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PHOTON ASSISTED COLLISIONS IN ATOMS AND MOLECULES

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INTRODUCTION TO PHOTON-ASSISTED  
ELECTRON-ATOM SCATTERING

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## INTRODUCTION TO PHOTON-ASSISTED ELECTRON-ATOM SCATTERING

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**Abstract** An introduction to the processes included in the photon-assisted electron-atom collision is given. The role of the theory is emphasized, critically examining the developments concerning utilization of perturbation theory. The major non-perturbative results are outlined and some of the evolving features in the area of research are critically examined.

### PROCESSES

Photon-assisted electron-atom collision (PEAC) is a recent field activity in atomic physics. The area of research called bremsstrahlung (and its inverse) can consider PEAC to be its natural progeny. The semiclassical papers of Goppert-Mayer<sup>1</sup>, Volkov<sup>2</sup>, and Bethe and Heitler<sup>3</sup> have anticipated and influenced much of the work in this area. The classic books of Sommerfeld<sup>4</sup>, Heitler<sup>5</sup> and Bethe and Salpeter<sup>6</sup> still serve as the finest introduction to the field.

The growing interest in PEAC has been due to the giant progress that continue to be made in producing powerful lasers. On one hand, this progress has spurred theorists in carrying out anticipatory calculations, which can only be checked ex-

perimentally when some of these powerful lasers are coupled to the sophisticated electron beam collisional apparatus. The class of the phenomena associated with PEAC has also considerable bearing on the plasma, where observables are average quantities obtained en passant distributions. In this introductory article on PEAC, we shall concentrate on the basic processes, thereby relegating this second aspect only to the cited bibliography.<sup>7-108</sup>

The field of electron atom collision involves study of the following processes:

$$e + A(i) \rightarrow e + A(i) \quad (1)$$

$$e + A(i) \rightarrow e + A(j) \quad (2)$$

$$e + A(i) \rightarrow e + e + A^+(j) \quad (3)$$

These are three processes: (i) elastic scattering, (ii) excitation and (iii) ionization of an atom (initially in a particular state defined by the index  $i$  in the bracket). Stripping of more than one electron can be considered to be generically included in the ionization process.

PEAC is the extension of these processes by absorption (or emission) of photons. (These processes are all presumed to be stimulated by a reasonably strong external electromagnetic field). We shall for simplicity write down only those involving the absorption of a single photon:

$$e + \gamma + A(i) \rightarrow e + A(i) \quad (4)$$

$$e + \gamma + A(i) \rightarrow e + A(j) \quad (5)$$

$$e + \gamma + A(i) \rightarrow e + e + A^+(j) \quad (6)$$

Notice that these processes have characteristic inprints

that distinguish them from those given in (1)-(3) in a fundamental manner. Let us discuss this in some detail, starting from the process (4).

For this process, in which the atom is left in its initial state, the energy of the photon is transferred to the electron. This increase can come by direct absorption of the photon by the electron which scatters elastically off the atom or the atom may absorb the photon, make a virtual transition (in general, the photon energy will not match the atomic frequencies), transfer the energy to the scattering electron, and drop back into the initial state. This process, i.e. (4), is well-known as a single photon bremsstrahlung and has a respectable history behind it, both from the classical and quantum treatment accorded to it. There is a multiphoton extension to the process, i.e. the electron may gain (or lose) the energy of  $N$  ( $N \geq 1$ ), photon, always leaving the atom in the initial state.

This process can be theoretically studied at various levels. Firstly, one may assume that since the atom begins and ends in its initial state, its spectrum may not enter in the problem at all. A suitable potential can mimic the electron-atom interaction. The process, then, may be treated either classically or quantum mechanically by means of scattering theory. The process can also be envisaged to occur at very high value of the intensity of the electro-magnetic field, so that the electron-field coupling cannot be treated perturbatively. The absorption may be, as pointed out

earlier, of multiphotonic kind as well. A great deal of attention has been paid to these questions recently.

An additional point of interest is the possibility that the atom-photon interaction is equally important as the electron-photon interaction. Note that this does not mean that both are very strong, i.e. the field-matter coupling can continue to be treated perturbatively. While for particular values of the parameters involved in the process one or the other coupling may dominate, it may very well be that both are important for many situations of interest. Physically, one may say that for the low-frequency photon, the atom is essentially 'transparent' to the field, so that one can ignore the photon-atom coupling, while for the resonant frequency, i.e. if the frequency of the photon matches two levels of the atom, the photon-atom coupling becomes extremely important, with the concomitant simplification that the atom may be described by a two level system. In the very broad region, where the frequency of the photon is neither low nor matches the atomic frequencies, the atomic structure will be crucial.

