INTERNATIONAL ATOMIC ENERGY AGENCY
UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION

INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
34100 TRIESTE (ITALY) - P.O.B. 586 - MIRAMARE - STRADA COSTIERA 11 - TELEPHONE: 2240-1
CABLE: CENTRATOM - TELEX 460392 - I

H4.SMR/381-5

COLLEGE ON ATOMIC AND MOLECULAR PHYSICS: PHOTON ASSISTED COLLISIONS IN ATOMS AND MOLECULES

(30 January - 24 February 1989)

PHASE CONTROL OF ATOMIC SCATTERING STATES IN TWO-PHOTON RADIATIVE COLLISIONS

M.H. NAYFEH
&
G.B. HILLARD

University of Illinois at Urbana-Champaign
Dept. of Physics
Urbana, Illinois

REPRINTED FROM PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON LASERS '82, DECEMBER 13-17, 1982

PHASE CONTROL OF ATOMIC SCATTERING STATES IN TWO-PHOTON RADIATIVE COLLISIONS

Munir H. Nayfeh and G. B. Hillard
Department of Physics
University of Illinois at Urbana-Champaign
1110 West Green Street
Urbana, IL 61801

We consider the two-photon interaction of two colliding atoms having transition frequencies ω_1 and ω_3 with a pulsed laser field of frequency ω which is not resonating with any of the transitions of either atom. It has been established that when $\omega \approx (\omega_1+\omega_3)/2$ the system of the two colliding atoms absorbs two photons and the atoms end up individually excited at ω_1 and ω_3 (two-photon radiative collision process). Our analysis shows that when one of the atoms receives both photons (one atom is radiatively active), the collisional interaction provides for the required coupling for the process, but at the same time causes some dephasing of the interaction (e.g. C_6/R^6) which limits the effectiveness of the coupling to a small range of internuclear separations. On the other hand when both atoms are radiatively active, we find that the collision provides an additional dephasing term which is proportional to the intensity of the laser (a.c. stark-collisional effect), thus making the overall C_6 of the interaction modulated by the intensity and detuning of the laser. This additional dephasing can be used to control and even eliminate the pure collisional dephasing (phase resonance), thus allowing the system to interact coherently over a wide range of internuclear separations (two-atom coherence). At phase resonance, the normal highly asymmetric line-shape for the process becomes symmetric. Moreover, accompanying this symmetry we find a large enhancement of the cross section. The enhancement is caused by the contribution from small impact parameters which, far away from phase resonance, would not have contributed.

I. Introduction

Considerable theoretical and experimental efforts have been devoted, in the last few years, to the investigation of absorption or stimulated emission resonances that are only present during collisions of excited atoms with ground state atoms of another element (radiative collision). In a radiative collision the initially excited atom having excitation energy ε returns to its ground state and leaves the second atom in an excited state of excitation energy nearly equal to $\varepsilon \pm \hbar\omega$. Large cross sections (i.e., several $Å^2$) are predicted[1] and were measured at power levels ~ 10^7 W/cm^2 in cases where the excitation transfer does not occur in the absence of the laser field.

Although the possibility of laser induced multiphoton radiative collisions has been previously suggested,[2] it is only recently that an experimental effort has dealt with it.[3] During the collision of Ba and Tl ground state atoms, two photons are absorbed which results in the simultaneous excitation of both atoms. The results of this experiment were analyzable in terms of a simple extension of the theoretical description used to explain the single photon radiative collision. In this article we show that this simple extension is not always the whole story, and report on a new coherent effect in the interaction: intensity induced 'two-atom' coherence.[4-7]

Our analysis of the interaction shows that when all the radiative interactions take place with only one of the atoms (see Fig. 1), as is the case in the Ba-Tl experiment, the process can be transformed to an equivalent form of a single-photon radiative process except for an ac Stark shift and the introduction of an effective two-photon coupling in place of the single photon coupling (simple extension of the single photon case which involves one photon and one collision couplings). The intensity normalized line shape is identical to the one photon case; it has an extended wing as is encountered in a single photon radiative collision.[8-9] However, in the case where both atoms couple to the electromagnetic field (see Fig. 2), we

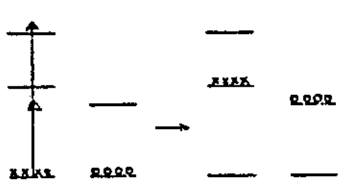

Figure 1. Two-photon-one collision interaction.

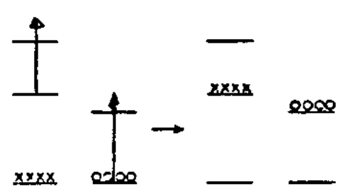

Figure 2. Two-photon-two collision interaction.

find that the above simple substitution does not hold. The overall radiative collision coupling is proportional to the product of an effective two photon coupling and an effective two-collision coupling. In addition to ac Stark shifts we find that a new intensity induced collisional shift is introduced. This induced shift can be used to control the overall phase between the initial and final scattering states for a wide range of internuclear separations. When the phase difference goes through zero (phase resonance) the 'two-atom' system interacts coherently with the electromagnetic field over a wide range of internuclear separations. As a result the cross section is enhanced and the two photon lineshape becomes both symmetric and highly sensitive to the intensity of the radiation.

