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LETTER TO THE EDITOR

Two-photon bremsstrahlung in electron collisions with noble gas atoms

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Abstract. We calculate the cross section for two-photon bremsstrahlung emission in the collision of keV electrons with noble gas atoms. In addition to bremsstrahlung from the projectile the two-photon bremsstrahlung emission arising from target electron excitation during the collision is taken into account explicitly. Such contributions turn out to be small, however, and do not explain the large discrepancy between previous calculations and the measurements of Hippler.

Recent experiments have shown that it is possible to detect the emission of two bremsstrahlung photons in a single collision of a fast incident electron on a neutral atom (Altman and Quarles 1985, Hippler 1991, Kahler *et al* 1992a, b). Calculations of the cross section for this process in which only the interaction of the projectile electron with the target nucleus is taken into account show very large discrepancies with experimental results. However, in the case of one-photon bremsstrahlung it has been shown (Amusia *et al* 1990) that it is necessary to consider the contribution from atomic electrons which are set in motion during the collision and can also give rise to bremsstrahlung. The aim here is to calculate the cross section for the two-photon bremsstrahlung in the collision of fast electrons with neutral atoms. In particular the intention is to see if the contribution from atomic electrons is sufficiently large to be able to explain the discrepancy between theory and experiment reported by Hippler (1991).

In the high impact-energy region to be considered, electron exchange between projectile and atomic electrons can be ignored. Then three distinct processes can be identified, namely, emission of two photons by the projectile electron, emission of one photon by the projectile and one by an atomic electron and emission of both photons by the atomic electrons. Only the first process has hitherto been treated theoretically (Smirnov 1977, Florescu and Djamoi 1986, Véniard *et al* 1987).

Since we are dealing with spontaneous emission, the interaction with the radiation field of the scattering complex of projectile plus atom can be treated in lowest (second) order of perturbation theory. Then it can be shown (Kracke *et al* 1993) that the transition matrix for a collision in which two photons of frequencies ω_1 and ω_2 with polarization vectors ε_1 and ε_2 are emitted is

$$T = (1 + P_{12})\langle\chi_f^-|a_2 \cdot (\boldsymbol{p} + \boldsymbol{P})G^+(\mathcal{E}_1)a_1 \cdot (\boldsymbol{p} + \boldsymbol{P})|\chi_i^+\rangle. \tag{1}$$

Here χ_f^-, χ_i^+ are two exact states of the scattering complex satisfying incoming and outgoing wave boundary conditions respectively and G^+ is the Green operator

$$G^{\pm}(\mathcal{E}) = \left[\mathcal{E} - H_A - T - V \pm i\delta\right]^{-1} \tag{2}$$

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with atomic Hamiltonian H_A, projectile kinetic energy operator T and V the interaction between them, i.e. G^+ is the full propagator for the scattering complex. The coupling between the electrons and the radiation field is given in the velocity form with $\boldsymbol{P}$ denoting the momentum operator of the projectile and $\boldsymbol{p}$ the sum over the momentum operators $\boldsymbol{p}_n$ of the electrons of the atom with nuclear charge Z

$$\boldsymbol{p} := \sum_{n=1}^{Z} \boldsymbol{p}_n. \tag{3}$$

Assuming atomic units throughout, the vectors $\boldsymbol{a}_1$ and $\boldsymbol{a}_2$ are given by

$$\boldsymbol{a}_1 = \frac{\boldsymbol{\varepsilon}_1^*}{2\pi\sqrt{\omega_1}} \qquad \text{and} \qquad \boldsymbol{a}_2 = \frac{\boldsymbol{\varepsilon}_2^*}{2\pi\sqrt{\omega_2}}. \tag{4}$$

The operator P_{12} in (1) simply permutes the order of the emission of the photons. The energy $\mathcal{E}_1 = \mathcal{E}_i - \omega_1$ is the energy of the scattering complex after the emission of one photon of frequency ω_1. The total initial energy of the scattering complex is

$$\mathcal{E}_i = \tfrac{1}{2}K_i^2 + E_i \tag{5}$$

where $\boldsymbol{K}_i$ is the projectile initial momentum and E_i is the (ground-state) atomic binding energy before the collision. It is also clear that the final energy of the scattering complex is

$$\mathcal{E}_f = E_f + \tfrac{1}{2}K_f^2 = \mathcal{E}_i - \omega_1 - \omega_2 \tag{6}$$

where $\boldsymbol{K}_f$ is the final projectile momentum and E_f is the energy of the final atomic state.

