

## Radiative collision-induced electron continuum-continuum scattering

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The photoionization of an atom is examined in the presence of a foreign atom. We find that a two-photon radiative collision process induces large cross sections for electron continuum-continuum scattering. The enhancement is caused by an intensity-induced collisional shift which, when used to cancel out the collisional dephasing between the initial and final scattering states at wide range of internuclear separations, enhances the electron continuum-continuum scattering process.

Recently, two-photon laser-induced radiative collisions have been observed.<sup>1</sup> During the radiative collision between Ba and Tl ground-state atoms, two photons are absorbed which are not in resonance with any of the transitions in the atoms and which result in the simultaneous excitation of both atoms. We recently analyzed the theoretical aspect of this process using a semiclassical approach.<sup>2</sup> We treated the two-photon—one-collision and two-photon—two-collision cases, and found in the latter case a new intensity-induced collisional phase shift. This shift is used to control the overall phase between the initial and final scattering states. When the phase difference goes through zero (phase resonance) the cross section is enhanced, and the two-photon line shape becomes both symmetric and highly sensitive to the intensity of the radiation.

In this paper we study the implications of the above two-photon—two-collision laser-induced radiative collision process<sup>2</sup> when it involves the continuum states of one of the colliding atoms. We shall examine the photoionization of this atom during its collision with a foreign atom in the presence of an applied radiation field which is nonresonant with either of the atom's discrete transitions.

The transfer of excitation from an excited atom  $B$  to an acceptor atom  $A$  accompanied by the simultaneous absorption of a single photon sufficient to photoionize the initially excited state of atom  $B$  was previously analyzed.<sup>3</sup> The excited acceptor atom  $A$  then ionizes atom  $B$  via a Penning ionization process resulting in an increase in the photoionization efficiency. In contrast to the one-photon—one-collision process, we find here that a second-order radiative process causes an intensity-induced collisional shift which can be used to cancel out the collisional dephasing shift between the initial and final scattering wave functions for all internuclear separations. As a

result of this cancellation we find that the collision causes a large enhancement in the cross section for electronic continuum-continuum scattering (i.e., the process which results in the production of electrons with different discrete energies) which is otherwise negligible.

The nature of the present process is quite different from the process involving only discrete states. In the discrete case, the frequency of radiation is taken not to coincide with any transition frequency in either atom. However, in the present case where continuum states are involved, the frequency of radiation coincides with some transitions to the continuum. Moreover, owing to the nature of the continuum states, the present process involves an infinite-level system.

We consider the collision of atoms  $A$  and  $B$  in the presence of a radiation field  $\vec{E} = \vec{E}_0 \cos(\omega t)$  via the process  $A + B^* + 2\hbar\omega \rightarrow A + B(\nu) + \hbar\omega \rightarrow A^* + B + \hbar\omega \rightarrow A^{**} + B \rightarrow A^* + B^+ + e(\epsilon)$ , where  $e(\epsilon)$  represents the distribution of electron energies produced for both of the continuum states involved in the scattering process. In describing the process we treat the motion of the nuclei classically. Moreover, we assume that the dominant contribution to the collisional cross section comes from large internuclear separations where electronic overlap is negligible. Hence we represent the system with a product of atomic states and write

$$\hat{H} = \hat{H}_A + \hat{H}_B + \hat{V}_{AB} - \vec{\mu}_A \cdot \vec{E} - \vec{\mu}_B \cdot \vec{E}, \quad (1)$$

where  $\hat{H}_A$  and  $\hat{H}_B$  are the electronic Hamiltonians of isolated atoms  $A$  and  $B$ ,  $\hat{V}_{AB}(t)$  is the atom-atom interaction, and the other terms are the laser-field—atom interaction terms in the dipole—classical-field approximation. We will treat the magnetic number degeneracy by treating the

atom-atom interaction in the rotating atom approximation, where  $\hat{V}_{AB}$  matrix elements are evaluated by assuming the dipole transition moments are always

$$\begin{aligned} \psi = & a_0(t) |0a\rangle |1b\rangle \exp(-i\omega_0 t) + \int a_\nu |0a\rangle |\nu b\rangle \exp[-i(\omega_0 + \omega_\nu)t] d\nu \\ & + a_1(t) |1a\rangle |0b\rangle \exp(-i\omega_1 t) + a_2(t) |2a\rangle |0b\rangle \exp[-i(\omega_1 + \omega_2)t] \\ & + \int a_{\nu'} |1a\rangle |\nu' b\rangle \exp[-i(\omega_{\nu'} + \omega_1)t] d\nu'. \end{aligned} \quad (2)$$