In Section II we analyze the two photon-one collision case, and in Section III we analyze the two photon-two collision case. The phase phenomenon is discussed in Section IV. Finally, in Section V we briefly discuss the concept of 'two-atom' coherence.

II. Two Photon-one Collision Case

We consider the collision of atoms A and B in their ground states in the presence of the radiation field $\vec{\varepsilon} = \vec{E}_o \cos\omega t$ which does not resonate with any of the transitions in either atom. We are interested in the process where both atoms emerge from the interaction excited. In describing the process we treat the motion of the nuclei classically. Moreover, we assume that the dominant contribution comes from large internuclear separations where electronic overlap is negligible. Hence we represent the system with a product of atomic states and write

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

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 V_{AB} matrix elements are evaluated by assuming the transition moments are always aligned along the line joining the nuclei.

Consider the first case where only one atom interacts with the field. The state vector of the system is taken to be of the form (see Fig. 3):

$$|\psi(t)\rangle = a_o(t)|0a\rangle|0b\rangle + a_1(t)|1a\rangle|0b\rangle\exp(i\omega_1 t) + a_2(t)|2a\rangle|0b\rangle\exp[i(\omega_1+\omega_2)t] + a_3(t)|1a\rangle|1b\rangle\exp[i(\omega_1+\omega_3)t] \qquad (2)$$

In the process the initial state $|0a\rangle|0b\rangle$ is virtually excited by the electromagnetic field to the state $|1a\rangle|0b\rangle$, which in turn is virtually excited by the electromagnetic field to the state $|2a\rangle|0b\rangle$. Finally, a collisional transfer from $|2a\rangle|0b\rangle$ to $|1a\rangle|1b\rangle$ nearly conserves the overall energy for the transition. Thus substituting Eqs. 1 and 2 in the time dependent Schrödinger equation gives the following equations for the time dependent coefficients in the rotating wave approximation: $da_o/dt = i\mu_{1A}E_o\exp(i\Delta_1 t)a_1$, $da_1/dt = i\mu^*_{1A}E_o\exp(-i\Delta_1 t)a_o + i\mu_{2A}E_o\exp(i\Delta_2 t)a_2$,

$da_2/dt = i\mu^*_{2A}E_o\exp(-i\Delta_2 t)a_1 + iV_2\exp(i\Delta_o t)a_3$,
$da_3/dt = iV_2^*\exp(-i\Delta_o t)a_2$,
where
$\Delta_1 = \omega_1 - \omega$, $\Delta_2 = \omega_2 - \omega$, $\Delta_o = \omega_3 - \omega_2$, μ_{1A} is the matrix element of the dipole moment $\hat{\mu}_{1z}$ of atom A in units of h, and V_2 is the matrix element $\langle 1a|\langle 1b|V_{AB_z}|2a\rangle|0b\rangle/\hbar$.

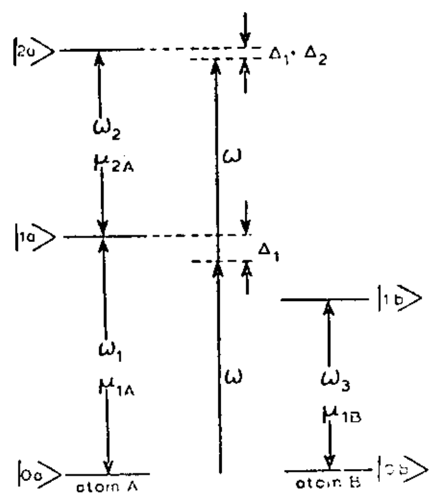

Figure 3. A partial energy-level diagram of the two-photon-one-collision interaction.

This system of four coupled levels can now be reduced to an effective two-level system comprised of the initial and final states. This aim is achievable since the intermediate states of amplitudes a_1 and a_2 are chosen not to interact strongly with the electromagnetic fields nor with the collisional field. In this limit the adiabatic condition applies where the amplitudes a_1 and a_2 adiabatically follow linear combinations of a_o and a_3. More quantiatively we take $|\Delta_1|$, $|\Delta_2|$ and $|\Delta_o|$ large enough that $\Delta_1 \gg \mu_{1A}E_o$, $\Delta_2 \gg \mu_{2A}E_o$, $\Delta_o \gg V_2$, and $|\Delta_1 + \Delta_2| \geq \mu_{2A}E_o$. These conditions also imply that $da_o/dt \ll \Delta_1$, $da_2/dt \ll \Delta_2$,

Δ_o; hence we can integrate the equation for a_1 by parts and keep only the leading terms:

$$a_1 = -\frac{\mu^*_{1A}E_o}{\Delta_1}\exp(-i\Delta_1 t)a_o + \frac{\mu_{2A}E_o}{\Delta_2}\exp(i\Delta_2 t)a_2$$