Consider next the process given in (5). The principle of observation of this process is simple. The electron gain of the photon energy is now modified by the energy difference between the relevant atomic energy levels, which depending on the kinematics, may either be a loss or a gain in the final electron energy. To make this clear, see the figure below.

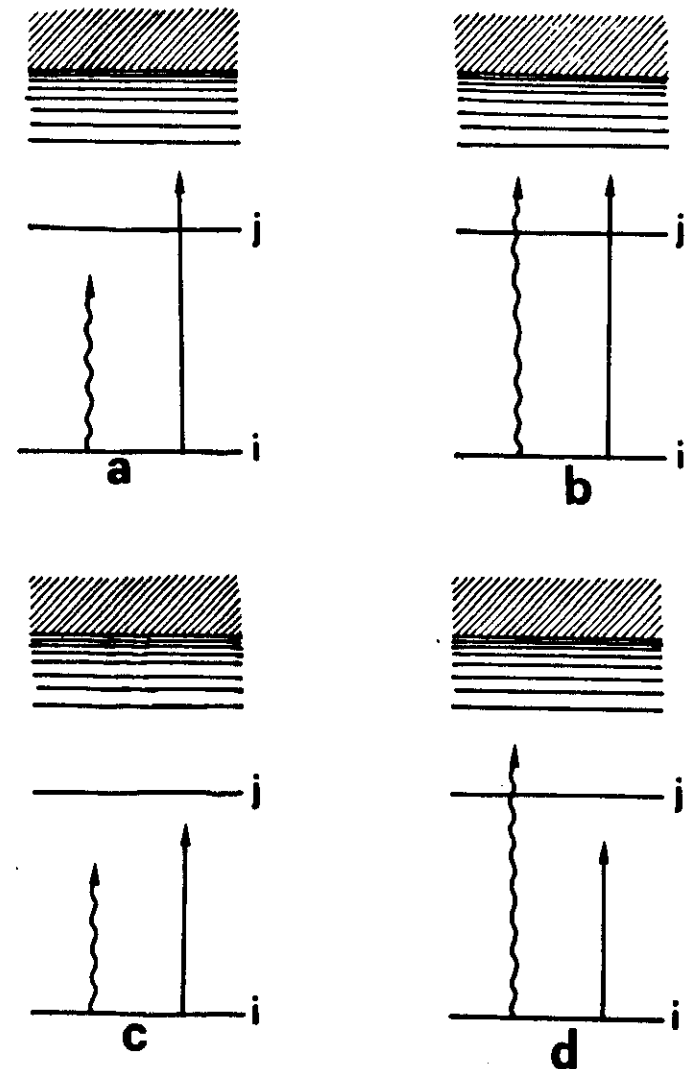


FIGURE 1. Schematic energy representations of the processes of PEAC.

On the drawn energy levels of an atomic system ending in a continuum, the energies of the photon and the electron are shown respectively by a wiggly and a straight vertical line, the height of each being the measure of the energy. Consider, for all the four cases, the excitation the first excited level, shown by  $j$ . For Fig. 1a, the electron will lose energy and for Fig. 1b, an electron with the same initial energy will gain energy (since the photon has excess energy for the excitation process). The electron lose energy for the scenario in Fig. 1c, which Fig. 1d, correspond to increase of the energy in the collision process. What we see here is a set of novel processes, which can only be seen with appreciable probability by utilizing a powerful photon source. Figs. 1b and 1d, correspond to superelastic scattering, but from the ground state of the atom! Secondly, the process shown in Fig. 1c, correspond to an excitation, which can only be achieved by this kind of photon-assisted collision.

Consider next the ionization i.e. (6). The kinematical arrangement shown in Fig. 1, can be extrapolated to the ionization case. Fundamentally, all the remarks above can be extended to this process as well, substituting excitation by ionization. The fact that the transitions involve two electrons in the continuum in the final state offers interesting experimental possibilities, while the theoretical treatments have to grapple with the subtleties of proper boundary conditions.

### PERTURBATION THEORY

The hamiltonian of electron-atom-radiation system can, in general, be written as

$$H = H_{AT} + H_e + H_{RAD} + H_{e-AT} + H_{AT-RAD} + H_{e-RAD} \quad (7)$$

where the terms are, in order, the atomic, the electronic and the radiation hamiltonians, the electron-atom interaction, the atom-radiation interaction and the electron-radiation interaction hamiltonians. From the proliferation of the terms in (7), one can suspect that the problems are non-trivial, which is indeed the case. Note at the outset, that the processes may be described by utilizing either classical or quantum description of the electromagnetic field, as long as the processes are stimulated at large photon density. This means that the  $H_{RAD}$  term can be dropped from our consideration.

