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Single and double ionization of helium by high-energy photon impact

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Abstract

Production of singly and doubly charged helium ions by impact of keV photons is studied. The ratio $R_{ph} = \sigma_{ph}^{++} / \sigma_{ph}^{+}$ for photoabsorption is calculated in the photon-energy range 2–18 keV using correlated initial- and final-state wave functions. Extrapolation towards asymptotic photon energies yields $R_{ph}(\omega \rightarrow \infty) = 1.66\%$ in agreement with previous predictions. Ionization due to Compton scattering, which becomes comparable to photoabsorption above $\omega \sim 3$ keV, is discussed.

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1 Introduction

Many-electron transitions in atomic systems induced by photon impact are of considerable interest since the Hamiltonian coupling of the electronic degrees of freedom to the electromagnetic field is built up of one-body operators. A transition involving more than one electron must therefore proceed via the interelectronic interaction (correlation). The simplest systems for studies of these processes are two-electron atoms and ions. Considerable work was carried out in the late 50s and early 60s on the dipole matrix elements for two-electron transitions in helium for the purpose of evaluating the Lamb shift of the ground state [1–3]. In the late 60s, when measurements of the ratio $R_{ph} = \sigma_{ph}^{++} / \sigma_{ph}^+$ of the double- to single-photoionization cross sections were reported from threshold up to 625 eV [4], it was realized that this quantity is very sensitive to the usage of highly accurate wave functions [5–10]. Apart from the theoretical efforts to obtain R_{ph} for photoabsorption in the low-energy regime, predictions of the non-relativistic asymptotic value $R_{ph}(\omega \rightarrow \infty)$ also became available [5–8]. The experimental verification of this fundamental quantity has become possible only very recently with the advent of synchrotron-light sources having sufficient intensity. This progress on the experimental side [11–13] has stimulated renewed theoretical interest [14–19] in double ionization of He at high photon energies.

A complication in the interpretation of the experiments arises, however, when the photon energy exceeds approximately 3 keV [20]. The photoionization cross section decays rapidly as $\omega^{-7/2}$ while the Compton scattering cross section is essentially independent of ω in this energy regime. The cross sections are equal at about 6 keV [21]. Based on the energy transfer to the atomic system, the approximate thresholds for single- and double-ionization due to inelastic Compton scattering are 2.5 and 4.5 keV, respectively. Since the present experiments cannot distinguish between these two competing processes, the measured ratio R is expected to be a weighted average of R_{ph} and the corresponding ratio for Compton scattering R_C . Above ~ 8 keV the experimentally measured R is exclusively determined by Compton scattering ($R = R_C$).

We here present calculations of ionization-excitation cross sections for photoabsorption in the 2–18 keV energy range. Using a sum rule we further predict the energy variation of R_{ph} at high energies and by extrapolation obtain the asymptotic value. Finally we discuss the single and double ionization process by Compton scattering and estimate the contribution to the apparent R as measured by the recent experiments.

2 Theory

The cross section for ionization of one electron into a continuum state labeled by the momentum k and angular momentum quantum numbers L and M and simultaneous excitation of the other electron to a $\text{He}^+(nlm)$ state by photoabsorption is, in the dipole approximation, given by (we use atomic units throughout unless otherwise stated)

$$\sigma_{ph}^{+*}(kLM, nlm) = \frac{2\pi^2}{c} \int dE \frac{df(kLM, nlm)}{dE} \delta(E + E_n - \omega + I_1), \quad (1)$$

where c is the speed of light, $df(kLM, nlm)/dE$ is the oscillator strength for the transition from ground state helium to a bound $\text{He}^+(nlm)$ state and a continuum state (kLM) with energy $E = k^2/2$, ω is the incident photon energy, I_1 is the first ionization potential of He, and E_n is the excitation energy of the n -manifold of He^+ measured from the ground state.

The *acceleration* form of the oscillator strength is

$$\frac{df^A(kLM, nlm)}{dE} = \frac{2k}{\omega^3} |\langle kLM, nlm | (\nabla_1 V + \nabla_2 V)_z | i \rangle|^2, \quad (2)$$

where V is the atomic potential energy and the polarization direction is taken along the z axis. For a two-electron atom or ion we have

$$(\nabla_1 V + \nabla_2 V)_z = Z \left(\frac{z_1}{r_1^3} + \frac{z_2}{r_2^3} \right), \quad (3)$$

where Z is the nuclear charge. Alternatively, the oscillator strength can be expressed in the *length* and *velocity* forms. With exact initial- and final-state wave functions the various forms of the oscillator strengths are equivalent while for approximate wave functions this is, in general,

not true. The sensitivity of the oscillator strength to the various forms provides in the latter case a measure of the quality of the wave functions. An investigation of the dependence of the transition amplitudes in many-body perturbation theory (MBPT) on the form of the dipole operator has been carried out by Hino *et al.* [19].