The resulting expression for a_1 is then substituted in the rest of the equations. Integrating the resulting equation for a_2 by parts in the same fashion, keeping the lowest order terms, and substituting back gives:

$$da_o/dt + i(b_1' E_o^2 + b_2' E_o^4)a_o = ic_o' E_o^2 V_2 a_3 \exp(i\delta t) \qquad (3)$$

and

$$da_3/dt + ib_3 V_2^2 a_3 = ic_3' E_o^2 V_2 a_o \exp(-i\delta t) \qquad (4)$$

where b_i' and c_i' are functions of the various detunings, and $\delta = \Delta_1 + \Delta_2 + \Delta_o$ is the detuning from exact resonance. Eqs. 3-4 are the effective two-level system describing the interaction of the four level system with the external field. It is to be noted that the ground state is stark shifted by the amount $b_1'E_o^2 + b_2'E_o^4$ which is over the time of collision for long-pulses can be taken to be flat. On the other hand the final state is collisionally shifted by an amount $b_3V_2^2$ which depends on the internuclear separation and hence on time since V_2 is a function of R. Figure 4 shows a schematic of the interatomic potential in this model. They are identical to those of single photon radiative collisions[1] except for the two photon coupling $C_o'E_o^2$ replacing the single photon coupling and the presence of the Stark shifts: $b_1' E_o^2 + b_2' E_o^4$ which do not exist in the single photon case.

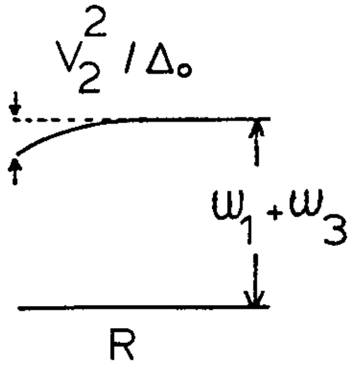

Figure 4. A schematic of the interatomic potential in the case of two-photon-one-collision case.

We now solve Eqs. 3 - 4 in the weak field limit by keeping terms of order E_o^2 only. In this limit the Stark shifts are neglected. Moreover the coupling to a_3 in Eq. 3 is neglected resulting in no depletion of the ground state population. Thus in the weak field limit, these two equations reduce to $a_o = 1$ and

$$da_3/dt - i(V_2^2/\Delta_o)a_3 = i\mu^*_{1A}\mu^*_{2A}E_o^2V_2^*[\Delta_1(\Delta_1+\Delta_2)]^{-1}\exp(-i\delta t) \qquad (5)$$

Integrating Eq. 5 gives:

$$a_3(t) = \frac{\mu^*_{1A}\mu^*_{2A}}{\Delta_1(\Delta_1+\Delta_2)}\exp[i\int_{-\infty}^{t}(V_2^2/\Delta_o)dt] \times \int_{-\infty}^{t} E_o^2 V_2^* \exp[i\int_{-\infty}^{t}(V_2^2/\Delta_o+\delta)dt]dt \qquad (6)$$

The collisional interaction has not yet been specified. Various possibilities can arise. The strongest of these are the dipole-dipole, dipole-quadrupole, and the quadrupole-quadrupole interactions. In this work we take the atoms to undergo dipole-dipole interactions. In the dipole-dipole interaction $V_2^* = \hbar\mu_{2A}\mu_{1B}/R^3$ where $R^2(t) = \rho^2 + V^2t^2$, ρ is the impact parameter and V is the relative speed of the atoms. When E_o changes very little over the time of collision Eq. (6) gives:

$$|a_3(\infty)|^2 = 4\alpha^2E_o^4 \left|\int_o^\infty R^{-3}(t)\cos[\int_o^t (CR^{-6}(t) + \delta)dt]dt\right|^2 \qquad (7)$$

where $\alpha^2 = \hbar^2\mu_{1A}^2\mu_{2A}^4\mu_{1B}^2[\Delta_1(\Delta_1+\Delta_2)]^{-2}$ and $C = \hbar^2\mu_{2A}^2\mu_{1B}^2/\Delta_o$.

We now discuss the line shape of the process. The absorption cross section σ, is calculated from the integration of $|a_3(\infty)|^2$ over the impact parameters. A thermal average of the cross section, $\bar{\sigma}$, then yields an absorption rate. For large C or C′, all impact parameters can be integrated over because the frequency shift becomes large for R values $\lesssim$ 15 Å and there is no change in a_3 at R values where overlap is important and deviations from a straight line trajectory occur. In fact a universal line shape exists for the two-photon-one collision case in analogy with the one photon-one collision case.[5] This line shape is given by the thermally averaged cross section,

$$\bar{\sigma} = \alpha^2E_o^4|C|^{-2/3}(2KT/\mu)^{-4/5}\, J(x) \qquad (8)$$

where T is the absolute temperature, μ is the reduced mass, $x = G|C|^{1/5}(2KT/\mu)^{-3/5}\delta$, G is the sign of C and J is a function which essentially gives the line shape. The line shape is asymmetric with an extended red or blue tail corresponding to G = +1 and G = -1 respectively.

