

Electron recombination in dense photonic, electronic and atomic environments

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Abstract. Free electrons can recombine with ions by either radiative, dielectronic or three-body recombination. In this contribution we discuss variants of these fundamental processes which can occur in dense photonic, electronic and atomic environments. First, dielectronic recombination is generalized to the case where two atomic centers participate in the process. In this situation, the incident electron is captured at one center with simultaneous excitation of a neighboring ion, atom or molecule which subsequently decays via photo-emission. Modifications of radiative recombination in the presence of a strong laser field are discussed afterward. Various relativistic effects, arising from a high energy of the incoming electron and its strong coupling to the intense laser field, are found to clearly manifest themselves in the photo-emission spectra. Finally, we consider three-body “recombination” (i.e. annihilation) of an electron and a positron in the presence of a spectator electron. The process leads to emission of just a single photon and can compete with the usual annihilation into two photons at very high electron densities.

1. Introduction

Recombination of free electrons with atomic or molecular ions is a fundamental quantum process of general interest to various fields of science, comprising atomic and molecular physics, plasma physics, and astrophysics [1, 2, 3]. Recombination into single atomic centers is known to proceed in three different ways: (i) The electron can be captured into a bound atomic state upon photo-emission. This process, which represents the time-inverse of photo-ionization, is referred to as radiative recombination. (ii) For certain energies of the incident electron, the resonant process of dielectronic recombination may occur. Here the electron capture leads to the formation of an autoionizing state (time-reversed Auger decay), which afterwards stabilizes radiatively. (iii) At very high particle densities, three-body recombination dominates where the electron capture is rendered possible by transferring the excess energy to another free electron.

In the present paper we discuss variations of these well-studied recombination processes which may occur in dense environments. As we will show, when an electron recombines with an ion which is not isolated in space but in close vicinity to another atomic center, resonant channels exist which rely on interatomic electron-electron correlations. Due to its resonant nature, this

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two-center dielectronic recombination can be remarkably efficient at internuclear distances up to the nanometer range [4, 5]. Characteristic effects also arise when electron-ion recombination takes place in the presence of an intense laser field, forming a dense background of low-frequency photons. While the total recombination cross section will remain essentially unchanged, the presence of the laser field can substantially modify the photo-emission spectra [6]. Besides, interparticle correlation effects may occur in very dense electron-positron plasmas, where they can enhance the relevance of some higher-order quantum electrodynamic reactions which are usually suppressed. As an example, we shall discuss the impact of a nearby spectator electron on the annihilation of an electron-positron pair [7]. A connection of this QED process with three-body recombination of electrons with ions will be drawn.

In order to put our studies into perspective it should be noted that the influence of the environment on microscopic processes has been under very active scrutiny in recent years. In particular, detailed investigations have been performed on electron transitions in systems of two (or more) atoms which are mediated by so-called interatomic Coulombic decay (see, e.g., [8, 9]), where the electronic excitation energy of one atom is transferred to a nearby partner atom, leading to ionization of the latter. This two-center autoionization mechanism can strongly accelerate molecular deexcitation and relaxation processes, as was demonstrated experimentally in helium dimers and water molecules, for instance [10, 11]. Similar interatomic correlations are also responsible for radiationless energy transfer processes in slow atomic collisions [12], cold quantum gases [13], and biomolecules [14]. The influence of background laser fields on scattering reactions and atomic processes has been studied in detail as well. Results have been obtained for laser-assisted scattering of electrons and x-rays on electrons, atoms and nuclei [15]. Also laser-assisted electron-ion recombination was studied in the regime of nonrelativistic parameters (see [16, 17, 18] and references therein). Superintense laser-matter interactions even allow for efficient generation of electron-positron samples [19], which offers future perspectives for laboratory studies on dense, relativistic e^+e^- plasmas. Therefore, the dynamics and evolution of these plasmas is currently being explored, including many-body correlation effects [20, 21, 22].

The paper is organized as follows. In the following Sec. 2 we discuss dielectronic recombination with participation of two atoms. Section 3 is devoted to radiative recombination of a relativistic electron with a highly-charged ion assisted by a strong laser field. A QED process analogous to three-body recombination is described in Sec. 4. The conclusions are presented in Sec. 5.

Atomic units (a.u.) are used throughout unless explicitly stated otherwise.

