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TITLE CALCULATION OF HARMONIC RADIATION AND NUCLEAR COUPLING ARISING FROM ATOMS IN STRONG LASER FIELDS

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Calculation of harmonic radiation and nuclear coupling arising from atoms in strong laser fields*

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ABSTRACT

A numerical, time-dependent quantum mechanical model is used to describe the interaction of an isolated ion with an intense applied laser field, including both electron and nuclear degrees of freedom. Calculated results are presented. We find that the model ion radiates in low odd harmonics of the laser frequency, in qualitative agreement with experimental observations. In addition, it radiates strongly in the x-ray region, at frequencies comparable with the electron Rydberg frequency. Such radiation should be possible to observe in future experiments. If it exists, it could provide a basis for a reasonably coherent x-ray source. We find that the probability of induced nuclear excitation is small for higher electric multipoles, although observable probabilities are obtained under appropriate circumstances for L=1.

1. INTRODUCTION

Triggering a gamma-ray laser will evidently require a mechanism for prompt nuclear excitation between neighboring states.[1] One possible such mechanism involves the dynamic electron-nucleus coupling, which would allow electron excitation to be transferred to the nucleus. This subject has been studied thoroughly for muonic atoms, where it is known as the dynamic hyperfine effect. The electronic case involves somewhat different considerations, however. An important complication is a great increase in the number of degrees of freedom. The result is that the problem cannot be solved completely. Significant approximations and simplifications must be made, and experimental tests are required in order to determine whether current theory is capable of a quantitative description.

The purpose of this paper is to present a theoretical laser-electron-nucleus model to describe this dynamic coupling, and to investigate related experimental consequences. The model is based upon solution of the single-particle time-dependent Dirac equation. The electron states are treated (relativistically) by numerical techniques in a finite realistic, self-consistent basis. The principal limitation is that only bound-state processes are included; ionization processes could be included only if the continuum is represented approximately by a discrete spectrum.

2. PHYSICAL PROBLEM

The problem to be addressed concerns an isolated ion in the field of a very intense ($I \geq 10^{16} W/cm^2$) ultraviolet ($\hbar\omega_0 \sim 5eV$) laser. We wish to describe two phenomena: (1) The photon spectrum re-radiated by the ion; and (2) The probability of indirect nuclear excitation induced by the moving electrons. In order to carry out the calculation, we make two main physical assumptions: (1) Initial ionization has occurred, so that a relatively stable ionization state has already been reached; and (2) Explicit e^-e^- correlations can be neglected, so that the system can be described through uninhibited single-particle transitions among specified states.

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3. ELECTRON-PHOTON MODEL

Our model is based upon a Dirac Hamiltonian with the time-dependent dipole approximation for the electron-photon interaction (laser assumed plane-polarized along the z-axis)

$$H(q,t) = \vec{\alpha}\cdot\vec{p}c + \beta m_e c^2 + V_0(r) + \frac{e^2}{a_0^2}\,\vec{E}_0\cdot\vec{r}\,\sin(\omega_0 t)$$

$$= H_0(q) + \frac{e^2}{a_0^2}\,\vec{E}_0\cdot\vec{r}\,\sin(\omega_0 t) \quad , \tag{1}$$

where q stands for all variables but the time t. This time-dependent wave equation is solved numerically in a finite static basis of dimension m

$$\{u_j(q)\} \quad j=1\ldots m \quad , \tag{2}$$

where j stands for $n\kappa\mu$, and the basis states are generated by $H_0(q)$

$$H_0(q)u_j(q) = E_j u_j(q) \quad , \quad E_j < 0 \quad , \tag{3}$$

and $H_0(q)$ $[V_0(r)]$ is obtained from a static self-consistent Dirac-Fock-Slater calculation.