For the ground-state of He we use a Hylleraas-type wave function

$$\Psi_i(\mathbf{r}_1, \mathbf{r}_2) = N \exp(-\beta s) \sum_{j,k,l} c_{jkl} s^j t^{2k} u^l, \quad (4)$$

where β and c_{jkl} are variationally determined parameters, N is the normalization constant, and $s = r_1 + r_2$, $t = r_1 - r_2$, $u = r_{12} = |\mathbf{r}_2 - \mathbf{r}_1|$ are the usual Hylleraas coordinates. Specifically, the 20-parameter wave function of Hart and Herzberg [22] is employed here. For the final state we use a wave function of the form

$$\Psi_{k,nlm}^{(-)}(\mathbf{r}_1, \mathbf{r}_2) = \frac{1}{\sqrt{2}} \left[\Phi_{nlm}(\mathbf{r}_1) \Phi_{\mathbf{k}}^{(-)}(\mathbf{r}_2) D_{\mathbf{k}_{12}}^{(-)}(\mathbf{r}_{12}) + \mathbf{r}_1 \leftrightarrow \mathbf{r}_2 \right], \quad (5)$$

where Φ_{nlm} and $\Phi_{\mathbf{k}}^{(-)}$ are bound and continuum wave functions defined in the unscreened field of the He^{2+} nucleus and

$$D_{\mathbf{k}_{12}}^{(-)}(\mathbf{r}_{12}) = \exp(-\pi\alpha/2) \Gamma(1 - i\alpha) {}_1F_1[i\alpha, 1, -i(k_{12}r_{12} + \mathbf{k}_{12} \cdot \mathbf{r}_{12})] \quad (6)$$

is a Coulomb distortion factor which accounts for the electron-electron interaction. The continuum states $\Phi_{\mathbf{k}}^{(-)}$ are normalized to a δ function on the momentum scale. In (6) $\mathbf{k}_{12} = \mathbf{k}/2$ is the interelectronic momentum and $\alpha = 1/(2k_{12})$. The states $|kLM, nlm\rangle$ in (2) are obtained by expanding $\Phi_{\mathbf{k}}^{(-)}$ and $D_{\mathbf{k}_{12}}^{(-)}$ in partial waves and recoupling to (LM) states.

Because of the explicit appearance of the interelectronic vector \mathbf{r}_{12} inside transcendental functions, the dipole matrix element cannot be reduced into a product of integrals over each radial coordinate. Three integrals out of six can be carried out analytically, while the remaining three must be performed numerically.

In order to obtain the total cross section for ionizing one electron and leaving the second electron bound to the nucleus, the ionization-excitation cross sections $\sigma_{ph}^{+*}(kLM, nlm)$ are

summed over all bound states and over angular momenta of the continuum electron

$$\sigma_{ph}^+(\omega) = \sum_{LM} \sum_{nlm} \sigma_{ph}^{+*}(kLM, nlm). \quad (7)$$

Due to the dipole selection rules, $L = l \pm 1$, and, since the polarization is taken along the the quantization (z) axis, $M + m = 0$.

In principle, when performing the summation in (7), k takes on different values for different excitations of the residual He^+ ion. However, as explained further below, if the photon energy is significantly higher than the double-ionization limit, the approximation $k = \sqrt{2(\omega - I_1)}$ for all terms in the sum is sufficiently accurate.

The cross section for double ionization with ejection of two electrons having energies $E = k^2/2$ and $E' = k'^2/2$ can be defined in analogy to (1) as

$$\frac{d\sigma_{ph}^{r+}(kLM, k'lm)}{dE'} = \frac{2\pi^2}{c} \int dE \frac{d^2 f(kLM, k'lm)}{dE dE'} \delta(E + E' - \omega + I_2), \quad (8)$$

where I_2 is the total ionization potential of He. The total double-ionization cross section is

$$\sigma_{ph}^{++}(\omega) = \sum_{LM} \sum_{lm} \int_0^{\omega - I_2} dE' \frac{d\sigma_{ph}^{++}(kLM, k'lm)}{dE'}. \quad (9)$$

In the acceleration form the oscillator strength in (8) is

$$\frac{d^2 f^A(kLM, k'lm)}{dE dE'} = \frac{2kk'}{\omega^3} |\langle kLM, k'lm | (\nabla_1 V + \nabla_2 V)_z | i \rangle|^2, \quad (10)$$

and the length and velocity forms are similarly defined.

The state $|kLM, k'lm\rangle$ in (10) is the analogue to (5) obtained by replacing the bound state Φ_{nlm} by $\Phi_{k'}^{(-)}$, by partial wave expanding the two continuum wave functions and the distortion factor, and by recoupling to angular momenta (LM) and (lm) of each electron.

