Two-center dielectronic recombination in slow atomic collisions

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A free electron can form a bound state with an atomic center A upon photoemission (radiative recombination). In the presence of a neighboring atom B, such a bound state can, under certain conditions, be also formed via resonant transfer of energy to B, with its subsequent relaxation through radiative decay (two-center dielectronic recombination). This two-center process is very efficient in the "static" case where A and B form a weakly bound system, dominating over single-center radiative recombination up to internuclear distances as large as several nanometers. Here we study its dynamic variant in which recombination occurs when a beam of species A collides with a gas of atoms B and show that, even though the average distance between A and B in collisions is orders of magnitude larger than the typical size of a bound system, the two-center recombination can still outperform the single-center radiative recombination.

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I. INTRODUCTION

Processes of recombination of free electrons with atomic or molecular ions are of general interest and relevance to various scientific disciplines [1,2]. In the case of single atomic centers there are three basic recombination processes. First, the electron can be captured into a bound state by emitting a photon (radiative recombination); this process is the time-inverse of photoionization. Second, for certain energies of the incident electron, the recombination can proceed resonantly via formation of an autoionizing state (time-reversed Auger decay) which then stabilizes to a bound state through spontaneous radiative decay. This process is especially important for low-charged ions. Third, an electron can be captured by an ion transferring excess energy to another free electron (three-body recombination); this process becomes very efficient in high-density plasma, especially when the energy transfer is small.

When an atom is not isolated in space but is close to another atom, recombination of a free electron with one of them can, under certain conditions, proceed—due to two-center electron-electron interaction—via resonant energy transfer to the other atom which afterwards stabilizes via spontaneous radiative decay [3,4]. This process, termed two-center dielectronic recombination (2CDR), is rather similar to the "standard" dielectronic recombination on a single center but, in contrast to the former, relies on the interaction between electrons of different centers. The 2CDR can also be viewed as a kind of three-body recombination in which an assisting free electron is replaced by an electron bound in a heavy atomic particle whose internal structure plays in this process a crucial role.

It is worth mentioning that the coupling of electronic structures at two spatially well-separated atomic centers by long-range electromagnetic interactions can lead to a variety of interesting phenomena. For example, interatomic electron-electron correlations are responsible for the population inversion in a He-Ne laser and the energy transfer in quantum optical ensembles [5] or cold Rydberg gases [6]. They also play an important role in biological systems such as Förster resonances between chromophores [7]. Another interesting realization of two-center electron-electron coupling is represented by a process in which the electronic excitation energy of one of the atoms cannot be quickly released through a forbidden (single-center) Auger decay and is instead transferred to the partner atom, resulting in its ionization (interatomic Coulombic decay). Stimulated by detailed theoretical predictions [8], this process has been observed in recent years in various systems such as van der Waals clusters [9], rare gas dimers [10], and water molecules [11]. Correlated electronic decay processes have also been observed in expanding nanoplasmas which were formed by irradiating clusters with intense laser pulses (see, e.g., Ref. [12]). In the process of the so-called interatomic Coulombic electron capture (ICEC), an electron is captured by one atomic center transferring the excess energy to a neigboring atom that results in its ionization [13].

Interatomic electron-electron correlations also drive the process of resonant two-center photoionization (2CPI) [14] in which ionization of a van der Waals dimer occurs via resonant photoabsorption by one of its atoms with subsequent transfer of excitation energy via two-center electron correlations to another atom, leading to its ionization. This two-center ionization channel can be remarkably effective, strongly dominating over the usual single-center photoionization. This was experimentally observed in Refs. [15,16].

It is known [3,4,14] that interatomic electron-electron correlations can greatly enhance recombination and ionization processes in a "static" situation in which two atomic centers constitute a (weakly) bound system. The strength of the two-center correlations rapidly decreases with increasing the size of the system. Nevertheless, it has recently been shown [17] that 2CPI can strongly dominate single-center photoionization also in collisions, even though the average interatomic distance in collisions exceeds by orders of magnitude the typical size of the corresponding bound system.

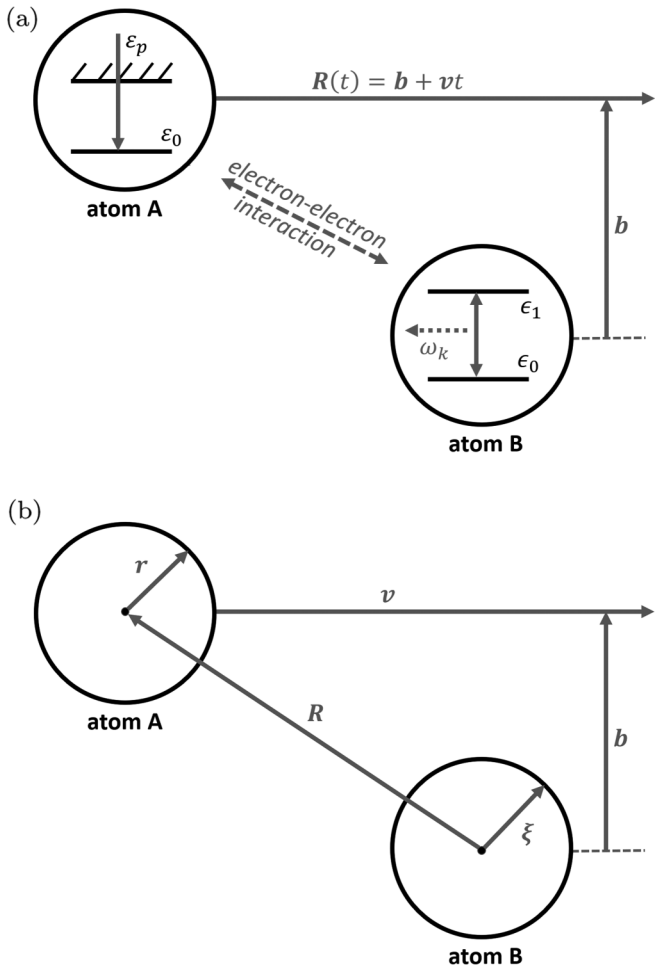

FIG. 1. Two-center dielectronic recombination in atomic collisions. (a) Scheme of the process. (b) Schematic representation of space coordinates characterizing the collision.