In the process the initial state  $|0a\rangle |1b\rangle$  is excited by the electromagnetic field to the continuum states  $|0a\rangle |\nu b\rangle$ . For continuum states nearly resonant with the applied photon, the interaction results in real excitations, and for those away from resonance the interaction results in virtual excitations. A virtual collision then transfers the excitation from  $|0a\rangle |\nu b\rangle$  to the state  $|1a\rangle |0b\rangle$  which in turn gets virtually excited by the electromagnetic field to  $|2a\rangle |0b\rangle$ . Finally, a collisional transfer from  $|2a\rangle |0b\rangle$  to  $|1a\rangle |\nu' b\rangle$  takes place.

During the process two sets of continuum states in atom  $B$  become excited, resulting in the emission of electrons with kinetic energies centered at two discrete values. One set of the continuum states becomes involved only as intermediate states in the overall process, while the other set helps comprise the final state of our system. The cross section for this process will be obtained by substituting the above wave function and Hamiltonian into the time-dependent Schrödinger equation and solving for  $a_\nu$ , the probability amplitude of the final state  $|1a\rangle |\nu' b\rangle$ . Later in the paper we will estimate the cross section for the process using a high-lying excited state of atom  $B$  for our initial state. The derivation which follows, however, is also applicable to the low-lying excited states of atom  $B$ .

In this model we have neglected the direct electron scattering process  $A + B^+ + e(\epsilon) + \hbar\omega \rightarrow A + B^+ + e(\epsilon')$  since this interaction is expected to be negligible in comparison to the radiative col-

lision process when using low-field intensities. The direct scattering process may become important, however, when using very intense fields.<sup>4</sup> In this case, a new set of high-lying continuum states becomes available to interact with the original continuum states. The interaction between these two overlapping electronic continua is predicted to become an important factor in the treatment of the collision dynamics. Furthermore, the participation of the high-lying continua due to the intense field is predicted to lead to interesting effects which include the emission of electrons having distributions in kinetic energies which are roughly shifted by  $\hbar\omega$  on either side of the laser field-free emitted electrons.

Substituting Eqs. (1) and (2) in the time-dependent Schrödinger equation gives

$$da_0/dt = i \int \mu_{\nu 1} E_0 \exp(i\Delta_\nu t) a_\nu d\nu, \quad (3)$$

$$\begin{aligned} da_\nu/dt = & i\mu_{1\nu} E_0 \exp(-i\Delta_\nu t) a_0 \\ & - iV_1 \exp(i\bar{\Delta}_\nu t) a_1, \end{aligned} \quad (4)$$

$$\begin{aligned} da_1/dt = & -i \int V_1^* \exp(-i\bar{\Delta}_\nu t) a_\nu d\nu \\ & + i\mu_{2A} E_0 \exp(i\Delta_2 t) a_2, \end{aligned} \quad (5)$$

$$\begin{aligned} da_2/dt = & i\mu_{2A}^* E_0 \exp(-i\Delta_2 t) a_1 \\ & - i \int V_2 \exp(i\bar{\Delta}_{\nu'} t) a_{\nu'} d\nu', \end{aligned} \quad (6)$$

$$da_{\nu'}/dt = iV_2^* \exp(-i\bar{\Delta}_{\nu'} t) a_2, \quad (7)$$

where

$$\Delta_\nu = \omega - \omega_\nu, \quad \Delta_2 = \omega - \omega_2, \quad \bar{\Delta}_\nu = \omega_0 + \omega_\nu - \omega_1, \quad \bar{\Delta}_{\nu'} = \omega_1 + \omega_2 - (\omega_{\nu'} + \omega_1) = \omega_2 - \omega_{\nu'},$$