In this section, we shall consider the case where  $H_{int} = H_{e-AT} + H_{AT-RAD} + H_{e-RAD}$  can be treated as a perturbation to the rest of the hamiltonian. Among these three terms, the first concerns field-free scattering. Treatment by means of perturbation theory, imply limitations on the radiation field: the e.m. field cannot be so strong as to dominate over the atomic potentials involved. In the processes that we are concerned, this will mean that the lower order photon processes, dominate over the higher order processes. This limitation, while a theoretical constraint, will allow us to consider a remarkably large domain of intensity. Indeed, the intensity at which the

breakdown of the perturbation theoretic treatment becomes apparent (in the limited sense of the utility of the lowest order terms) has not yet been established. The lowest order treatment is useful for calculating the probability of processes for a large region of intensity, and additionally, serve as the limiting value for non-perturbative theories.

If the approximation that the  $H_{e-AT}$  term can be replaced by a local potential  $V(r)$  the lowest order theory gives rise to two terms shown in Fig. 2. The two diagrams (a) and

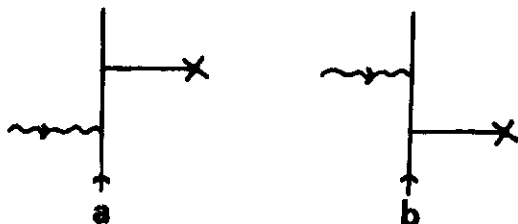


FIG. 2. Potential scattering description of PEAC.

(b) represent the amplitudes in which, for (a), the electron absorbs the photon and subsequently interacts via the potential, while for (b), the electron interacts with the potential followed by photon absorption. The vertical line represents the electron propagating from bottom to top, the wavy line is the photon, (the arrow showing absorption) and the horizontal straight line ending in a cross, represents the potential. The cross-section of this process is given

essentially in all texts, (e.g. Sommerfeld, Bethe and Salpeter). The point to note is that with a powerful laser, the probability of these processes, being proportional to the intensity of the field, can be significantly boosted, a point not taken into consideration in the classics.

This appealing picture of the process have to be modified with the increase of the intensity (standard perturbation theoretic treatment breaks down). But, also at lower intensities, the treatment is non-applicable as the frequency of the photon increases from very low frequencies. The photon interacting with the atom begins to be important with increasing frequencies, and this interaction can be taken into account by replacement of  $V(r)$  by the exact  $H_{e-AT}$ . The photon now couples to both the scattering electron and the atom, and diagrammatically, we can write the amplitude, as shown in Fig. 3 (next page).

Here the double line correspond to the atom. Notice that the scattering description has not been modified, i.e. we still have single scattering. However, now, to the same order of matter-field coupling, one has four terms, the two new terms arising from absorption of the photon by the atom.

The crossed line in Fig. 3 (for hydrogen atom) correspond to:

$$H_{e-AT} = \frac{1}{r_{12}} - \frac{1}{r_1}$$

where 1 and 2 correspond to the scattering electron and the atomic electron respectively. Generalization to other atoms

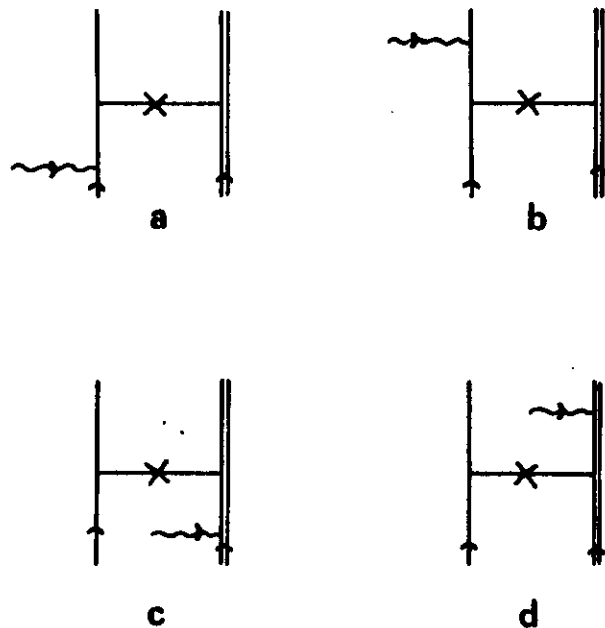


FIG. 3. The four lowest order terms of PEAC.

is obvious. If one computes the four terms in Fig. 3, one sees that with the increase in  $\omega$ , the effect of the last two terms increases with frequency in a uniform manner. Now, the atomic hydrogen problem can be worked out exactly, since the Green's function of a single particle in the Coulomb field gives rise to closed form analytical functions. Thus, the effect of atomic structure become quite transparent. However, approximate calculations, involving many-body techniques also show that for non-hydrogenic atoms, this

effect is present.

