Model studies of collective atomic excitations by intense laser fields

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A quantum version of the Drude-Lorentz model of the atom is used to study abnormally large energy transfers found in recent ionization experiments by strong laser fields. It is argued that collective excitations greatly enhanced by parametric resonance may lead to an exponential growth of the atomic energy with time. It is shown that the repulsion between the electrons plays an essential role, reducing the frequency at which parametric resonance occurs.

I. INTRODUCTION

Recent experiments\(^1\)\(^-\)\(^6\) on multiphoton ionization of heavy atoms by intense laser fields (up to \(10^{17}\) W/cm\(^2\)) seem to indicate that many-electron response of the atom plays a role in its interaction with high-intensity radiation. Collective, many-electron excitations are probably responsible for the abnormal abundance of ions with high-charge multiplicity. These experiments also show a sensitive dependence of the energy transferred from the light beam to the atom on the atomic number \(Z\).

The theory of many-electron excitations in atoms developed over the last 20 years by many authors,\(^7\) although very successful in high-frequency spectroscopy as exemplified by the prediction of the giant-dipole resonance, is restricted to the weak-field (linear-response) regime. The exact theory of many-electron excitations induced by intense laser fields requires solving the many-body Schrödinger equation without resorting to perturbation theory and has, therefore, not lead to any quantitative results. Under these circumstances, simple models were proposed to explain the experimental data. Crance\(^8\) used a statistical model to calculate, by pure combinatorics, relative abundances of ions with different charge multiplicities and the energy spectra of photoelectrons. She obtained a reasonably good agreement with experiments for laser intensities up to \(10^{14}\) W/cm\(^2\). In her model the dynamics of the laser-atom interaction is not specified and electrons are treated as independent, indistinguishable particles. The same purely statistical reasoning permeates the recent work by Geltman.\(^9\) Using the Hartree model of the atom, he expressed all multiple ionization probabilities by the probabilities to eject one electron from any given shell and he next fitted his formulas to the experimental data.\(^10\) In the papers by Crance and Geltman only the outer-shell electrons are taken into account. In view of the preliminary character of the experimental data, it is difficult to judge the success of the statistical approach and to decide whether the correlations between the electrons play a significant role.

In turn, Rhodes\(^2\)\(^,\)\(^3\) suggested that inelastic collisions between the outer and the inner atomic shells may be responsible for the observed inner-shell excitations induced by high-intensity lasers.

Still, the exact mechanism of many-electron, multiphoton atomic excitations remains unknown. The interesting questions are: Can collective modes of the electron cloud be excited by intense laser fields? Do these collective excitations play a significant role in increasing the rate of the energy transfer to the atom? What is the role of the mutual interaction between the electrons?

The purpose of the present paper is to study the dynamics of the interaction of a many-electron atom with a strong laser field using a simplified, exactly soluble model. Our model has all the features of the realistic situation (electron-nucleus attraction as well as electron-electron repulsion, Fermi-Dirac statistics, and the minimal electromagnetic coupling to radiation) with the only exception that all Coulombic forces have been replaced by harmonic forces. Thus, our model may be viewed as a quantum version of the classic Drude-Lorentz model of the atom.\(^1\)\(^1\)

The main result of our analysis is the discovery of parametric resonances in collective excitations that lead, under some conditions, to a greatly enhanced energy transfer from the wave field to the atom. The location of the resonance regions in the frequency-amplitude plane of the electromagnetic wave depends in a crucial way on the electron-electron repulsion.

