PHYSICAL REVIEW A

VOLUME 35, NUMBER 1

Two-photon bremsstrahlung

V. Véniard and M. Gavrila

Fundamenteel Onderzoek der Materie (FOM), Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

A. Maquet

Laboratoire de Chimie Physique, Université Pierre et Marie Curie, 11 rue Pierre et Marie Curie, F-75231 Paris Cedex 05, France (Received 8 September 1986)

We present a calculation of the cross sections for two-photon bremsstrahlung in a Coulomb potential. The calculation was done in second-order nonrelativistic perturbation theory and in the dipole approximation. The potential was taken into account exactly by using the Coulomb Green's function. The matrix element was integrated analytically as far as possible and then computed numerically quite accurately. Limiting cases are discussed. Results are presented here for a special emission geometry. These are compared with the recent experiment of Altman and Quarles, and large discrepancies are found.

One or more bremsstrahlung photons can be emitted by an electron decelerated in an atomic collision, though the corresponding probability decreases rapidly with the number of photons. This is why all practical interest has been focused on the one-photon process, which has been studied in quite some detail.¹ On the other hand, the two-photon bremsstrahlung has attracted a different kind of interest, being regarded as an example of a multiple-production process since the early days of quantum mechanics, when it was first envisaged by Heitler and Nordheim.² Besides, it was realized that it is a typical quantum effect, in the sense that its probability cannot be estimated unambiguously from classical theory by application of the correspondence principle. Because of the complexity of the calculations involved, it was only much later that advances were made toward a quantitative description. Moreover, after many years in which the effect was an impossible challenge for the experimental detection capabilities, quite recently an effort was finally made toward its identification.

All calculations done so far refer to the Coulomb potential, and were carried out in the relativistic Born approximation, which is valid at high initial and final electron energies. Thus, even at high incident electron energies, the Born approximation is not apt to describe the entire photon spectrum (namely, its high-energy end, corresponding to low final electron energies). Within this approximation, special emission geometries were considered by Zazunov and Fomin,³ and by Nadzhafov,⁴ but subsequently Smirnov⁵ presented an intricate analytic formula to cover the general case. The situation is confused by the fact that these results do not agree among themselves in the cases of overlap, neither do their nonrelativistic limits agree with the simple direct evaluation of the nonrelativistic Born approximation.

The experiment was performed by Altman and Quarles⁶ and was carried out at x-ray energies. It is of the pioneering kind, at the limit of present detection capabilities. The results turned out to be almost three orders of magnitude larger than the prediction obtained from Smirnov's Bornapproximation cross section.⁶ This striking discrepancy raises questions concerning the applicability of the Born approximation at the relatively low energies of the experiment, about the correctness of Smirnov's formula, and about the experiment itself.

In an attempt to shed light on these issues, we now present the results of a nonrelativistic calculation of the two-photon bremsstrahlung in a Coulomb potential, done within the dipole approximation. The validity of the calculation is therefore restricted to photon energies and electron kinetic energies sufficiently small with respect to the electron rest energy mc^2 , and to sufficiently small nuclear charge Z $[(\alpha Z)^2 \ll 1]$, where α is the fine-structure constant].⁷ This calculation is a natural extension of one we have recently completed for the case of two-photon stimulated emission and absorption.⁸ It evaluates the matrix element given by second-order perturbation theory with no approximations, so that the initial and final velocities of the electron can be arbitrarily low. We have pushed the analytical calculation as far as possible and have eventually resorted to a very accurate numerical computation. Our analytical result represents the analog of that obtained by Sommerfeld⁹ for the one-photon case.

We assume that we are dealing with an electron transition from initial momentum \mathbf{p}_i to final momentum \mathbf{p}_f contained in $d \Omega_e$, and that the two photons have energies ω_o , polarization vectors \mathbf{e}_o , and propagation unit vectors $\mathbf{\hat{k}}_o$ contained in $d \Omega_o$ ($\sigma = 1,2$). The basic cross section for the process, defined as transition probability per unit time interval, per differential energy intervals and solid angles, divided by the incoming electron current, can be written¹⁰

$$\frac{d^5\sigma}{d\omega_1 d\omega_2 d\Omega_1 d\Omega_2 d\Omega_e} = \frac{\alpha^2 (\alpha Z)^4 \omega_1 \omega_2}{16(2\pi)^6} \frac{p_f}{p_i} |M|^2 , \quad (1)$$