4. GENERAL CALCULATIONAL METHOD

Time-dependent wave equations are usually nontrivial to solve numerically because they are stiff, i.e., many independent frequencies are present in the solution vector. We avoid this problem by an intermediate expansion in a basis of adiabatic eigenstates. This has the advantage of factoring out certain frequencies explicitly, so that the basis itself provides the solution in the limit of an infinitely slowly-varying Hamiltonian. We wish to solve

$$H(q,t)\Psi(q,t) = i\hbar\dot{\Psi}(q,t) \quad . \tag{4}$$

The adiabatic basis is defined by

$$H(q,t)\phi_k(q,t) = \epsilon_k(t)\phi_k(q,t) \quad . \tag{5}$$

where $\epsilon_k(t)$ and $\phi_k(q,t)$ are the adiabatic eigenvalues and eigenfunctions. The solution is expanded

$$\Psi(q,t) = \sum_{k=1}^{m} c_k(t) e^{-i\theta_k(t)} \phi_k(q,t) \quad , \tag{6}$$

which is exact in principle as $m \to \infty$. If we define the phase

$$\hbar\dot{\theta}_k(t) = \epsilon_k(t) \quad , \tag{7}$$

then the exponential factor in Eq. (6) is the time-evolution operator for the adiabatic solution $\phi_k(q,t)$. Inserting Eq. (6) into Eq. (4) and using this definition, we obtain the differential equations obeyed by the coefficients $c_k(t)$:

$$\sum_{k=1}^{m} e^{-i\theta_k(t)} [\dot{c}_k(t)\phi_k(q,t) + c_k(t)\dot{\phi}_k(q,t)] = 0 \quad . \tag{8}$$

Equations (7) and (8) may be expanded to first order as

$$\theta_k(t\pm dt) = \theta_k(t) \pm \frac{dt}{2\hbar} [\epsilon_k(t\pm dt) + \epsilon_k(t)] \tag{9}$$

and

$$c_\ell(t\pm dt) = \sum_{k=1}^{m} e^{\mp i[\theta_\ell(t) - \theta_k(t)]} c_k(t) \langle \phi_\ell(t) | \phi_k(t \mp dt) \rangle \quad . \tag{10}$$

The algorithm defined in Eq. (10) has some interesting properties. In particular, it is <u>exactly</u> unitary, time-reversal invariant, and gauge invariant, independent of the basis size m, the size of dt, or the form adopted to solve Eq. (7). This useful property is not shared by other first-order algorithms, nor by higher-order algorithms we have investigated. The proof involves some simple algebra; it may be motivated by the interpretation of Eq. (10) as defining a sequence of sudden approximations in time-dependent perturbation theory.

5. RELATION TO STATIC BASIS

The adiabatic solutions are found by diagonalizing the Hamiltonian $H(q,t)$ at each time t in the static Dirac basis. They are represented by the matrix expansions

$$\phi_k(q,t) = \sum_{j=1}^{m} a_{kj}(t) u_j(q) \quad , \tag{11}$$

where the hermiticity of H(q,t) allows us to choose real coefficients $a_{kj}(t)$. In terms of these coefficients, the scalar products appearing in Eq. (10) are represented

$$\langle\phi_\ell(t)|\phi_k(t\mp dt)\rangle = \sum_{j=1}^{m} a_{\ell j}(t)a_{kj}(t\mp dt) \quad , \tag{12}$$

and an arbitrary matrix element between two states α and β at times t and t' is

$$\langle\Psi_\beta(t')|Q|\Psi_\alpha(t)\rangle = \sum_{\substack{k=1\\ \ell=1}}^{m} c_k^{*(\beta)}(t')c_\ell^{(\alpha)}(t)\, e^{i[\theta_k^{(\beta)}(t')-\theta_\ell^{(\alpha)}(t)]} \times \sum_{\substack{i=1\\ j=1}}^{m} a_{ki}(t')a_{\ell j}(t)\langle u_i|Q|u_j\rangle \quad . \tag{13}$$

6. RADIATED POWER

The power radiated by an atom described by the wave function $\Psi(t)$ is given in first-order time-dependent perturbation theory in terms of the multipole moments

$$n_{LM}(\omega) = \frac{2}{N\tau_0}\int_0^{N\tau_0} dt\, e^{i\omega t}\langle\Psi(t)|\, r^L\, Y_{LM}^*(\hat{r})\, |\Psi(t)\rangle \quad , \tag{14}$$

where the time integration is carried out for N fundamental periods τ_0. The total power radiated for each multipole LM and frequency ω is

$$P_{LM}(\omega) = \frac{2\pi e^2 c}{[(2L+1)!!]^2}\,\frac{L+1}{L}\,\left(\frac{\omega}{c}\right)^{2L+2}\,|n_{LM}(\omega)|^2 \quad . \tag{15}$$

Because the integration is carried out for a finite time $N\tau_0$, the Fourier components in $\Psi(t)$ will appear as peaks in the power spectrum with characteristic width $1/N\tau_0$.

