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INTENSE LASER-MATTER INTERACTIONS: AN APPROACH TO LASER DRIVEN ELECTRONIC AND NUCLEAR ENERGY TRANSFER

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A semiclassical model treating the interaction of an intense laser field with a non-radiatively coupled system is developed. The model is applied to laser-electron-nuclear energy transfer in a simple single particle model. Results from an initial series of runs on an illustrative example, as reported in conferences and the open literature, are summarized. The work is of ultimate interest in excitation of low-energy nuclear transitions for isotope separation, examination of a laser triggered isotropic gamma-source, or, with extensions, laser damage studies.

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FOREWORD

This paper documents work performed on a subtask under DARPA Task Assignment A-107, "Tactical Applications of Advanced Electromagnetic Devices," sponsored by the Office of Directed Energy, Defense Advanced Research Projects Agency. It describes two stages of work in the development of a new area of laser science. This memorandum report summarizes work of the authors which has been presented or published in open literature conferences or proceedings. This work is of interest in intense laser-matter interactions pertinent to possible applications as a laser-triggered gamma source or for nuclear isotope separation; it amplifies an idea discussed in the Task's mid-year report (IDA Paper P-1970, June 1987).

ACKNOWLEDGMENTS

PREFACE

Non-radiative energy transfer between coupled systems in the presence of intense laser fields is of interest in the laser-driven electronic excitation of nuclei in laser plasmas. Research in the United States in this area is relatively recent; two years ago we proposed a classical and semiclassical approach to the problem. At that time a group at Los Alamos began a different approach utilizing a perturbative quantum mechanical treatment. Since collective motions may ultimately play some role in the dynamics of coupled electronic-nucleonic energy transfer and partially due to the wide applicability of the basic model described herein to other applications of intense laser beam interactions--such as laser polymer damage--we developed this approach at IDA in collaboration with Oak Ridge National Laboratory and some collaboration with the Sandia National Laboratory at Livermore. Although the model parameters thus far treated are for general nuclear systems and a trivial one electron atom, the single-particle models provided a good starting point.

ABSTRACT

A semiclassical model treating the interaction of an intense laser field with a non-radiatively coupled system is developed. The model is applied to laser-electron-nuclear energy transfer in a simple single particle model. Results from an initial series of computer-based analyses of an illustrative example, as reported in conferences and the open literature, are summarized. The work is of ultimate interest in excitation of low-energy nuclear transitions for isotope separation, examination of a laser triggered isotropic gamma-source, or, with extensions, laser damage studies.

CONTENTS

I. INTRODUCTION

With the development of intense laser sources operating at higher powers, shorter pulse widths or shorter wavelengths than previously available, the study of intense laser-matter interactions is a growing area of science of interest to the study of new phenomenon. Much of the recent work centers around the use of laser produced plasmas as x-ray or x-ray laser sources (Ref. 1). In addition, new and interesting areas are the study of above threshold ionization, ultraviolet multiphoton picosecond processes, multiphoton ionization and, of some interest here, the production of harmonic radiations from atoms subjected to strong laser fields (Ref. 2). In all of these areas of research the intense laser field interacts with the electronic charge distribution of the atom.

In addition to *transferring* energy to the electronic cloud of the atom, the possibility also exists for energy transfer to low-lying excited states of the nucleus. In general, the energy transfer could proceed directly from the laser to the nucleus, or most likely, indirectly via non-radiative energy transfer from the electronic motions to nuclear excitations. A brief overview of this area was presented in the mid-year report (Ref. 3) of this task assignment.

Laser driven nuclear electromagnetic excitations is a relatively new area of study in the United States. Some work has been done by Soviet researchers. Nuclear-electronic coupled interactions have additionally been pursued experimentally in Japan (Ref. 4).

In 1985, Biedenharn, Rinker, Solem and Baldwin (Los Alamos) (Ref. 5), having keen interests in exploring the possibilities for lasers using nuclear electromagnetic transitions, approached an understanding of the energy transfer process by focusing on the laser-electron part and, using the perturbative approach of Morita (Ref. 6) or Rinker, Solem, and Biedenharn (Ref. 7), to estimate the excitation probability.

At the same time, Noid, Hartmann and Koszykowski attempted an alternate approach based on studying the coupled dynamics of the system in a semiclassical approach (Ref. 8). This work initially looked at the electron-nucleus system and, in continuation at Oak Ridge National Laboratory, has now looked at a simple model

1

containing electron-nucleus-laser terms. Within the past year (1987), Gogny, Berger and Weiss (Ref. 9) (Livermore National Laboratory) examined the physics using a classical motion of the electron in the laser field and a perturbative approach to the nuclear matrix element.

In Chapter II we present our semiclassical dynamical approach to the question of laser driven electron-nuclear energy transfer. We have constructed a mathematical model and applied it to an example problem. The appendices document the results we have obtained and reported thus far at meetings, conferences and in proceedings. This work is still in the primitive stages; we are working towards the development of a more suitable, improved model (expected completion October 1988). Appendix A describes the work on electron-nucleus coupling presented at the American Physical Society (Topical Group on Laser Science) and Optical Society of America 1986 International Laser Science Conference, published in the proceedings book *Advances in Laser Science*. Appendix B describes the work presented on the electron-nucleus-laser system at the Annual Meeting of the Optical Society of America Optics '87 in Rochester, and the Third International Laser Science Conference sponsored by the APS Topical Group on Laser Science in Atlantic City, NJ (proceedings to be published).

The excitation of nuclei in plasmas can proceed by a number of mechanisms other than those where the electron cloud is driven by a coherent light source. Estimates of these additional excitation mechanisms were made in a previous report (IDA Paper P-1970, June 1987); the unclassified excerpt is available as IDA Memorandum Report M-291.

II. SEMICLASSICAL DYNAMICAL APPROACH TO ELECTRONIC-NUCLEAR ENERGY TRANSFER IN INTENSE LASER FIELDS

A. INTRODUCTION

There has been considerable interest recently in the exchange of energy between electronic excitations of the atom and its nucleus in an intense laser field. Experiments have shown that the possibility for nuclear excitations exists due to transitions of atomic electrons to inner-shell vacancies. Additionally, laser driven excitation of the first excited state of ^{235}U has been reported (Ref. 10) and a second experiment is ongoing (Ref. 11). These experiments show the possibility of exciting ultra-low energy nuclear transitions of possible interest in isotope separation or generating controlled gamma emissions from isomeric levels, by transition from long- to shorter-lived states (as discussed at DARPA in 1987).

In this chapter, we examine the transfer of energy to the nucleus from the laser beam in a simple model treating electron and nucleon dynamics. The emphasis, here, is to describe our method for calculating energy transfer between two coupled systems in the presence of a laser field. This semiclassical approach is one which solves Hamilton's dynamical equations of motion for the electron and nucleon from initial quantum conditions, and treats the laser electric field classically and as an explicit function of time. This approach is one which will provide a route to models which include both nuclear as well as electronic collective degrees of freedom.

B. MODEL

Consider a single particle nucleon model where the nucleon moves in a Woods-Saxon potential well. Parameters for the well can be selected from those appropriate to the magic nuclei (those having both closed neutron and closed proton shells). This model will have dynamics characteristic of a generic nucleus (Blatt-Weisskopf transition rates); the nuclear frequencies will, however, be unrealistically high for nuclei of ultimate interest.

3

The electron is assumed to interact in a Coulomb potential well; this potential involves both the nuclear core and the free proton, as described below.

The time dependent Hamiltonian, H, for the coupled electronic-nucleonic system is given by:

$$H(p,r) = H_n(p_n, r_n) + H_e(p_e, r_e) + H_c(r_e, r_n) + H_{laser}(r_e, r_n, t)$$

where H_n is the nuclear Hamiltonian, H_e is the electronic Hamiltonian, H_c is the electro-nuclear coupling term, and H_{laser} is the time dependent laser Hamiltonian. The nuclear Hamiltonian is:

$$H_n(p_n, r_n) = m_n c^2 \left[\left(\frac{\tilde{p}_n^2}{m_n^2 c^2} + 1 \right)^{1/2} - 1 \right] + V_0 \left\{ 1 + \exp \left[\frac{(r_n - R_0)}{a_n} \right] \right\}^{-1}$$

Here m_n is the nucleon rest mass, V_0 is the Woods-Saxon well depth, a_n is the well diffusivity and R_0 is the nuclear radius. The electron Hamiltonian is given by :

$$H_e(p_e, r_e) = m_e c^2 \left[\left(\frac{\tilde{p}_e^2}{m_e^2 c^2} + 1 \right)^{1/2} - 1 \right] - \frac{e^2 \eta (Z - k)}{r_e}$$

and the Coulombic coupling term is given by:

$$H_{coup} = - \frac{\eta k e^2}{|\tilde{r}_e - \tilde{r}_n|}$$

where m_e is the electron rest mass, Z is the nuclear charge, η is a screening parameter, and k is an artificial coupling parameter. When k is equal to zero, the system is "uncoupled." When k is equal to one, the system is "coupled." (Currently no model explicitly incorporates the effects of "spin" completely, see discussion on page 8.) The laser Hamiltonian is given by:

$$H_{laser}(r,t) = \bar{\mu}(r) \bullet \bar{E} \cos \omega t \ .$$

This term contains the time-dependent contribution for the laser, characterized by electric field strength E and frequency ω. The quantity of $\mu(r)$ is the dipole moment given by the electron-nucleon distance; the nuclear core is fixed at the origin of the coordinate system.