$$\mu_{1A} = \frac{1}{\hbar} \langle 1a | \hat{\mu}_{A_z} | 0a \rangle, \quad \mu_{2A} = \frac{1}{\hbar} \langle 2a | \hat{\mu}_{A_z} | 1a \rangle, \quad \mu_{0\nu} = \frac{1}{\hbar} \langle \nu b | \hat{\mu}_{B_z} | 0b \rangle,$$

$$\mu_{0\nu'} = \frac{1}{\hbar} \langle \nu' b | \hat{\mu}_{B_z} | 0b \rangle, \quad \mu_{\nu 1} = \frac{1}{\hbar} \langle \nu b | \hat{\mu}_{B_z} | 1b \rangle,$$

$$V_1 = \frac{1}{\hbar} \langle 0b | \langle 1a | \hat{V}_{AB_z} | 0a \rangle | \nu b \rangle, \quad \text{and} \quad V_2 = \frac{1}{\hbar} \langle \nu' b | \langle 1a | \hat{V}_{AB_z} | 2a \rangle | 0b \rangle.$$

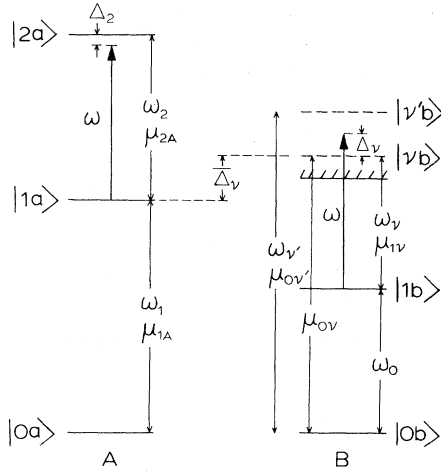


FIG. 1. Restricted energy-level diagram for the two-photon radiative collision involving continuum states.

We now proceed to reduce this system of infinite coupled levels to an effective two-level system involving only the initial and final states by sequentially eliminating  $a_\nu$ ,  $a_1$ , and  $a_2$ . This approach is justified if these intermediate states are chosen not to interact strongly with the electromagnetic field nor with the collisional field. In the elimination of the continuum states, however, the electromagnetic field will necessarily resonate with a portion of the continuum. We will show later that this radiatively resonant set of continuum states is responsible for a portion of the ionization current while the non-resonant states contribute primarily to the efficient transfer of excitation.

The first step we carry out is the elimination of the amplitudes of the continuum states  $a_\nu$  by considering them as intermediates to the transition between  $a_0$  and  $a_1$ . We integrate Eq. (4) in order to solve for  $a_\nu$  and hence eliminate it from the rest of the equations. Note that if  $|\Delta_\nu| = |\omega - \omega_\nu|$  were always large, we could integrate the first term of Eq.

(4) by parts and keep only the zeroth-order term. However,  $|\Delta_\nu|$  can be small, and therefore this procedure is not accurate in evaluating the integral. The same argument holds in the contribution from the second integral of Eq. (4). Both cases must be considered in order to treat the problem accurately. In order to do this, we will use the following procedure.<sup>5</sup>

Since transitions from  $|0a\rangle |1b\rangle$  to the continuum obey the Frank-Condon principle, we separate the continuum states into two sets; the first set is close to the resonance condition ( $|\Delta_\nu| \leq |\Delta_\nu^0|$ ) where  $|\Delta_\nu^0|$  is a small quantity, and the second set is that which is sufficiently far away from resonance ( $|\Delta_\nu| \geq |\Delta_\nu^0|$ ).