An estimate of the cross section at the peak of the lineshape can be arrived at analytically from Eq 7. Taking $\delta = 0$, and noting that the dephasing $\int CR^{-6}dt$ kills all the contribution coming for small impact parameters, we get

$$\left|a_3(\infty)\right|^2 = 4\alpha^2E_o^4\left|\int_o^\infty R^{-3}dt\right|^2 \quad \text{or} \quad \left|a_3(\infty)\right|^2 = \frac{4\alpha^2E_o^4}{v^2\rho^4} \tag{9}$$

Integrating over the impact parameter using a small impact parameter cutoff as a result of the dephasing effect gives:

$$\sigma_1 = \int_{\rho_c}^\infty \left|a_3(\infty)\right|^2\rho d\rho = 8\pi\alpha^2E_o^4\left(V^2\rho_o^2\right)^{-1} \tag{10}$$

where $\rho_o = \left(\frac{3\pi}{8}\frac{1}{V}\frac{c}{\hbar}\right)^{1/5}$ is the Weisskopf radius and V is the relative speed. Numerical estimates of this cross section will be given in Section IV.

III. Two Photon-two Collision Case

Consider now the second case where both atoms interact with the radiation field. The state vector of the system is taken to be of the form (see Fig. 5):

$$\begin{aligned}\psi(t) = &|0a\rangle|0b\rangle\, a_o(t) + |0a\rangle|1b\rangle \exp(i\omega_3t)a_1(t) + |1a\rangle|0b\rangle \exp(i\omega_1t)a_2(t)\\ &+ |2a\rangle|0b\rangle \exp(i(\omega_1+\omega_2)t)a_2'(t) + |1a\rangle|1b\rangle \exp(i(\omega_1+\omega_3)t)a_3(t)\ .\end{aligned} \tag{11}$$

In the process, the initial state $|0a\rangle|0b\rangle$ is virtually excited by the electromagnetic field to the state $|0a\rangle|1b\rangle$. A virtual collision then transfers the excitation from $|0a\rangle|1b\rangle$ 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 $|1a\rangle|1b\rangle$ nearly conserves the overall energy for the transition. Thus the time dependent Schrödinger equation gives in the rotating wave approximation:

$da_o/dt = i\mu_{1B}E_o \exp(i\Delta_1't)a_1$, $\quad da_1/dt = i\mu^*_{1B}E_o \exp(-i\Delta_1't)a_o + iV_1 \exp(-i\Delta_o't)a_2$,

$da_2/dt = iV_1^*\exp(i\Delta_o't)a_1 + i\mu_{2A}E_o \exp(i\Delta_2t)a_2'$,
$da_2'/dt = i\mu^*_{2A}E_o \exp(-i\Delta_2t)a_2 + iV_2 \exp(-i\Delta_ot)\ a_3$,
$da_3/dt = iV_2^* \exp(i\Delta_ot)a_2'$,

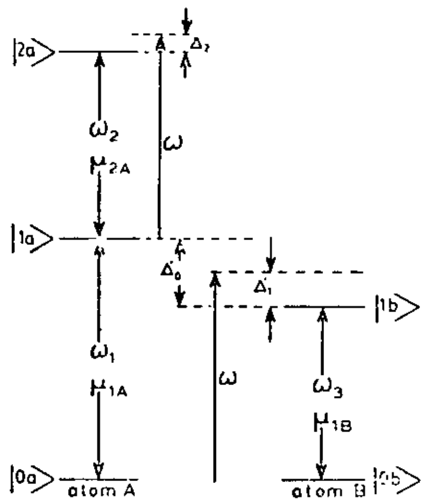

Figure 5. A partial energy-level diagram of the two-photon-two-collision interaction.

where $\Delta_1' = \omega_3 - \omega$, $\Delta_o' = \omega_3 - \omega_1$, $\Delta_2 = \omega_2 - \omega$, $\Delta_o = \omega_2 - \omega_3$, $\mu_{1B} = \langle 0b|\mu_{B_z}|1b\rangle/\hbar$, $\mu_{2A} = \langle 1a|\mu_{A_z}|2a\rangle/\hbar$, $V_1 = \langle 0b|\langle 1a|V_{AB_z}|0a\rangle|1b\rangle/\hbar$ $V_2 = \langle 1a|\langle 1b|V_{AB_z}|0b\rangle|2a\rangle/\hbar$. This system of five coupled levels can now be reduced to an effective two level system comprised of the initial and final states. The adiabatic approximation we used above in reducing the previous case to a two level system will be used. For this purpose we take $\mu^*_{1B}E_o << |\Delta_1'|$, $V_1 << |\Delta_o'|$, $\mu_{2A}E_o << |\Delta_2|$, $V_2 << |\Delta_o|$, $|\Delta_1' - \Delta_o'| = |\Delta_1| \geq V_1$ and eliminate a_1, a_2, and a_2' sequentially by integrating their equations by parts and keeping the lowest order contribution. The resulting equations have the form

$$da_o/dt + iS_1a_o = C_oE_o^2V_1V_2 \exp(i\delta t)a_3 \tag{12}$$

$$da_3/dt + iS_2a_3 = C_4E_o^2V_1^*V_2^* \exp(-i\delta t)a_o \tag{13}$$

where $S_1 = b_1E_o^2 + b_2E_o^2V_1^2 + b_3E_o^4V_1^2$, $S_2 = b_4V_2^2, \delta = \Delta_1+\Delta_2-\Delta_0-\Delta_0'$ is the detuning from exact resonance and the coefficients b_i and c_i depend on the various detunings and the dipole moment matrix elements.