The Woods-Saxon potential well is depicted in Figure 1 as a function of the nuclear radial distance. In this figure we depict the energy of the quantum levels and their orbital angular momenta as found using the WKB solution to the well. Spin-orbit coupling terms, (not treated here) are also indicated for reference. The WKB solution is used to fix the outer turning points for the initial conditions of the nucleon trajectory. The parameters for one series of runs are listed in Table 1.
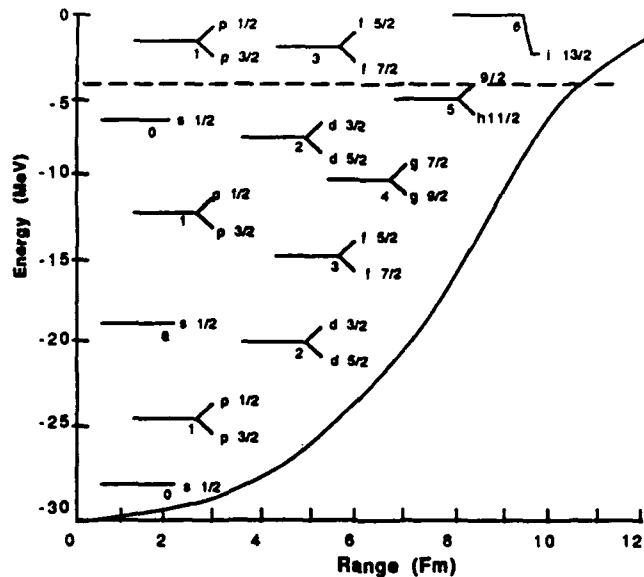

FIGURE 1. The Woods-Saxon Potential as a Function of the Nucleon Distance. Parameters are given in Table 1.

TABLE 1. Parameters for the Pb Systems

Woods-Saxon parameters

$V_O = -30$ MeV
$R_O = 1.25$ Fm $A^{1/3} = 8.66$ Fm for $A = 208$
$a_n = 0.65$ Fm

Coulomb Well Parameters

$Z = 82$

System Parameters

$m_e = 0.511$ MeV
$m_n = 938$ MeV

5

The parameters which we use to show our approach here are those for Z = 82 and one odd proton. This system has a convenient valence nucleon level of angular momentum 1 = 3, which together with a higher lying 1 = 1 state provide a convenient two-level valence proton transition with which to run our model. This transition is still too high in energy for reasonable laser frequencies and still of a single particle nature which makes the quantitative results of the model not useful except for qualitative interpretation and tests of progress thus far.

Hamilton's equations:

$$\dot{p} = -\frac{\partial H}{\partial r} \ , \ \dot{x} = \frac{\partial H}{\partial p}$$

are used to generate trajectories.

For the case where the laser intensity is zero, we can apply the spectral analysis method (Ref. 12) to study the dynamics of the electron and nucleon coupling. The spectral analysis method is used to obtain the power spectrum of the dynamical variables. The classical trajectories are used to obtain the classical auto-correlation function and the power spectrum is obtained via a Fourier transform. When the spectral lines compare quite closely with the quantum mechanical transitions, they are generally relatively narrow; in the classically ergodic regime, the spectral lines broaden.

Initial conditions for the trajectories of coupled systems not permitting separation of variables are obtained by quantizing the action variables, as first proposed by Einstein (Ref. 13):

$$J_i = \oint p \bullet dq = n_i h$$

where the different J_i's are obtained by integrating over topologically independent paths; $q \ (\equiv q_1, q_2, ...q_N)$ and $p \ (\equiv p_1, p_2, ...p_N)$ denote canonically conjugate coordinate and momenta. Keller (Ref. 14) showed that fractional terms arose:

$$J_i = \oint p \bullet dq = (n_i + \delta) h$$

where δ_i is usually 0 or 1/2.

The trajectories are used to calculate auto correlation functions. The auto correlation function of a dynamical variable x(t) is:

$$c(t) = <x(0) \ x \ (t)>$$

6

where the brackets indicate an ensemble average. The absorption band shape $I(\omega)$, or power spectrum or spectral density, is given by the Fourier transform of the auto correlation function:

$$I(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} c(t) \exp(-i\omega t) \, dt$$

According to Bohr's correspondence principle, the mechanical frequencies are equal to the differences of eigenvalues $(E_m - E_n)/h$.

In our calculations of the coupled electron-nuclear system, without the laser term, we found that with the application of the Langer correction (Ref. 15) for the initial condition for the electron's trajectory, the trajectory evolved in an elliptic fashion as opposed to the circular orbit; this is as expected. As Z was varied through a range from 29 to 83, the coupling was seen to increase: with the strongest coupling the trajectory became chaotic (the power spectrum broadened). This is seen in Figure 2. Such features are

Z=51 (Uncoupled)   Z=51 (Coupled)
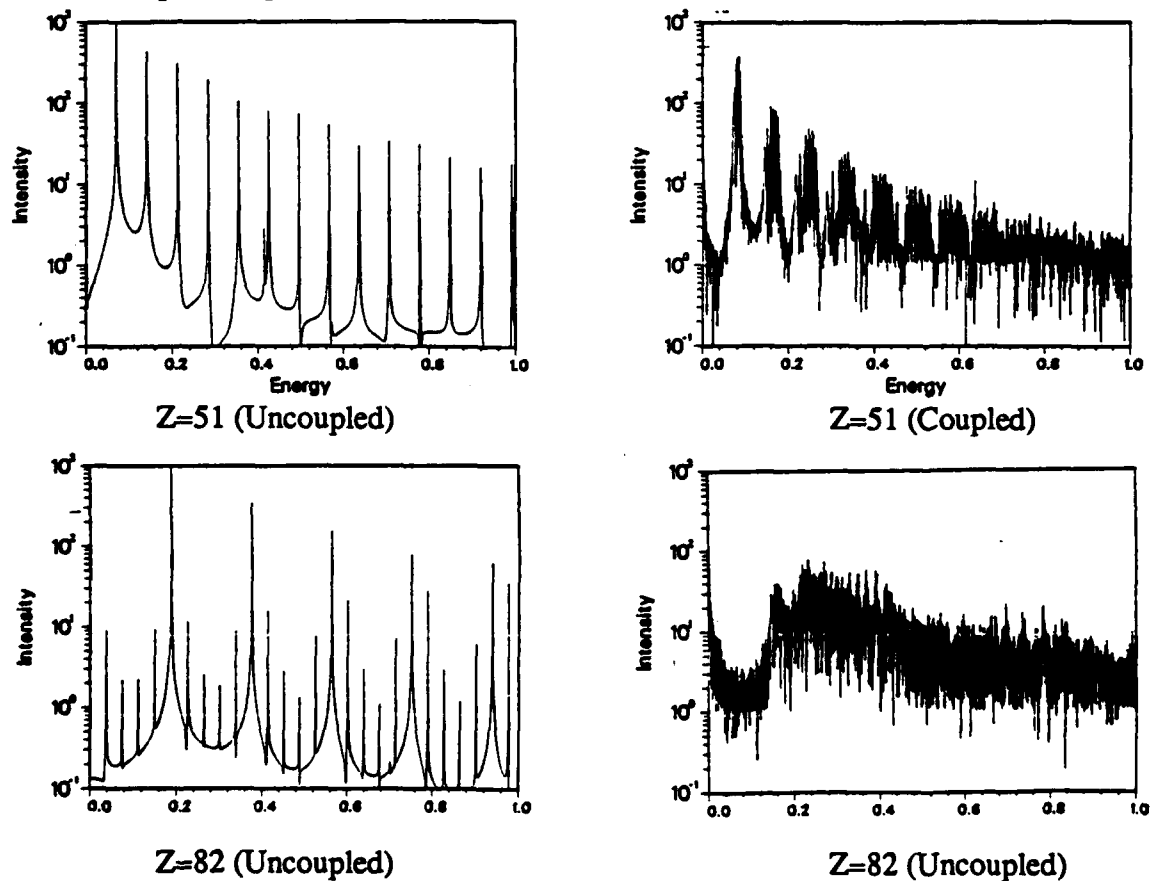
Z=82 (Uncoupled)   Z=82 (Uncoupled)

FIGURE 2. Power spectra for the non-relativistic case.

sometimes seen in nonseparable anharmonic systems, such as observed in the astronomical literature (Ref. 16), where quasi-periodic behavior ensues; such behavior is supported by the fundamental theory of Kolmogorov, Arnold and Moser (Ref. 17). At higher energies an ergodicity occurs, the action-angle variables are no longer good variables, the coupling is stronger and quasi-periodicity breaks down--the spectral lines broaden.