In this paper, a dynamic variant of two-center dielectronic recombination, which occurs in atomic collisions (see Fig. 1), is studied. We show that although, compared to collisional 2CPI, collisional 2CDR turns out to be much less efficient, it still can outperform single-center radiative recombination (RR).

The paper is organized as follows. Section II is devoted to considering two-center dielectronic recombination in slow collisions of two atomic centers and to a derivation of formulas for the rates of this process. Besides, in this section we also very briefly discuss (single-center) radiative recombination and the collisional version of the ICEC. Section III contains numerical results and discussion, and the main conclusions are summarized in Sec. IV.

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

II. THEORETICAL CONSIDERATION

A. Two-center dielectronic recombination

Let us consider recombination of an electron with atomic center A occurring in the presence of a neighbor center, B. In the static case of 2CDR [3] A and B are supposed to be at rest with a fixed distance R_0 between them. If the energy release in the process of $e^- + A$ recombination is close to an excitation energy of a dipole-allowed transition in atom B, then the recombination can proceed by transferring—via the (long-range) two-center electron-electron interaction—the energy excess to atom B which, as a result, undergoes a transition into an excited state. Afterwards, atom B can either radiatively decay to its initial (ground) state, which means that 2CDR has occurred, or the system can undergo two-center autoionization with re-emission of the captured electron (the channel of resonance electron scattering, see Ref. [4]). Due to its resonant nature, 2CDR may enhance the rate for recombination by orders of magnitude compared to the case when center B is absent [3].

The state of the total system consisting of the ground state of $(e^- + A)$ and an excited state of B is unstable. Its width is determined by the sum of the radiative width Γ_r^B of the excited state of B and the width Γ_{2c-A} due to two-center autoionization caused by the electron-electron interaction. When the distance between A and B is not very large ($\lesssim 10$ a.u.) the width Γ_{2c-A} begins to exceed the radiative width Γ_r^B [14] and has to be taken into account when considering the static 2CDR.

Suppose now that free electrons and a beam of atomic centers A (represented by, e.g., ions or atoms) move in a (relatively dense) gas of atoms B. As was just mentioned, two-center recombination relies on the energy transfer resonant to a transition in B. However, the relative motion of A and B leads to uncertainty in electron transition energies (as they are viewed by the collision partner), effectively broadening them. Therefore, the efficiency of this recombination channel is expected to be restricted to low-velocity collisions and only such collisions, where the velocity v of A with respect to B is much less than 1 a.u. (1 a.u. $= 2.18 \times 10^8$ cm/s), are considered here.

Even though the collision velocity is low, we shall assume that one can still use the semiclassical approximation in which the relative motion of the nuclei of A and B is treated classically. According to the applicability conditions of the semiclassical approximation (which are discussed, e.g., in Ref. [18]), this can be safely done down to impact energies as low as $\gtrsim 1$ eV/u ($v \gtrsim 0.01$ a.u.). Moreover, only (very) distant collisions are considered in which the colliding subsystems $(e^- + A)$ and B do not overlap, the interaction between them is rather weak, and the relative motion of their nuclei can be approximated by straight lines.

Compared to the static case, in collisional 2CDR the role of two-center autoionization is greatly diminished by the relative motion of A and B. In particular, as our estimates show, even at $v \simeq 0.01$ a.u. the time T which A and B spend at those distances, where the autoionization width Γ_{2c-A} becomes close or even exceeds the radiative width Γ_r^B, is so short that $\Gamma_{2c-A}\, T \ll 1$ and, thus, the two-center autoionization simply does not have enough time to unveil itself in the collision.

Let us choose a reference frame in which B is at rest and take the position of its nucleus as the origin. In this frame A moves along a classical straight-line trajectory, $\boldsymbol{R}(t) = \boldsymbol{b} + \boldsymbol{v}t$, where $\boldsymbol{b} = (b_x, b_y, 0)$ is the impact parameter and $\boldsymbol{v} = (0, 0, v)$ is the collision velocity (see Fig. 1 for illustration).

The recombination process is described by the Schrödinger equation

$$i\frac{\partial\Psi(t)}{\partial t} = \hat{H}\Psi(t). \tag{1}$$

Here the total Hamiltonian $\hat{H}$ reads

$$\hat{H}(t) = \hat{H}_A + \hat{H}_B + \hat{H}_\gamma + \hat{V}_{AB} + \hat{V}_\gamma, \tag{2}$$

where $\hat{H}_A$ is the Hamiltonian of the subsystem consisting of A and an electron (initially incident and finally bound with A), $\hat{H}_B$ is the Hamiltonian of the free (noninteracting) atom B, and

$$\hat{V}_{AB} = \frac{\boldsymbol{r}\cdot\boldsymbol{\xi}}{R^3(t)} - \frac{3[\boldsymbol{r}\cdot\boldsymbol{R}(t)][\boldsymbol{\xi}\cdot\boldsymbol{R}(t)]}{R^5(t)} \tag{3}$$

is the dipole-dipole interaction between $(A + e^-)$ and B. In Eq. (3) the coordinate $\boldsymbol{r}$ refers to the electron recombining with A and is given with respect to the nucleus of A, whereas the coordinate $\boldsymbol{\xi}$ refers to the electron bound in B and is given with respect to the nucleus of B [see Fig. 1(b) for illustration]. We note that the "electrostatic" approximation (3) for the interatomic interaction can be used if the distance R is not too large: $R \ll c/\omega_{\rm tr}$, where c is the speed of light and $\omega_{\rm tr}$ is the frequency of the virtual photon transmitting the interaction (see, e.g., Refs. [3,14,19]).

