effects have been neglected when calculating the excitation cross sections. The information on the excitation of higher states indicates that the population of the studied lowest excited states dominates by at least one order of magnitude in the studied energy range [8, 16, 17]. The linear polarisation of the $^2P_{3/2} \rightarrow {}^2S_{1/2}$ transition is generally small (see Fig. 1 and [7, 18, 15]) and the polarization correction to the cross sections is negligible.

3. Results

As an example we show in Fig. 1 the excitation cross sections for the Rb-inert gas systems [12]. In Fig. 1a $Q^e_{3/2}$, the cross section for exciting Rb $5\,^2P_{3/2}$, is presented as a function of the c.m. energy; for $E_{c.m.} < 80$ eV $Q^e_{3/2}$ should better be taken from $Q^q_{3/2}$ in Fig. 2 via Eq. (2). Aside from Rb–Ne the polarisation fractions in Fig. 1b are $< 3\%$, but nevertheless show a strong energy dependence. A qualitative explanation for such a kind of structure has been tried in [18]. Fig. 1c shows the ratio $Q^e_{3/2}/Q^e_{1/2}$ of the cross sections for populating Rb $5\,^2P_{3/2}$ and Rb $5\,^2P_{1/2}$. Even at 500 eV this ratio has not reached yet the statistical value 2.0 in all cases. At the lowest energies accessible the ratios are however close to two. A qualitative explanation for the structure observed at intermediate energies has been offered in [19]; more detailed considerations can be found in [12].

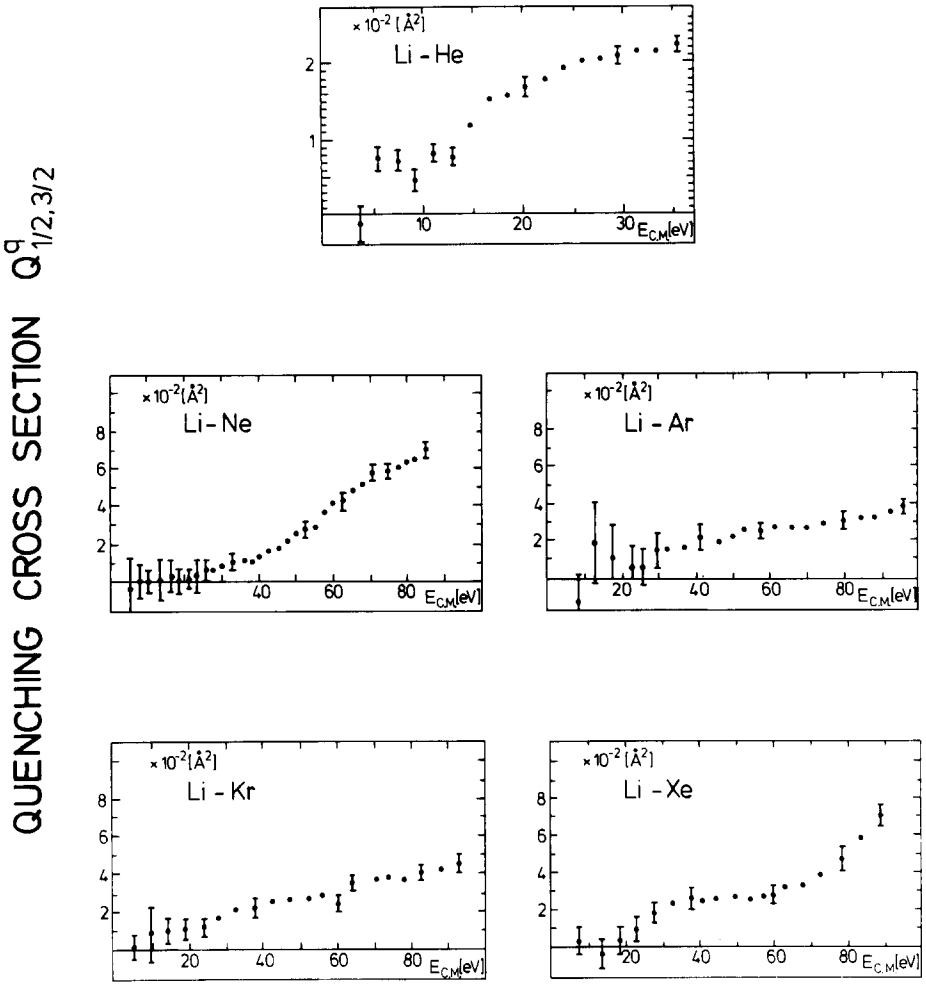

Fig. 5. Quenching cross section $Q^q_{1/2,\,3/2}$ versus $E_{c.m.}$ for the Li-inert gases

Figures 2 to 4 show the cross sections $Q^q_{3/2}$ for quenching $^2P_{3/2}$ to the ground state $^2S_{1/2}$ for collisions of Rb, K, and Na with the inert gases. The error bars solely give the uncertainty due to counting statistics; in all three cases we estimate that the absolute scale should be accurate within 30% (see also 2.2). For very low energies ($E_{c.m.} < 20$ eV) the systematic error may be larger because the background correction to the photon count rate is more difficult to apply. This is illustrated for Rb – He: we obtain a finite $Q^q_{3/2} = 1.0 \cdot 10^{-3}$ Å^2 for $E \to 0$. However, the pseudopotential calculations [5] give no indication for a curve crossing at very low energies which would explain the finite value of $Q^q_{3/2}$ for $E = 0$ (see also Sect. 4).