The broadening of the spectral lines arises, in a classical interpretation, from the arbitrary alignment of the nuclear charge dipole when the electron flies by in its "near" pass in the elliptic orbit. During the time of this classical "near" pass, significantly all of the classical energy transfer takes place due to the coupling term. In the quantum case, this suggests that the energy transfer matrix elements depend on that part of the electronic wave function "over the nuclear volume." For lower Z, the electron is classically further from the nucleus and no chaotic behavior is observed. Further aspects of this chaos have not been pursued. The differences in such coupled and uncoupled trajectories are shown in Figure 3. The associated coupled and uncoupled dipole moments are shown in Figure 4.

Now an interesting problem arises. With increasing Z, the electron classical velocity increases and starts to approach light speed. This necessitates the introduction of relativistic kinetic energy terms in our Hamiltonian. We then find, for the relativistic Hamiltonian, that the Langer correction does not work for Z in excess of 67.5. Classically, the electron's mass increases appreciably and it flies into the center of the nucleus--of course, this cannot happen. This singularity can easily be seen in Sommerfield's relativistic treatment of the Bohr atom, whose energies are given by:

$$E = mc^2 \left[1 + \frac{\alpha^2 Z^2}{\left(n_r + \sqrt{k^2 - \alpha^2 Z^2} \right)^2} \right]^{-1/2} - mc^2$$

where α is Sommerfield's fine structure constant; and n_r, k are the two integer quantum numbers replacing the usual principal quantum number $n = n_r + k$ in the non-relativistic classical treatment when quantized using the Bohr condition.

In order to avoid this singularity, we use the original $\delta = 0$ as we enter the relativistic regime, or "old quantum theory". Thus, in contrast to Ref. (8), we do not start with elliptic trajectories. (In light of the recent comments on "spin" by Jackson (Ref. 18), it is interesting to note that relativistic classical dynamics applied to the quantum atom contain some surprising results and this is being examined further.)
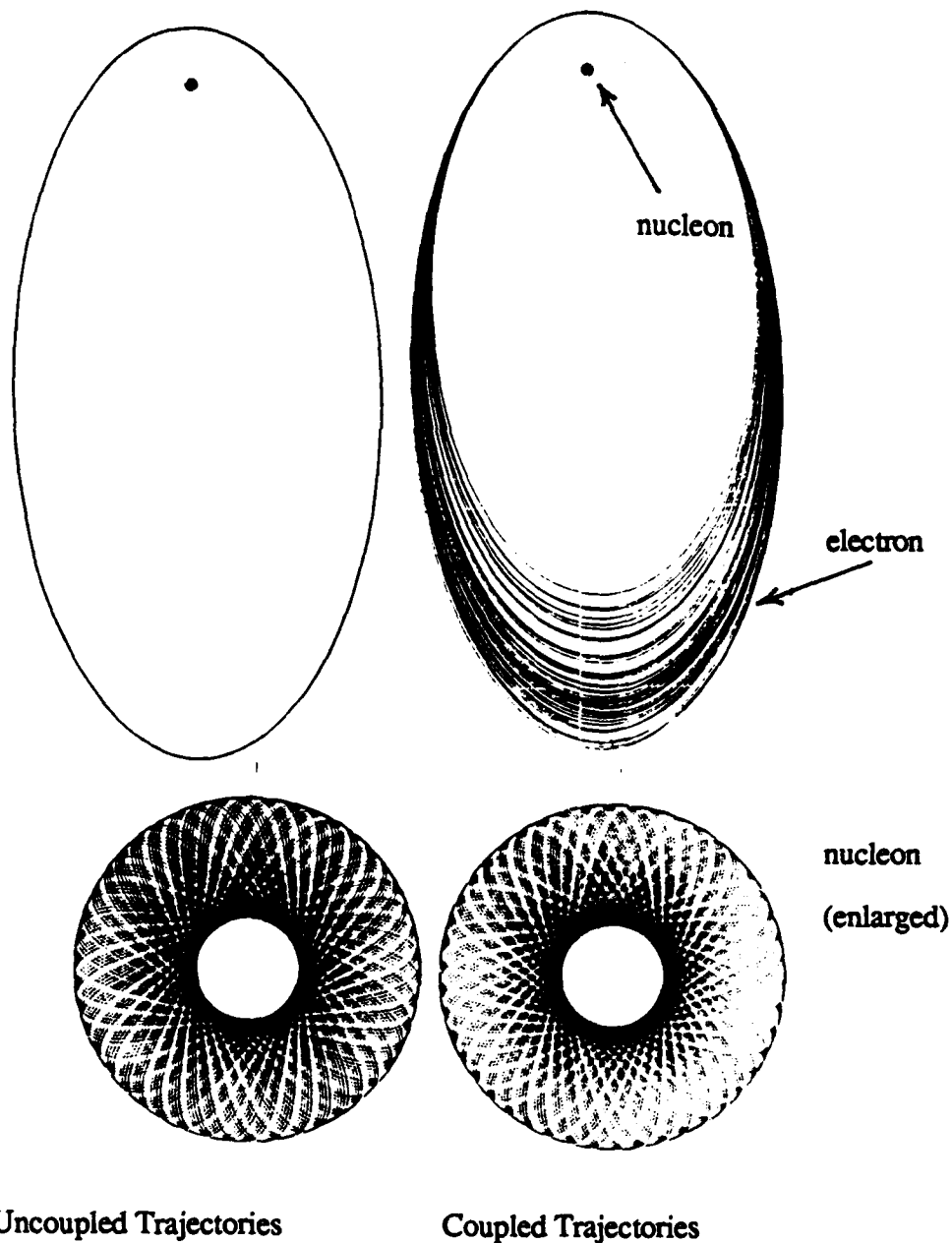
8

FIGURE 3. The two-dimensional uncoupled and coupled electron and nucleon trajectories are depicted above. In the lower portion of the figure we see enlargement of coupled and uncoupled nucleon trajectories for the $Z = 82$ non-relativistic case. An interesting feature is the chaotic behavior of the electron trajectory in the coupled case.
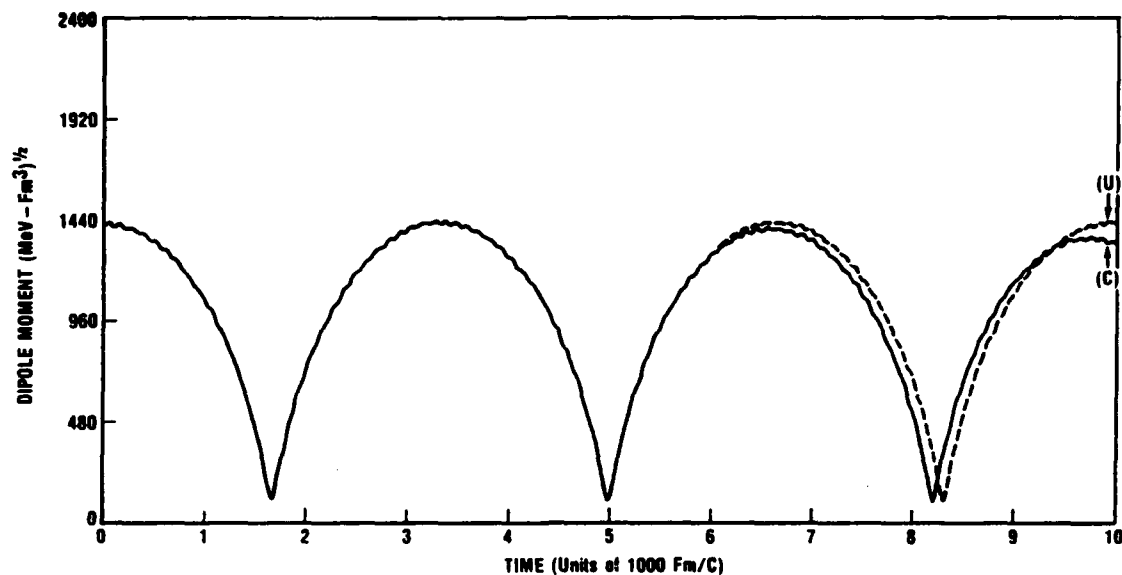
9

FIGURE 4. Uncoupled and coupled evolution of the system dipole moment.