Before leaving this aspect of the problem, it is worthwhile to underline the fact that the modification of the standard expressions in the texts is of a fundamental nature. At high energy, single scattering predominates. This single scattering, allows one, to clarify the problem, i.e. it produces four unambiguous terms in the amplitude as shown in Fig. 3. Each are of the same order in perturbation series. The physically reasonable hypothesis of the predominance of the diagrams of Fig. 3a and Fig. 3b over Fig. 3c and Fig. 3d is limited to only quite low frequencies. At higher frequencies, the other two terms contribute substantially and one must make use of this before arriving at comparison of theory with experiment.

It is expected that experiments are performed not with hydrogen atoms but with other atoms, such as rare gases. Therefore, calculations of the terms in Fig. 3 for these atoms are of value. The calculations will be of similar nature as the second order Born approximation, i.e. in the final analysis, one would need to make further approximation on the Green's function of the atom. The important frequency dependence is, however, indirectly seen in the low energy calculations of Argon.<sup>107</sup> The much more transparent high energy calculation, with the rare gases, therefore, would be an importance to guide experiments in this area.

Consider now the case where the scattering cannot be

calculated perturbatively, but the photonic absorption can. The need for such calculation has long been felt for calculation of the absorption coefficient at a given temperature for the electron in various atomic surrounding. These are useful for astrophysical problems, as well as for plasma experiments. A perusal of the Boltzmann factor immediately show that the cross-section needed are at low energies. These calculations are now quite valuable for possible beam experiments, since in order to obtain the various probabilities one needs only to scale the cross-section with the appropriate intensity factor. What is lacking, however, are calculations done at the frequencies of the various lasers. This is a gap that can be filled without any additional theoretical problem. From the experience of the history of the electron-atom scattering calculations, it seems, however, it would be best to take the appropriate photon energies, and then compute the cross-sections involved in Fig. 3a, b, c and d at all energies. These would be the bench mark calculations, useful at high energies, serve as a preliminary indicator for experimental purposes and will also be useful to show their limitations when and if low energy calculations are done.

Let us now consider the case where the atom is left in a state different from the initial state, limiting ourselves again to the region where  $H_{int}$  can be treated perturbatively. Buimistrov suggested that this may indeed be new method for atomic excitation, especially when the

frequency of the photon is near resonant to the atomic transition. While it is indeed simple to treat the near-resonant case, as far as atomic level structure is concerned a theory for all frequencies was developed somewhat later. The problem simplifies in the high energy to the consideration of the terms of Fig. 3 only, with the only difference that the final atomic state is different from the initial one. Calculation for these has been performed with the treatment of the intermediate virtual state by means of a Sturmian expansion for the 1S-2S and 1S-3S transitions of the hydrogen atom. The dependance on the frequency as well as the energy has been reported. The frequency dependence near resonance points up essentially the suggestion by Buimistrov, i.e. the cross-section rises enormously. These resonances are very broad. The process of excitation remains an interesting phenomenon at non-resonant frequencies, i.e. those available with powerful lasers, since here truly a new atomic phenomenon is involved. It is worthwhile, therefore, that calculations involving non-hydrogenic atoms are also attempted, since these would surely be of more interest vis-a-vis experiments.

There are various ancillary questions related to the processes of this kind. These include the angular distribution, with respect to both the scattering angle as well as the polarization direction, the Raman-like aspect of the electron energy, the possibility of super-elastic scattering etc. Some of these questions have by now amply discussed.

The low energy scattering involving photon absorption, (i.e.  $H_{e-AT}$  cannot be treated perturbatively) involves various interesting questions. For example, there exists kinematic region where the excitation possible is only by means of the electron-photon joint excitation. This is an experimentally interesting area since the signals can only be due to a unique process. The calculations in this region while difficult are comparable to the usual scattering calculations and therefore, to be welcomed. Finally, the threshold of these excitations are situated at a different region than that of the ordinary electronic excitation. The behaviour of the cross-section at the threshold remains one of the other unexplored area.