In the case of two continuum electrons the Sommerfeld parameter α in $D^{(-)}$ depends on the relative angle between the emission directions of the two electrons. The evaluation of σ_{ph}^{++} is therefore more difficult than the evaluation of ionization-excitation cross sections. However, if the photon energy is significantly larger than the ionization potential the largest contribution to the oscillator strength comes from the phase-space region where one electron is ejected in a p

state carrying nearly all the available energy $\omega - I_2 \approx \omega$, and the second electron is ‘shaken up’ to an s state with a continuum energy close to zero [17, 19]. The error introduced by fixing the energy of the fast electron and extending the upper limit in the integration in (9) to infinity is therefore small at high photon energies. The sum of single- and double-ionization cross sections

$$\sigma_S = \sigma_{ph}^+ + \sigma_{ph}^{++} = \sum_{LM} \sum_{lm} \left[\sum_n \sigma_{ph}^{+*}(kLM, nlm) + \int_0^{\omega - I_2} dE' \frac{d\sigma_{ph}^{++}(kLM, k'lm)}{dE'} \right] \quad (11)$$

can be evaluated with these approximations by using the closure property of the He^+ eigenfunctions. The double ionization cross section can therefore be obtained without reference to the two-electron continuum states, as $\sigma^{++} = \sigma_S - \sigma^+$. In the limit $\omega \rightarrow \infty$ this procedure becomes exact [2, 23].

Compton scattering of bound electrons is usually treated in terms of the coherent and incoherent cross sections [24]. For the problem at hand, this approach cannot be applied since we are concerned with final-state specific processes. The evaluation of double ionization by Compton scattering is significantly more difficult than for photoabsorption because three liberated particles share the energy in the final state. A high-energy approach as described above for photoabsorption is not justified because the dominant energy transfers ΔE from the photon to the electron(s) ranges from zero to an upper limit ΔE_{max} approximately given by the value for Compton scattering off free electrons

$$\Delta E_{max} = \frac{2\omega^2}{mc^2 + 2\omega} \quad (12)$$

and the distribution of energy transfers is essentially independent of ΔE in this range. One other important distinction between Compton scattering and photoabsorption is the distribution of angular momenta in the final state. While for photoabsorption only the final-state P sector is reached from the ground state of He (in the dipole approximation), a large number of final-state angular momenta will contribute to the transition amplitude for Compton scattering.

In order to estimate the influence of Compton scattering on the measured R we make here an impulse (or ‘binary-encounter’) approximation to obtain the Compton cross section differential

in the energy transfer ΔE

$$\frac{d\sigma_C(\omega)}{d\Delta E} = \int d^3q \int_0^{p_{\max}} dp \frac{p}{c} \delta(\mathbf{q} \cdot \mathbf{p} + p^2/2 + \epsilon_B - \Delta E) |\phi(\mathbf{q})|^2 \frac{d\sigma_{KN}(\omega)}{dp} \quad (13)$$

where p_{\max} is the electronic momentum corresponding to maximum energy transfer (see Eq. 12) in a binary encounter between the photon and one electron, ϵ_B is the orbital binding energy of one electron, $\phi(\mathbf{q})$ is the momentum-space wave function of the one electron in the ground state, and $d\sigma_{KN}(\omega)/dp$ is the free-electron Compton cross section differential in the momentum transfer to the electron for which we use the Klein-Nishina formula. The single-ionization Compton cross section is then

$$\sigma_C^+(\omega) = 2 \int_{I_1}^{\omega} d\Delta E \frac{d\sigma_C(\omega)}{d\Delta E}, \quad (14)$$

where the 2 in front of the integral accounts for the two electrons in He. For double ionization we use

$$\sigma_C^{++}(\omega) = 2 \int_{I_2}^{\omega} d\Delta E \frac{d\sigma_C(\omega)}{d\Delta E} R_C(\Delta E), \quad (15)$$

where $R_C(\Delta E)$ specifies the ratio of double to single ionization at a given energy transfer. Of course, the exact knowledge of $R_C(\Delta E)$ would imply that the problem at hand was solved. We make here the following approximation: for final states in the P sector $R_C(\Delta E)$ is assumed to equal the photoabsorption ratio at the photon energy ΔE , for higher angular momenta in the final state the shake-off value 0.73% [15] is used. The justification for this approximation relies on calculations for ionization-excitation by Compton scattering [25]. These indicate that the branching ratios for ionization of one electron and excitation of the second to an ns state of He^+ are approaching universal functions of the energy transfer but are only weakly dependent on the primary photon energy. Further, the branching ratios for ejection of a p electron follow closely the corresponding results for photoabsorption, while ejection of electrons with higher angular momenta result in smaller branching ratios close to the shake-off value.

3 Results and discussion

The cross sections for ionization-excitation to the final states (Ep, ns) and (Es, np) are shown in Fig. 1 for $n \leq 3$ in the energy range 2-18 keV. The dominant (Ep, ns) channels exhibit the well known $E^{-7/2}$ high-energy behavior and the (Es, np) channels decay as $E^{-9/2}$ [2] and do not substantially contribute to the total single-ionization cross section (7). Our results are close to the results obtained by Salpeter and Zaidi [3] and Brown [9] for the (Ep, ns) channels. Brown also determined oscillator strengths for the $(Es, 2p)$ channel. We find disagreement with the latter calculation, which employed an uncorrelated final state.