With the laser in the model turned on, we integrate Hamilton's equations as a function of time for an ensemble of 64 trajectories whose phases are evenly distributed in the intervals $(0, 2\pi)$. In the calculation of the energy transfer, the ensemble average of the energy of the electron and nucleon system is calculated as a function of time. The electron and nucleon Hamiltonians are clearly time independent; our treatment of the explicit time dependence of the laser easily allows us to vary frequency (ω) and intensity (E), and even, if desired, the envelope.

The trajectory calculations are carried out on a Cray computer at Oak Ridge National Laboratory under operation by the Department of Energy, in collaboration with this project. We chose 64 trajectories for the phase ensemble since the vector capability of the Cray allowed us to run up to 64 trajectories in parallel.

Typical relativistic trajectories for coupled and uncoupled particles are depicted in Figure 5. The basic frequencies are depicted in Figure 6 in the absence of the laser beam. The basic frequencies give some insight into the physical motions in this particular model. The lowest frequency is associated with the classical orbital motion of the electron. In a crude sense, the nucleon trajectory has a "narrow" elliptic orbit close to the appearance of a vibration; this then precesses. The two highest frequencies arise from the "vibrational" frequency of the nucleon in the potential well plus and minus half the "rotational" frequency

of this "intrinsic" vibration. This rotational frequency appears as the smallest significant peak between the electronic orbital and the nucleonic vibrational frequencies in Figure 6. When the total dipole moment in the uncoupled case is Fourier-transformed, components of the electronic frequency appear superimposed on the nucleonic frequencies. For the relativistic Hamiltonian (in the old quantum theory) the total dipole moment shows a stronger admixture of the electron and nucleon motion; with coupling, the energy transfer, in the absence of the laser field, is not sufficient to show up as frequency shifts in these spectra.
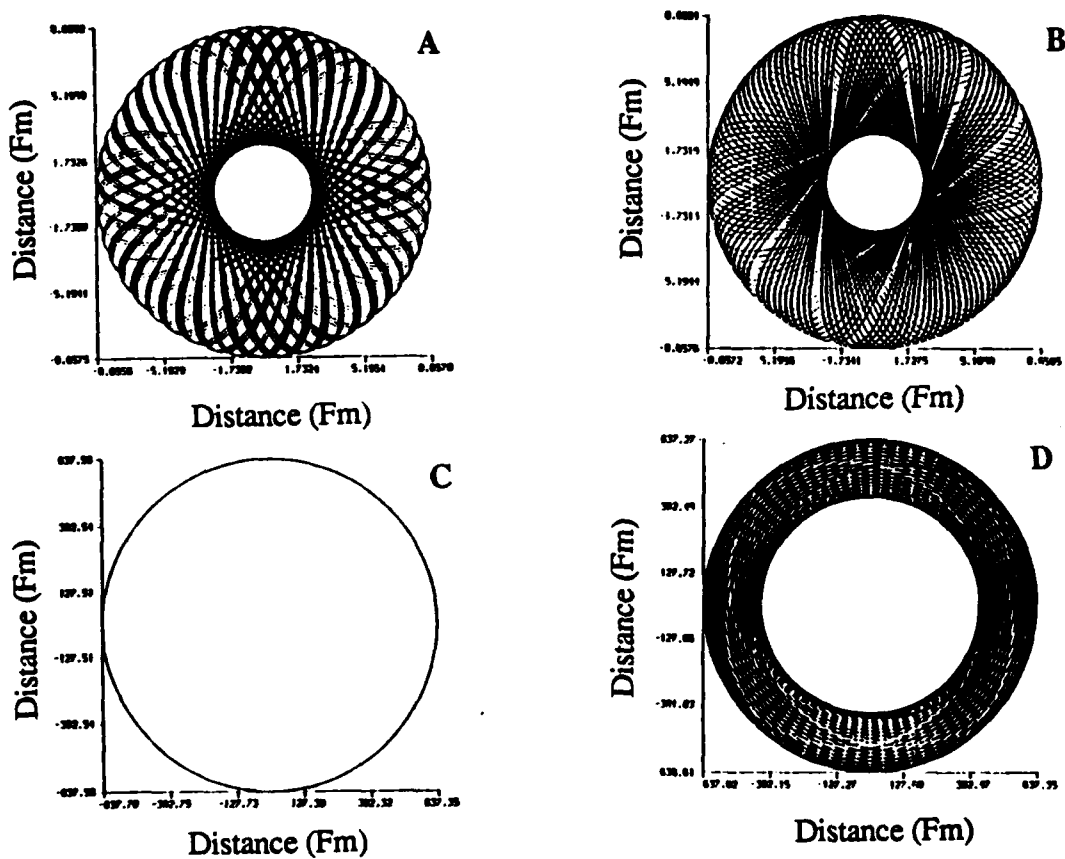

FIGURE 5. Illustrative trajectories for relativistic nucleonic (A and B) and electronic (C and D) motion for uncoupled (left column) and coupled (right column) cases.
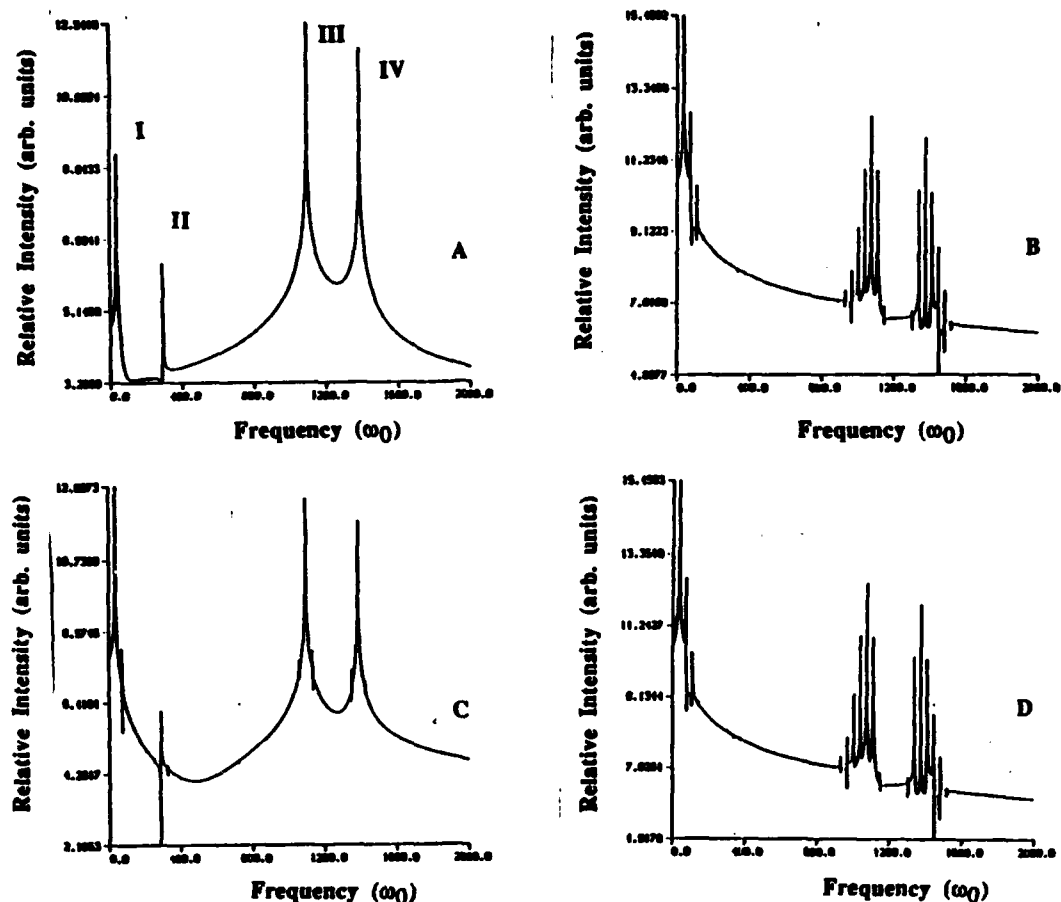
FIGURE 6. Frequencies of the coupled and uncoupled motions. The four characteristic frequencies in this example are denoted in part A of the figure: I is associated with electronic motion, II is associated with the nuclear "rotation" (or precession, as defined in the text), III and IV are associated with the nuclear "vibration" (a narrowed elliptic orbit as defined in the text) with sum and differences due to the nuclear "rotation". Parts A and B are uncoupled, non-relativistic and relativistic, respectively. Parts C and D are the corresponding spectra with coupling. For the latter case, electronic frequencies become superimposed on the nuclear region of the spectra. The laser intensity is zero in this case, and the coupling term has an unnoticeable effect on the basic frequency characteristics. The unit frequency ω_0 is 2.5 × 10(−5) inverse time units.