Further, in Eq. (2) $\hat{H}_\gamma$ is the Hamiltonian of the (quantized) radiation field and

$$\hat{V}_\gamma = \frac{1}{c}\hat{\boldsymbol{A}}(\boldsymbol{\xi},t)\cdot\hat{\boldsymbol{p}}_{\boldsymbol{\xi}} + \frac{1}{2c^2}\hat{\boldsymbol{A}}^2(\boldsymbol{\xi},t) \tag{4}$$

is the interaction of B with this field. Here, $\hat{\boldsymbol{p}}_{\boldsymbol{\xi}}$ is the momentum operator for the electron bound in atom B and

$$\hat{\boldsymbol{A}}(\boldsymbol{\xi},t) = \sqrt{\frac{2\pi c^2}{V_{\rm ph}\omega_k}}\boldsymbol{e}_{\boldsymbol{k}\lambda}[\hat{a}_{\boldsymbol{k}\lambda}e^{i(\boldsymbol{k}\cdot\boldsymbol{\xi}-\omega_k t)} + \text{H.c.}] \tag{5}$$

is the vector potential of the radiation field, where $\boldsymbol{k}$ is the wave vector, $\boldsymbol{e}_{\boldsymbol{k}\lambda}$ ($\lambda = 1$ and 2) are the unit polarization vectors ($\boldsymbol{e}_{\boldsymbol{k}1}\cdot\boldsymbol{e}_{\boldsymbol{k}2} = 0$, $\boldsymbol{e}_{\boldsymbol{k}\lambda}\cdot\boldsymbol{k} = 0$), $\omega_k = ck$ is the frequency, and $V_{\rm ph}$ is the normalization volume for the field. In what follows the interaction $\hat{V}_\gamma$ is treated in the dipole approximation, i.e., $\boldsymbol{k}\cdot\boldsymbol{\xi} \approx 0$.

The initial ($\Psi_{\boldsymbol{p}0}$), intermediate (Ψ_{01}), and final (Ψ_{00}) states of the total system—$(A + e^-) + B$ + radiation field—are given by

$$\begin{aligned}
\Psi_{\boldsymbol{p}0}(\boldsymbol{\xi},\boldsymbol{\rho},t) &= \phi_{\boldsymbol{p}}[\boldsymbol{\rho}-\boldsymbol{R}(t)]e^{-i\varepsilon_p t}\alpha(\boldsymbol{\rho},t)\chi_0(\boldsymbol{\xi})e^{-i\epsilon_0 t}\\
&\quad\times|0_{\boldsymbol{k}\lambda}\rangle,\\
\Psi_{01}(\boldsymbol{\xi},\boldsymbol{\rho},t) &= \phi_0[\boldsymbol{\rho}-\boldsymbol{R}(t)]e^{-i\varepsilon_0 t}\alpha(\boldsymbol{\rho},t)\chi_1(\boldsymbol{\xi})e^{-i\epsilon_1 t}\\
&\quad\times|0_{\boldsymbol{k}\lambda}\rangle,\\
\Psi_{00}(\boldsymbol{\xi},\boldsymbol{\rho},t) &= \phi_0[\boldsymbol{\rho}-\boldsymbol{R}(t)]e^{-i\varepsilon_0 t}\alpha(\boldsymbol{\rho},t)\chi_0(\boldsymbol{\xi})e^{-i\epsilon_0 t}\\
&\quad\times|1_{\boldsymbol{k}\lambda}\rangle.
\end{aligned} \tag{6}$$

Here $\phi_{\boldsymbol{p}}$ is the state of the electron incident on A with an asymptotic momentum $\boldsymbol{p}$ (as is seen in the rest frame of A), ϕ_0 (χ_0) is the ground state of the subsystem $(e^- + A)$ (atom B) with an energy ε_0 (ϵ_0), χ_1 is the excited state of B with an energy ϵ_1, $\boldsymbol{\rho} = \boldsymbol{r} + \boldsymbol{R}(t)$, and $\alpha(\boldsymbol{\rho},t) = e^{i\boldsymbol{v}\cdot[\boldsymbol{\rho}-\boldsymbol{R}(t)]}e^{-i\frac{v^2}{2}}$ is the translational factor. Finally, $|0_{\boldsymbol{k}\lambda}\rangle$ ($|1_{\boldsymbol{k}\lambda}\rangle$) represents the state of the radiation field before (after) the spontaneous radiative decay in B.

Using the second order of time-dependent perturbation theory and taking into account the selection rules for dipole-allowed transitions in atom B ($\Delta l = 1$ and $\Delta m = 0$ and ±1) for the orbital (l) and magnetic (m) quantum numbers we obtain that the transition amplitude for collisional two-center dielectronic recombination is given by

$$\mathcal{S}_{2C}^{\rm DR} = \sum_{\Delta m=-1}^{1}\mathcal{S}_{2C}^{{\rm DR},\Delta m}. \tag{7}$$

Here,

$$\mathcal{S}_{2C}^{{\rm DR},\Delta m} = \frac{1}{i^2}\int_{-\infty}^{\infty}dt\,\mathcal{M}_2^{\Delta m}(t)\int_{-\infty}^{t}dt'\,\mathcal{M}_1^{\Delta m}(t'), \tag{8}$$

where $\mathcal{M}_1^{\Delta m}(t') = \langle\Psi_{01}|\hat{V}_{AB}|\Psi_{\boldsymbol{p}0}\rangle$ and $\mathcal{M}_2^{\Delta m}(t) = \langle\Psi_{00}|\hat{V}_\gamma|\Psi_{01}\rangle$. Integrating in Eq. (8) by parts results in

$$\mathcal{S}_{2C}^{{\rm DR},\Delta m} = \sqrt{\frac{2\pi}{V_{\rm ph}\omega_k}}\frac{\mathcal{W}_{01}^{B,\Delta m}}{\frac{\Gamma_r^B}{2}+i\delta}\int_{-\infty}^{\infty}dt\,\mathcal{M}_1^{\Delta m}(t)e^{-i\delta t}, \tag{9}$$

where $\mathcal{W}_{01}^{B,\Delta m} = \langle\chi_0(\boldsymbol{\xi})|\,\boldsymbol{e}_{\boldsymbol{k}\lambda}\hat{\boldsymbol{p}}_{\boldsymbol{\xi}}\,|\chi_1(\boldsymbol{\xi})\rangle$, Γ_r^B is the width of the excited state χ_1 due to its spontaneous radiative decay, and $\delta = \epsilon_1 - \epsilon_0 - \omega_k$.