In order to carry out the sum in (7) cross sections for $n \leq 8$ were calculated and the residual summation over bound states was carried out by extrapolation assuming that an n^{-3} dependence has been reached at $n = 8$. In Fig. 2 are shown the branching ratios for the (Ep, ns) channels $B(n) = \sigma_{ph}^{+*}(n)/\sigma_S$ and for double ionization $\sigma_{ph}^{++}/\sigma_S$.

While it has been established that the ionization-excitation and double ionization cross sections are independent of correlation in the final state as $\omega \rightarrow \infty$, provided an accurate wave function of the initial state is used [5,6,15], this is not true at finite ω . We illustrate this in Fig. 3 where the difference in the branching ratios $\Delta B = B_{corr} - B_{uncorr}$ calculated with and without the final-state distortion $D^{(-)}$ are shown for $\omega = 2$ keV. The effect of final-state correlation is to redistribute probability for ionization without excitation to the ionization-excitation and double-ionization channels. In the language of MBPT this corresponds to the two-step-one (TS1) process where the fast photoelectron scatters at the second electron on the way out of the collision.

In Fig. 4 we show our result for R_{ph} as function of ω^{-1} together with the recent calculations by Teng and Shakeshaft [17], Hino [18], and the MBPT calculation by Hino *et al.* [19]. In the calculations of Hino and of Teng and Shakeshaft the double-ionization cross section was calculated directly using the two-electron continuum analogue to the final state (5). Teng and Shakeshaft used the velocity form of the dipole operator, while Hino used the acceleration form but took only the monopole contribution from the distortion factor $D^{(-)}$ into account. The

MBPT calculation used various forms of the dipole operator. The acceleration form, shown in Fig. 4, the length and velocity forms all give similar results in the high-energy region [19].

Our present result reaches an ω^{-1} behavior for $\omega > 5$ keV and extrapolation to infinite photon energy yields $R_{ph}(\infty) = 1.66\%$ as was obtained early by other authors [5, 6] using only a correlated initial state. The value of the coefficient of the leading ω^{-1} term is 0.90 keV. The short-dashed line in Fig. 4 represents an extrapolation of this linear behavior in ω^{-1} to both larger and smaller energies.

The various calculations [17-19] do not converge to the correct high-energy limit, even though they differ by relatively small amounts. We attribute this discrepancy to inaccurate initial-state wave functions used in the calculations. For $\omega^{-1} > 0.2$ the results start to diverge significantly. It is important to realize that the final state (5) and its two-electron continuum analogue constitute high-energy approximations and their use is not justified for lower photon energies. An indication for this problem can be found in the work by Teng and Shakeshaft who continued their calculation down to the double-ionization threshold. Their resulting R_{ph} reaches a maximum of 10% [17], which is a factor two higher than the experimental value. Our present result and the result of Teng and Shakeshaft have very similar slopes from 8 down to about 4 keV. At lower ω our result has a much stronger dependence on ω^{-1} . We have traced this strong dependence to the contributions from the $l \neq 0$ multipoles of $D^{(-)}$. The fact that Hino only retained the monopole term is likely the reason why his R has a significantly smaller slope than the present as well as Teng and Shakeshaft's results. Apart from the validity of the final state (5), the accuracy of the present result relies on the accuracy of the closure approximation. This approximation breaks down if the energy sharing between the two electrons is not highly asymmetric. We have verified for an uncorrelated final state that the accuracy is sufficient at least down to $\omega = 2$ keV. However, the validity of the closure method for correlated final states at low energies remains to be verified. Even if the energy distribution is highly asymmetric, the correlated final state introduces angular correlations in the emission pattern of the electrons which are not accounted for by the closure method. The $l \neq 0$ multipole contributions from $D^{(-)}$ are likely to be most strongly affected by these angular correlations. We are currently

investigating the possibility of incorporating these effects in the closure approximation.

In Fig. 5 is shown the ratio R_{ph} for photoabsorption (dash-dotted curve) together with the corresponding ratio for Compton scattering R_C (dotted curve) and the weighted mean of both processes (solid curve) which should be compared to the experimental points. The agreement is, considering the simplicity of the approximation, satisfactory. We also show the linear extrapolation of the photoabsorption ratio (dashed curve) which appears to improve the agreement with the experiments below $\omega = 3$ keV. Further work on two-electron processes by photoabsorption and inelastic scattering of photons is in progress.