The states of the scattering complex can be written,

$$|\chi_i^+\rangle = |\Psi_i\rangle + G^+(\mathcal{E}_i)V|\Psi_i\rangle \tag{7}$$

and

$$|\chi_f^-\rangle = |\Psi_f\rangle + G^-(\mathcal{E}_f)V|\Psi_f\rangle \tag{8}$$

where $|\Psi_i\rangle$, $|\Psi_f\rangle$ are the initial and final states of the scattering complex in the absence of projectile-target interaction, with energies $\mathcal{E}_i$, $\mathcal{E}_f$ respectively. The amplitude (1) can be separated into three terms representing the three processes enumerated above. These are as follows.

(i) Two-photon emission from the projectile

$$T_P = (1 + P_{12})\langle\chi_f^-|\boldsymbol{a}_2 \cdot \boldsymbol{P}\, G^+(\mathcal{E}_1)\boldsymbol{a}_1 \cdot \boldsymbol{P}|\chi_i^+\rangle. \tag{9}$$

(ii) One photon from the projectile, one from the atom ('mixed' projectile–atom contribution)

$$T_M = (1 + P_{12})(1 + P_{AP})\langle\chi_f^-|\boldsymbol{a}_2 \cdot \boldsymbol{p}\, G^+(\mathcal{E}_1)\boldsymbol{a}_1 \cdot \boldsymbol{P}|\chi_i^+\rangle \tag{10}$$

where P_{AP} is an operator permuting the momentum operators of the atom and the projectile.

(iii) Two-photon emission from the atomic electrons

$$T_A = (1 + P_{12})\langle\chi_f^-|\boldsymbol{a}_2 \cdot \boldsymbol{p}\, G^+(\mathcal{E}_1)\boldsymbol{a}_1 \cdot \boldsymbol{p}|\chi_i^+\rangle. \tag{11}$$

Thus far the treatment of the scattering complex is exact, however the evaluation of the corresponding T-matrix elements is hardly feasible. As restriction is made to high (keV) incident and final energies the exact propagator $G^{\pm}$ and the exact scattering states $|\chi_i^+\rangle$ and $|\chi_f^-\rangle$ will be expanded in the projectile–atom interaction V. Keeping only terms of first order in V the lowest-order contributions become

$$T_{\mathrm{P}} = (1 + P_{12})\big[T(\boldsymbol{a}_2 \cdot \boldsymbol{P}, \boldsymbol{a}_1 \cdot \boldsymbol{P}, V) + T(\boldsymbol{a}_2 \cdot \boldsymbol{P}, V, \boldsymbol{a}_1 \cdot \boldsymbol{P}) + T(V, \boldsymbol{a}_2 \cdot \boldsymbol{P}, \boldsymbol{a}_1 \cdot \boldsymbol{P})\big] \quad (12)$$

$$T_{\mathrm{M}} = (1 + P_{12})(1 + P_{\mathrm{AP}})\big[T(\boldsymbol{a}_2 \cdot \boldsymbol{P}, \boldsymbol{a}_1 \cdot \boldsymbol{p}, V) + T(\boldsymbol{a}_2 \cdot \boldsymbol{P}, V, \boldsymbol{a}_1 \cdot \boldsymbol{p}) + T(V, \boldsymbol{a}_2 \cdot \boldsymbol{P}, \boldsymbol{a}_1 \cdot \boldsymbol{p})\big] \quad (13)$$

$$T_{\mathrm{A}} = (1 + P_{12})\big[T(\boldsymbol{a}_2 \cdot \boldsymbol{p}, \boldsymbol{a}_1 \cdot \boldsymbol{p}, V) + T(\boldsymbol{a}_2 \cdot \boldsymbol{p}, V, \boldsymbol{a}_1 \cdot \boldsymbol{p}) + T(V, \boldsymbol{a}_2 \cdot \boldsymbol{p}, \boldsymbol{a}_1 \cdot \boldsymbol{p})\big] \quad (14)$$

where

$$T(X, Y, Z) := \langle \Psi_f | X G_0^+(\mathcal{E}_1) Y G_0^+(\mathcal{E}_1) Z | \Psi_i \rangle \quad (15)$$

and

$$G_0^+(\mathcal{E}) = \big[\mathcal{E} - H_{\mathrm{A}} - T + \mathrm{i}\delta\big]^{-1}. \quad (16)$$