II. MODEL OF THE ATOM

Our model is most conveniently formulated in the language of second quantization, which is best suited for handling collective phenomena. In this description, the basic quantities are the operators \(\psi_\sigma(r,t)\) and \(\psi_\sigma^\dagger(r,t)\) that annihilate and create an electron at the point \((r,t)\) with the spin projection \(\sigma (\sigma = \frac{1}{2}, -\frac{1}{2})\). Since the electrons obey the Fermi-Dirac statistics, their field operators obey the anticommutation relations

\[
[\psi_\sigma(r,t),\psi_\sigma^\dagger(r',t)]_+ = \delta_{rr'}\delta(t-t') .
\]

The field operator \(\psi_\sigma(r,t)\) obeys the Heisenberg equation of motion \(\hat{\mathbf{H}} = 1\),

\[
\frac{\partial}{\partial t}\psi_\sigma(r,t) = [\psi_\sigma(r,t),\hat{H}_A]_+ ,
\]

where the atomic Hamiltonian \(\hat{H}_A\) is chosen in the following form:
where $m$ is the electron mass and

$$p = -i \nabla .$$  \hspace{1cm} (4)$$

The first part of this Hamiltonian describes the motion of electrons in an external potential of the isotropic harmonic oscillator which simulates the Coulomb potential of the nucleus. The second part describes the mutual interaction between the electrons. Here also, the Coulombic forces have been replaced by (repulsive) harmonic forces.

Following the analysis of a closely related model, described in an earlier publication, we introduce the following set of collective operators:

$$N = \int \psi_r^\dagger(r,t) \psi_r(r,t) ,$$  \hspace{1cm} (5a)$$

$$\mathbf{R}(t) = \int \psi_r^\dagger(r,t) \mathbf{r} \psi_r(r,t) ,$$  \hspace{1cm} (5b)$$

$$\mathbf{P}(t) = \int \psi_r^\dagger(r,t) \mathbf{p} \psi_r(r,t) ,$$  \hspace{1cm} (5c)$$

$$U_\mu(t) = \frac{1}{2} \int \psi_r^\dagger(r,t) \mathbf{P} \mathbf{P} \psi_r(r,t) - \frac{1}{2N} R_i(t) R_i(t) ,$$  \hspace{1cm} (5d)$$

$$T_\mu(t) = \frac{1}{2} \int \psi_r^\dagger(r,t) \mathbf{P} \mathbf{P} \psi_r(r,t) - \frac{1}{2N} P_i(t) P_i(t) ,$$  \hspace{1cm} (5e)$$

$$W_\mu(t) = \frac{1}{4} \int \psi_r^\dagger(r,t) [ R_i(t) P_j(t) + P_j(t) R_i(t) - 2 R_j(t) P_i(t)] ,$$  \hspace{1cm} (5f)$$

where the integral sign denotes both the integration over $r$ and the summation over $\sigma$.

These operators have the following physical interpretation. The scalar operator $N$ is the operator of the total number of electrons in the atom and is a constant of motion. The vector operators $\mathbf{R}(t)$ and $\mathbf{P}(t)$ describe the position of the center of mass and the total momentum of the atomic electrons. The remaining (tensor) operators describe the quadrupole degrees of freedom of the electron cloud. The Hamiltonian $H_A$ expressed in terms of these collective operators has the form

$$H_A = \frac{1}{2} \sum_{\sigma} \int d^3r \psi_\sigma^\dagger(r,t) \left[ \mathbf{p} - \frac{e \mathbf{a} \mathbf{e}}{c} \sin(\omega_L t - \mathbf{k} \cdot \mathbf{r}) \right] - \frac{1}{2} \frac{e \omega_L m c^2}{\omega_L} \sin^2(\omega_L t) ,$$  \hspace{1cm} (10)$$

where $\omega_L$, $\mathbf{k}$, and $\mathbf{e}$ are the frequency, the wave vector, and the polarization vector of the wave.

We have neglected the coupling of the electrons to the wave through their magnetic moments since it is of the order of higher relativistic corrections.