where M is the transition matrix element. In the nonrela-

448

tivistic dipole approximation M is given by

$$M = \langle u_{\mathbf{p}_{f}}^{(-)} | (\mathbf{e}_{2} \cdot \mathbf{P}) G^{(+)}(\Omega_{1}) (\mathbf{e}_{1} \cdot \mathbf{P}) | u_{\mathbf{p}_{i}}^{(+)} \rangle$$
$$+ \langle u_{\mathbf{p}_{f}}^{(-)} | (\mathbf{e}_{1} \cdot \mathbf{P}) G^{(+)}(\Omega_{2}) (\mathbf{e}_{2} \cdot \mathbf{P}) | u_{\mathbf{p}_{i}}^{(+)} \rangle . \quad (2)$$

Here **P** is the electron momentum operator, $u_{\mathbf{p}_{l}}^{(-)}, u_{\mathbf{p}_{l}}^{(+)}$ are continuum Coulomb states corresponding to the indicated asymptotic momenta and having incoming or outgoing spherical wave behavior, respectively, and $G^{(+)}(\Omega)$ is the Coulomb Green's operator for energy parameter $\Omega + i\epsilon$ (for definitions, see Ref. 8). The values of interest for Ω are $\Omega_{\sigma} = E_{i} - \omega_{\sigma}$ ($\sigma = 1, 2$). Conservation of energy requires that $E_{i} = E_{f} + \omega_{1} + \omega_{2}$. Our formulas are written in Z-scaled atomic units, i.e., momenta are expressed in units of Z a.u., and energies in units of $Z^{2} \mathcal{R}$, where \mathcal{R} is the rydberg except when otherwise noted.¹¹

Rotational invariance requires that M can be written as

$$= P(\mathbf{e}_1 \cdot \mathbf{e}_2) + Q(\mathbf{e}_1 \cdot \mathbf{v}_i)(\mathbf{e}_2 \cdot \mathbf{v}_i) + R(\mathbf{e}_1 \cdot \mathbf{v}_i)(\mathbf{e}_2 \cdot \mathbf{v}_f)$$

+
$$S(\mathbf{e}_1 \cdot \mathbf{v}_f)(\mathbf{e}_2 \cdot \mathbf{v}_i) + T(\mathbf{e}_1 \cdot \mathbf{v}_f)(\mathbf{e}_2 \cdot \mathbf{v}_f)$$
, (3)

where $\mathbf{v}_{\alpha} = \mathbf{p}_{\alpha}/p_{\alpha}$ ($\alpha = i, f$), and P, Q, R, S, T are invariant amplitudes.

We have carried out the integrations involved in Eq. (2) in momentum space by applying techniques developed earlier.^{8,12} After a tedious analytical calculation the amplitudes of Eq. (3) were expressed as one-dimensional integrals over Gauss hypergeometric functions. A typical example is P, equal to $P = P(\Omega_1) + P(\Omega_2)$, where $P(\Omega)$ is given in Ref. 8, Eqs. (8) and (9).

This rather complicated analytic result simplifies considerably in a number of *limiting cases*.

(1) High initial and final electron energies $(E_i, E_f \gg 1)$. The analytic result reduces in this limit to¹³

$$\frac{\frac{d^{5}\sigma}{d\omega_{1}d\omega_{2}d\Omega_{1}d\Omega_{2}d\Omega_{e}}}{=\frac{a^{2}(aZ)^{4}(\mathbf{e}_{1}\cdot\boldsymbol{\Delta})^{2}(\mathbf{e}_{2}\cdot\boldsymbol{\Delta})^{2}}{16\pi^{4}\omega_{1}\omega_{2}}\frac{p_{f}}{p_{i}}|f_{B}|^{2}, \quad (4)$$

with $f_B = 2\Delta^{-2}$ and $\Delta = \mathbf{p}_f - \mathbf{p}_i$. Equation (4) can be obtained also directly from Eqs. (1) and (2) by applying the Born approximation to the initial and final states of the electron, as well as to the Green's function. By summing over the photon polarizations in Eq. (4) we get¹³

$$\frac{d^{2}\overline{\sigma}}{d\omega_{1}d\omega_{2}d\Omega_{1}d\Omega_{2}d\Omega_{e}} = \frac{\alpha^{2}(\alpha Z)^{4}}{4\pi^{4}} \frac{1}{\omega_{1}\omega_{2}} \frac{p_{f}}{p_{i}} [1 - (\hat{\mathbf{k}}_{1} \cdot \boldsymbol{\delta})^{2}] [1 - (\hat{\mathbf{k}}_{2} \cdot \boldsymbol{\delta})^{2}] , \quad (5)$$

where $\delta = \Delta/\Delta$. Equation (5) disagrees with the nonrelativistic limits of Zazunov and Fomin,³ and of Smirnov.^{5,14}