Acknowledgements

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Figure Captions

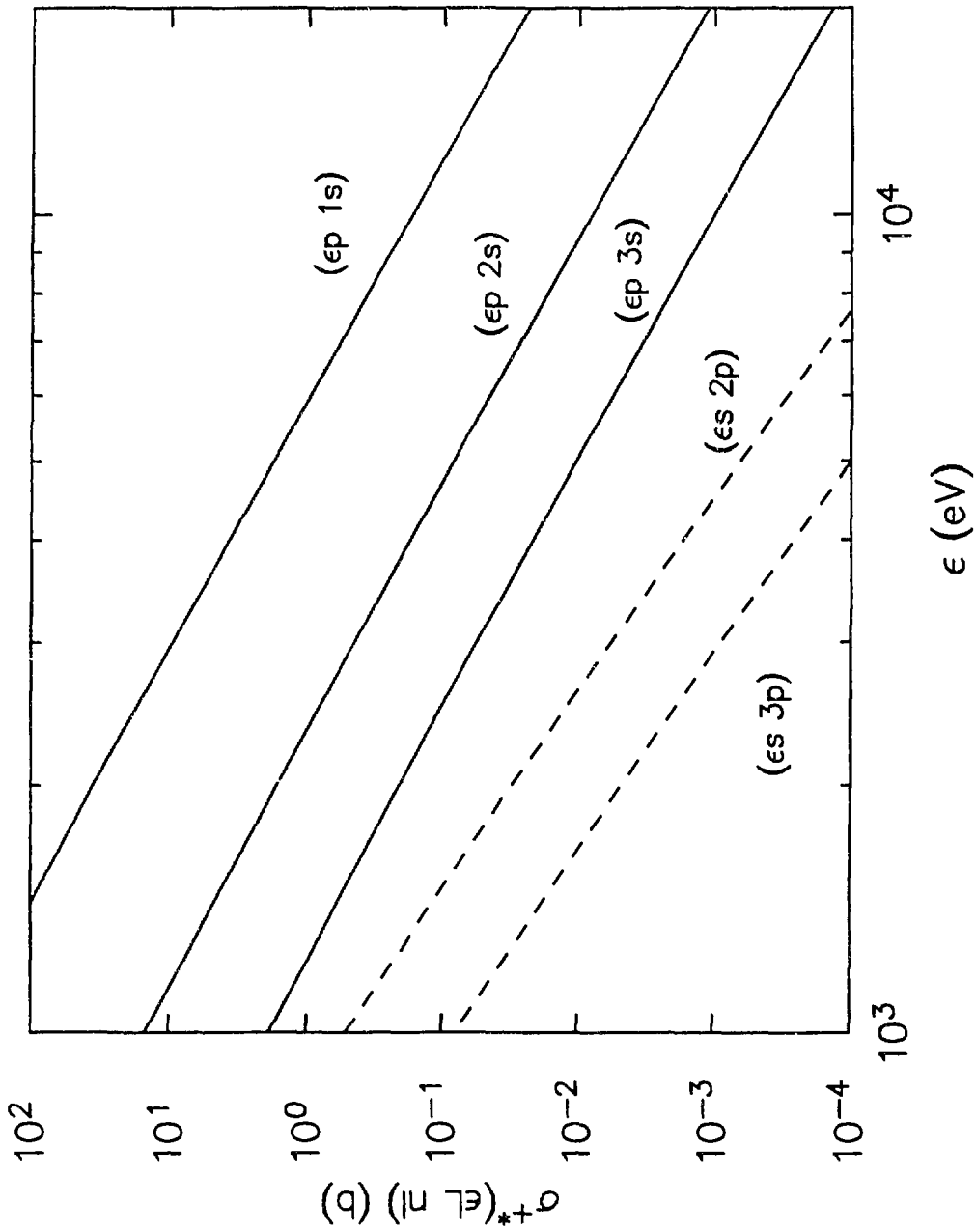
Figure 1: Cross sections for ionization-excitation of He by photoabsorption as function of the energy of the continuum electron.