As in the previous case, this effective two state system is similar to the single photon process except that an effective two photon coupling replaces the single photon coupling and an effective two collision coupling replaces the single collision coupling, and except for some additional shifts. The ground state has additional shifts which are the ordinary ac Stark shifts ($b_1E_o^2$) and new intensity induced collisional shifts ($b_2E_o^2V_1^2 + b_3E_o^4V_1^4$). This intensity induced collisional shift is absent in the two photon-one collision case. It arises here from a coherent nonresonant interaction of the field followed by a coherent nonresonant collisional interaction. The final state on the other hand is only collisionally shifted in this model by an amount $b_4V_2^2$. These induced shifts can be used to control the overall shift between the initial and final states making the lineshape highly dependent on the intensity. In fact, one can conceive of situations where most of the shift cancels out in this case. In this paper we will consider the weak field case to explain the effects of these shifts, and leave the strong field case for a later study.

In the weak field limit we are interested in a solution with no depletion in a_o and to first order in E_o^2. Thus the coupling of a_o to a_3 in Eq. 12 will be neglected. Although the coupling of a_3 to a_o in Eq. 13 is sufficient to give a solution to order E_o^2, one cannot neglect completely the field dependence in the phase shift despite the fact if kept it will enter to all orders. We keep the shift proportional to $E_o^2 V_1^2$ in Eqs. (12)-(13) since even in the weak field limit this shift may be of the same order as V_2^2/Δ_0. Thus in the weak field limit, the process is described by the following equations:

Figure 6 shows a schematic of the interatomic potential

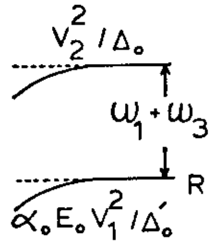

Figure 6. A schematic of the interatomic potential in the case of two-photon-two-collision case.

$$da_o/dt + i\alpha_o E_o^2(V_1^2/\Delta_o')a_o = 0 \tag{14}$$

$$da_3/dt + i(V_2^2/\Delta_o)a_3 = i\alpha_4 E_o^2 V_1^* V_2^* e^{-i\delta t} a_o \tag{15}$$

where $\alpha_o = \mu_{1B}^2[\Delta_1'(\Delta_o'-\Delta_1')]^{-1}$; $\alpha_4 = \mu_{1B}^*\mu_{2A}[\Delta_1'(\Delta_0'-\Delta_1')(\Delta_1'+\Delta_2-\Delta_0')]^{-1}$ $\Delta_0'-\Delta_1' = \omega-\omega_1$, and $\Delta_1'+\Delta_2-\Delta_0' = \omega_1+\omega_2-2\omega$.

Integrating Eq. 14 with the assumption that E_o changes very little during the interaction gives:

$$a_o(t) = \exp[-\alpha_o E_o^2 \int_{-\infty}^{t}(V_1^2/\Delta_o')dt] . \tag{16}$$

Substituting this expression in Eq. 15 and integrating with the same assumption on E_o gives:

$$|a_3(\infty)|^2 = 4\alpha_4^2 E_o^4 \,|\int_o^{\infty} V_1^* V_2^* \,\mathrm{Cos}S\, dt|^2 \tag{17}$$

$$S = \int_{-\infty}^{t} (\frac{V_2^2}{\Delta_o} - \alpha_o E_o^2 \frac{V_1^2}{\Delta_o'} - \delta)dt. \tag{18}$$

In the dipole-dipole interaction, the interactions V_1^* and V_2^* are: $V_1^* = \hbar\mu_{1A}^*\mu_{1B}^*/R^3$ and $V_2^* = \hbar\mu_{2A}^*\mu_{1B}^*/R^3$, and the phase difference S and the transition probability become

$$S = \int_o^t (C'R^{-6}-\delta)dt, \qquad C' = \hbar^2\mu_{2A}^2\mu_{1B}^2\Delta_0^{-1} - \hbar^2\alpha_o E_o^2\mu_{1A}^2\Delta_0'^{-1} \quad \text{and} \tag{19}$$

$$|a_3(\infty)|^2 = 4|\alpha'|^2\, E_o^4|\int_o^{\infty} R^{-6}\mathrm{Cos}S\, dt|^2 \tag{20}$$

where $\alpha' = \hbar^2\mu_{1B}^3\mu_{2A}^2\mu_{1A}/[\Delta_1'(\Delta_0'-\Delta_1')(\Delta_1'+\Delta_2-\Delta_0')] = \hbar(\mu_{1B}^2/\Delta_1')\alpha$.