(2) Low photon-electron energy ratio for one of the photons only $(\omega_2/E_f \ll 1, \text{ although } \omega_2 \text{ itself needs not to be small with respect to 1; <math>\omega_1$ unrestricted). To lowest order in ω_2 the analytic result for M is $M = 2\omega_2^{-1}(\mathbf{e}_2 \cdot \Delta) \times M'(\mathbf{p}_f, \mathbf{p}_i, \mathbf{e}_1)$, where M' is the one-photon bremsstrahlung matrix element connecting the electron states of momentum \mathbf{p}_i and \mathbf{p}_f , with the emitted photon having polarization \mathbf{e}_1 . Thus

$$\frac{d^{5}\sigma}{d\omega_{1}d\omega_{2}d\Omega_{1}d\Omega_{2}d\Omega_{e}} = \frac{\alpha(\alpha Z)^{2}}{4\pi^{2}} \frac{(\mathbf{e}_{2}\Delta)^{2}}{\omega_{2}} \frac{d^{3}\sigma}{d\omega_{1}d\Omega_{1}d\Omega_{e}},$$
(6)

where $d^3\sigma/d\omega_1 d\Omega_1 d\Omega_e$ is the one-photon bremsstrahlung cross section corresponding to M'.¹⁵ Equation (6) is an extension to our case of the low-energy theorem of Low¹⁶ for one-photon bremsstrahlung.

(3) Low photon-electron energy ratios for both photons $(\omega_1/E_f \ll 1, \omega_2/E_f \ll 1, although \omega_1, \omega_2$ themselves need not be small with respect to 1). In this case, to lowest order in ω_1 and for $\theta \neq 0$, the one-photon bremsstrahlung matrix element can be written

$$M'(\mathbf{p}_f, \mathbf{p}_i) = \frac{4\pi}{\omega_1} (\mathbf{e}_1 \cdot \mathbf{\Delta}) f_C(\mathbf{p}_i, \mathbf{\Delta}) \quad , \tag{7}$$

where $f_C(\mathbf{p}_i, \Delta)$ is the Coulomb elastic scattering amplitude for energy E_i [see Ref. 8, Eq. (11)]. Since $|f_C|^2 = |f_B|^2$, the fivefold differential cross section Eq. (6) coincides with Eq. (4) and by summing over the photon polarizations we again get Eq. (5).¹⁷

The number of variables in the fivefold differential cross section Eq. (1) is too large to keep track of all of them. In experiment⁶ the direction of the final electron momentum was not recorded, neither were the photon polarizations. We denote the corresponding fourfold cross section [i.e., Eq. (1) summed over $\mathbf{e}_1, \mathbf{e}_2$ and $d\Omega_e$] by $d^4\overline{\sigma}/d\omega_1 d\omega_2 d\Omega_1 d\Omega_2$.¹⁰

We have first computed the cross section Eq. (1) with great accuracy (to better than 10^{-5} relative accuracy) and then $d^4\overline{\sigma}/d\omega_1 d\omega_2 d\Omega_1 d\Omega_2$. We have checked that in the limiting cases (1), (2), and (3) considered above our numerical results go over smoothly into the predictions obtained from the analytical formulas given there.

Of the variety of emission directions $\hat{\mathbf{k}}_1$ and $\hat{\mathbf{k}}_2$ possible, we shall consider here the *special geometry* of the Altman and Quarles experiment:⁶ $\hat{\mathbf{k}}_1$ and $\hat{\mathbf{k}}_2$ are perpendicular to \mathbf{p}_i , and opposite to each other ($\hat{\mathbf{k}}_1 = -\hat{\mathbf{k}}_2$). For reference, we give the fourfold differential cross section corresponding to Eq. (5) in this special geometry:

$$\frac{d^{4}\overline{\sigma}}{d\omega_{1}d\omega_{2}d\Omega_{1}d\Omega_{2}} = \frac{\alpha^{2}(\alpha Z)^{4}}{2\pi^{3}} \frac{x}{\omega_{1}\omega_{2}} \left[\frac{27}{16} - \frac{5}{8}x^{2} + \frac{3}{16}x^{4} + (3x^{2} - 5)\frac{(1 - x^{2})^{2}}{64x} \ln\left[\frac{(1 - x)^{2}}{(1 + x)^{2}}\right] \right],$$
(8)

which is relevant for the limiting cases (1) and (3) above. We have denoted here $x = p_f/p_i$.