Since all matrix elements now involve only free propagation of the projectile between radiative emission or non-radiative scattering it is a simple matter to integrate over projectile coordinates. If $\boldsymbol{R}$, $\boldsymbol{r}_n$ are the projectile- and atomic-electron coordinates with respect to the nucleus the interaction potential is

$$V = \sum_{n=1}^{Z} \frac{1}{|\boldsymbol{r}_n - \boldsymbol{R}|} - \frac{Z}{R}. \quad (17)$$

Then one finds that the various contributions can be put into the form (Kracke *et al* 1993)

$$T_{\mathrm{P}} = \frac{(\boldsymbol{a}_2 \cdot \boldsymbol{q})(\boldsymbol{a}_1 \cdot \boldsymbol{q})}{\omega_1 \omega_2} \langle \psi_f | F(\boldsymbol{q}) | \psi_i \rangle \quad (18)$$

$$T_{\mathrm{M}} = -(1 + P_{12}) \frac{(\boldsymbol{a}_2 \cdot \boldsymbol{q})}{\omega_2} \langle \psi_f | \boldsymbol{a}_1 \cdot \boldsymbol{p}\, G_{\mathrm{A}}^+(E_f + \omega_1) F(\boldsymbol{q}) + F(\boldsymbol{q}) G_{\mathrm{A}}^+(E_i - \omega_1) \boldsymbol{a}_1 \cdot \boldsymbol{p} | \psi_i \rangle \quad (19)$$

$$\begin{aligned} T_{\mathrm{A}} = (1 + P_{12}) \langle \psi_f | & \boldsymbol{a}_2 \cdot \boldsymbol{p} G_{\mathrm{A}}^+(E_f + \omega_2) \boldsymbol{a}_1 \cdot \boldsymbol{p}\, G_{\mathrm{A}}^+(E_f + \omega_1 + \omega_2)\, F(\boldsymbol{q}) \\ & + \boldsymbol{a}_2 \cdot \boldsymbol{p}\, G_{\mathrm{A}}^+(E_f + \omega_2) F(\boldsymbol{q}) G_{\mathrm{A}}^+(E_i - \omega_1) \boldsymbol{a}_1 \cdot \boldsymbol{p} \\ & + F(\boldsymbol{q}) G_{\mathrm{A}}^+(E_i - \omega_1 - \omega_2) \boldsymbol{a}_2 \cdot \boldsymbol{p} G_{\mathrm{A}}^+(E_i - \omega_1) \boldsymbol{a}_1 \cdot \boldsymbol{p} | \psi_i \rangle \end{aligned} \quad (20)$$

with $\boldsymbol{q}$ being the net momentum change of the projectile and

$$F(\boldsymbol{q}) := \frac{1}{2\pi^2 q^2} \left(\sum_{n=1}^{Z} \mathrm{e}^{\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{r}_n} / Z \right). \quad (21)$$

The final and initial states of the atom are $|\psi_f\rangle$, $|\psi_i\rangle$ and integration is now over atomic coordinates only. The Green operator in (19) and (20) is that for propagation of the atom alone, i.e.

$$G_{\mathrm{A}}^{\pm}(E) = \big[E - H_{\mathrm{A}} \pm \mathrm{i}\delta\big]. \quad (22)$$

The cross section for emission of two photons with frequencies ω_1 and ω_2 into solid angles $d\Omega_1$, $d\Omega_2$ respectively is given by

$$\frac{d\sigma}{d\omega_1\, d\omega_2\, d\Omega_1\, d\Omega_2} = \frac{(2\pi)^4}{c^6}\omega_1^2\omega_2^2\frac{K_f}{K_i}\sum_{\varepsilon_1,\varepsilon_2}\int d\Omega_f\, |T|^2. \tag{23}$$

The sum $\varepsilon_1, \varepsilon_2$ is a sum over all independent polarizations and the integration is over the scattering angles of the projectile. The velocity of light is denoted by c and T is either T_P, T_M, T_A for the separate contributions or the coherent sum $T_P + T_M + T_A$ for the total emission cross section. The T-matrices T_M and T_A which involve the atomic Green function have been calculated using the implicit summation method of Dalgarno and Lewis (1955) with an effective Herman–Skillman Hamiltonian. This independent-particle Hamiltonian was also used to generate the eigenstates of the krypton atom. The restriction has been made that the final state is the ground state of the krypton atom.