When  $\Delta_\nu$  is large, the exponential in the first integral of Eq. (4) oscillates swiftly. If, in addition, the field amplitude  $E_0$  is a slowly varying function of time, we may approximate the integral by keeping the first-order term in an integration by parts, i.e.,  $-(\mu_{\nu 1} E_0 / \Delta_\nu) \exp(-i\Delta_\nu t) a_0$ . However, when near-resonant states are considered in the integral,  $\Delta_\nu$  is small, and this part of the contribution to  $a_\nu$  can only be represented by a full integral as  $i \int_{-\infty}^t \mu_{\nu 1} E_0 \exp(-i\Delta_\nu t') a_0 dt'$ . Similarly, we can write expressions for the second integral of Eq. (4) corresponding to the cases where  $\Delta_\nu$  is large and  $\Delta_\nu$  is small.

Thus substituting these expressions for  $a_\nu$  in Eqs. (3) and (5) gives

$$\frac{da_0}{dt} + \gamma_0 a_0 = iG_1 \exp(i\Delta_1 t) a_1, \quad (8)$$

$$\begin{aligned} \frac{da_1}{dt} - iV_1'^2 / \Delta_0' a_1 = iG_2 \exp(-i\Delta_1 t) a_0 \\ + i\mu_{2A} E_0 \exp(i\Delta_2 t) a_2, \end{aligned} \quad (9)$$

where

$$\begin{aligned} \gamma_0 = \pi \mu_R^2 E_0^2 \eta + i \frac{\mu_{1B}^2 E_0^2}{\Delta_1'}, \quad \eta = \frac{d\nu}{dE}, \quad \frac{\mu_{1B}^2}{\Delta_1'} = \int \frac{\mu_{\nu 1}^2}{\Delta_\nu} d\nu, \\ G_1 = \alpha_1 + i\beta_1, \quad G_2 = \alpha_2 + i\beta_2, \quad \beta_1 = \pi E_0 \mu_r V_r \eta, \quad \beta_2 = \beta_1^*, \\ \alpha_1 = -E_0 \int \frac{\mu_{1\nu} V_1}{\Delta_\nu} d\nu, \quad \alpha_2 = E_0 \int \frac{\mu_{\nu 1} V_1^*}{\Delta_\nu} d\nu, \quad \frac{V_1'^2}{\Delta_0'} = \int \frac{V_1^2}{\Delta_\nu} d\nu. \end{aligned}$$

Here  $\mu_R^2 = \int \mu_{1\nu}^2 d\nu$  and  $\mu_r V_r = \int \mu_{1\nu} V_1 d\nu$  represent integrals over the radiatively resonant continuum states,  $V_R^2 = \int V_1^2 d\nu$  represents an integral over the collisionally resonant states,  $\Delta_1 = \omega_1 - \omega$ , and  $\eta = d\nu/d\Delta$  is the density of the continuum states. We also note that we used the quantities  $\mu_{1B}^2 E_0^2 / \Delta_1'$  and  $V_1'^2 / \Delta_0'$  as a compact way of describing, respectively, the Stark and collisional frequency shifts covered by all the non-resonant intermediate continuum states. Later in the paper we will, for some cases, show that the resonant

states' contribution to  $G_1$  and  $G_2$ ,  $\pi E_0 \mu_r V_r \eta$ , is much smaller than the contribution of the nonresonant states.

We now discuss the nature of the couplings  $G_1$  and  $G_2$ . For this purpose we concentrate on the system comprised of  $a_0$ ,  $a_1$ , and  $a_\nu$  by dropping the coupling to  $a_2$  in Eq. (9). In this case one can easily show that

$$\frac{d}{dt}(|a_0|^2 + |a_1|^2) + 2(\text{Re}\gamma_0)|a_0|^2 + 4\text{Re}(\beta_1 a_1 a_0^* e^{i\Delta_1 t}) = 0.$$

This result shows that the total population  $|a_0|^2 + |a_1|^2$  of the closely coupled system of  $a_0$  and  $a_1$  has, as expected, the decay rate  $2(\text{Re}\gamma_0)|a_0|^2$  as a result of the direct photoionization losses from  $|0a\rangle$   $|1b\rangle$ . In addition to this rate, the system has an oscillatory coherent rate which does not depend on either population  $|a_0|^2$  or  $|a_1|^2$  but upon the coherent product of  $a_0^*$  and  $a_1$ . This rate is a result of the participation of the resonant continuum states.