2. Dielectronic recombination involving two atomic centers

Dielectronic recombination can be generalized to the case where two atomic centers participate in the process [4, 5]. In this situation, the incident electron is captured at one center with simultaneous excitation of a neighboring ion, atom or molecule which subsequently decays via photo-emission. The process relies on resonant electron-electron correlations between the two neighboring atomic centers and may be called two-center dielectronic recombination (2CDR). An illustration is given in Fig. 1. We note that this interatomic channel represents an additional pathway for recombination which interferes with the channel of radiative recombination.

Let R denote the internuclear distance between the two atoms in Fig. 1. We shall assume that R is not too small such that the atoms keep their individuality, but not too large either so that the interaction between the electrons may be treated as instantaneous Coulombic, disregarding retardation effects. That is, $a_0 \ll R \ll c/\omega_{fi}$, where a_0 is the atomic size, ω_{fi} the electron transition frequency, and c the speed of light. Under such conditions, restricting our attention to dipole-allowed transitions, the interaction between the two atoms can be described as

$$V_{AB} = \frac{\mathbf{r}_A \cdot \mathbf{r}_B}{R^3} - \frac{3(\mathbf{r}_A \cdot \mathbf{R})(\mathbf{r}_B \cdot \mathbf{R})}{R^5}, \quad (1)$$

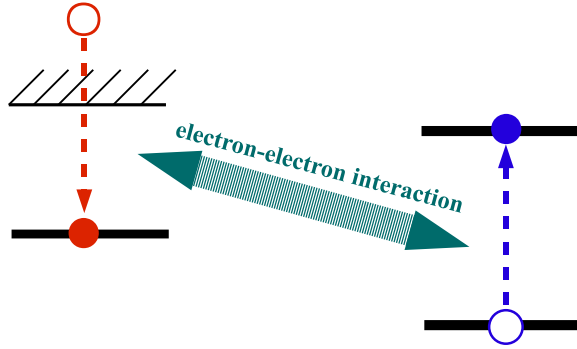


Figure 1. Schematic illustration of two-center dielectronic recombination (2CDR). Shown is the first step, where an electron is captured from the continuum at center A (left) with simultaneous excitation of a neighbouring atom at center B (right). Afterwards, the system stabilizes by radiative deexcitation of atom B.

where $\mathbf{r}_{A,B}$ are the coordinates of each participating electron with respect to its own nucleus.

The dipole-dipole interaction (1) leads to a coupling of the initial two-electron state $\psi_{\mathbf{p}_i}$ – where the incident electron with momentum \mathbf{p}_i is in the continuum, while the atomic electron at center B is in the ground state – to the resonant intermediate state ψ_a – where the incident electron is in a bound state at center A and the atomic electron is in an excited state. This intermediate state is unstable and can either decay backwards via two-center Auger decay (resp. interatomic Coulombic decay) or it can stabilize via radiative deexcitation at center B, this way completing the recombination.

The cross section for the two-center recombination pathway is

$$\sigma_{2\text{CDR}} = \frac{\pi}{p_i^2} \frac{\Gamma_a \Gamma_{\text{rad}}^{(B)}}{(E_{\mathbf{p}_i} - E_a)^2 + \Gamma^2/4}, \quad (2)$$

where $\Gamma_a = 2\pi p_i \int |\langle \psi_a | V_{AB} | \psi_{\mathbf{p}_i} \rangle|^2 d\Omega_{\mathbf{p}_i}$ and $\Gamma_{\text{rad}}^{(B)}$ are the widths due to two-center Auger decay and spontaneous radiative decay of the excited state at center B, respectively, and $\Gamma = \Gamma_a + \Gamma_{\text{rad}}^{(B)}$ is the total width. Besides, $E_{\mathbf{p}_i}$ and E_a denote the energies of the two-electron states $\psi_{\mathbf{p}_i}$ and ψ_a , respectively.

It is instructive to consider the ratio between the cross sections for two-center dielectronic recombination and the usual single-center radiative recombination at center A (without participation of atom B). Exactly on the resonance, where $E_{\mathbf{p}_i} = E_a$, one obtains the very simple relation

$$\frac{\sigma_{2\text{CDR}}}{\sigma_{\text{RR}}^{(A)}} \sim \left(\frac{c}{R\omega_{fi}} \right)^6, \quad (3)$$

provided that $\Gamma_a < \Gamma_{\text{rad}}^{(B)}$ is satisfied which will always hold true at sufficiently large values of R . The $1/R^6$ behaviour reflects the resonant dipole-dipole nature of the two-center process. Since the typical transition frequency ω in atoms and ions is related to their spatial size a by $\omega \sim \alpha c/a$, an alternative representation of Eq. (3) is $\sigma_{2\text{CDR}}/\sigma_{\text{RR}}^{(A)} \sim (a/\alpha R)^6$, where $\alpha \approx 1/137$ denotes the fine-structure constant. Hence, it is the ratio of atomic size to interatomic distance which mainly determines the relative importance of 2CDR.