In the dipole approximation, the Hamiltonian $H$ expressed in terms of collective operators has the form

$$H = H_A - \frac{e \omega_L m c}{2mc^2} N \sin^2(\omega_L t) .$$  \hspace{1cm} (11)$$

The last term does not influence the dynamics of the system, because $N$ is a constant of motion. The second term is a source of the driving force in the equation for the component of the $\mathbf{R}$ operator in the direction of the wave polarization. We shall fix the coordinate system by assuming that the wave propagates in the $z$ direction and is polarized in the $y$ direction. Using the commutation relations (9), we obtain the following Heisenberg equations of motion:

III. ATOM-WAVE INTERACTION
IN THE DIPOLE APPROXIMATION

We will now study the interaction of our model atom with the laser light, treated as an external field. This field will be represented by a monochromatic, linearly polarized, plane electromagnetic wave. Assuming minimal electromagnetic coupling, we obtain the following Hamiltonian:
\[ \dot{R}_y = \frac{e}{m} a N \sin(\omega_L t), \quad \text{(12a)} \]
\[ \dot{P}_y = -m \Omega^2 R_y, \quad \text{(12b)} \]

From Eqs. (12) we get the following equation for \( R_y \):
\[ \ddot{R}_y + \Omega^2 R_y = -\frac{e \mathcal{E}}{m} N \cos(\omega_L t), \quad \text{(13)} \]

where \( \mathcal{E} \),
\[ \mathcal{E} = \frac{a \omega_L}{c}, \quad \text{(14)} \]
is the amplitude of the electric field of the wave.

The solutions of this equation exhibit the well-known giant dipole resonance. At the resonance frequency \( \omega_L = \Omega \) the energy of the dipole grows as \( t^2 \). Off the resonance, the energy of the atom oscillates with a constant amplitude. In the optical and in the uv region we shall always be in this purely oscillatory regime. The interaction of an atom with a wave taken in the dipole approximation leaves the other degrees of freedom unaffected.

IV. QUADRUPOLE APPROXIMATION

In order to study the excitations of the internal degrees of freedom characterized by the operators \( U_{ij} \), \( T_{ij} \), and \( W_{ij} \), we shall consider the quadrupole approximation to the interaction of our model atom with the wave. The most interesting quantity to be determined is the energy transferred from the wave to the atom through these degrees of freedom.

The total Hamiltonian of the system in the quadrupole approximation has the form

\[ H = H_A + H_I, \]

where

\[ H_I = -\frac{ea}{mc} \mathbf{P} \sin(\omega_L t) - \frac{e^2 a^2}{2mc^2} \mathbf{k} \cdot \mathbf{R} \sin(2\omega_L t) \]
\[ + \frac{ea}{mc} \frac{(k \cdot R)(e \cdot P)}{N} \cos(\omega_L t) + \frac{e^2 a^2}{2mc^2} \frac{N}{N} \sin^2(\omega_L t) \]
\[ + \frac{2ea}{mc} \mathbf{k} \cdot \mathbf{W} \cdot \mathbf{e} \cos(\omega_L t). \quad \text{(15)} \]

The first three terms in the interaction Hamiltonian \( H_I \) influence only the motion of the atomic dipole, but the last term changes also the evolution of the operators \( U_{ij} \) and \( T_{ij} \), thus changing the energy \( H_Q \) of the quadrupole degrees of freedom. Also, in this case, the evolution of the quadrupole is completely decoupled from the evolution of the dipole, and one can study these two problems separately.

The Heisenberg equations of motion for the dipole operators \( \mathbf{R} \) and \( \mathbf{P} \) are changed in the quadrupole approximation as compared to Eqs. (12). Now, the \( z \) component also (in the direction of the wave propagation) are driven by the wave field, due to the second term in the Hamiltonian \( H_I \). The third term in \( H_I \) couples the \( y \) and the \( z \) components. As a result, the oscillator frequency is modulated by the field and parametric resonance may occur. However, we shall not study this resonance any further, because the frequencies of the electromagnetic wave in the optical or in the uv region are far from the dipole resonance region in real atoms.