We now derive an effective system coupling the initial and final amplitudes  $a_0$  and  $a_\nu$ , respectively. We take  $\Delta_1 > |G_1|$ ,  $\Delta_2 \geq \mu_{2A} E_0$  and eliminate  $a_1$  and  $a_2$  sequentially from Eqs. (8) and (9) and (6) and (7). We then integrate their equations by parts and keep the lowest terms. The resulting equations are

$$da_0/dt + \Gamma a_0 = ig_1 \int g_2 \exp(i\delta_\nu t) a_\nu d\nu', \quad (10)$$

$$da_\nu/dt + iV_2 \exp(-i\bar{\Delta}_\nu t) \int g_2 \exp(i\bar{\Delta}_\nu t) a_\nu d\nu'' = ig_3 \exp(-i\delta_\nu t) a_0, \quad (11)$$

where

$$\begin{aligned} \Gamma &= \pi\mu_R^2 E_0^2 \eta + i\mu_{1B}^2 E_0^2 / \Delta_1' + iG_1 G_2 (\Delta_1 + V_1'^2 / \Delta_0')^{-1} \\ &\quad - \mu_{2A}^2 E_0^2 G_1 G_2 (\Delta_2 - V_1'^2 / \Delta_0')^{-1} (\Delta_1 + V_1'^2 / \Delta_0')^{-1} [\Delta_1 + \Delta_2 - \mu_{2A}^2 E_0^2 (\Delta_2 - V_1'^2 / \Delta_0')^{-1}]^{-1}, \\ g_1 &= \mu_{2A} E_0 G_1 (\Delta_2 - V_1'^2 / \Delta_0')^{-1}, \quad g_2 = V_2 [\bar{\Delta}_\nu - \mu_{2A}^2 E_0^2 (\Delta_2 - V_1'^2 / \Delta_0')^{-1}]^{-1}, \\ g_3 &= V_2 \mu_{2A} E_0 G_2 (\Delta_1 + V_1'^2 / \Delta_0')^{-1} [\Delta_1 + \Delta_2 - \mu_{2A}^2 E_0^2 (\Delta_2 - V_1'^2 / \Delta_0')^{-1}]^{-1}, \end{aligned}$$

and

$$\delta_\nu = \Delta_1 + \Delta_2 + \bar{\Delta}_\nu.$$

We now make the following comments about Eqs. (10) and (11). This system of equations describes the close coupling of the ground state with the final scattering continuum state  $|\nu' b\rangle$ ; it is an effective two-photon-two-collision radiative coupling. The couplings  $g_1$  and  $g_2$  are complex. The real parts of the couplings are responsible for the actual excitation to the final state via the coupling to the nonresonant intermediate continuum states. The imaginary parts of these couplings, however, describe a coherent rate similar to the rate discussed after Eq. (9). It results from the participation of the intermediate resonant continuum states.

For a given radiation frequency, only the state  $|\nu' b\rangle$  which satisfies the overall energy conservation of the process can be appreciably populated. Consequently, only a few states which are in the neighborhood of the resonating state need to be kept in the integrals of the right- and left-hand sides of Eqs. (10) and (11), respectively. Hence Eqs. (10) and (11) become

$$\frac{da_0}{dt} + \Gamma a_0 = ig_1 g_2 \exp(i\delta_\nu t) a_\nu, \quad (12)$$

$$\frac{da_\nu}{dt} + iV_2 g_2 a_\nu = ig_3 \exp(-i\delta_\nu t) a_0. \quad (13)$$

The function  $\Gamma$  is complex. Its real part gives an induced decay rate of the ground-state population. The decay rate includes a number of terms. One of these terms is due to direct photoionization ( $\pi\mu_R^2 E_0^2 d\nu/d\Delta$ ). The other terms are collision-induced photoionization rates. They depend on the product of the field intensity and the square of the collisional coupling. For example, the term

$$-\pi\mu_r V_r \left[ \int \frac{\mu_{\nu 1} V_1^*}{\Delta_\nu} d\nu \right] E_0^2 \frac{d\nu}{d\Delta}$$

is a new collision-induced ionization which depends simultaneously on the intensity and the collisional coupling. We note that this rate enhances or weakens the direct photoionization rate when the sign of  $\Delta_\nu$  is negative and positive, respectively. The real part of the last term of  $\Gamma$  gives a higher-order effect of this collision-induced photoionization.