Since typical atomic transition energies lie in the range of $\omega_{fi} \sim 1\text{--}10\text{ eV}$, relation (3) demonstrates that the resonant two-center channel may dominate over the nonresonant single-center channel at interatomic distances of $R \sim 1\text{--}10\text{ nm}$, while it can still be competitive at $R \sim 10\text{--}100\text{ nm}$.

As an example, let us consider recombination in a system initially consisting of a free electron with energy close to 27.2 eV , a proton at center A and a singly-charged helium ion at center B. In this situation, the electron can recombine with the proton either radiatively or via the

two-center mechanism $e^- + \text{H}^+ + \text{He}^+(1s) \rightarrow \text{H}(1s) + \text{He}^+(2p_m) \rightarrow \text{H}(1s) + \text{He}^+(1s) + \gamma$. Here, m denotes the magnetic quantum number of the intermediate $2p$ state in the helium ion. Both recombination channels can interfere, so that the shape of the resonance shows a characteristic Fano profile, as shown in Fig. 2. While the cross section for radiative recombination with the proton amounts to about 30 barn, the resonant two-center channel can strongly enhance the probability for recombination by two orders of magnitude at an internuclear separation of 3 nm.

We note that in our calculation the positions of the nuclei are assumed to be fixed. To account for nuclear motion, an average over the internuclear distance in Eq. (2) could be carried out. It is important to note that the electron capture, which represents the first step of the 2CDR process, occurs on a very short time scale of about 1 a.u. during which the positions of the nuclei practically do not change. The second step of 2CDR (i.e., the radiative stabilization) does not depend on whether the nuclei move or not. Hence, taking an average in Eq. (2) would essentially mean to average the two-center Auger rate in the numerator, provided that $\Gamma_a < \Gamma_{\text{rad}}^{(B)}$ holds.

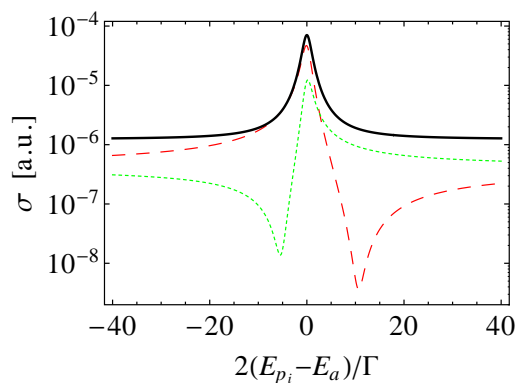


Figure 2. Recombination cross section in the system $e^- + \text{H}^+ + \text{He}^+(1s)$, as a function of the normalized detuning from the 2CDR resonance. The interatomic distance is $R = 3$ nm and the decay width amounts to $\Gamma \approx 7 \mu\text{eV}$. The red dashed and green dotted curves show the separate Fano profiles when the intermediate $2p_0$ and $2p_1$ states, respectively, in the helium ion are involved. The solid black curve refers to the whole process, including a summation over all intermediate states.

Two-center dielectronic recombination can also be of relevance in more complex systems ranging from dense plasmas to (bio)chemical environments. As an example from daily life, let us consider an alkali salt such as NaCl dissolved in water. In this case, a hydrated $\text{Na}^+(\text{H}_2\text{O})_n$ complex forms where on average $n = 6$ water molecules shield the cation at a mean distance of about $R \approx 3 \text{ \AA}$ (and similarly for the Cl^- anion). The ionization potential of neutral Na is 5.14 eV. The first photo-absorption band in water is relatively broad; it lies around 7.5 eV and has a width of a few eV. Hence, electrons with resonant energies in the range of about 1.5–4 eV can recombine with the Na^+ ion through 2CDR involving the assistance of one of the surrounding water molecules. Assuming that Eq. (3) also applies approximately to this more complex system, we may roughly estimate that the electron recombination is largely enhanced by ten orders of magnitude due to 2CDR. The corresponding cross section amounts to $\sigma_{\text{2CDR}} \sim 10^{-10} \text{ cm}^2$ in the resonant region. Note that, even after integration over energies in an incident electron beam, 2CDR may have an enormous effect on the total number of recombination events since the absorption band in water is quasi-continuous and rather broad.