#### NON-PERTURBATIVE REGIME

As the sub-heading indicates,  $H_{AT-RAD} + H_{e-RAD}$  cannot be treated perturbatively all the time. Two approaches have generally been used to confront this case. Firstly, and this is overwhelmingly the majority approach, one stipulates that  $H_{AT-RAD}$  can be suppressed. The radiation coupling with the scattering electron is presumed to describe most of the physics. The interaction of the electron with the atom is treated by means of a potential  $V(r)$ .

The Schrödinger equation or a charged particle coupled to a plane wave e.m. field has an exact solution. (The above is true even for the Klein-Gordon and the Dirac equations). The solution goes under the name of Volkov solution (V.S.).

For the Schrödinger equation, the V.S. is given by

$$\begin{aligned}\psi_p(\vec{r}, t) &= \exp. \left[ \frac{-i p^2}{2m} t + i \vec{p} \cdot \vec{r} - i \phi_p(t) \right] \\ \phi_p(t) &= \int_0^t dt' \left[ \frac{-e}{mc} \vec{A} \cdot \vec{p} + \frac{e^2 A^2}{2mc^2} \right]\end{aligned}\quad (8)$$

The symbols all have the usual meaning. Note that this is the semiclassical solution, (i.e. the e.m. field is treated classically). While we can utilize this solution only for all our subsequent discussion, the fully quantum mechanical solution also exist, given by<sup>11</sup>

$$\psi_{np} = \sum_{e=-\infty}^{\infty} \gamma_e(\vec{p}) |n+\ell\rangle |\vec{p}-\ell\hbar\vec{k}\rangle \quad (9)$$

where  $n+\ell$  are the number states of photons, and  $|\vec{p}-\ell\hbar\vec{k}\rangle$  are the electronic wave function with momentum  $\vec{p}-\ell\hbar\vec{k}$ ,  $\vec{k}$  being the momentum of the photon.  $\gamma_e(p)$  are coefficients that depend on the density of the photons as well as its frequency and polarization.

The interpretation of Eq. (9) and also of Eq. (8) (after a Fourier decomposition of the exponential term involving the field has been made) can be seen simply by means of the diagrams shown below in Fig. 4. Since the diagrams tell the story in a concise and far easier manner than algebraic expressions, we shall from now on resort to them. The top diagram which is a vertical line propagating from bottom to top and a square embedded on it represents the V.S. This consists of a sum of an infinite member of amplitudes, the first few of which are shown in an increasing

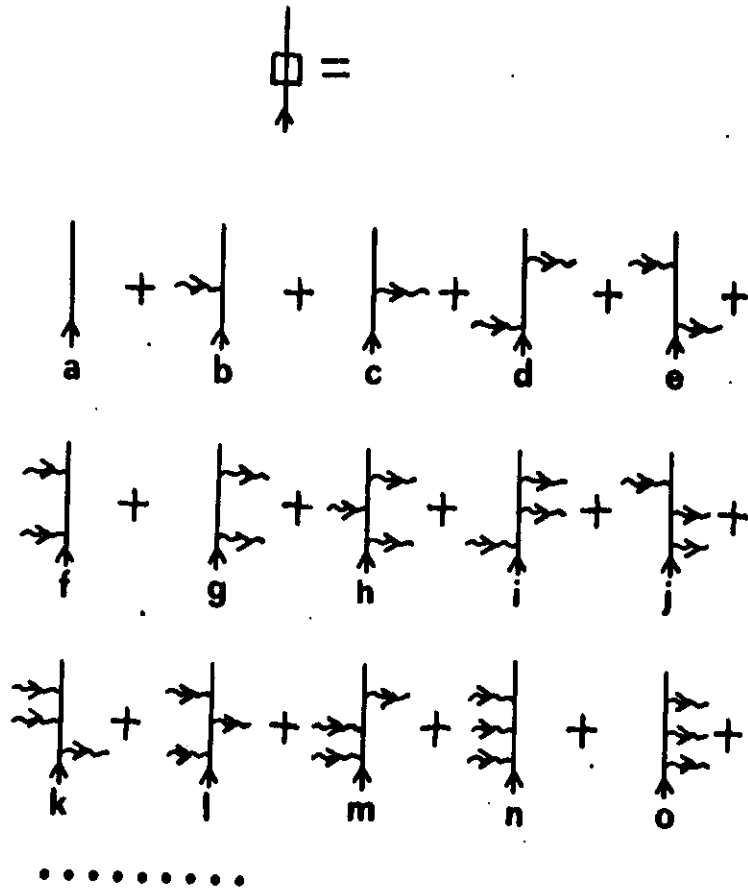


Fig. 4. Content of the Volkov solution