Performing the time integration in Eq. (9), we arrive at

$$\begin{aligned}
\mathcal{S}_{2C}^{{\rm DR},\Delta m} = \sqrt{\frac{2^3\pi}{V_{\rm ph}\omega_k}}\frac{|\Delta|}{bv^3}\frac{\mathcal{W}_{01}^{B,\Delta m}}{\frac{\Gamma_r^B}{2}+i\delta}\Big\{&vK_1(\eta)\mathcal{W}_{01\boldsymbol{p}0}^{\Delta m}(\boldsymbol{\xi}_\perp\cdot\boldsymbol{r}_\perp)\\
&-\frac{|\Delta|}{b}K_2(\eta)\mathcal{W}_{01\boldsymbol{p}0}^{\Delta m}((\boldsymbol{\xi}_\perp\cdot\boldsymbol{b})(\boldsymbol{r}_\perp\cdot\boldsymbol{b}))\\
&+b|\Delta|K_0(\eta)\mathcal{W}_{01\boldsymbol{p}0}^{\Delta m}(\xi_z r_z)+i\Delta K_1(\eta)\\
&\times\mathcal{W}_{01\boldsymbol{p}0}^{\Delta m}[(\boldsymbol{\xi}_\perp\cdot\boldsymbol{b})r_z+(\boldsymbol{r}_\perp\cdot\boldsymbol{b})\xi_z]\Big\},
\end{aligned} \tag{10}$$

where $\eta = |\Delta|\frac{b}{v}$, $\Delta = \varepsilon_p - \varepsilon_0 - \omega_k$, $\mathcal{W}_{01\boldsymbol{p}0}^{\Delta m}(x) = \langle\phi_0(\boldsymbol{r})\chi_1(\boldsymbol{\xi})|\,x\,|\phi_{\boldsymbol{p}}(\boldsymbol{r})\chi_0(\boldsymbol{\xi})\rangle$ and K_n ($n = 0$, 1, and 2) are the modified Bessel functions of the second kind [20]. $\boldsymbol{r}_\perp$ ($\boldsymbol{\xi}_\perp$) is the transverse part of the coordinate $\boldsymbol{r}$ ($\boldsymbol{\xi}$), which is perpendicular to the collision velocity $\boldsymbol{v}$.

The spectra of emitted photons can be calculated from the following quantity

$$\frac{d^3\sigma_{2C}^{\rm DR}}{d\boldsymbol{k}^3} = \frac{V_{\rm ph}}{(2\pi)^3}\sum_\lambda\int_{b_{\rm min}}^{\infty}dbb\int_0^{2\pi}d\varphi_{\boldsymbol{b}}\left|\mathcal{S}_{2C}^{\rm DR}\right|^2, \tag{11}$$

where the integrations run over the absolute value b and the azimuthal angle $\varphi_{\boldsymbol{b}}$ of the impact parameter $\boldsymbol{b}$ and we assume that $b_{\rm min} \gg 1$ a.u. The total number of the two-center recombination events is proportional to

$$\sigma_{2C}^{\rm DR} = \int d^3k\frac{d^3\sigma_{2C}^{\rm DR}}{d\boldsymbol{k}^3}. \tag{12}$$

One should note that since two-center recombination is a three-body collision process (incident electron + A + B) the quantities (11) and (12) strictly speaking are not cross sections (how they are normally defined).

Although analytical expressions for Eqs. (11) and (12) can be obtained for bound states ϕ_0 and χ_0 with arbitrary principal

and orbital quantum numbers (see Ref. [21]), they in general turn out to be quite cumbersome. Therefore, in this paper we present results only when ϕ_0 and χ_0 are s states.

The frequency (energy) spectrum of emitted photons is proportional to the "cross section":

$$\frac{d\sigma_{2C}^{DR}}{d\omega_k} = \frac{1}{6\pi}\frac{\omega_B\omega_k r_A^2 r_B^2}{v^2 b_{\min}^2 p^2}\frac{\Gamma_r^B}{\frac{(\Gamma_r^B)^2}{4}+\delta^2}\eta_m^2\big\{\sin^2\vartheta_{\boldsymbol{p}}K_1^2(\eta_m) + (1+\cos^2\vartheta_{\boldsymbol{p}})\eta_m K_0(\eta_m)K_1(\eta_m)\big\}. \quad (13)$$

Here, p and $\vartheta_{\boldsymbol{p}}$ are the absolute value and the polar angle, respectively, of the momentum $\boldsymbol{p}$ of the incident electron, and $\eta_m = |\varepsilon_p - \varepsilon_0 - \omega_k|b_{\min}/v$. Further, $r_A = \int_0^\infty dr r^3 g_0(r) g_{p1}(r)$ is the radial matrix element for the transition of the incident electron into the ground state ϕ_0, where g_{p1} and g_0 are the radial parts of the continuum and bound states, respectively. Similarly, $r_B = \int_0^\infty d\xi\, \xi^3 h_1^*(\xi) h_0(\xi)$ denotes the radial matrix element for transitions between the ground state and the excited state of atom B with h_0 and h_1 being their radial parts.

The total cross section is obtained by integrating Eq. (13) over the photon frequency ω_k. In order to perform this integration we remark that the right-hand side of Eq. (13) contains the factor $\Gamma_r^B/(\delta^2 + (\Gamma_r^B)^2/4)$ which varies with ω_k much more rapidly than the rest: it has a maximum at $\omega_k = \omega_B = \epsilon_1 - \epsilon_0$, very quickly decreases when the detuning $|\omega_k - \omega_B|$ increases, and is already strongly suppressed when the detuning exceeds just several Γ_r^B's, whereas the other ω_k-dependent factors in Eq. (13) vary on much broader scales. By exploiting this feature we obtain

$$\sigma_{2C}^{DR} = \frac{4\pi^2}{3}\frac{r_B^2}{v^2 b_{\min}^2}\frac{r_A^2}{p^2}\tilde{\eta}^2\big\{\sin^2\vartheta_{\boldsymbol{p}}K_1^2(\tilde{\eta}) + (1+\cos^2\vartheta_{\boldsymbol{p}})\tilde{\eta}K_0(\tilde{\eta})K_1(\tilde{\eta})\big\}, \quad (14)$$

where $\tilde{\eta} = |\varepsilon_p - \varepsilon_0 - \omega_B|b_{\min}/v$ and r_B^2 can be expressed via the radiative width Γ_r^B of atom B according to