IV. The Phase Resonance

The phase of the final scattering state relative to the initial scattering state is given in Eq. 18. The phase difference at $R = \infty$ is δt which is controlled by the frequency of the laser excitation. When the collisional couplings V_1 and V_2 are of the same type, that is both are due to dipole-dipole, or to dipole-quadrupole, etc., then both will have the same dependence on the internuclear separation, and hence have the same time dependence (apart from an additional time dependence as a result of the laser field envelope). By choosing the detunings in S appropriately, the intensity induced shift can be chosen to have the opposite sign of the pure collisional shift hence allowing for possible cancellation. Finally, for laser pulses which change very little during the collision (i.e. long pulses compared to the time of collision) complete cancellation of the collisional shift can be achieved by choosing the appropriate laser intensity, thus achieving phase resonance for all internuclear separations and hence all times.

Let us consider the dipole-dipole interaction for both V_1 and V_2. In this case the phase difference is given in Eq. 19 and the transition probability is given in Eq. 20. The situation where C' is very small suggests a large coupling coefficient in the absence of any dephasing effect for all internuclear separations $R \geq 4Å$. This could lead to extremely large cross sections for the process. Moreover, because of the absence of the shift, the line shape is expected to be symmetric. 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. (20) by taking $C' = 0$ and $\delta = 0$. In this case $|a_3(\infty)|^2 = 1.5\,\pi\alpha'^2 E_0^4/(\rho^{10}V^2)$. A lower limit on the estimate can be found by calculating the contribution from impact parameters where orbiting is not important; that is

$$\sigma_2 > \int_{\rho_c}^{\infty} 2\pi\ \rho d\rho\,|a_3(\infty)|^2 = 3\pi^2\alpha'^2 E_0^4 (V\rho_c^4)^{-2}. \tag{21}$$

The enhancement in the cross section as a result of the phase resonance can now be compared to the two photon-one collision case which has no phase resonance. Dividing Eq. 21 by Eq. 10 gives

$$\frac{\sigma_2}{\sigma_1} > \hbar^2 \mu_{1B}^4 \rho_o^2 (\Delta_1'^2 \rho_c^8)^{-1}\ .$$

The above expressions for $|a_3(\infty)|^2$ were numerically integrated using cautious adaptive Romberg extrapolation. Since it is necessary to integrate over time and square the result before integrating over impact parameter, two versions of a one-dimensional algorithm working though common were employed. The computer codes were part of a library leased from IMSL Inc. No attempt was made to average over a thermal velocity distribution as this would have greatly increased cost and would not have altered the basic physics. Previous workers analyzing a one-photon radiative collision in this approximation have shown that the maximum cross section is underestimated by about 50%.

In the numerical calculations we took the following numerical values: $\mu_{1A} = 1.25$ a.u., $\mu_{2A} = .25$ a.u., $\mu_{1B} = 5$ a.u., $\Delta_o' = -6000\ \text{cm}^{-1}$, $\Delta_1 = 5000\ \text{cm}^{-1}$, $\Delta_2 = -9000\ \text{cm}^{-1}$

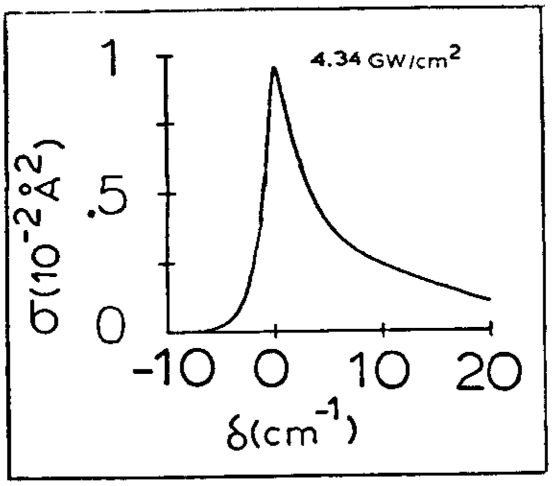

Figure 7. The absolute line shape of the two-photon-one collision case taken at laser intensity $I_o = 4.3\ \text{GW/cm}^2$.

and $(\mu_{2A}/\mu_{1A})_2^2\ \Delta_o'/\Delta_o = .03$. With these numerical values, the quantity C' goes to zero at laser intensity $I_o = 4.337\ \text{GW/cm}^2$.