The imaginary part of  $\Gamma$  gives a shift in the energy of the ground state. The shift consists of three types. One type is the ordinary Stark shift caused by the electromagnetic field interaction with the nonresonant continuum states,  $\mu_{1B}^2 E_0^2 / \Delta_1' = E_0^2 \int \mu_{1\nu}^2 / \Delta_\nu d\nu$ . The second type of

shift is an intensity-induced collisional shift  $ib_1E_0^2V_1'^2 + ib_2E_0^4V_1V_1'$ , where  $b_1$  and  $b_2$  are functions of the detunings and the effective dipole matrix elements of the discrete-discrete transition and discrete-nonresonant continuum transitions. The third type of shift is an intensity-induced collisional shift with only the resonant continuum state participating. The sign of this shift, as well as the other two types, will depend on the sign of the various detunings.

The intensity-induced collisional shifts are interesting since their magnitude can be controlled by the intensity of the electromagnetic field. Hence the overall shift of the final state with respect to the ground state can be controlled by the intensity. In fact, it is possible to achieve complete cancellation of the relative phase shift between the initial and final transition probability amplitudes. In this paper we analyze the process in the weak-field limit, leaving the strong-field effects for a later work. In the weak-field limit, Eqs. (10) and (11) reduce to

$$\frac{da_0}{dt} + i \operatorname{Re} \left[ \frac{G_1 G_2}{\Delta_1} \right] a_0 = 0, \quad (14)$$

$$\frac{da_{\nu'}}{dt} + i \frac{V_2^2}{\Delta_{\nu'}} a_{\nu'} = \frac{i\mu_{2A} V_2 E_0 G_2}{\Delta_1(\Delta_1 + \Delta_2)} \exp(-i\delta_{\nu'} t) a_0, \quad (15)$$

where we kept the lowest order of the intensity-induced collisional shift since even in the weak-field limit, this shift may be of the same order as the collisional shift of the final state. When  $E_0$  changes very little during the time of collision, Eq. (14) gives

$$a_0 = \exp \left[ -i \int_{-\infty}^t \operatorname{Re} \left[ \frac{G_1 G_2}{\Delta_1} \right] dt \right] \quad (16)$$

and hence

$$|a_{\nu'}(\infty)|^2 = \frac{4\mu_{2A}^2 E_0^2}{\Delta_1^2(\Delta_1 + \Delta_2)^2} \left| \int_0^\infty V_2 G_2 e^{iS} dt \right|^2, \quad (17)$$

$$S = \int \left[ \frac{V_2^2}{\Delta_{\nu'}} + \frac{\alpha_1 \alpha_2 - \beta_1^2}{\Delta_1} - \delta_{\nu'} \right] dt. \quad (18)$$

We now calculate the cross section for the continuum-continuum scattering process. Taking  $\delta_{\nu'} = 0$ , in the dipole-dipole interaction we find  $S = \int_0^t CR^{-6} dt$ , where

$$C = \frac{\hbar^2 v_{0\nu'}^2 \mu_{2A}^2}{\Delta_{\nu'}} + \frac{\mu_{1A} E_0^2}{\Delta_1} \left[ \int \frac{\mu_{1\nu} \mu_{0\nu}}{\Delta_{\nu'}} d\nu \right] \left[ \int \frac{\mu_{\nu 1} \mu_{\nu 0}}{\Delta_{\nu'}} d\nu \right] - \frac{\pi^2 \hbar^2 \mu_{1A}^2 E_0^2 \left[ \int^r \mu_{1\nu} \mu_{\nu 0} d\nu \right]^2 \eta^2}{\Delta_1} \quad (19)$$