$$r_B^2 = \frac{9c^3}{4\omega_B^3}\Gamma_r^B. \quad (15)$$

Taking into account that species A move in a gas of atoms B, the total decay rate per unit of time for 2CDR per one $(e^- + A)$ pair reads

$$\mathcal{R}_{2C}^{DR} = \sigma_{2C}^{DR} n_B v = 3\pi^2\frac{n_B}{v b_{\min}^2}\frac{c^3\Gamma_r^B}{\omega_B^3}\frac{r_A^2}{p^2}\tilde{\eta}^2\big\{\sin^2\vartheta_{\boldsymbol{p}}K_1^2(\tilde{\eta}) + (1+\cos^2\vartheta_{\boldsymbol{p}})\tilde{\eta}K_0(\tilde{\eta})K_1(\tilde{\eta})\big\}, \quad (16)$$

where n_B is the density of atoms B. The functions $K_n(x)$ ($n = 0, 1, \ldots$) diverge at $x \to 0$ and decrease exponentially at $x > 1$ [20]. Therefore, in distant low-velocity collisions ($b_{\min} \gg 1$, $v \ll 1$) the most favorable conditions for 2CDR, according to Eq. (16), are realized when the energy of the incident electrons is within the small interval centered at $\varepsilon_{p,r} = \varepsilon_0 + \omega_B$ with the width $\sim\delta\varepsilon_p \sim v/b_{\min}$. Since the quantity $v/b_{\min}$ is typically orders of magnitude larger than the natural width Γ_r^B, we see that the collision strongly smears out the static resonance conditions $\varepsilon_0 + \omega_B - \Gamma_r^B \lesssim \varepsilon_p \lesssim \varepsilon_0 + \omega_B + \Gamma_r^B$, leading to a much broader range of "quasiresonance" energies of the incident electron.

If the incident electrons do not have a fixed momentum $\boldsymbol{p}$, the rate (16) should be averaged over their momentum distribution function $f(\boldsymbol{p})$. This, in general, can be done only numerically.

However, a simple formula for the averaged rate, which enables one to establish a direct correspondence with the case of 2CDR at a fixed distance between A and B, can be derived if we suppose the following: (i) the function $f(\boldsymbol{p})$ can be factorized as $f(\boldsymbol{p}) = f_\varepsilon(\varepsilon_p) f_\Omega(\Omega_{\boldsymbol{p}})$; and (ii) the function $f_\varepsilon(\varepsilon_p)$ is distributed over an energy range which covers the interval of the quasiresonance energies, $\varepsilon_0 + \omega_B - v/b_{\min} \lesssim \varepsilon_p \lesssim \varepsilon_0 + \omega_B + v/b_{\min}$, and is much broader than this interval, with $f_\varepsilon(\varepsilon_p)$ noticeably varying on a scale much larger than $\delta\varepsilon_p \sim v/b_{\min}$ (i.e., within the energy interval essential for 2CDR, $f_\varepsilon(\varepsilon_p)$ is roughly a constant). Then, taking into account that the "width" of the continuum (i.e., the energy range on which the quantity r_A^2/p^2 noticeably varies: typically $\sim$10 eV for atoms and $\sim$1 eV for negative ions) is much larger than $\delta\varepsilon_p \sim v/b_{\min}$, we obtain that the averaged rate is approximately given by

$$\langle\mathcal{R}_{2C}^{DR}\rangle = \frac{9\pi^4}{16}\frac{n_B}{b_{\min}^3}\frac{\Gamma_r^B c^3}{\omega_B^3}\left(\frac{r_A^2}{p^2}\right)_{p=p_r} f_\varepsilon(\varepsilon_{p,r}) \times \int d\Omega_{\boldsymbol{p}} f_\Omega(\Omega_{\boldsymbol{p}})\left(1+\frac{1}{2}\sin^2\vartheta_{\boldsymbol{p}}\right), \quad (17)$$

where $p_r = \sqrt{2\varepsilon_{p,r}} = \sqrt{2(\varepsilon_0 + \omega_B)}$. Assuming for simplicity that all electrons are incident under the angle $\vartheta_{\boldsymbol{p}} = \pi/2$ and are homogeneously distributed over the energy interval ΔE, we get

$$\langle\mathcal{R}_{2C}^{DR}\rangle = \frac{3^3\pi^4}{2^5}\frac{n_B}{b_{\min}^3}\frac{c^3}{\omega_B^3}\frac{\Gamma_r^B}{\Delta E}\left(\frac{r_A^2}{p^2}\right)_{p=p_r}. \quad (18)$$

B. Single-center radiative recombination

Single-center radiative recombination is a very well-known process, which has been studied for decades with energies of the incident electrons ranging from below 1 eV to relativistic values (see, e.g., Refs. [1,2,22] and references therein).

The (total) rate per unit time for radiative recombination of the $(e^- + A)$ pair reads

$$\mathcal{R}_{1C}^{RR} = \frac{4\pi}{3}\frac{\omega_A^3}{c^3}\frac{r_A^2}{p^2}, \quad (19)$$

where p, as before, is the momentum of the incident electron, $\omega_A = \varepsilon_p - \varepsilon_0$ is the transition energy, and r_A is the radial matrix element (which was already defined in the previous subsection).

If the energy of the incident electrons is not fixed one should average the rate (19) over their energy distribution. Assuming that the width of this distribution is much smaller than the energy range on which the quantity r_A^2/p^2 noticeably varies, we obtain that the averaged rate for RR, $\langle\mathcal{R}_{1C}^{RR}\rangle$, simply coincides with $\mathcal{R}_{1C}^{RR}$ given by formula (19):

$$\langle\mathcal{R}_{1C}^{RR}\rangle = \frac{4\pi}{3}\frac{\omega_A^3}{c^3}\frac{r_A^2}{p^2}. \quad (20)$$