Concluding this section we point out that the electron capture at center A may also be accompanied by the ionization (rather than excitation) of a neighbouring atom B . This process, which has been studied in [23], effectively represents an interatomic electron exchange, letting the total charge at the two centers remain unchanged. Moreover, we note that the time-reversed process of 2CDR is two-center resonant photo-ionization in the limit of low field intensities [5, 24, 25]. Here, an incident photon resonantly excites an atom which subsequently deexcites via interatomic Coulombic decay. This process shows particularly interesting properties for larger photon field strengths due to the Rabi flopping dynamics induced in the assisting atom. Very special effects also arise in the resonant two-photon ionization of a system consisting of

two identical (e.g. hydrogen) atoms [26]. The energy transfer between two spatially separated hydrogen atoms exposed to a nonresonant laser field has been investigated in [27].

3. Radiative electron-ion recombination assisted by a strong laser field

The availability of high-intensity lasers has led to sustained interest in radiative electron-ion recombination in the presence of an external laser field [16, 17, 18]. The latter can strongly modify the field-free properties of the process. The corresponding theoretical description, which so far was always restricted to the nonrelativistic interaction regime (with the laser field being treated in dipole approximation), has recently been extended to the fully relativistic domain [6]. Relativistic effects may arise from a high incident electron energy, a strong coupling to the applied laser field, and a deeply bound final electron state.

Within the relativistic theory, the transition amplitude for laser-assisted radiative recombination may be written as

$$S_{fi} = -i \int_{-\infty}^{+\infty} \langle \Psi_f(t) | \hat{W} | \Psi_i(t) \rangle dt, \quad (4)$$

where $\hat{W} = \boldsymbol{\alpha} \cdot \hat{\mathbf{A}}_\gamma$ denotes the interaction with the quantized radiation field $\hat{\mathbf{A}}_\gamma$ which is responsible for the spontaneous photo-emission during the process. The initial and final states are of product form, $|\Psi_i(t)\rangle = \psi_i(t) |0\rangle$, $|\Psi_f(t)\rangle = \psi_f(t) |\mathbf{k}'\lambda\rangle$, with $\psi_{i,f}(t)$ denoting the respective states of the electron, whereas $|0\rangle$ and $|\mathbf{k}'\lambda\rangle$ represent states of the radiation field containing, respectively, no photons and one non-laser photon of momentum \mathbf{k}' and polarization λ .

The laser field is taken as a classical plane wave of frequency ω , wave vector \mathbf{k} and field strength F_0 . For mathematical simplicity, it is assumed to be circularly polarized. The laser four-potential may be written as $A^\mu = A_0(1, \mathbf{c}\mathbf{k}/\omega)$, with $A_0 = F_0(\mathbf{e}_2 \cdot \mathbf{r} \cos \varphi - \mathbf{e}_1 \cdot \mathbf{r} \sin \varphi)$ and the laser phase $\varphi = \omega t - \mathbf{k} \cdot \mathbf{r}$. This gauge offers the advantage that the hydrogen-like final bound state of the electron, $\psi_f(t)$, can be taken to a good approximation as a pure Coulomb-Dirac wave function which is undistorted by the laser field. The initial state of the electron may be approximated by $\psi_i(t) = \exp[i(F_0/\omega)(\mathbf{e}_1 \cdot \mathbf{r} \cos \varphi + \mathbf{e}_2 \cdot \mathbf{r} \sin \varphi)]\psi_{\mathbf{p}_i}$, where $\psi_{\mathbf{p}_i}$ denotes a Dirac-Volkov state in the usual representation for an electron with asymptotic momentum \mathbf{p}_i . The latter is adapted to the chosen gauge by the additional phase factor. These approximations are justified as long as the nuclear charge satisfies the condition $Z \ll v_i$ (with the incident electron velocity v_i) and the laser field strength is much lower than the nuclear Coulomb field experienced by the bound electron.