A scan in frequency of the laser is then made at a fixed high intensity. For each fixed frequency the ensemble averaged nucleon energy, electron energy and total energy are computed as a function of time. Fourier transforms of these time dependent ensemble averages are used to extract sum and difference frequency information on the location of significant resonances for laser absorption in the coupled system. This is useful to sufficiently map out gross features of the spectrum.

12

Figure 7 shows plots of the maximum changes in nuclear energy, electron energy and total energy over a range in frequencies. Figures 8, 9, and 10 depict example time dependent ensemble averages of the nuclear, electron, and total energy for the laser scan through the resonance at 12×10^{-4} in Figure 7 at an electric field strength of 1×10^{-6}.
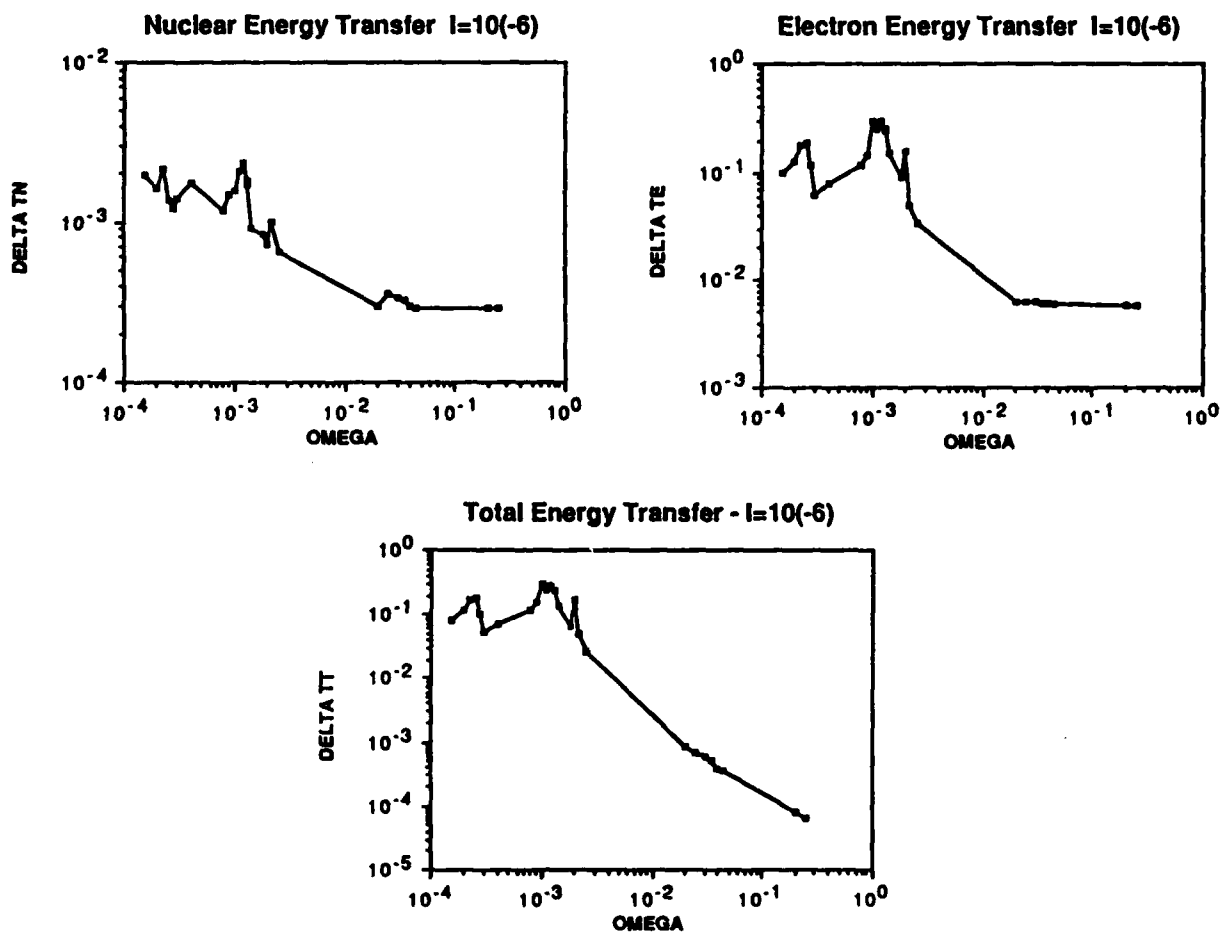