C. 2CDR-to-RR ratios

The relative effectiveness of collisional 2CDR and single-center RR can be characterized by the ratios

$$\mu_{2C,1C} = \frac{\mathcal{R}^{DR}_{2C}}{\mathcal{R}^{RR}_{1C}} = \frac{9\pi}{4}\frac{n_B}{v b^2_{min}}\frac{c^6\Gamma^B_r}{\omega^3_A\omega^3_B}\tilde{\eta}^2\left\{\sin^2\vartheta_{\boldsymbol{p}}K^2_1(\tilde{\eta}) + (1+\cos^2\vartheta_{\boldsymbol{p}})\tilde{\eta}K_0(\tilde{\eta})K_1(\tilde{\eta})\right\} \quad (21)$$

and

$$\overline{\mu}_{2C,1C} = \frac{\langle\mathcal{R}^{DR}_{2C}\rangle}{\langle\mathcal{R}^{RR}_{1C}\rangle} = \frac{3^3\pi^3}{2^6}\frac{n_B}{b^3_{min}}\frac{c^6\Gamma^B_r}{\omega^3_A\omega^3_B}f_\varepsilon(\varepsilon_{p,r}) \times \int d\Omega_{\boldsymbol{p}} f_\Omega(\Omega_{\boldsymbol{p}})\left(1+\frac{1}{2}\sin^2\vartheta_{\boldsymbol{p}}\right) = \frac{3^4\pi^3}{2^7}\frac{n_B}{b^3_{min}}\frac{c^6}{\omega^3_A\omega^3_B}\frac{\Gamma^B_r}{\Delta E}, \quad (22)$$

where in obtaining the last line of Eq. (22) it was assumed that the electrons are incident under the angle $\vartheta_{\boldsymbol{p}} = \pi/2$ and we set $f_\varepsilon(\varepsilon_{p,r}) = 1/\Delta E$.

D. Collisional 2CDR versus "static" 2CDR

In the case of 2CDR occurring at a fixed distance R_0 between the centers A and B, the ratio of this process to the single-center radiative recombination is given by [3]

$$\overline{\mu}^{static}_{2C,1C} \simeq \frac{c^6}{R^6_0\omega^3_A\omega^3_B}\frac{\Gamma^B_r}{\Delta E}. \quad (23)$$

Comparing Eq. (23) with (the last line of) Eq. (22) we see that in collisions the role of the fixed interatomic distance R_0 is overtaken by $R_{eff} = (b_{min}\overline{R})^{1/2}$, where $\overline{R} \approx n_B^{-1/3}$ is the average distance between the atoms. Thus, the quantity R_{eff} plays the role of an effective interatomic distance in the collisions.

Since for not very dense gases one has $b_{min} \ll \overline{R}$, we obtain that $R_{eff} \ll \overline{R}$. Due to a steep dependence of the two-center channel on the interatomic distance, the colliding atoms interact mainly in the vicinity of their closest rapprochement ($R \sim b$), which is much less than the averaged distance $\overline{R}$ between them. This explains why the effective distance R_{eff} is strongly reduced as compared to the average distance $\overline{R}$. Because of the same reason the "electrostatic" form (3) of the two-center electron-electron interaction may be used provided $b_{min} \ll c/\omega$.

E. Interatomic Coulombic electron capture

If we consider a three-body collision—incident electron + A + B—in the same way as for 2CDR, but now the energy of the incident electron is sufficient to ionize atom B, this process is called interatomic Coulombic electron capture (ICEC). It was already studied for the static case, in which A and B constitute a bound system [13,23]. A detailed consideration of this process in slow atomic collisions is given in Ref. [24] and here we only quote our results for the total rate per unit time $\mathcal{R}^{EC}_{2C}$ for ICEC and the ratio $\mu^{EC}_{1C,2C}$ between the total rates for ICEC and single-center RR, which are given by

$$\mathcal{R}^{EC}_{2C} = \frac{1}{2^{11}\pi}\frac{n_B}{b^3_{min}}[5+\cos^2(\vartheta_{\boldsymbol{p}})]\frac{r^2_A}{p^2}\left(\frac{r^2_B}{p_B}\right)_{p_B=p_s} \quad (24)$$

and

$$\mu^{EC}_{1C,2C} = \frac{\mathcal{R}^{EC}_{2C}}{\mathcal{R}^{RR}_{1C}} = \frac{9\pi}{2^{11}}\left(\frac{c}{\omega_A}\right)^4\frac{n_B}{b^3_{min}}[5+\cos^2(\vartheta_{\boldsymbol{p}})]\sigma^B_{PI}(\omega_A). \quad (25)$$

Here, r_B is the radial matrix element for the bound-continuum transition in atom B, p_B is the momentum of an electron emitted from B, $p_s = \sqrt{2(\epsilon_0+\omega_A)}$, and σ^B_{PI} is the photoionization cross section of atom B.

III. RESULTS AND DISCUSSION

In order to illustrate our theoretical findings here we discuss the relationship between 2CDR and single-center RR for a few collision systems.

As it follows from the consideration given in the previous section, the results for 2CDR depend on the parameter b_{min}, which is assumed to be sufficiently large but whose (exact) magnitude cannot be strictly defined within our approach. Estimates show that collisions with small impact parameters from $b = 0$ up to several atomic units yield a very substantial contribution to 2CDR [25]. Therefore, in order to account for as much of the total rate as possible, we have set $b_{min} = 5$ a.u., which is close to the minimum possible value of the impact parameter which still enables one, as estimates show, to fulfill the main assumptions of our approach: the nuclei of the colliding particles move along straight-line trajectories, the electrons of the colliding $(e^- + A)$ and B subsystems essentially do not overlap, and the interaction between these subsystems (the two-center electron-electron interaction) can be treated within the first order of perturbation theory.

Let us first consider the collision system $(e^- + K^+)$ ($|\varepsilon_0| \approx 4.34$ eV)–Be($2s^2$) (atom B) in which electron capture by center A is accompanied by excitation of the $2s_{1/2} \to 2p_{3/2}$ dipole transition in Be ($\omega_B = 5.28$ eV, $\Gamma^B_r = 1.66\times10^{-8}$ eV). Using Eq. (21) and choosing $v = 0.01$ a.u. (corresponding to 2.5 eV/u), $\varepsilon_p = 0.938$ eV, and $\vartheta_{\boldsymbol{p}} = \pi/2$, we obtain that $\mu_{2C,1C} \geqslant 1$ if $n_B \gtrsim 1.17\times10^{14}$ cm^{-3}. An atomic density of $n_B = 10^{15}$ cm^{-3} (which is more than 4 orders of magnitude smaller than the density of air $n_{air} \approx 3\times10^{19}$ cm^{-3} under normal conditions) would yield a ratio of $\mu_{2C,1C} \approx 8.60$.