The cross sections for quenching $^2P_{1/2}$ can be obtained from Eq. (3) at energies where the ratios $Q^e_{3/2}/Q^e_{1/2}$ are known. The data for the K-inert gases can be found in [11, 20, 19], those for the Na-inert gases in [7], and for the Rb-inert gases in Fig. 1c.

For the Li-inert gas collisions Fig. 5 displays merely $Q^q_{3/2,\,1/2}$ since the fine structure components cannot be separated. Data were obtained from Eq. (4) employing the excitation cross sections of [15] and assuming that $Q^e_{3/2}/Q^e_{1/2} = 2$. The error bars are due to the counting statistics; the absolute scale is again estimated to be accurate within 30% (see 2.2).

Table 1 lists upper limits for the quenching cross sections $\langle Q^q_{1/2,\,3/2}\rangle$ at thermal energies (column (b)). The justification for these values is given in Sect. 4 after discussing the quenching mechanism. Table 1 also gives previous upper limits for $\langle Q^q_{1/2,\,3/2}\rangle$ (column (a)); the brackets indicate that these values are averaged over the maxwellian distribution of the velocities at the reported temperature.

4. Discussion

Since the first efforts to explain excitation mechanisms for alkali-inert gas collisions [6], the following picture has emerged:

Table 1. Upper limits for $\langle Q^q_{1/2,\,3/2}\rangle$ at thermal energies
(a) Upper limits at thermal energies from previous experiments.
(b) Upper limits at thermal energies derived from the threshold behavior of $Q^q_{1/2,\,3/2}$.
(c) Estimates for $\langle Q^q_{1/2,\,3/2}\rangle$ under thermal conditions at a temperature corresponding to 0.05 eV via Eq. (5).

	(a)	(b)	(c)
Li – He	<0.05 [38]	$<1.5\cdot10^{-3}$	$2\cdot10^{-7}$
– Ne	–	$<6\cdot10^{-3}$	$2\cdot10^{-18}$
– Ar	<0.05 [38]	$<2\cdot10^{-3}$	$2\cdot10^{-9}$
– Kr	–	$<8\cdot10^{-3}$	–
– Xe	–	$<4\cdot10^{-3}$	–
Na – He	$<10^{-2}$ [36]	$<1.5\cdot10^{-3}$	$2\cdot10^{-6}$
– Ne	$<10^{-2}$ [36]	$<1.5\cdot10^{-3}$	$3\cdot10^{-7}$
– Ar	$<10^{-3}$ [35]	$<2.5\cdot10^{-3}$	$2\cdot10^{-8}$
– Kr	$<10^{-2}$ [36]	$<1.5\cdot10^{-3}$	–
– Xe	$<10^{-2}$ [36]	$<3\cdot10^{-3}$	–
K – He	<0.04 [34]	$<3\cdot10^{-3}$	–
– Ne	–	$<2.5\cdot10^{-3}$	–
– Ar	<0.07 [34]	$<3\cdot10^{-3}$	–
– Kr	–	$<2.5\cdot10^{-3}$	$4\cdot10^{-10}$
– Xe	–	$<2\cdot10^{-3}$	–
Rb – He	<0.11 [28]	$<1.5\cdot10^{-3}$	$1\cdot10^{-6}$
– Ne	<1.0 [37]	$<1.0\cdot10^{-3}$	$1\cdot10^{-5}$
– Ar	<0.14 [34]	$<1.5\cdot10^{-3}$	–
– Kr	–	$<2.2\cdot10^{-3}$	–
– Xe	<0.21 [34]	$<1.5\cdot10^{-3}$	–

All values in Å^2

At energies below a few keV the alkali atom excitation is dominated by electron promotion if curve crossings of the molecular orbital emerging from the ns-atomic orbital ($n=2, 3, \ldots$ for Li, Na, ...) do occur during the collision. This mechanism predicts differential cross sections which show a very pronounced angular threshold [21, 22]. In the integral excitation cross section a sudden rise appears at the energy where these crossings become accessible energetically. Rb – Kr (see Fig. 1) shows this behavior clearly: the strong rise of $Q^e_{3/2}$ near $E_{c.m.}=60$ eV is due to excitation via a curve crossing mechanism. The other systems where electron promotion is mainly responsible for the excitation process between about 50 and 1,000 eV are Rb – Ar, Xe (see Fig. 1), K – Ar, Kr, Xe [6], Na – Ne, Ar, Kr, and Xe [7], and Li – Ne, Ar, Kr [15]; the relevant molecular orbital crossings can readily be found from diabatic orbital correlation diagrams [6, 7]. In this electron promotion process the inert gas electrons play an important role; usually it is found that close to the energy where promotion becomes possible aside from alkali atom excitation also excitation of the inert gas atom becomes important [6, 7, 15]. Calculations for Na – Ne based on this mechanism [21, 23] were able to reproduce both the differential cross section for Na excitation [21] and the integral cross sections for Na and Ne excitation [7].