FIGURE 7. Frequency dependency of the energy transfer. These three figures show the total energy transfer, electron energy transfer and the nucleon energy transfer at selected values of the laser frequency. The key qualitative feature is the presence of resonances in the coupled system at which energy transfer is enhanced. Lines in the figure are drawn to guide the eye, as each point results from a number of trajectory runs. (Fourier transforms of the time-dependent energy transfer for a particular trajectory assist in locating the resonances. The units of OMEGA are inverse time units, defined in the text, and the energy depicted here, when divided by the number of trajectories (64), has units of MeV. The electric field is fixed at $1 \times 10(-6)$ as discussed in the text.
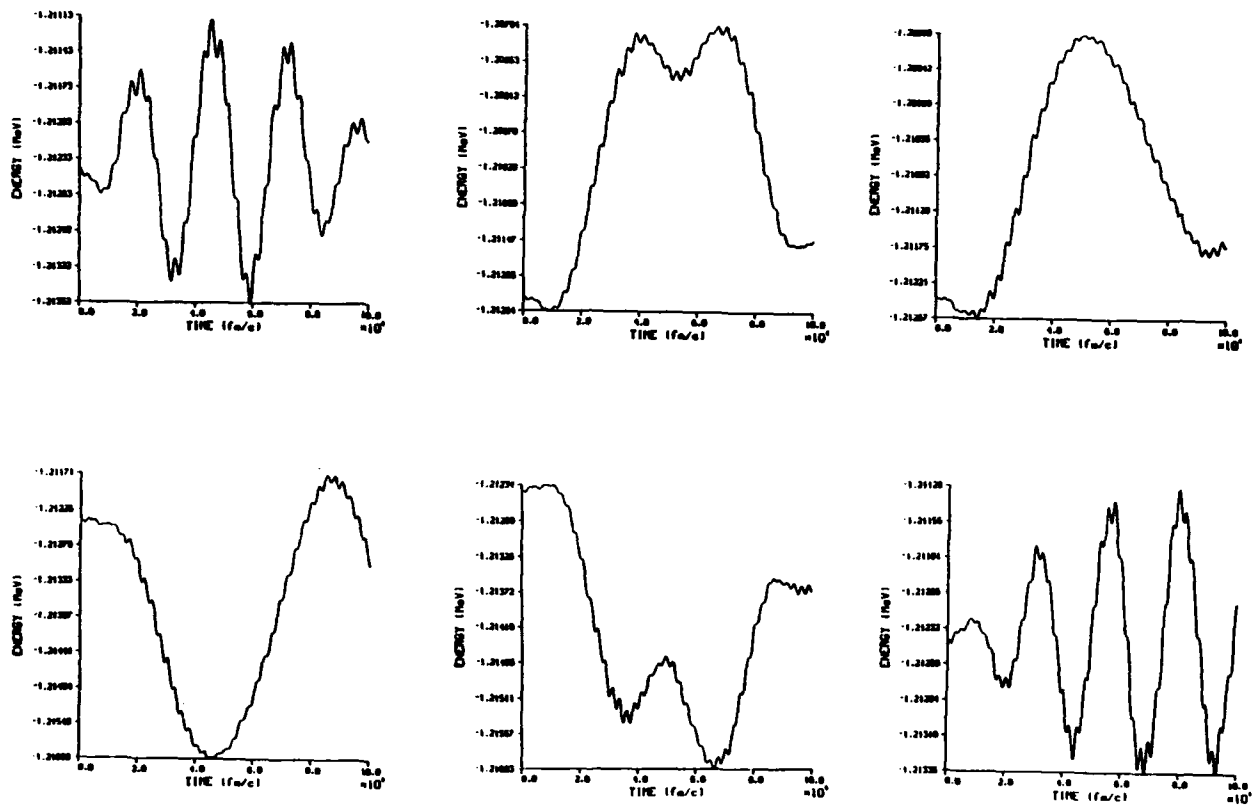
FIGURE 8. Time-dependent total energy transfer. The behavior of the ensemble averaged energy of the total system (including the coupling term) is depicted for six successive values of the laser frequency: 9,10,11,12,13 and 14 × 10(−4) (inverse time units) from left to right, top to bottom at a fixed electric field strength of 1 × 10(−6). The behavior through a resonance is illustrated by this sequence.
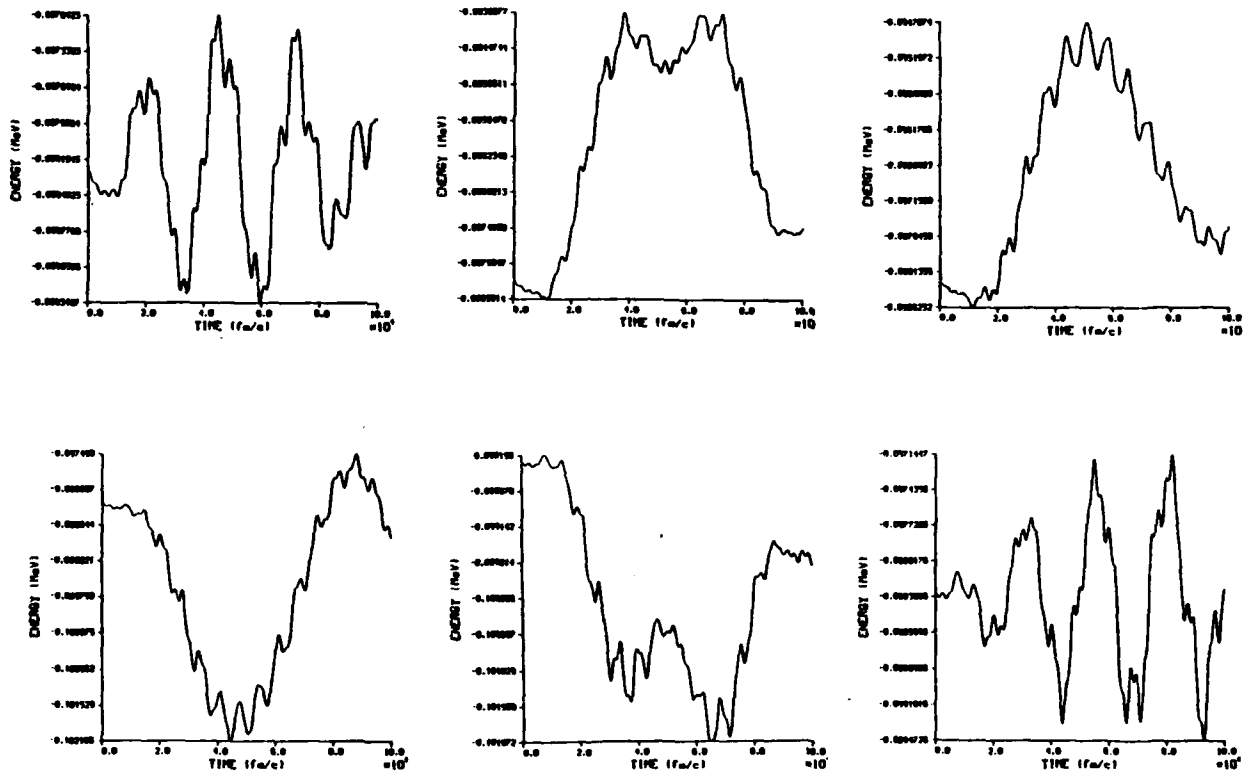
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FIGURE 9. Time-dependent electron energy transfer. The electron energy component of the total energy is illustrated here for the same cases treated in Figure 8.
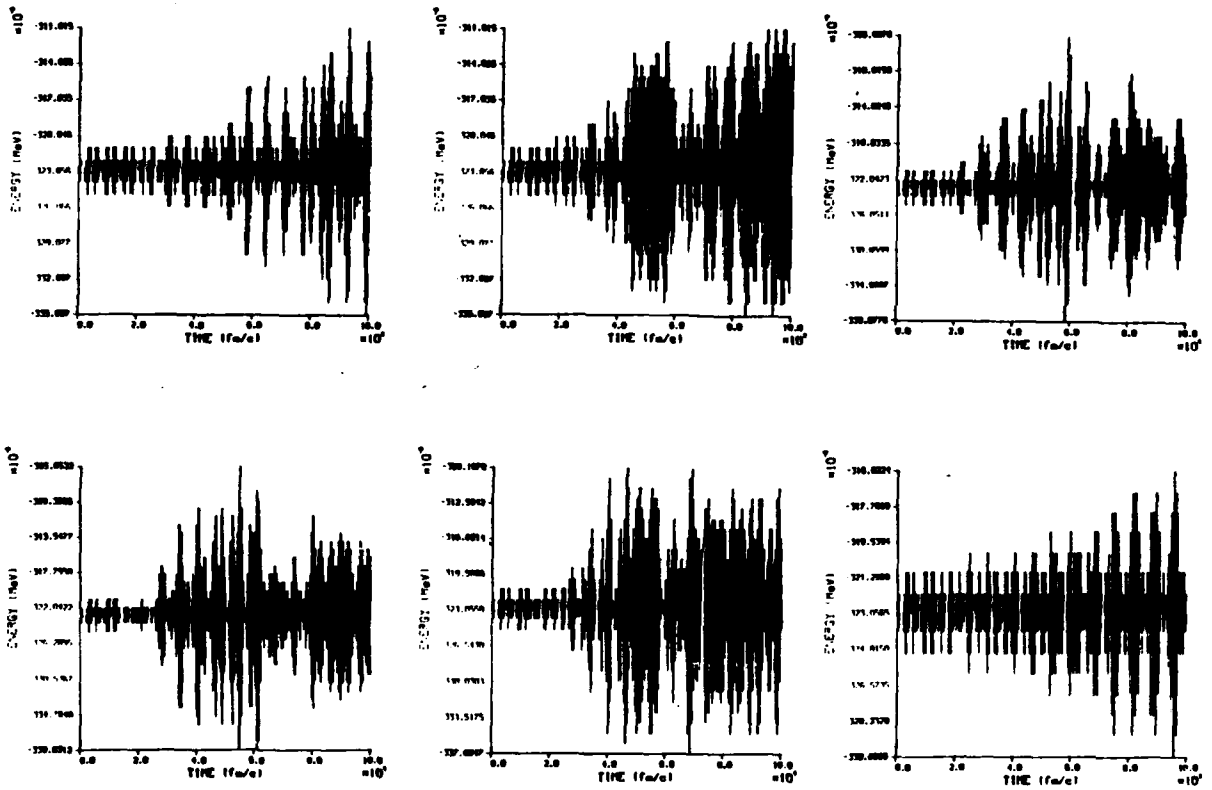
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FIGURE 10. Time-dependent nucleon energy transfer. The nucleon energy component of the total energy is illustrated here for the same cases treated in Figures 8 and 9.

16

The frequency is in units of inverse time units, where 1 time unit is the time for light to travel one Fermi. The frequency is converted to energy units of MeV by multiplying by 197. Hence, the dipole moments have units of $MeV^{1/2} \cdot Fm^{3/2}$ and eE has units of MeV/Fm. These units are convenient since $e^2 = 1.44$ MeV $\cdot$ Fm and $e^2/2\pi hc$ is 1/137.

The intensity dependence of the energy transfer is depicted in Figure 11 at the fixed frequency of $\omega = 12 \times 10^{-4}$, for electric field strengths of 10^{-6}, 10^{-7}, and 10^{-8}; the intensity is proportional to the square of the electric field strength. Clearly the energy transfer falls with decreasing electric field strength when on or near resonance.
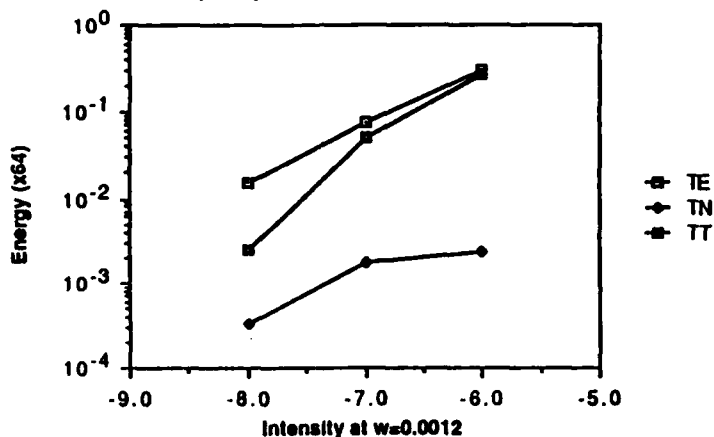

FIGURE 11. Laser intensity dependence of the energy transfer. The total energy transfer (TT), electron energy transfer (TE) and nucleon energy transfer (TN) in units of MeV are depicted at three electric field strengths of the laser [1 × 10(-6), 1 × 10(-7) and 1 × 10(-8)] at a fixed laser frequency.