Within this framework, the transition amplitude can be evaluated analytically. It adopts the form

$$S_{fi} \approx \sum_{n=-\infty}^{\infty} M_n \delta(n\omega + \varepsilon_0 + \omega' - U_p - E_{\mathbf{p}_i}), \quad (5)$$

where $E_{\mathbf{p}_i}$ is the initial electron energy outside the laser field, U_p is the relativistic ponderomotive energy describing the “dressing” of the electron by the laser field, and ε_0 is the final electron energy in the bound state. The summation index n counts the number of laser photons absorbed (if $n < 0$) or emitted (if $n > 0$) during the recombination step. The structure of the transition amplitude (5) implies that the recombination cross section

$$\sigma = \frac{1}{v_i} \int \frac{d\mathbf{k}'}{(2\pi)^3} |S_{fi}|^2 = \sum_{n=-\infty}^{\infty} \sigma_n \quad (6)$$

also decomposes into a sum of partial cross sections σ_n which give the probability for emission of a photon of energy $\omega' = E_{\mathbf{p}_i} + U_p - \varepsilon_0 - n\omega$. The energy spectrum of emitted photons will

thus consist of a comb of lines, with a spacing of ω between neighbouring lines. The height of each line is determined by the squared magnitude of the matrix elements M_n . One can show that the relevant range of photon numbers is bounded by $\pm n_{\max}$ with

$$n_{\max} \approx \frac{F_0}{\omega^2} \frac{|\mathbf{p}_i^\perp|}{E_{\mathbf{p}_i}/c^2 - (\mathbf{k}/\omega) \cdot \mathbf{p}_i}, \quad (7)$$

where \mathbf{p}_i^\perp is the component of the initial electron momentum perpendicular to \mathbf{k} .

As an example we consider the recombination of a relativistic electron of momentum $p_i = 3mc$ into the ground state of a bare Zn ion (nuclear charge number $Z = 30$). In the absence of any laser field, the spectrum of emitted photons consists of a single line at the free-bound transition energy given by $\omega' = 1.117$ MeV. In contrast, in the presence of an 800-nm laser beam of 10^{16} W/cm² intensity which is irradiated under 90 degrees with respect to the incident electron velocity, the emission spectrum will comprise a multitude of lines forming a quasi-continuous distribution with a total width of about $2n_{\max}\omega \approx 50$ keV and very pronounced side wings (see Fig. 3). The total cross section amounts to ~ 0.1 barn.

The distinct features of the photo-emission spectrum in Fig. 3 find a semiclassical explanation in terms of the instantaneous electron energy inside the field. For the considered beam geometry, the classical electron energy reads

$$E(\varphi) = E_{\mathbf{p}_i} + U_p + \frac{F_0 v_i}{\omega} \cos \varphi. \quad (8)$$

Since the interaction with the laser field renders the electron energy time dependent, the emitted photon energy depends on the moment of recombination. Maximum photon energies will result when the recombination occurs at laser phases around $\varphi = 0$, whereas $\varphi = \pi$ leads to minimum photon energies. The spectral width, accordingly, amounts to $2F_0 v_i/\omega$ which agrees with the prediction from quantum mechanics. Besides, the center of the spectrum is slightly shifted upwards by the ponderomotive energy $U_p \approx 0.2$ keV.

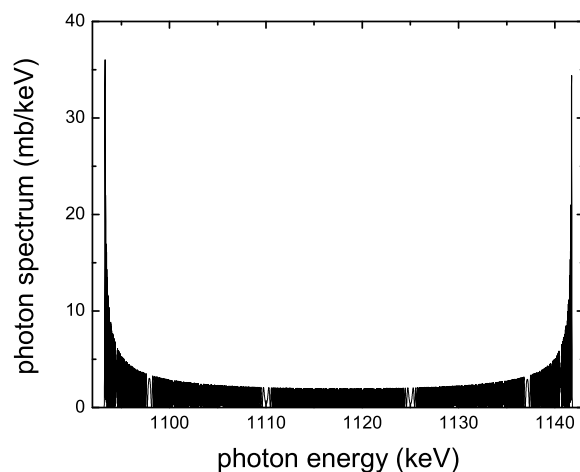


Figure 3. Energy spectrum of the photons which are emitted when a relativistic electron ($p_i = 3mc$) radiatively recombines with a bare Zn ion in the presence of a circularly polarized laser field of frequency $\omega = 1.5$ eV and intensity 10^{16} W/cm². The laser beam is assumed to cross the incident electron beam under an angle of 90° .

Genuinely relativistic signatures arise in the angular distribution of the emitted photons. They are caused by the light pressure exerted by the laser field, leading to an electron momentum component along the laser beam axis [6].