We turn now to the study of the evolution of the quadrupole operators. We shall use the following equal-time commutation relations to write the Heisenberg equations of motion. These commutation relations are obtained from the definitions (5) of the \( U_{ij} \), \( T_{ij} \), and \( W_{ij} \) operators and from the anticommutation relation (1) for the field operators \( \psi_\alpha(r,t) \) and \( \psi_\beta^\dagger(r,t) \):

\[ [U_{ij}, T_{kl}] = \frac{1}{2} i (\delta_{ik} W_{jl} + \delta_{il} W_{jk} + \delta_{jk} W_{il} + \delta_{jl} W_{ik}), \quad \text{(16a)} \]
\[ [U_{ij}, W_{kl}] = \frac{1}{2} i (\delta_{lj} U_{ik} + \delta_{lj} U_{jk}) \]
\[ [T_{ij}, W_{kl}] = -\frac{1}{2} i (\delta_{ij} T_{lk} + \delta_{ij} T_{lk}) \]
\[ [W_{ij}, W_{kl}] = \frac{1}{2} i (\delta_{li} W_{kj} - \delta_{lk} W_{ij}). \quad \text{(16d)} \]

The Heisenberg equations of motion for \( U_{ij} \), \( T_{ij} \), and \( W_{ij} \) are governed only by that part of the Hamiltonian which contains the quadrupole operators. We shall write it in the form

\[ H_{xx} + H_{yy} + H_{zz} + 2g(t)W_{xy}, \]

where

\[ H_{xx} = \frac{1}{m}(T_{xx} + \kappa U_{xx}), \quad \text{(17)} \]

etc., and

\[ g(t) = \frac{e \mathcal{E}}{mc} \cos(\omega_L t). \quad \text{(18)} \]

Since in the quadrupole approximation there is no coupling of the atom to the magnetic field of the linearly polarized wave, the part of the energy corresponding to the motion in the \( x \) direction, namely \( H_{xx} \), remains constant. In what follows we shall disregard \( H_{xx} \) and study only the varying part \( E \) of the quadrupole energy,

\[ E = H_{yy} + H_{zz}. \quad \text{(19)} \]

The rate of change of this energy is

\[ \frac{dE}{dt} = \frac{2}{m} g(t)(- T_{xy} + \kappa U_{xy}). \quad \text{(20)} \]

The two components of the quadrupole operators appearing on the right-hand side of this equation are coupled to other components through the following ten equations of motion:
\[ \begin{align*}
\dot{T}_{yy} &= -\frac{\kappa}{m}(W_{yy} + W_{yz}) - g(t)T_{yy}, \\
\dot{U}_{yy} &= -\frac{1}{m}(W_{yy} + W_{yz}) + g(t)U_{yy}, \\
\dot{T}_{yy} &= -\frac{2}{m}\kappa W_{yy}, \\
\dot{U}_{yz} &= -\frac{2}{m}W_{yz}, \\
\dot{W}_{yy} &= \frac{1}{m}(T_{yy} - \kappa U_{yy}), \\
\dot{W}_{yz} &= \frac{1}{m}(T_{yy} - \kappa U_{yy}) + g(t)(W_{zz} - W_{yy}), \\
\dot{W}_{yz} &= \frac{1}{m}(T_{yy} - \kappa U_{yy}) - g(t)W_{yz}, \\
\dot{W}_{yz} &= \frac{1}{m}(T_{yy} - \kappa U_{yy}) + g(t)W_{yy}, \\
\dot{T}_{zz} &= -\frac{2}{m}\kappa W_{zz} - 2g(t)T_{yy}, \\
\dot{U}_{yy} &= \frac{2}{m}W_{zz} + 2g(t)U_{yy}.
\end{align*} \]

(21a) 
(21b) 
(21c) 
(21d) 
(21e) 
(21f) 
(21g) 
(21h) 
(21i) 
(21j)

Fortunately, by choosing proper linear combinations, we may reduce this large set to a number of independent sets of three equations. To this end, we define the dimensionless quantities,

\[ K_\pm = \frac{2}{m\omega}(-T_{yy} + \kappa U_{yy}) \pm (W_{zz} - W_{yy}), \]

(22a) 
\[ L_\pm = \frac{2}{m\omega}(T_{yy} + \kappa U_{yy}) \mp T_{yy} \pm \frac{1}{m}(W_{yy} + W_{yz}), \]