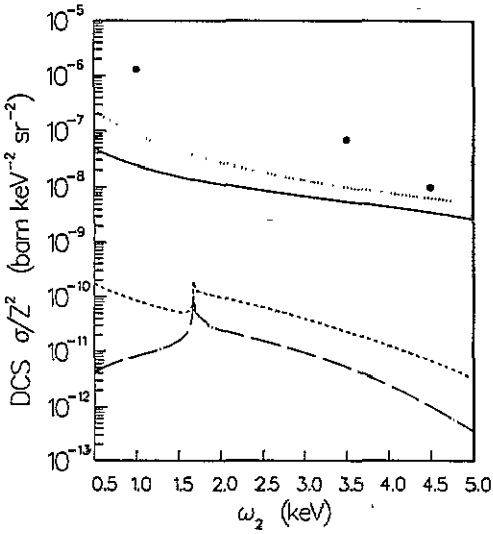

Figure 1. Two-photon bremsstrahlung cross section (full curve) divided by Z^2 for krypton plotted against photon energy ω_2. The incident electron energy is 8.82 keV and the photon energy $\omega_1 = 2.8$ keV. The photons are emitted in opposite directions perpendicular to the beam. The 'atomic' (chain curve) and 'mixed' (broken curve) contributions are also shown. The dotted curve is a distorted wave calculation of the two-photon projectile bremsstrahlung and the points are the experimental values of Hippler (1991).

The result for the particular case $\frac{1}{2}K_i^2 = 8.82$ keV, $\omega_1 = 2.8$ keV, $\theta_1 = \theta_2 = 90°$, $\phi_2 - \phi_1 = 180°$ and ω_2 variable are shown in figure 1 and compared to the measurements of Hippler (1991). One sees that the 'atomic' contributions are less than one per cent of the total emission cross section and cannot explain the discrepancy between calculated projectile two-photon bremsstrahlung and experiment. The pronounced resonance structure to be seen in the 'mixed' and 'atomic' contributions are due to the virtual excitation of the 2s and 2p subshell electrons into empty bound orbitals followed by a radiative decay into the initial state, i.e. they are simply due to electron impact excitation of atomic line radiation.

Since our estimates of the two contributions from processes involving atomic electrons cannot explain the discrepancy between the two-photon projectile bremsstrahlung and experiment one might question the three major approximations made here.

(a) The treatment of the projectile electron in lowest order, i.e. as a plane wave.

(b) The use of an effective one-particle mean field potential for the real and virtual atomic states.

(c) The restriction to the atomic ground state as the final state.

The first approximation (a) is justified by the high impact energy of the electron. To check this we have performed a calculation of the two-photon projectile bremsstrahlung of an electron scattered off the static potential of the charge distribution of the ground-state krypton atom, where the projectile electron is represented by a distorted wave. The resulting cross section is shown in figure 1 and differs only slightly from the plane wave result.

The second approximation (b) effects mostly the spectral distribution of the photons emitted from continuum target atom states excited by the colliding electron. The inverse radiative process, i.e. single photoionization, is known (Amusia and Cherepkov 1975) to be poorly described by a Herman–Skillman potential in the photon energy region up to some 100 eV. Here orders of magnitude error in the photoionisation cross section can be made. However in the keV photon energy region considered here the single particle approximation should be valid.

The third approximation (c) involves firstly the neglect of processes in which a bound electron is excited to a continuum state and then radiates back, not into the initial state but into some other empty bound or free state. These contribution could feasibly increase the atomic contributions by a factor of two or three but they can hardly explain the three orders of magnitude difference between the atomic contributions and experiment. Secondly, processes in which a bound electron is ionized by the projectile and the resulting hole is filled radiatively by another atomic electron have also been neglected. However these processes give rise to atomic line radiation and there are no prominent lines from such processes at the energies of the Hippler experiment.

In all cases involving atomic electrons, the excitation of the atomic electron by the projectile electron is the precursor of a radiative transition of the former. Hence the cross section for all such radiative processes is governed ultimately by the probability of inelastic scattering leading to virtual excitation or ionization of the target. The basic reason for the smallness of the contribution of atomic electrons to the bremsstrahlung cross section is that at these impact energies the probability of inelastic scattering is of the order of one percent (this also justifies the Born expansion used). However, although the atomic contributions are negligible here and cannot explain the Hippler measurement, they are expected to be significant at lower impact electron and photon energies, as has been shown in the case of one-photon bremsstrahlung (Amusia *et al* 1990). A detailed discussion of these lower energy processes in the simpler case of atomic hydrogen is in preparation (Kracke *et al* 1993). Here, a first estimate of the contribution of atomic bremsstrahlung to two-photon emission from many-electron atoms has been made.

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