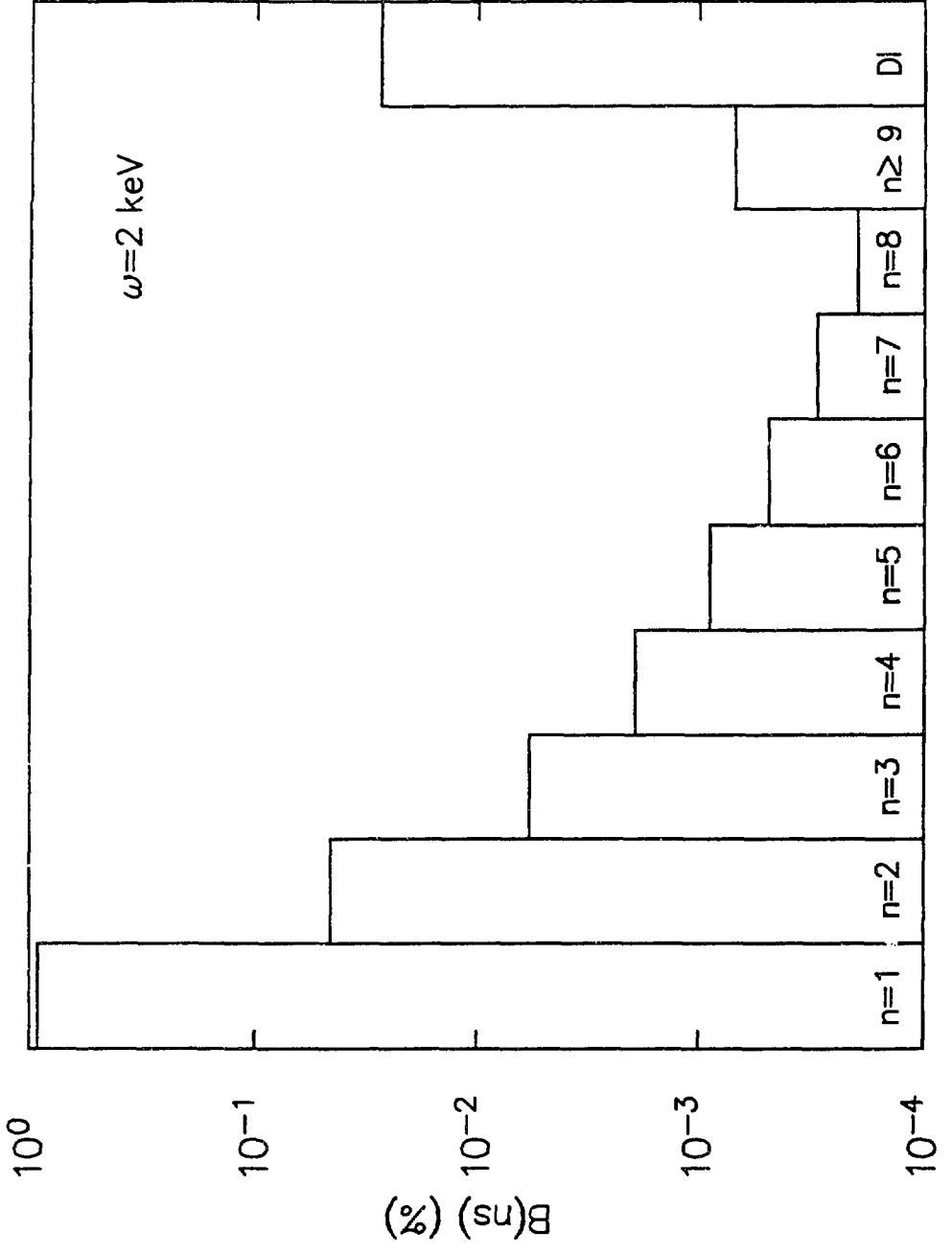
Figure 2: Branching ratios $B(n) = \sigma_{ph}^{+*}(n)/(\sigma_{ph}^+ + \sigma_{ph}^{++})$ at $\omega = 2$ keV. The numbers in the bars indicate n and DI denotes double ionization.

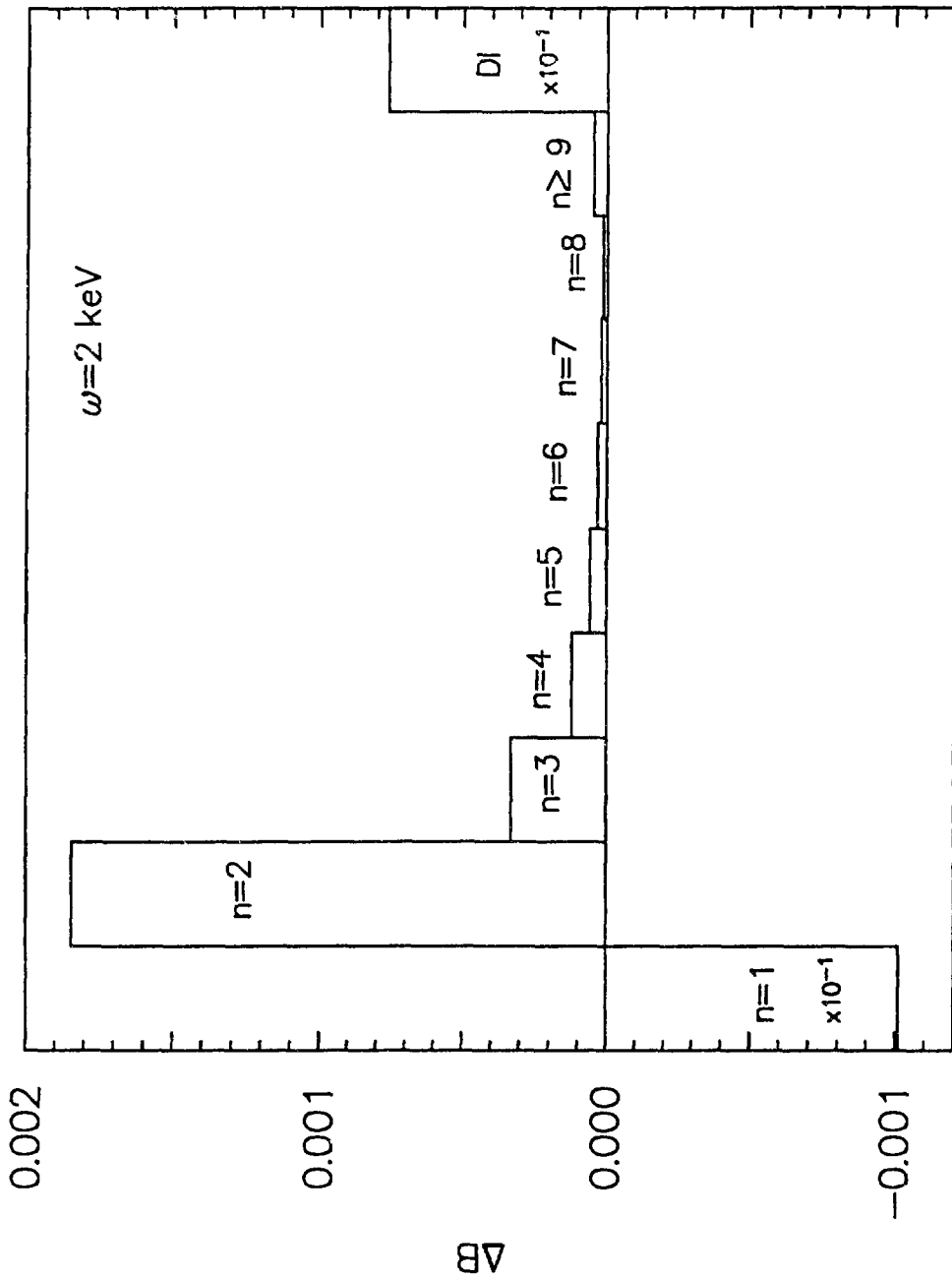
Figure 3: The change ΔB in the branching ratios $B(n) = \sigma_{ph}^{+*}(n)/(\sigma_{ph}^+ + \sigma_{ph}^{++})$ calculated with and without the distortion factor $D^{(-)}$ in the final-state wave function. The bars for $n = 1$ and double ionization (DI) have been multiplied by 0.1 for clarity.