As a second example let us take the collision system $(e^- + Cs^+)$ ($|\varepsilon_0| \approx 3.89$ eV)–Mg($3s^2$) (atom B) assuming that the dipole transition $3s \to 3p$ in Mg ($\omega_B = 4.35$ eV, $\Gamma^B_r = 3.35\times10^{-7}$ eV) is involved. Applying Eq. (21) with $v = 0.01$ a.u., $\varepsilon_p = 0.458$ eV, and $\vartheta_{\boldsymbol{p}} = \pi/2$, we obtain $\mu_{2C,1C} \geqslant 1$ if $n_B \gtrsim 4.90\times10^{13}$ cm^{-3}. Choosing an atomic density of $n_B = 10^{15}$ cm^{-3} would lead to $\mu_{2C,1C} \approx 20.4$ (see Fig. 2).

Finally, we consider the collision system $(e^- + Li^+)$ ($|\varepsilon_0| \approx 5.39$ eV)–H($1s$) (atom B) in which electron capture by Li is assisted by the dipole transition $1s \to 2p$ in H

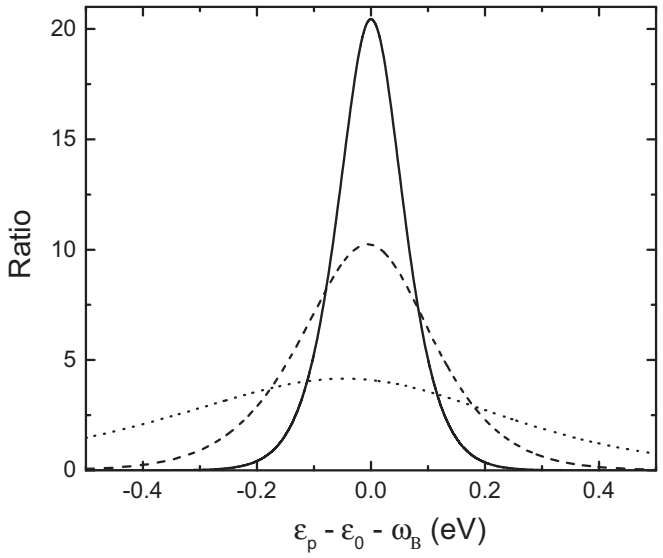

FIG. 2. The ratio (21) as a function of $\Delta\varepsilon_p = \varepsilon_p - \varepsilon_0 - \omega_B$ at $\vartheta_{\boldsymbol{p}} = \pi/2$ for collision velocities $v = 0.01$ a.u. (solid line), $v = 0.02$ a.u. (dashed line), and $v = 0.05$ a.u. (dotted line) for the collision system $(Cs^+ + e^-)$–$Mg(3s^2)$. $b_{\min} = 5$ a.u.; $n_B = 10^{15}$ cm^{-3}.

($\omega_B = 10.2$ eV, $\Gamma_r^B = 7.44 \times 10^{-6}$ eV). Employing Eq. (21) with $v = 0.01$ a.u., $\varepsilon_p = 4.81$ eV, and $\vartheta_{\boldsymbol{p}} = \pi/2$, we obtain that $\mu_{2C,1C} \geqslant 1$ if $n_B \gtrsim 1.06 \times 10^{15}$ cm^{-3}. An atomic density of $n_B = 3 \times 10^{15}$ cm^{-3} yields $\mu_{2C,1C} \approx 2.85$.

Thus, the 2CDR channel can dominate single-center RR of a free electron with atomic center A for relatively low densities of atoms B (as compared to n_{air}). One reason for the good performance of the 2CDR channel is that $R_{\text{eff}} \ll \overline{R}$. For example, using $b_{\min} = 5$ a.u. and $n_B = 10^{15}$ cm^{-3}, we obtain $R_{\text{eff}} \approx 5$ nm $\ll \overline{R} \approx 100$ nm.

For more insight, in Fig. 2 we show the ratio $\mu_{2C,1C}$ given by Eq. (21) as a function of the detuning $\Delta\varepsilon_p = \varepsilon_p - \varepsilon_0 - \omega_B$ from the resonance energy of the incident electron at a fixed $\vartheta_{\boldsymbol{p}} = \pi/2$. Since the shape of $\mu_{2C,1C}$ turns out to be very similar for all the collision systems considered above, in Fig. 2 it is presented just for one of them, $(e^- + Cs^+)$–$Mg(3s^2)$, for impact velocities ranging from 0.01 a.u. (2.5 eV/u) to 0.05 a.u. (62.5 eV/u). It follows from the figure that the function $\mu_{2C,1C}$ reaches a maximum at the position of the resonance, $\varepsilon_{p,r} = \varepsilon_0 + \omega_B$, and is roughly symmetric with respect to this point. The maximum is rather broad: its width is caused by the relative motion of centers A and B, and even for the lowest velocity considered in Fig. 2 it exceeds the corresponding radiative width Γ_r^B ($\Gamma_r^B \sim 10^{-7}$ eV) by many orders of magnitude.

Throughout the above examples different atomic species $(e^- + A)$ and B were considered. One should note that the main requirement for two-center dielectronic recombination to occur—the energy, which is released in the formation of a bound state of the $(e^- + A)$ subsystem, should be close to an excitation energy of B—can also be fulfilled if $(e^- + A)$ and B are of the same species but electron capture takes place in an excited state of the $(e^- + A)$ subsystem (for example, the process $e^- + p^+ + H(1s) \to H(2s) + H(2p) \to H(2s) + H(1s) + \hbar\omega_k$, see Ref. [26]).

At this point one more remark can be appropriate. In our treatment of collisional 2CDR we use bound states of free (noninteracting) centers A and B. In distant collisions, which are considered here, the interaction between them is quite weak. Nevertheless, as estimates show, even in such collisions this interaction may influence these states shifting their energies by noticeable amounts. Since the latter ones can be much larger than the radiative and Auger widths of B, the neglect of them would clearly be unjustified for considering 2CDR in the static situation in which a very fine tuning (within Γ_r^B or $\Gamma_r^B + \Gamma_{2c-A}$) of the transition energies on both centers is necessary in order to reach the highest possible effectiveness of the two-center process [3]. However, in the case of collisional 2CDR, the relative motion so strongly broadens the resonance (see Fig. 2) that the neglect of the energy shifts is not expected to have a substantial impact on the result.