4. Electron-assisted annihilation: three-body recombination with the QED vacuum

Finally, we briefly discuss a process where an electron recombines not with an ion but rather with a hole in the vacuum state of quantum electrodynamics. This is, the electron recombines

with a vacancy in the Dirac sea of negative-energy states or, in other words, it annihilates with a positron. However, contrary to the usual annihilation into two photons, $e^+e^- \rightarrow 2\gamma$, in the process under consideration the annihilation proceeds in the presence of a nearby spectator electron which is capable of absorbing recoil momentum. In this situation the pair can annihilate by emitting just a single photon according to

$$e^+e^- + e \rightarrow \gamma + e'. \quad (9)$$

The triple interaction (9) resembles three-body electron-ion recombination because also there the recombining electron transfers its energy excess to a nearby partner electron. In both processes, the recoil absorbed by the assisting electron reduces the number of emitted photons by one, as compared to radiative recombination and two-photon annihilation, respectively.

Reaction (9) has been studied in some detail with respect to the decay of positronium ions [28, 29]. In this case the assisting electron is loosely bound to the neutral positronium core. For the ratio of single-photon to two-photon annihilation in Ps^- , the small value $R_{1\gamma}/R_{2\gamma} \sim 10^{-10}$ was found. The reason for this small ratio is that the annihilation channel (9) requires very high particle densities to become sizeable. Its relative contribution with respect to $e^+e^- \rightarrow 2\gamma$ may be estimated by order of magnitude as

$$\frac{R_{1\gamma}}{R_{2\gamma}} \sim \alpha \rho \lambda_C^3, \quad (10)$$

where ρ is the number density of electrons and λ_C the Compton wavelength. Besides, a factor of the fine-structure constant α appears, because (9) is of higher order than $e^+e^- \rightarrow 2\gamma$ in the QED coupling parameter. For the case of positronium ions, Eq. (10) becomes $R_{1\gamma}/R_{2\gamma} \sim \alpha^4$.

Equation (10) implies that the single-photon annihilation channel will become prominent at extremely high densities of the order of $\rho \sim \lambda_C^{-3} \approx 10^{31} \text{ cm}^{-3}$ and above. In fact, we have shown [7] that reaction (9) starts to dominate over $e^+e^- \rightarrow 2\gamma$ in terms of total rate at e^+e^- plasma densities of $6.5 \times 10^{32} \text{ cm}^{-3}$, corresponding to a plasma temperature of about 3 MeV. Such enormous densities are generated in astrophysical processes such as the initial stage of gamma-ray bursts [30]. They also existed in the lepton era of the early universe between $\sim 10^{-4} \text{ s}$ and 4 s during which the density dropped from $\sim 10^{38} \text{ cm}^{-3}$ down to $\sim 10^{28} \text{ cm}^{-3}$ [31]. Note for comparison that also three-body recombination of electrons with ions dominates over the usual radiative recombination when the electron density is sufficiently high.

Besides, under certain conditions the photon produced in reaction (9) may exhibit very special properties [7]. The latter might allow for an identification of the single-photon annihilation process in a dedicated laboratory experiment. Suppose that a relativistic electron beam of few-MeV energy penetrates through a dense, cold e^+e^- plasma target. Then the photon from the three-body process (9) will be emitted predominantly into the backward direction with respect to the incident electron beam. Moreover, the photon is polarized to a high degree, with the polarization vector being orthogonal to the plane spanned by the momentum vectors of the photon and incident electron. These peculiar features of the photo-emission can be explained in intuitive terms based on a combination of classical electrodynamics and the familiar properties of two-photon annihilation [7].

Note that the inverse process of (9) is photo-production of an e^+e^- pair on an electron which has been studied for a long time. In recent years the interest in this kind of trident pair creation has been revived in view of its generalization to the multiphoton case in strong laser fields [32, 33].

5. Conclusion

Three different electronic recombination processes have been considered which take place in dense environments consisting of atoms, electrons or photons. First, it was shown that dielectronic

recombination can occur with participation of two spatially well-separated atomic centers. This interatomic recombination process can dominate over the competing single-center radiative recombination channel at atomic densities of the order or above $\sim 10^{18} \text{ cm}^{-3}$. Second, we demonstrated that radiative recombination can be strongly modified by background laser fields of high intensity, corresponding to photon densities of $\sim 10^{24} \text{ cm}^{-3}$. Finally, we discussed the process of electron-assisted annihilation of an electron-positron pair into a single photon, which may be viewed as a three-body recombination with a hole in the vacuum state of QED. It can compete with the usual annihilation into two photons at electronic densities around $\sim 10^{32} \text{ cm}^{-3}$. While the latter process can be of relevance in astrophysical e^+e^- plasmas, the first two may be tested in the laboratory by modern experimental techniques available today.

Acknowledgments

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