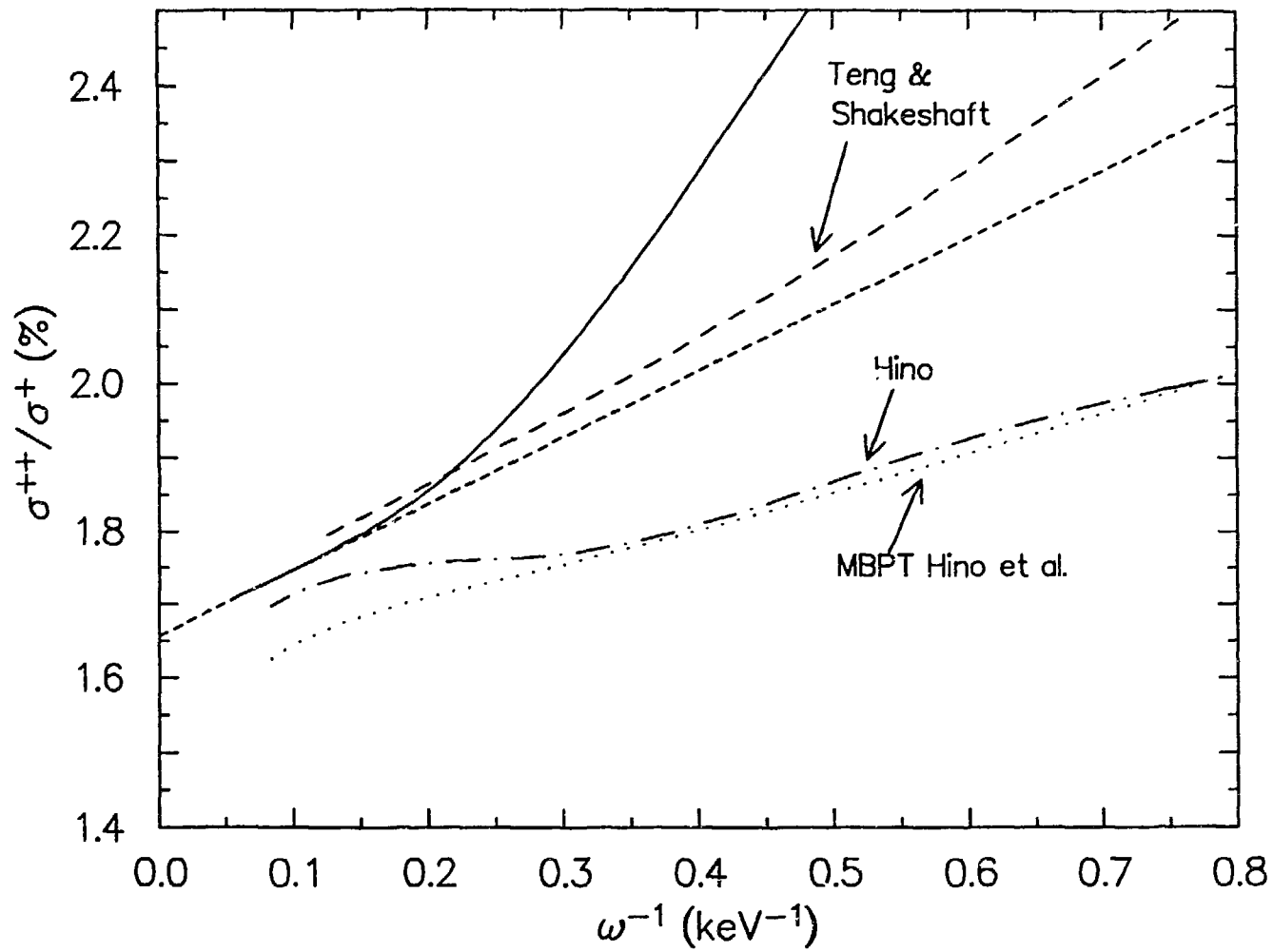
Figure 4: The ratio $R_{ph} = \sigma_{ph}^{++}/\sigma_{ph}^+$ for photoabsorption as function of ω^{-1} . Solid curve: present result; long-dashed curve, Teng and Shakeshaft [17]; dash-dotted curve, Hino [18]; dotted curve, Hino *et al.* [19]. Short-dashed curve: the high-energy behavior of the present result accurate to order ω^{-1} .