In Figs. 1 and 2 we represent $\log_{10}(Z^{-4}d^{4}\overline{\sigma}/d\omega_{1}d\omega_{2} \times d\Omega_{1}d\Omega_{2})$ for some fixed values of E_{1} and ω_{1} , as function of ω_{2} . A common trait of these figures is that all cross sections are monotonically decreasing with ω_{2} . The fastest

decrease occurs at small ω_2 , where the cross sections behave as $1/\omega_2$ [see Eq. (6)]. The cross sections are always enhanced by diminishing ω_1 . In a few cases we have also shown the limiting formula Eq. (8) (dashed curves). Its validity is quite restricted, and the error increases toward the upper end of the photon spectrum.¹⁸

449

M =

450

V. VÉNIARD, M. GAVRILA, AND A. MAQUET



FIG. 1. Two-photon bremsstrahlung cross section $\log_{10}(Z^{-4} \times d^4 \overline{\sigma}/d \omega_1 d \omega_2 d \Omega_1 d \Omega_2)$ dependence on ω_2 , at given E_i and ω_1 (as marked), for the special geometry considered in the text. All energies are given in units of $Z^2 \mathcal{R}$, and the cross section $d^4 \overline{\sigma}/d \omega_1 d \omega_2 d \Omega_1 d \Omega_2$ in $Z^{-6}(a_0/\mathcal{R})^2$ units (see Ref. 11). The dashed line represents Eq. (8).

In Fig. 3 we represent the ω_2 dependence of $(Z^{-4} \times \omega_1 \omega_2 d^4 \overline{\sigma} / d \omega_1 d \omega_2 d \Omega_1 d \Omega_2)$ at $\omega_1 = 0.25$ and various E_i . The dashed straight line represents the limiting case of very large E_i , when Eq. (8) becomes valid over the finite interval ω_2 shown (with $x \rightarrow 1$).

Let us now compare our results with experiment.⁶ This was carried out with gold (Z = 79) as a target, at incident electron energy of 75 keV ($E_i = 0.883Z^2\mathcal{R}$) and one of the photon energies fixed at 20 keV ($\omega_1 = 0.236Z^2\mathcal{R}$). In Fig 4 we represent the experimental ω_2 spectrum recorded, and a number of theoretical results. The latter are unscreened Coulomb calculations done for Z = 79. Curve (a) represents the results of our numerical calculation, based on Eqs. (1) and (2); curve (b) represents the nonrelativistic Born approximation, Eq. (8); curve (c) was computed⁶ from the relativistic Born approximation formula given by Smirnov.⁵

In general, curve (b) differs from (a) by a factor varying between 2 and 3, the disagreement being larger at the high-energy end of the spectrum.¹⁸ The two Born approximations (b) and (c) differ from each other by some 30-40%. This may be expected, since the main relativistic corrections should be of order $(\alpha Z)^2$, i.e., about 33%. Striking, however, is the discrepancy between experiment and our curve (a) [and curves (b) and (c) as well], the experimental results lying nearly three orders of magnitude higher. Surely, in order to get the true theoretical cross section for comparison with experiment, our curve (a) should be corrected for relativity and screening. At the low electron energies involved, relativistic corrections



FIG. 2. Same as for Fig. 1.

TWO-PHOTON BREMSSTRAHLUNG



FIG. 3. ω_2 dependence of cross section $(Z^{-4}\omega_1\omega_2d^4\bar{\sigma}/d\omega_1d\omega_2d\Omega_1d\Omega_2\times 10^{15})$ at $\omega_1=0.25$ and various E_i (as marked). Same units as in Figs. 1 and 2. The dashed straight line is obtained from Eq. (8), with x = 1.

should be of order $(aZ)^2$ [see the variance of curves (b) and (c)], whereas screening effects should depress to some extent the result (a) (as a consequence of replacing Z by some lower effective Z_e), thereby increasing the discrepancy with experiment. Thus, it does not appear possible to bridge the gap between theory and experiment by improving the theory. We believe that this either leaves a question mark on the experiment, or may possibly indicate the presence of some physical effect not accounted for.