(22b) 
\[ M_\pm = \frac{1}{m}\omega[(T_{yy} - T_{yy}) + \kappa(U_{yy} - U_{zz})] \pm (W_{yy} + W_{yz}). \]

(22c)

The equations of motion for the operators \( K_+, L_+, \) and \( M_+ \) have a similar form to equations for \( K_-, L_-, \) and \( M_- \) we shall write them together in a compact form,

\[ \begin{align*}
\dot{K}_\pm &= \pm \omega M_\mp + g(t)\dot{L}_\pm, \\
\dot{L}_\pm &= \pm \omega K_\mp, \\
\dot{M}_\pm &= \mp \omega K_\pm + g(t)\dot{K}_\pm.
\end{align*} \]

(23a) 
(23b) 
(23c)

This set of three equations can also be replaced by one third-order equation,

\[ L_\pm \pm \omega L_\pm \pm 2g(t)L_\pm = 0. \]

(24)

Equations (23) have a constant of motion,

\[ K_\pm^2 = L_\pm(L_\pm + 2M_\pm). \]

The energy \( E \) and its rate of change can be expressed as follows:

\[ E = \omega(L_+ + M_+ + L_- + M_-)/2, \]

(25) 
\[ \frac{dE}{dt} = \frac{\omega}{2}g(t)(K_+ + K_-). \]

(26)

Equations (23) together with (25) and (26) will be used as a basic tool in our analysis. We shall show in Sec. V that they exhibit parametric resonance near the frequency \( \omega \).

V. PARAMETRIC RESONANCE

In the absence of the field, when \( g(t) = 0 \), all quadrupole variables of the atom oscillate with the frequency \( \omega \). In this case, Eqs. (23) for \( K_\pm \) and \( M_\pm \) describe simply a one-dimensional oscillator. When \( g(t) \neq 0 \), the evolution equations (21), (23), or (24) involve a parametric coupling between the atom and the field. This parametric coupling introduces field-dependent modulations of the atomic quadrupole oscillations with the amplitude and period of modulation dependent on the field intensity and the detuning. It is of interest to locate the regions of parametric resonance where the oscillatory modulations turn into an exponential growth. We expect that in these regions the energy transferred to the atom will increase dramatically. The location of these regions in the frequency-amplitude plane can be found analytically or numerically.

Since we cannot solve Eq. (24) exactly, we shall resort to well-established methods of studying the parametric resonance which employ the existence of two time scales. These methods eliminate the divergencies occurring in the straightforward perturbation theory that are due to the appearance of secular terms.

We expect parametric resonance to occur when the laser frequency \( \omega_L \) is close to the frequency \( \omega \) of the atomic quadrupole oscillations. In addition to the time scale determined by this resonant frequency, there is another, much larger time scale associated with an analog of the Rabi frequency in our problem, namely by

\[ \omega_R = \frac{eB}{mc}. \]

(27)

The ratio \( \delta \) of these two frequencies,

\[ \delta = \frac{\omega_R}{\omega_L}, \]

(28)

will serve as a small parameter in our perturbative analysis of the instability regions. We shall use the Whittaker method, which employs the ansatz,

\[ L(t) = e^{\gamma t}l(t), \]

(29)

where \( l(t) \) is a rapidly changing part of the solution, while \( \exp(\gamma t) \) is a slowly changing envelope. The idea of the Whittaker method is to absorb all secular terms into the slowly varying part of the solution. This can be done in every order of perturbation with respect to \( \delta \). In the first order, we obtain three linearly independent solutions for \( L(t) \), characterized by the following three values of \( \gamma \):

\[ \gamma_{1,2} = \pm \frac{1}{2} \left[ \omega_R^2 - 4(\omega_L - \omega)^2 \right]^{1/2}, \]

(30)

\[ \gamma_3 = 0. \]

The first solution grows exponentially for the detunings \( \Delta = \omega_L - \omega \) satisfying the condition:

\[ |\Delta| < \omega_R/2. \]

(31)

This condition gives the boundary of the instability region.
in the frequency-amplitude plane in the lowest order of perturbation theory.