7. NUCLEAR EXCITATION

We may similarly treat nuclear excitation in first-order time-dependent perturbation theory, as we expect these rates to be small in general. The Coulomb interaction at nuclear coordinate $\vec{R}$ generated by an electron at coordinate $\vec{r}$ is

$$V(\vec{R},\vec{r}) = \frac{-e^2}{|\vec{R}-\vec{r}|} = -\sum_{LM} \frac{4\pi e^2}{2L+1} \frac{r_<^L}{r_>^{L+1}} Y^*_{LM}(\hat{R})\, Y_{LM}(\hat{r}) \quad . \tag{16}$$

The time-dependent expectation value of this operator produces a time-dependent interaction potential at $\vec{R}$:

$$V_{LM}(\vec{R},t) = \langle\Psi(t)| \frac{-4\pi e^2}{2L+1} \frac{r_<^L}{r_>^{L+1}} Y^*_{LM}(\hat{R})\, Y_{LM}(\hat{r}) \,|\Psi(t)\rangle \quad . \tag{17}$$

In terms of this potential, the first-order probability for an induced nuclear transition from state Im to state I'm' is given by

$$W(Im\rightarrow I'm';\omega) = \frac{1}{\hbar^2} \left| \int_0^{t_0} dt\, e^{i\omega t} \langle I'm'|V_{LM}(t)|Im\rangle \right|^2 \quad , \tag{18}$$

where t_0 is the time over which the perturbation acts, normally the length of the laser pulse. It should be pointed out that the Fourier components of $V_{LM}(t)$ produce peaks in the excitation spectrum with height $\propto t_0^2$ and width $\propto 1/t_0$. If the nuclear level width is large compared with $1/t_0$, then the relevant physical quantity is the peak area, $\propto t_0$. This situation forms the physical basis of Fermi's Golden Rule No. 2, resulting in a constant transition rate. For other cases of interest, the nuclear level width can be small, and the relevant physical quantity is the height at frequency ω.

8. MODEL PARAMETERS

The figures display results for some sample calculations. These were carried out for Uranium (Z=92) using a basis consisting of the n=4 shell (32 states). Initial conditions consisted of a uniform wave packet spread over the odd parity (p, f) spherical basis states. The laser frequency was $\hbar\omega_0$ = 5eV, with a total pulse length of 1ps (1200 cycles). Calculations were carried out for electric field strengths of E_0=1, 10, and 100 atomic units, corresponding to intensities of 3.5×10^{16}, $\times10^{18}$, and $\times10^{20}$W/cm^2. The dipole radiation from the atom was calculated, as were nuclear excitation multipoles L = 1, 2, and 3. Nuclear transition strength was assumed to be 1 Weisskopf unit, with the transition charge located at the nuclear surface. The calculation was carried out for 20 laser cycles, with 512 time steps per cycle. In order to extrapolate to a 1ps pulse, the nuclear excitation probabilities were multiplied by a factor 60. This gives an approximate measure of the total transition probability in the sense that the peak areas are correct, but the peak heights are too small by a factor 60 and too wide by the same factor. Alternatively, the calculation corresponds to a laser pulse which is 60 times less monochromatic than the square-pulse theoretical limit.

9. RESULTS

Figure 1 shows as dashed lines radiative dipole moments [Eq. (14)] calculated for (L,M)=(1,0) (parallel to the driving field $\vec{E}_0$); and as solid lines, moments for (L,M)=(1,1) (perpendicular to the driving field $\vec{E}_0$). Electric field strength is E_0=10 atomic units. Response to the fundamental laser frequency is parallel to $\vec{E}_0$, and fluorescence is mainly perpendicular, in qualitative agreement with available experimental evidence.[2] Figure 2 shows results of the same calculation with a factor 10 increase in driving field strength. Increased fluorescence and parallel odd harmonics up to the 7th (close examination reveals the 9th and 11th as well) appear. Figure 3 shows the dipole power spectrum corresponding to Fig. 1, at E_0=10. Three principal regions of fluorescence appear both parallel and perpendicular to $\vec{E}_0$, at the approximate energies 200eV, 650eV, and 850eV. Figure 4 shows the dipole power spectrum corresponding to Fig. 2, at E_0=100. At this strong driving field, the main fluorescence has shifted to over 1keV, comparable with the electron binding energy. This is a evidently a quantum-mechanical analogue of electron synchrotron radiation. Because ionization channels are closed in our model, it is not certain whether this radiation will be produced by real atoms, or whether the electrons responsible will instead be ionized. Calculations involving higher n-shells suggest that ionization may not be important for these states. Experimental tests are indicated.