The energy transfer values are proportional to the frequency spread of the particles' motion. Typically, the ensemble energy transfer, which is proportional to that frequency spread, reaches a constant after some time. We have run extended time durations to verify that this ensemble frequency spread is roughly constant for an extended time duration; thus, the laser absorption is essentially a quasi-periodic function of time.

C. CONCLUSIONS

An approach for exploring non-radioactive energy transfer in coupled systems and intense laser fields has been set up and used to study energy transfer in a single-particle electron and single-particle nucleon model.

It is demonstrated that energy transfer to the nuclear motion occurs via coupling to the electronic motion in a laser field for a simplistic single-particle model of a valence nucleon and an inner shell electron at a laser resonance. Energy transfer in this system not only depends on the laser frequency but also on intensity. Electrons closest to the nucleus have greater coupling; however, electrons further from the nucleus will be most affected by the laser.

The nucleon model sets the energy scale of the frequency response of the coupled system--currently this is too high for practical applications since the single-particle nucleon transitions are too high in energy. The specific features of the nucleon model can be scaled to examine lower frequency transitions and collective motions included now that the codes are basically set up. The electron part of the model basically describes a tightly bound electron and with suitable changes can be extended to treat outer shell electrons or collection motions (in the laser field) as appropriate. Muonic atoms can also be studied.

APPENDIX A

CLASSICAL AND SEMICLASSICAL CALCULATION
OF ELECTRON-NUCLEON COUPLING

A. INTRODUCTION

The main text describes the most recent results of work in progress on the laser-electron-nucleon model. This chapter documents the work on electron-nucleon coupling in the absence of a laser field as presented at the International Laser Science Conference and published in the proceedings book: *Advances in Laser Science-II*. The summary is taken from *Optics News* and the Abstract from the Bulletin of the American Physical Society.

B. SUMMARY

Classical and Semiclassical Calculation of Nuclear-Electron Coupling
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Oak Ridge National Laboratory, Institute for Defense Analyses,
and Sandia National Laboratory.

Energy transfer processes between an excited nuclear proton and an inner core-electron are studied semiclassically. The coupled independent particle models exhibit large spectral perturbations.

C. PAPER

CLASSICAL AND SEMICLASSICAL CALCULATION OF NUCLEAR-ELECTRON COUPLING

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ABSTRACT

Energy transfer processes between an excited nuclear proton and an inner core electron are studied semiclassically. The coupled independent particle models exhibit large spectral perturbations.

INTRODUCTION

Interactions between electronic transitions of an atom and nucleonic transitions of its nucleus are of interest in the excitation of nuclei from ground states to low-lying excited states or the excitation of short-lived states from long-lived isomeric states. Such interactions can proceed via nonradiative energy transfer between the excited electronic state and the initial nuclear state.

The excitation of low energy isomeric states in nuclei from their ground states has been observed by Gol' danskii and Namiot [1] in hot plasmas. They reported the laser plasma production of 235mU (73 eV 7/2- to 1/2+) as a result of the capture of continuum electrons to the outer shell in ionized uranium atoms. Their explanation of this excitation differs from that of Izawa and Yamanaka[2] who initially attributed the excitation to a $6p_{3/2}$ (-32.5 eV) to $5d_{5/2}$ (-103.1 eV) atomic transition. Additional evidence for the excitation of nuclear ground states is given by the observation of de-excitation gamma-rays in 237Np by Saito, Shinohara and

21

Otozai[3] and the excitation of 189Os by Otozai, Arakawa and Morita[4]. The possibility of nuclear excitation by laser driven coherent outer electron oscillations is described by Biedenharn, Baldwin, Boyer and Solem[5,6].

In general, nuclear-electron interactions occur in the processes of nuclear orbital electron capture or internal gamma-ray conversion. Ultra-low energy nuclear transitions, in particular, lead to excitations in the outer electron shells; e.g. the 73 eV electromagnetic decay in 235mU is a highly converted transition which leads to ejection of a bound electron near the valence shell; the 2.6 keV weak decay in 163Ho leads to atomic excitations in the M and higher shells (<2 keV regime)[7]. The first case illustrates the predominance of the multipole components of the near Coulombic field of the nucleus over the direct radiation process (which plays the same role in the inverse process); the second example suggests an alternate approach to achieving specific electronic excitations in atoms. The analogous muonic excitations of nuclei have been extensively reviewed by Borie and Rinker[8].

We have recently devoted our attention to the study of excitation pathways available for the transfer of energy from highly excited long-lived isomeric nuclear states to much shorter-lived states in laser plasmas.[9] In many non-radiative energy transfer processes one treats the nuclear and electron quantum system as states of a separable Hamiltonian and the coupling term is treated as a perturbation. This is illustrated by the model presented by Morita.[10] In this paper we briefly highlight results of our non-perturbative approach used to study coupling between electronic and nucleonic transitions applied to a simple single-particle model for both electrons and nucleons.

22

SPECTRAL ANALYSIS METHOD

A quantization of systems not permitting separation of variables was first proposed by Einstein:[11] one finds canonical invariants, namely the action variables J_i, and quantizes them:

$$J_i = \oint \tilde{p} \, d\tilde{q} = (n_i + \delta_i) \, h$$

such that the different J_i's are obtained by integrating over topologically independent paths. Here $\tilde{q}$ (q_1, q_2, ... q_N) and $\tilde{p}$ (p_1, p_2, ... p_N) denote canonical coordinates and momenta and n_i is a quantum number. Keller[12] introduced the fractional term δ_i, usually 0 or 1/2. Eastes and Marcus[13], and Noid and Marcus[14] showed how to evaluate these action integrals in nonseparable systems having smoothly varying potential energy functions and evaluated eigenvalues semiclassically. Other methods have been developed for systems having degeneracies. These latter results are for cases which are quasiperiodic (systems having action-angle variables).

The differences in eigenvalue frequenices for complicated systems can be obtained by using the coupled trajectory to compute the appropriate autocorrelation function. This is then Fourier-transformed to obtain the spectrum. According to Bohr's correspondence principle the resultant mechanical frequenices are equal to the differences of eigenvalues $(E_i - E_j)/h$. A more complete discussion of the semiclassical method and many numerical tests of these theoretical techniques are discussed in ref. (15).

MODEL HAMILTONIAN

We consider a mathematical model characteristic of nuclear and electronic single particle dynamics. Nuclear transitions are described by single particle transitions of an odd proton in a Woods-Saxon potential well[16]:

$$V(r_n) = V_0 [1 + \exp (r_n - R_0)/a_n]^{-1}$$

where r_n is the nucleon position, a_n the well diffusivity and R_0 is the nuclear radius. We use $a_n = 0.65$ fm and $R_0 = 1.25$ $A^{1/3}$ where A is the atomic mass number. Such a picture might best approach reality for a doubly-magic nucleus (such as $209Bi = 208Pb + 1p$) with the neglect of spin-orbit effects. Mathematically we can, of course, scale any parameter. The most convenient parameters to scale are the atomic mass A and the proton number Z. The well depth V_0 is chosen such that the density of states $\rho = (4\pi/3) (p_n/2\pi h)^3$ [given by a square well approximation to the nuclear volume] leads to a binding energy of the odd nucleon comparable to the experimental value, when filled by the other nucleons. The single particle electronic transitions are modelled by an electron which moves in orbits of a screened nuclear core of charge $\eta(Z - k)e$ and the screened odd proton has charge ηke. The parameter $k = 1$ models the coupled system; $k = 0$ models the uncoupled system. Here η is a screening parameter which scales the hydrogenic orbits. The complete Hamiltonian, restricted for convenience to a planar geometry is written as:

$$H = \frac{\tilde{p}_n^2}{2M_n} + V_0 \{ 1 + \exp[(r_n - R_0) / a_n] \}^{-1} +$$

$$\frac{\tilde{p}_e^2}{2M_e} - \frac{e^2 \eta (Z-k)}{r_e} - \frac{\eta k e^2}{|r_e - r_n|}$$

24

This Hamiltonian is separable thus all but the last coupling term is easily quantizable. Hamilton's equations of motion are used to numerically generate the classical trajectories for both the coupled and uncoupled system. Initial conditions for the nucleon trajectory are found using a WKB approximation to the Woods-Saxon well to fix the position-momentum at the classical outer turning point. Initial conditions for the Coulombic well are given in ref. (17).