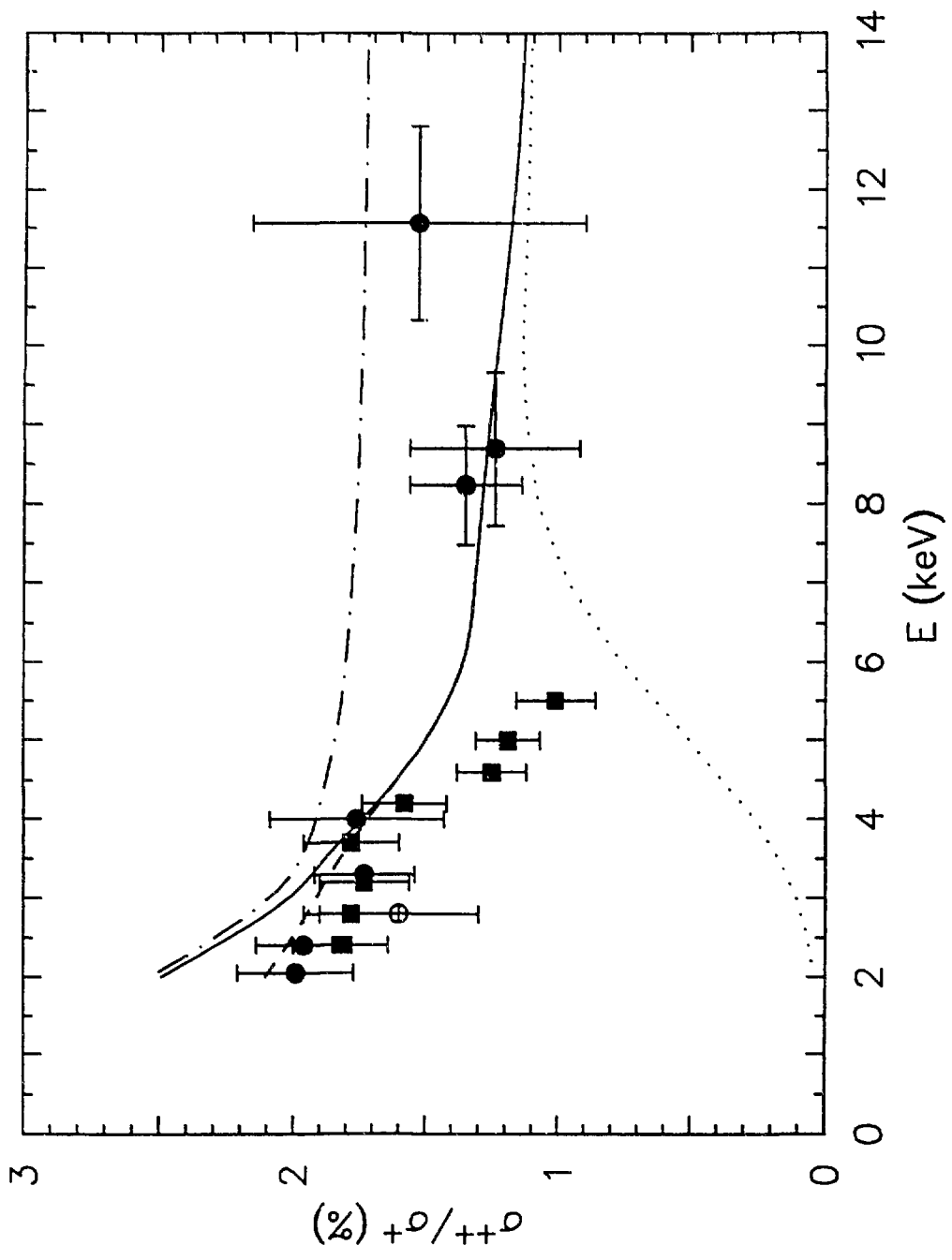
Figure 5: The ratio of double to single ionization of He by photon impact. Dash-dotted curve, photoabsorption; dotted curve, Compton scattering; solid curve, weighted mean of both processes; Dashed curve, linear extrapolation of the high-energy behavior of R_{ph} to order ω^{-1} , corrected for Compton scattering. Experimental data: open circle, Levin *et al.* [11]; solid circles, Levin *et al.* [12]; solid squares, Bartlett and Samson [13].











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