Figure 5 shows nuclear excitation probabilities for (L,M)=(1 0) (no change in nuclear angular momentum component parallel to $\vec{E}_0$), and E_0=10. Significant excitation occurs only at the fundamental laser frequency. Figure 6 shows the same for (L,M)=(1,1) (change in nuclear angular momentum component parallel to $\vec{E}_0$). Low odd harmonics appear, as well as fluorescence effects between 200 and 300 eV. Figure 7 shows excitation probabilities for (L,M)=(1,0) and E_0=100. Dominant are low odd harmonics of the driving field, reaching a maximum of 2%. Figure 8 shows the same for (L,M)=(1,1). Excitation shows both low odd harmonics of the driving field and combined harmonics/fluorescence at 200-300eV. Maximum excitation is smaller by a factor 10.

Figures 9-12 show similar results for L=3. The main qualitative differences are large decreases in absolute excitation probabilities, and a shift in relative excitation strength to higher energies.

It is clear from these results that observable induced nuclear excitation is possible for L=1, with the appropriate combination of atomic and nuclear levels and modest increases in laser power over what is currently available. For applied fields less than E_0=10, we find an approximately linear dependence of excitation probability on laser power (the fundamental peak in Fig. 5 is reduced to 10^{-4} with E_0=1). It is also clear that excitation probability decreases dramatically for higher multipoles. Thus it may not be possible to observe, for example, the 77eV L=3 state in Uranium.[1] An encouraging note is that excitation is not a monotonically decreasing function of energy, so that somewhat higher nuclear states may be considered in future experiments.

10. REFERENCES

1. See, e.g., P. L. Dyer, J. A. Bounds, R. C. Haight, and T. S. Luk, preceeding paper (this conference); J. A. Bounds, P. Dyer, and R. C. Haight, in Proceedings of the Second International Laser Science Conference, Seattle, Washington, 1986, ed. M. Lapp, W. C. Stwalley, and G. A. Kenney-Wallace (AIP, 1987), p. 87; G. A. Rinker, J. C. Solem, and L. C. Biedenharn, p. 75; J. F. Berger, D. Gogny, and M. S. Weiss (this conference), and J. Quant. Spectrosc. Rad. Transfer (to be published); L. C. Biedenharn, J. C. Solem, and G. A. Rinker, Bull. Am. Phys. Soc. 32, 1043 (1987); Optics News 13, 124 (1987); Technical Digest OSA Annual Meeting, 66 (1987); and to be published.