At energies above a few keV direct transitions between the molecular orbitals correlating with the valence orbital of the ground and excited state of the alkali atom take place with increasing probability [24]. This mechanism is effective at larger internuclear distances than the promotion process, and the differential excitation cross section does not show a pronounced angular threshold at higher energies anymore [21].
For systems where electron promotion is improbable due to the lack of molecular orbital crossings this direct interaction is the only mechanism also at low energies. From the systems shown in Fig. 1 Rb – He and Ne are examples for this category: the excitation cross section shows an ill-defined threshold and continues to rise smoothly towards higher energies. The other systems of this class are K – He [11], Li – He, and Xe [15], and Na – He [7].
Even for systems with curve crossings direct transitions are responsible for the tails of the excitation cross sections at very low energies (<40 eV) which are found for all studied systems. This was first suggested in [6] for the K-inert gases and is now being put on solid ground by the calculations performed for Na – Ne [23]: for this system the weak tail below $E_{c.m.}=60$ eV is due to direct transitions between the σ Na $3s$ and both the σ and π Na $3p$ orbitals at relatively large internuclear distances.
For this mechanism the excitation and therefore also the quenching cross sections should decrease monotonously with decreasing energy: at low energies which are not influenced by curve crossing phenomena the cross sections should behave like [10, 31–33].

$$Q^q = B\exp(-2\Delta E\cdot a/(v_i+v_f)\,h) \tag{5}$$

where v_i, v_f are the initial and final velocities of the colliding atoms; a is of the order of the spatial extent of the interaction between the molecular states and may itself be weakly energy dependent [41].
The upper limits for $Q^q_{1/2,\,3/2}$ appearing in column (b) of Table 1 were obtained under the assumption based on Eq. (5) that Q^q will continue to fall off even at energies which are not yet accessible experimentally. Our upper limits are equal to the upper end of the error bar at the smallest measured cross section value. The calculations of [23] give $Q^q_{1/2,\,3/2}=1.3\cdot10^{-19}$ cm^2 for Na – Ne at $E_{c.m.}=40$ eV.
It is likely that $\langle Q^q\rangle$ at thermal energies will still be several orders of magnitude lower than our upper limits in column (b). A rough estimate for $\langle Q^q\rangle$ can be obtained in the following way: For Li – He, Ne, Ar, for Na – He, Ne, Ar, K – Kr, and Rb – He, Ne the low energy tail seems not to be influenced much by contributions from electron promotion because Q^q can reasonably well be fitted by (5). For Rb – He the

zero of the Q^q-scale has been shifted up by an amount of $1.3 \cdot 10^{-3}$ Å^2 before applying (5). Thus $(\Delta E \cdot a)/h$ can be determined from the data of Figs. 2 to 5 yielding values of the order of $2.5 \cdot 10^6$ cm/s. In these cases an extrapolation of Q^q has been attempted towards lower energies. $\langle Q^q \rangle$ is finally obtained after averaging over a Maxwellian distribution of the relative velocities of the colliding atoms (see the procedure described in [32]). The values for $\langle Q^q_{1/2,\,3/2} \rangle$ in column (c) correspond to thermal collisions at a temperature equivalent to 0.05 eV.

The above conclusions for the alkali-inert gas systems cannot be generalized to collisions of alkali atoms with other atoms: The size of the quenching cross sections depends critically on the coupling and the energetic separation between the involved molecular states and on the location of the relevant curve crossings. Likely candidates which presumably have much larger quenching cross sections at thermal energies are the alkali-alkali-systems. Both experiments on collisional excitation [25, 29] and recent pseudopotential calculations [26, 30] have shown the existence of a $^3\Sigma - {}^3\Pi$ curve crossing which could be effective even at thermal energies. Efficient quenching in these systems is due to rotational coupling between these states [40]. Q^q for K – K and K – Na may be estimated by applying (2) to the data of [25].

Quenching in thermal collisions between atoms has recently been reported for collisions of B with Kr and Xe atoms [27]. The unexpectedly large cross sections were tentatively attributed to a curve crossing between the involved molecular states which is accessible at thermal energies [39].

5. Summary

The cross sections for quenching of the lowest n^2P-alkali states by inert gases are presented as a function of the collision energy. In most studied cases improved upper estimates for the size of the quenching cross sections at thermal energies could be given. Mechanisms for quenching in alkali-inert gas collisions are discussed.

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