As was mentioned in the Introduction, an incident electron can also form a bound state with an ion by interacting with another free electron, which carries away the energy excess. To get a rough idea about the relative effectiveness of this process we take, as an example, electrons recombining with protons into the ground state of hydrogen (since in this case the data can be easily found for three-body and single-center radiative recombination). Assuming that 2CDR occurs in collsions with helium (atom B), we obtain that the energy of the incident electrons is $\simeq 7.6$ eV and that three-body recombination would outperform radiative recombination starting with densities of the free electrons of $\simeq 5 \times 10^{17}$ cm^{-3} while 2CDR begins to dominate radiative recombination at $n_B \approx 3 \times 10^{16}$ cm^{-3}.

It is of interest to compare collisional 2CDR with collisional 2CPI which was studied very recently in Ref. [17]. In the static situation, where the two centers constitute a bound system, both 2CDR and 2CPI show about the same effectiveness compared to the single-center radiative recombination and photoionization, respectively. It turns out, however, that in collisions 2CDR becomes substantially less effective compared to 2CPI (which at first glance might seem unexpected since these processes can be thought of as the inverse of each other).

The collision influences the processes of two-center recombination and photoionization in two main ways. First, in collisions the effective distance between A and B is greatly increased compared to the interatomic distance in the static case. This equally impacts both collisional 2CDR and collisional 2CPI making them less effective than their static counterparts. Second, the relative motion of the centers A and B effectively broadens their internal transition energies (as they are "viewed" by the collision partner), which in general diminishes the role of resonances (both 2CDR and 2CPI are resonant processes).

However, since in the collisional 2CPI the source of photons is at rest with respect to resonating atoms B [17], the relative motion of centers A and B does not affect the resonant character of the first step of this process—the interaction between B and the external laser field: like in the static case, atom B acts as a very efficient "antenna", absorbing energy from the laser field and transferring it to the subsystem $(e^- + A)$. Although from the "point of view" of the latter

the transfer involves a rather broad range of energies, this does not affect its effectiveness since transitions in $(e^- + A)$ are between bound and continuum states and, thus, are not resonant.

In contrast, in collisional 2CDR, its first step—the energy transfer between the internal states of the subsystems $(e^- + A)$ and B—is strongly affected by the relative motion. From the point of view of atom B this motion broadens the energy of electron transitions in $(e^- + A)$ and even at low collision velocities this broadening is much larger than the natural width of the excited state of B. As a result, there is a very low probability that, for a given change $\varepsilon_p - \varepsilon_0$ in the internal energies of $(e^- + A)$, the corresponding energy transfer $\omega_{\rm tr}$ to B will fit into the resonance conditions $\omega_B - \Gamma_r^B \lesssim \omega_{\rm tr} \lesssim \omega_B + \Gamma_r^B$ for the spontaneous radiative decay of B. That is why the two-center process studied in the present paper is less effective compared to collisional two-center photoionization.

Let us now very briefly consider the correspondence between single-center RR and ICEC in collisions. In order to compare these processes we need incident electron energies which are higher than those in 2CDR since now atom B is ionized. Taking the collision system $(e^- + {\rm Cs}^+)$–${\rm Mg}(3s^2)$ and choosing $b_{\rm min} = 5$ a.u., $v = 0.01$ a.u., and $\varepsilon_p = 3.81$ eV, we obtain that the transition frequency for the electron capture, ${\rm Cs}^+ + e^- \to {\rm Cs}$, is $\omega_A = 7.70$ eV, which is slightly above the ionization threshold for ${\rm Mg}(3s^2)$ ($|\epsilon_0| = 7.65$ eV). Then, we can use the experimental photoionization cross section [27] to get $\sigma_{\rm PI}^{{\rm Mg}(3s^2)}(7.70\,{\rm eV}) \approx 1.2 \times 10^{-18}\,{\rm cm}^2$. For $\vartheta_{\boldsymbol{p}} = \pi/2$ we obtain $\mu_{2C,1C} \geqslant 1$ if $n_B \gtrsim 5.2 \times 10^{18}\,{\rm cm}^{-3}$. Thus, compared to 2CDR, an increase in the atomic density n_B by more than 4 orders is necessary to make the ICEC channel comparable in strength to the corresponding single-center radiative recombination.

Such a large difference between the effectiveness of 2CDR and ICEC is caused mainly by two reasons. One of them (minor) is larger transition frequencies involved in the ICEC, but the main reason is that a dipole-allowed transition from the ground state to an excited bound state, which is characteristic for 2CDR, may be comparable (or even exceed) in its strength transitions from the ground state to the whole (single-electron) continuum (we remark that ICEC involves just a tiny fraction of this continuum).

IV. CONCLUSIONS

We have considered two-center dielectronic recombination occurring in slow atomic collisions. Our consideration was based on the semiclassical approximation and the first order of perturbation theory in the interatomic interaction. Only contributions to this process from relatively large impact parameters were taken into account, which means that the present results should in fact be viewed as yielding a lower boundary for the effectiveness of this process in the collisions.

We have shown that two-center dielectronic recombination, in which the capture of an incident free electron by center A is driven by dynamic two-center electron-electron correlations involving quasiresonant dipole-allowed bound-bound transitions in center B, can outperform the direct single-center process of radiative recombination also in collisions, provided the density of atoms B is not too low. Thus, 2CDR can "survive" even in collisions where the mean distance between A and B exceeds by orders of magnitude the typical size of a bound A-B system.

Compared to the process of collisional two-center photoionization [17] the process considered in the present paper is more depreciated by the relative motion of the colliding centers. This motion affects quite differently the resonance conditions on which both these processes heavily rely: while these conditions are essentially not influenced by this motion in the case of collisional 2CPI, it does wash out the resonant character of collisional 2CDR.

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Correction: Minor typographical errors in Sec. II were found. The indices in the label for the intermediate state in the text before Eq. (6) have been fixed. Numerical prefactors in Eqs. (14), (16), and (21) have been fixed.