and

$$|a_{\nu'}(+\infty)|^2 = 4F_1^2 E_0^4 \left| \int_0^\infty R^{-6} \cos S dt \right|^2, \quad (20)$$

where

$$F_1^2 = \frac{\hbar^4 \mu_{2A}^4 \mu_{0\nu'}^2 \mu_{1A}^2}{\Delta_1^2 (\Delta_1 + \Delta_2)^2} \left[ \pi^2 \left| \int^r \mu_{\nu 1} \mu_{\nu 0} d\nu \right|^2 \eta^2 + \left| \int \frac{\mu_{\nu 1} \mu_{\nu 0}}{\Delta_{\nu'}} d\nu \right|^2 \right]. \quad (21)$$

The ionization cross section  $\sigma$  is calculated from the integration of  $|a_{\nu'}(\infty)|^2$  over the impact parameters. A thermal average of the cross section  $\bar{\sigma}$  then yields an ionization rate. For large  $C$ , all impact parameters can be integrated over because the frequency shift becomes large for  $R$  values less than or approximately equal to 15 Å, and there is no change in  $a_{\nu'}$  at  $R$  values where overlap is important and deviations from straight-line trajectory occur.

At some intensities, however,  $C$  can become very small even in the weak-field limit. The situation where  $C$  is very small suggests a large coupling coefficient in the absence of any dephasing effect for all internuclear separations  $R \geq 4$  Å. This could lead to extremely large cross sections for the process. However, because of the detuning at small  $R$ , orbiting phenomena play a significant role. An estimate of the magnitude of the cross section at the peak of the resonance can be determined from Eq. (9) by taking  $C=0$ . In this case  $|a_{\nu'}(\infty)|^2 = 1.5\pi F_1^2 E_0^4 / (\rho^{10} v^2)$ . A lower limit on the estimate can be found by calculating the contribution from impact parameters where orbiting is not important; that is,  $\sigma > \int_{\rho_c}^\infty 2\pi \rho d\rho |a_{\nu'}(\infty)|^2 = 3\pi^2 F_1^2 E_0^4 (v \rho_c^4)^{-2}$ , where  $F_1$  is given by Eq. (21) along with the condition  $C=0$ .

We now examine the cross section for a realistic case. The degree of contribution to the phase from the resonant and nonresonant continuum states depends on the atom in question (structure of the continuum) and the position of the resonant state with respect to the ionization limit as well as to the states

of the atoms. Here we are interested in arriving at an order-of-magnitude estimate of the cross section. We take the case where the initial excited state of atom  $B$  is regarded as high lying and hydrogenlike with a principle quantum number  $n$ . In this case the Kramers formulas, when applied to the absorption from bound states to free states, give the following expression for  $\mu_{1\nu}^2\eta$  (Ref. 6):

$$\pi\mu_{1\nu}^2\eta = \frac{1}{\pi\sqrt{3}} \left[ \frac{2R_y}{\hbar\omega} \right]^4 \frac{a_0^2 e^2}{\hbar R_y} \frac{1}{n^3}, \quad (22)$$

where  $R_y$  is the Rydberg energy,  $a_0$  is the Bohr radius, and  $e$  is the electronic charge. The couplings  $G_1$  and  $G_2$  can now be estimated by taking the effective nonresonant continuum state to be a high Rydberg state.<sup>3</sup> Thus

$$\begin{aligned} G_2 &= -E_0 \int \frac{\mu_{\nu 1} V_1^*}{\Delta_\nu} d\nu + i\pi\mu_{1\nu} E_0 V_1^* \eta \\ &= -\frac{B_1 E_0}{R^3} \left[ 1 - \frac{i}{2} \frac{B_2}{B_1} \right], \\ G_1 &= -\frac{B'_1 E_0}{R^3} \left[ 1 - \frac{i}{2} \frac{B'_2}{B'_1} \right] \end{aligned} \quad (23)$$