We shall illustrate different types of behavior near and at the resonance by numerically integrating Eqs. (23) in the strong-field regime \((\omega_R/\omega_L=0.1)\). For simplicity, we have assumed the initial shape of the atom to be spherically symmetric \([K_+(0)=0=M_+(0)\) and \(L_+(0)=L_-(0)\). In Fig. 1 we show the time dependence of that part of the quadrupole energy which is affected by the light wave.

Far from the resonance region, the energy oscillates rather mildly; few photons are exchanged between the wave and the atom. As we approach the region of parametric resonance, both the period and the amplitude of modulations increase dramatically. Finally, in the resonance region the energy transferred to the atom grows exponentially with time. As seen in Fig. 1, after only 15 wave periods the atom absorbs as many as 25 photons.

VI. DISCUSSION

Since experimental evidence seems to suggest that collective behavior of electrons is responsible for abnormally high-energy transfers, we have chosen a model in which the dynamics of the atom is fully collectivized. By substituting the harmonic forces for the Coulombic forces we have effectively replaced the atom by its dipole and quadrupole moments. In realistic atoms all the multipole moments will be dynamically relevant, but we expect the lowest two moments to be most important for the coupling to the electromagnetic wave in the optical or in the UV region.

One should observe the essential role of the repulsion between the electrons. In our model it leads to a softening of the characteristic frequency of the quadrupole oscillations. This may lead to the frequency \(\omega\) being much lower than the frequency of the dipole oscillations. Were it not for this softening, we could not argue that the resonance region for the quadrupole oscillations lies at much lower energies than about 100 eV, which is typical for the giant dipole resonance. As seen in Eq. (8), the characteristic quadrupole frequency \(\omega\) results from subtracting two large quantities. In the absence of electronic repulsion, the quadrupole frequency, as expected, would be twice the dipole frequency. However, at this stage we have no way to predict the values of the two free parameters of our model: \(\Omega\) and \(\Omega_L\). This would require either a fit to experimental data when they become available (frequency dependence of the ionization rates) or a derivation of our model from the proper theory with Coulombic forces.

In order to excite the quadrupole degrees of freedom, we had to resort to the quadrupole approximation in the wave-atom coupling. Even though the coupling of the quadrupole oscillations to the wave field is, in some sense, weaker than that of the dipole, the energy transferred to the atom through the quadrupole degrees of freedom can be much higher if it all happens in the region of parametric resonance. Let us also note that the standard arguments for the smallness of the quadrupole coupling, based on the smallness of the atomic size as compared to the wavelength, may lose their validity for the multielectron atom which expands rapidly when "heated" by an intense laser field.

If one views our model more realistically than we had originally intended, two clear predictions can be made. First, the interaction has a resonant character. Therefore, one should try experimentally to locate the resonance regions for different elements. Second, the quadrupole excitations play the crucial role. This fact should show up in the correlations of the ejected electrons characteristic of such excitations.

Note added in proof: The model of the atom with all Coulomb forces replaced by the harmonic forces has been also studied by Moshinsky and his collaborators [M. Moshinsky, O. Novaro, and A. Calles, J. Phys. (Paris), Colloq. 31, C4-125 (1970); M. Moshinsky, N. Mendez, and E. Murow, Ann. Phys. (N.Y.) 163, 1 (1985)], but they explored different aspects of this model.

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2C. K. Rhodes, in Multiphoton Processes, edited by P. Lambro-


10 Geltman's fit for Xe has been recently improved by J. Zakrzewski (unpublished).


13 These methods were developed mostly in connection with the Mathieu equation, which like our Eq. (24) is also an equation with periodic coefficients. For a thorough review of perturbative methods see, for example, A. H. Nayfeh, *Introduction to Perturbative Techniques* (Wiley, New York, 1981), especially Chap. 11.