2. A. McPherson et al., J. Opt. Soc. Am. B4, 595 (1987).

FIG. 1 Dipole Moments $E_0 = 10$

FIG. 2 Dipole Moments $E_0 = 100$

FIG. 3 Power Spectrum $E_0 = 10$

FIG. 4 Power Spectrum $E_0 = 100$

FIG. 5 Nuclear Excitation

FIG. 6 Nuclear Excitation

FIG. 7 Nuclear Excitation

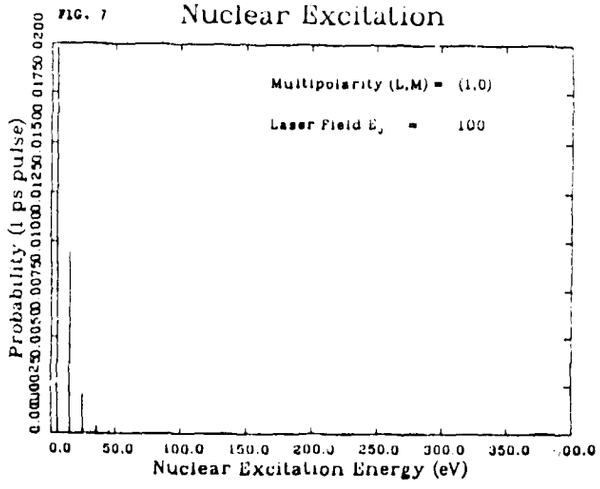

FIG. 8 Nuclear Excitation

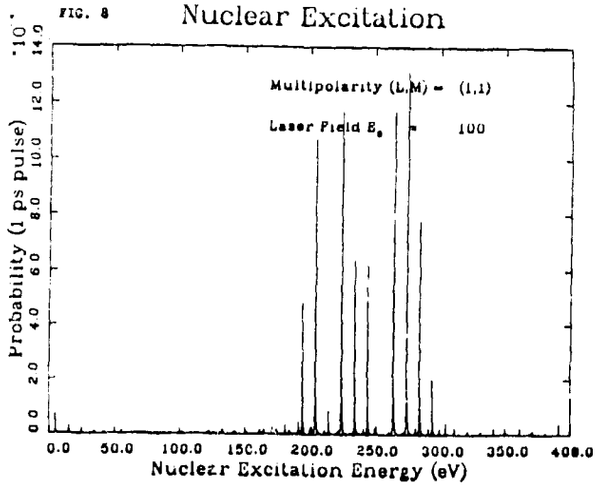

FIG. 9 Nuclear Excitation

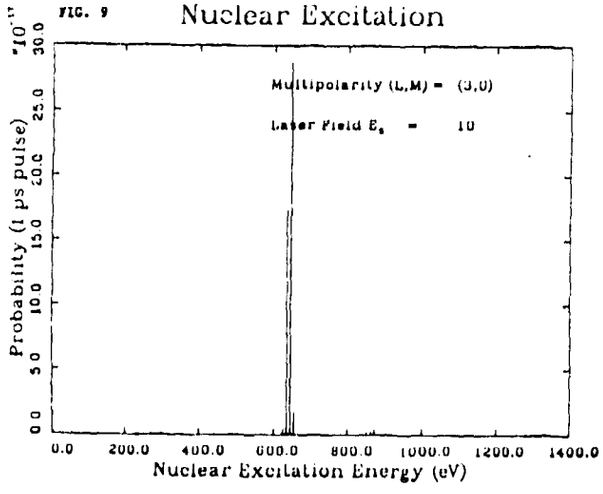

FIG. 10 Nuclear Excitation

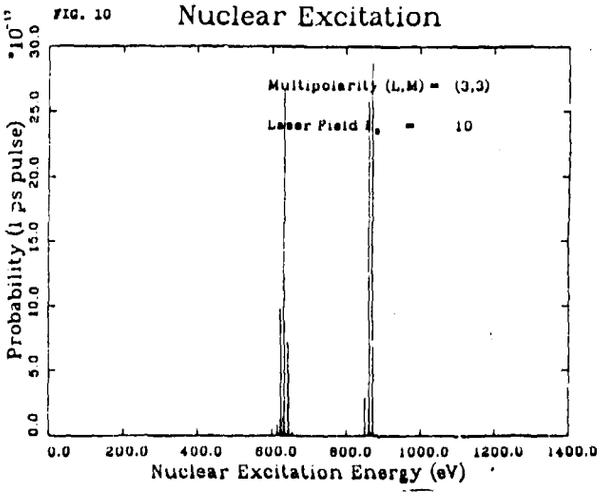

FIG. 11 Nuclear Excitation

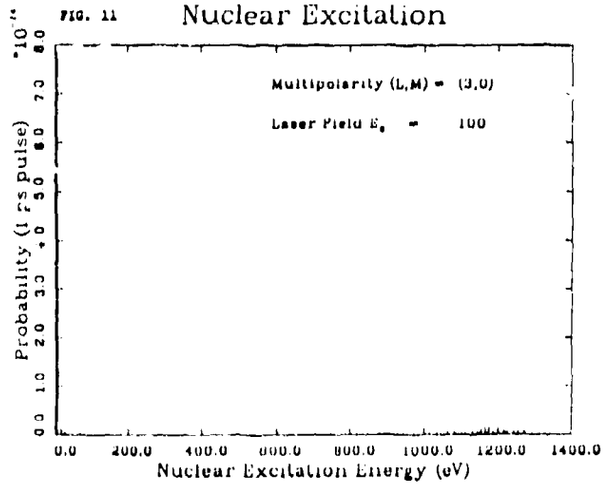

FIG. 12 Nuclear Excitation