with

$$\begin{aligned} B_1 &= \hbar(\mu_{1A}\mu_{0n'}\mu_{nn'})/\omega \quad \text{and} \quad \frac{B_2}{B_1} = \frac{\omega_1}{\omega_0 n^2}, \\ B'_1 &= \hbar(\mu_{1A}\mu_{0n'}\mu_{nn'})/\omega' \quad \text{and} \quad \frac{B'_2}{B'_1} = \frac{\omega_1}{\omega_0 n^2}, \end{aligned} \quad (24)$$

where  $\mu_{0n'}$  and  $\mu_{nn'}$  are the matrix elements of the transition  $0 \rightarrow n'$  and  $n \rightarrow n'$ . Taking  $\mu_{1A}^2 \sim e^2 a_0^2$ ,  $\mu_{0n'}^2 \sim e^2 a_0^2/n^3$ , and  $\mu_{nn'}^2 \sim e^2 a_0^2/n^4$  gives  $B_1 = e^6 a_0^6/(\omega n^7)$  and  $B'_1 = e^6 a_0^6/(\omega' n^7)$ . The fact that  $B_2/B_1 \ll 1$  and  $B'_2/B'_1 \ll 1$  for  $n > 5$  allows us to neglect the imaginary part of  $G_1$  and  $G_2$ ; hence

$$G_1 = -\frac{B'_1 E_0}{R^3}, \quad G_2 = -\frac{B_1 E_0}{R^3}, \quad (25)$$

and consequently

$$C = \frac{\hbar^2 \mu_{0\nu'}^2 \mu_{2A}^2}{\Delta_\nu} - \frac{B_1 B'_1 E_0^2}{\Delta_1} \quad \text{and} \quad F_1^2 = \frac{\hbar^2 \mu_{2A}^4 \mu_{0\nu'}^2 B_1^2}{\Delta_1^2 (\Delta_1 + \Delta_2)^2}. \quad (26)$$

These estimates show, in the case of a high-lying initial state, that the contribution of the resonant continuum states to the intensity-induced shift and to the amplitude of  $a_\nu$  is negligible compared to that of the nonresonant states. However, these contributions are expected to become more important when the initial scattering states are low lying. We will examine the effect of these contributions in a later work.

We now numerically estimate the cross section using typical values for a general system. Taking  $E_0^2 = (2.5 \times 10^6 \text{ V/cm})^2$  (corresponding to a power density of  $8.3 \times 10^9 \text{ W/cm}^2$ ) gives  $\mu_{1A}^2 E_0^2/\omega^2 = 2 \times 10^{-3}$ ,  $\mu_{0n'}^2 = 0.25 \text{ a.u.}^2$ ,  $\mu_{nn'}^2 = 6.25 \times 10^{-2} \text{ a.u.}^2$  with  $n = 4$ . Choosing  $\mu_{2A}^2 = 9 \times 10^{-2} \text{ a.u.}^2$ ,  $\mu_{0\nu'}^2 = 1.4 \times 10^{-3} \text{ a.u.}^2$ , and  $\Delta_\nu/\Delta_1 = 4.05$  then gives  $C = 0$ . We choose  $\omega = 1.5 \times 10^4 \text{ cm}^{-1}$ ,  $\Delta_1 = 1000 \text{ cm}^{-1}$ ,  $\Delta_2 = 1000 \text{ cm}^{-1}$ ,  $V = 5 \times 10^4 \text{ cm/s}$ , and  $\rho_c = 3 \text{ \AA}$  yielding  $\sigma > 2.8 \times 10^{-5} \text{ \AA}^2$ .

We note that when other discrete states of atom  $B$  are involved, other electrons will be ejected with appropriate energy to conserve the overall energy of the process. Hence owing to this effect, a discrete spectrum of electrons is produced.

In conclusion, we have shown that the phase of the wave functions of the initial and final collisional states can be externally controlled, thereby allowing a large continuum-continuum scattering cross section. Owing to the change in the energy of the electrons, this effect should be amenable to experimental investigations using energy analysis techniques.

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