We start by describing the results of the two-photon-one collision case.[7] The intensity dependence of the response enters only through the factor E_o^4 which gives a quadratic dependence on the intensity. This factor results from the two-photon nature of the process. The intensity normalized line shape $\sigma(\delta)/I^2$ is shown in Fig. 7. The peak of the response occurs at $\delta=0$ that is when the atoms are at infinite internuclear separations.[3-6]

We now describe the results of the two-photon-two collision case.[7] In this case the intensity dependence is far more involved. Figures 8a-8e give the lineshape of the two collision cases at laser intensities 1.02, 1.87, 2.68, 4.34, $8.84\times10^9\ \text{w/cm}^2$ respectively. These figures show that as the intensity rises from $1.02\times10^9\ \text{w/cm}^2$ the asymmetry on the red wing becomes less pronounced. At intensity $I_o = 4.34\times10^9\ \text{w/cm}^2$ the lineshape becomes completely symmetric indicating what we call intensity induced symmetry (phase resonance). As the intensity rises to a value above the intensity that produces the phase resonance, the lineshape becomes again asymmetric, developing a blue wing.

It is to be noted that the lineshape exhibits a line shift as the intensity of the radiation is varied. The shift of the peak of the response is plotted as a function of the intensity in Fig. 9 for values around the value I_o (at phase resonance). The shift is found to be linear at small intensities, and also linear in the neighborhood of I_o. At intensities larger than I_o, the shift becomes a nonlinear function of the intensity. Only at phase resonance the response peaks at $\delta=0$, that is it occurs when the atoms are at infinite internuclear separations (R_∞). At intensities different from I_o, the resonance occurs at $\delta\neq0$, that is occurs at finite internuclear separations.

The cross section of the two-collision case was also studied as a function of the intensity of the laser. The nonnormalized cross section is shown in Fig. 10. At higher intensities the cross section continues to rise, while at lower intensities, the cross section vanishes. Figure 11 gives the intensity normalized peak of the cross section σ_o/I^2 as a function of the intensity in the neighborhood of I_o. It shows that the cross section exhibits the phase resonance. Below $I = 1.3\times10^7\ \text{w/cm}^2$ the normalized cross section approaches a constant value as the intensity decreases, while above $I = 10^9\ \text{w/cm}^2$, the cross section

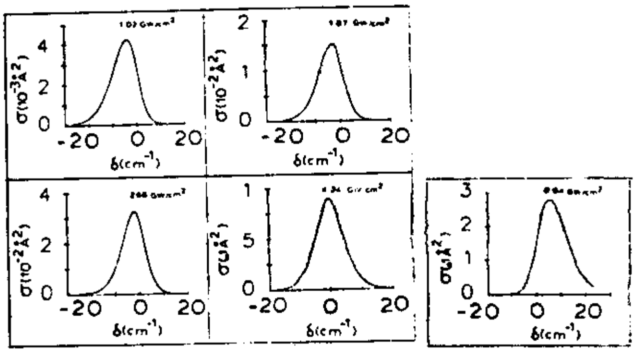

Figure 8. The absolute line shape of the two-collision interaction as a function of the intensity.

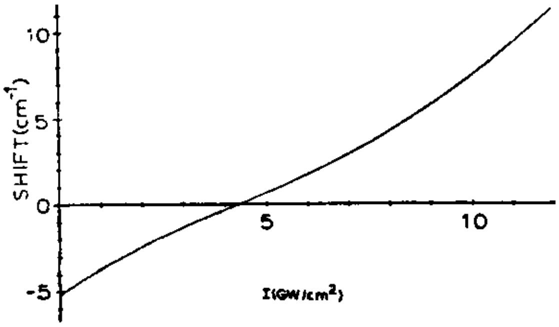

Figure 9. The shift of the peak of the cross section of two-collision interaction as a function of the intensity.

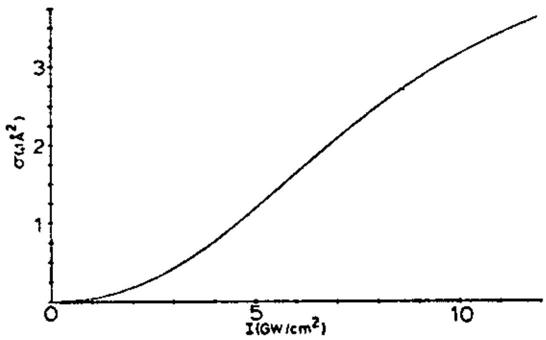

Figure 10. The peak of the cross section of the two collision case as a function of the intensity.

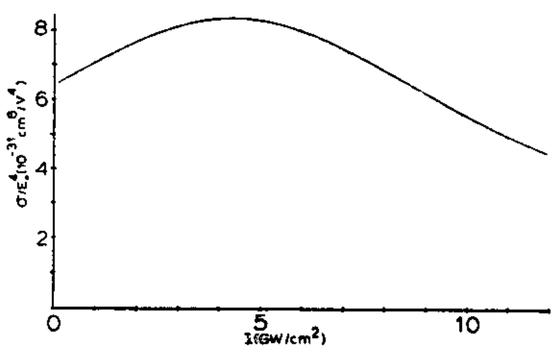

Figure 11. The intensity normalized peak of the cross section of the two collision interaction as a function of the intensity.

continues to decrease as the intensity increases. Finally Fig. 12 gives the ratio of the peak of the cross section of the two-collision case at phase resonance to that of the one-collision case as a function of the minimum impact parameter. The figure shows the enhancement resulting from the interaction at small impact parameter due to the phase resonance.