- ¹R. H. Pratt and I. J. Feng, in *Atomic Inner Shell Physics*, edited by B. Crasemann (Plenum, New York, 1983).
- ²W. Heitler and L. Nordheim, Physica 1, 1059 (1934).
- ³L. G. Zazunov and P. I. Fomin, Zh. Eksp. Teor. Fiz. **46**, 1392 (1964) [Sov. Phys. JETP **19**, 941 (1964)].
- ⁴I. M. Nadzhafov, Yad. Fiz. 11, 416 (1970) [Sov. J. Nucl. Phys. 11, 232 (1970)].
- ⁵A. I. Smirnov, Yad. Fiz. **25**, 1030 (1977) [Sov. J. Nucl. Phys. **25**, 548 (1977)].
- ⁶J. C. Altman and C. A. Quarles, Phys. Rev. A **31**, 2744 (1985).
- ⁷The limitations of our results in comparison to an exact relativistic calculation should be the same as for the usual bremsstrahlung case. For details, see Ref. 1.
- ⁸M. Gavrila, A. Maquet, and V. Véniard, Phys. Rev. A **32**, 2537 (1985); and (unpublished).
- ⁹A. Sommerfeld, Ann. Phys. (Leipzig) 11, 257 (1931).
- ¹⁰The dimensions of the cross section are thus (length)²/ (energy)².
- ¹¹Consequently, Eq. (1) is given in units of $Z^{-6}(a_0/\Re)^2$. The conversion to the experimental units can be made by using $(a_0/\Re)^2 = 1.513 \times 10^{17} \,\mu$ barns/keV².
- ¹²M. Gavrila, Phys. Rev. A 6, 1348 (1972).
- ¹³The fact that we are using Z-scaled atomic units yields the factor Z^4 in Eqs. (4) and (5). When passing to the usual atomic units Eq. (4) contains only the factor Z^2 , as should be



FIG. 4. Two-photon bremsstrahlung cross section $d^4\bar{\sigma}/d\omega_1 d\omega_2 d\Omega_1 d\Omega_2$ [in μ barns/(keV)² (sr)²] vs ω_2 (in keV), for Z = 79, $E_i = 75$ keV, $\omega_1 = 20$ keV, from the experiment of Altman and Quarles (Ref. 6), and theory: (a) present numerical results; (b) nonrelativistic Born approximation, Eq. (8); (c) relativistic Born approximation (Refs. 5 and 6).

This work was jointly sponsored by the Centre National de la Recherche Scientifique (CNRS), France, and by The Netherlands Organization for the Advancement of Pure Research (ZWO) through its foundation Fundamenteel Onderzoekder Materie (FOM). The Laboratoire de Chimie Physique of the Université Pierre et Marie Curie is a "Laboratoire Associé au CNRS."

in Born approximation.

- ¹⁴In the nonrelativistic limit, Smirnov finds an angle independent result [see Ref. 5, Eq. (7); this formula differs from our Eq. (5) in that it does not contain the last two factors]. On the other hand, Zazunov and Fomin [Ref. 3, Eq. (8)] find nonvanishing results in what they call the "parallel" and "antiparallel" cases, whereas from Eq. (5) we find zero.
- ¹⁵The infinite rise of the cross sections Eqs. (6) and (7) for vanishing photon energies is yet another aspect of the infrared divergences of quantum electrodynamics [e.g., Jauch and Rohrlich, *The Theory of Electrons and Photons* (Springer, New York, 1976)].
- ¹⁶F. E. Low, Phys. Rev. **110**, 974 (1958).
- ¹⁷Note that for the Coulomb potential, Eq. (7) does not hold for forward scattering $\theta = 0$, even though $\Delta \neq 0$ and f_C is finite; consequently, neither does Eq. (6) hold in this case. However, since the error extends only over angles of order ω_2 this will not affect an integrated cross section, such as Eq. (8).
- ¹⁸At the upper end of the ω_2 spectrum (x = 0), Eq. (8) vanishes, whereas the exact result does not. Such large discrepancies may be expected, since the criteria of validity of Eq. (8), from both points of view under which it was derived [limiting cases (1) or (3) above], are strongly violated: now $p_f \rightarrow 0$, and $\omega_2/E_f \rightarrow \infty$.