V. Two-atom Coherence

This phenomenon can be viewed from the point of view of what we call 'two-atom' coherence. It is known that the coherence in single or multiphoton interactions of coherent radiation with isolated atoms is destroyed by collisions with other atoms. This study shows the existence of a new coherent effect in the two-photon interaction of coherent radiation with atoms undergoing binary collisions: intensity induced 'two-atom' coherence. In this effect, the dephasing effects in the 'two-atom' system caused by their collisional interaction are eliminated, thus allowing the electromagnetic field to interact coherently with the system over a wide range of interatomic distances.

The present effect occurs only when both partners of the system are driven by the field (Fig. 2). The nature of the phenomenon lies in the fact that the system in Fig. 2 undergoes energy nonconserving sequencies of radiative and collisional interactions which result in the familiar a.c. Stark shift and collisional shift and in a new mixed a.c. Stark-collisional shift. Whereas the familiar collisional shift is responsible for the dephasing of the coherence, the new mixed shift can be used to eliminate this dephasing effect for a wide range of internuclear distances (phase resonance) if the intensity of the radiation and other detunings are chosen appropriately.

When phase resonance is achieved, the transition frequency of the system which is ordinarily dependent on the internuclear distance relevant to the process becomes constant (parallel potential curves) and consequently the 'two-atom' system may then interact coherently with the electromagnetic field without interruption (see Fig. 13). It is found that such coherent interaction will result in the excitation of the

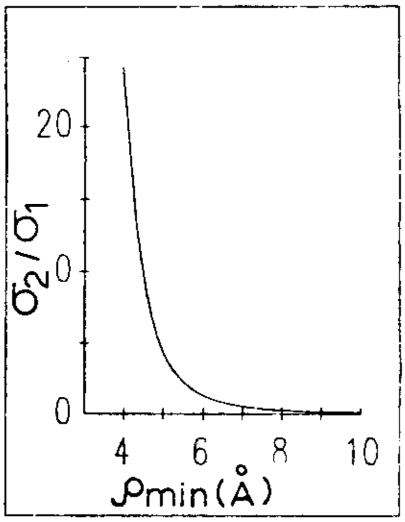

Figure 12. The ratio of the peak of the cross section of two-collision case to that of the one-collision case at phase resonance as a function of the minimum impact parameter ρ_m.

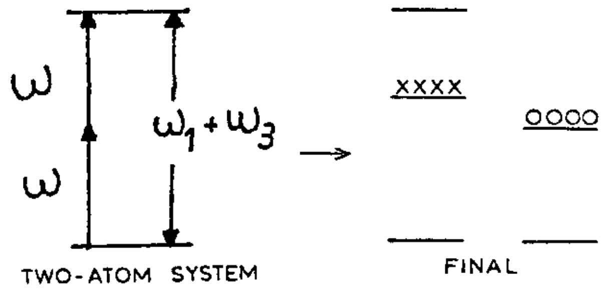

Figure 13. A schematic of the "two-atom" system.

relevant transitions of the individual atoms of the 'two-atom' system at frequencies ω_1 and ω_3 if the frequency of the excitation ω is in near resonance with half the frequency of the 'two-atom' system $(\omega_1 + \omega_3)/2$. Our results also indicate that the coherent absorption by the coherent 'two-atom' system will dominate the absorption by the incoherent 'two-atom' system of Fig. 1.

This work was supported by NSF Grant NSF PHY 81-09305.

REFERENCES

1. L. I. Gudzenko and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. 62, 1686 (1972) [translation: Sov. Phys. JETP 35, 877 (1972)]; S. E. Harris, R. W. Falcone, W. R. Green, D. B. Lidow, J. C. White, and J. F. Young, in Tunable Lasers and Applications, S. Mooradian, T. Jaeger, and P. Stoketh, eds. (Springer-Verlag, New York, 1976), p. 193.

2. S. E. Harris and D. B. Lidow, Phys. Rev. Lett. 33, 674 (1974); 34, 1972 (E) (1975).

3. J. C. White, Opt. Lett. 6, 242 (1981)

4. M. H. Nayfeh and G. B. Hillard, Phys. Rev. A 24, 3, 1409 (1981)

5. M. H. Nayfeh and D. B. Geohegan, Phys. Rev. A (in press).

6. M. H. Nayfeh, G. B. Hillard, and D. B. Geohegan, in Photon-Assisted Collisions and Related Topics, N. Rahman and C. Guidotti eds., Harwood Academic Publishers (London & New York), 1982.

7. M. H. Nayfeh and G. B. Hillard, to be published.

8. S. E. Harris and J. C. White, IEEE J. Quant. Electron. QE-13, 972 (1977).

9. M. G. Payne, C. W. Choi and M. H. Nayfeh, International Conference on Multiproton Processes, Rochester, New York, June 1977 (unpublished).