RESULTS

Sample electron and nucleon trajectories are depicted in the left parts of figs. (1) and (2) for both uncoupled and coupled cases. The distance scales are on the order of a hundred fermis. The nucleon trajectories shown there comprise approximately 5000 points; each point is integrated over two "time-unit" time steps. The natural "time-unit" which we use is defined as the time for light to travel one fermi in a vacuum. Trajectories up to a million points are used for the actual computation of autocorrelation functions. In the same figures, the electron orbits are depicted for 10,000 points integrated in 25 time-unit steps. Electron trajectories have drastically visible differences for electron trajectories involving the K shell, the differences in the nucleon trajectories for coupled and uncoupled cases are more subtle. Enlargements of the nuclear centers are shown on the right of figs. (1) and (2) with increases to 10,000 points for both coupled and uncoupled trajectories. The precession rates of the orbits in both cases are not identical.

The time-dependent dipole moment for the total system is used to study the dynamics of the system. Initially, the largest component of the total dipole moment is of lower frequency associated with the electronic motion. Superimposed on the electron contribution is the nucleon contribution which is initially higher in frequency and smaller in magnitude. When the coupling is turned-on, the system dipole moment

25

deviates from its regular evolution in intensity and frequency. Typical Fourier-transforms of such dipole moment autocorrelation functions are depicted in figs.(3) for Z=51 and (4) for Z=83. For Z=29, the coupled spectrum looks identical to the uncoupled spectrum. By scaling Z and A we can examine the dynamics of coupling by examining the eigenvalue spectra. In figs. (3) and (4), we look at Z and A corresponding to near magic nuclei for this mathematical model. With increasing A, the spectra change and can be characterized by a transition from quasiperiodic to chaotic behavior [see ref. (15)].

DISCUSSION

The frequencies in the spectra are understood as follows. In the uncoupled system we see spectral intensities at frequencies characteristic of the nuclear motions and spectral intensities characteristic of the electron motions. In the coupled system we expect to see additional spectral intensity in electron-nucleon sum and difference bands. These spectral lines must be assigned to specific coupled single-particle electron-nucleon transitions to examine a particular transition of interest. All possible transitions are computed for the given nuclear parameters and not all (such as energetically high lying hole states) are of practical interest. The initial starting conditions serve to fix the total energy and total orbital angular momentum of the system.

The relationships of the slight nucleon orbit precession differences and the chaotic electron behavior at higher A has a physical (classical) interpretation. When the electron is traveling fastest, it is nearest the nucleus. In this region the rotating nuclear moment can be in any angular position relative to the electron's near pass. At this distance, the effects of the energy transfer on the spatial alteration of the trajectory are subtle. It is not until the electron travels furthest from the nucleus, that the radial

26

energy dependence of the potential reveals the significant deviations in the electron's orbital trajectory. Since the nucleon-electron energy transfer occurs mostly for the brief time the electron is nearest the nucleus and the nuclear moment is essentially randomly oriented, the electron achieves a chaotic orbit. Ultimately it is the spectral features of the Hamiltonian which correspond to the true observables in the spectral analysis technique, and clearly any classical interpretation of the intermediate steps is only to give physical insight.

Theoretically, in the limit of extreme coupling, the electromagnetic emission associated with the spectrum of such a system is strictly neither a gamma ray nor an x-ray but a coupled emission from a single wavefunction with electron and nucleon components.

CONCLUSION

We have calculated absorption frequencies of coupled and uncoupled electron-nucleon motion in a simple single-particle model. The dynamics of the coupled and uncoupled systems are qualitatively different, and the difference is more pronounced with increasing A and Z. Transition probabilities, frequencies and intensities are significant signs of differences between the coupled models and the uncoupled models. We have found some instances of strong coupling and instances of chaotic motion in this simple model. The spectral analysis method is a powerful technique for studying non-linear non-separable systems which model physical systems.

This model is being expanded to treat the deformed rotor and single-particle rotor models to examine behavior of nuclear-electron coupling in more complicated systems. These results should prove useful to the study of upconverted nucleon transitions from isomeric levels. Such transitions, expected to be driven from highly excited electronic states produced in laser plasmas, could lead to enhanced decay rates

from nuclear isomers. Experiments are now under consideration which may demonstrate this effect.

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16.A. Bohr and B. R. Mottelson, Nuclear Structure.(Benjamin, Reading,1969) Vol. 1.

17. R. Langer, Phys. Rev. $\underline{51}$, 669 (1937).
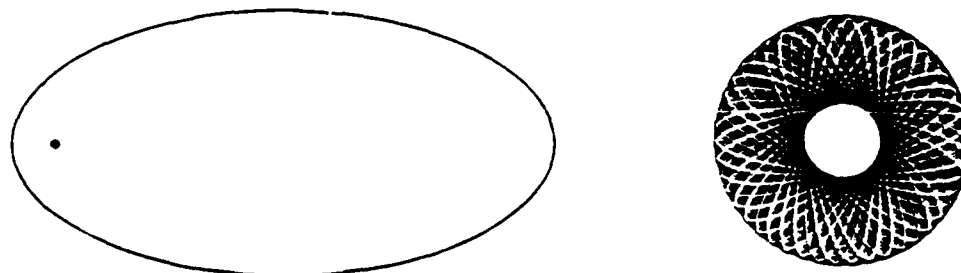
Fig. 1. Uncoupled electron (max r=1188 fm) and nucleon (r=8.66 fm) trajectories.
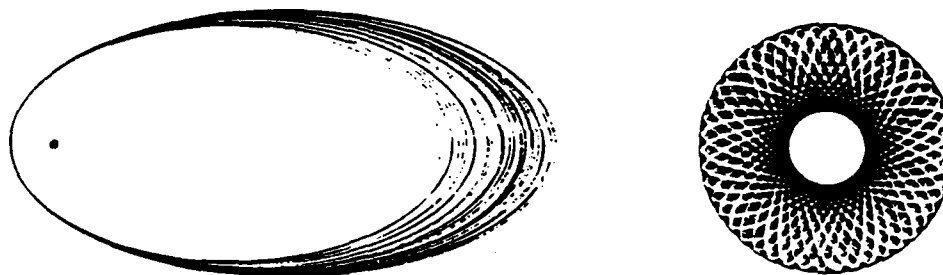
Fig. 2. Coupled electron and nucleon trajectories for comparison to fig. 1.
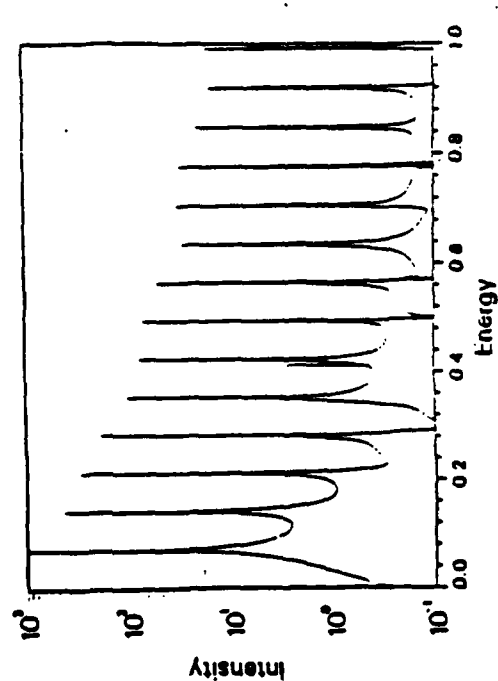
Fig. 3. Uncoupled and coupled spectra for a case having Z=51.
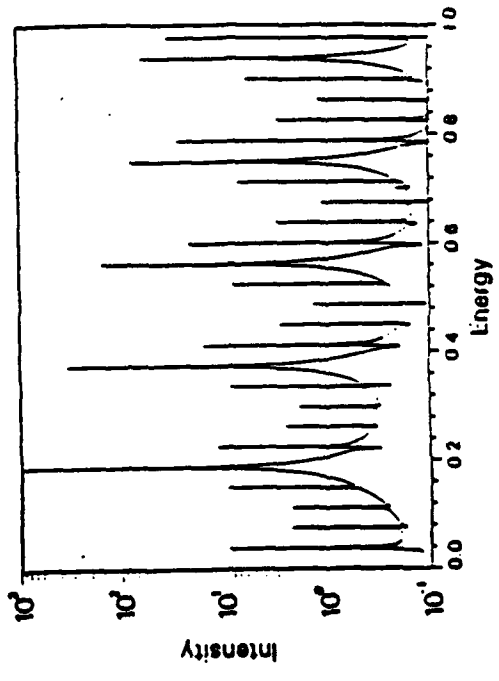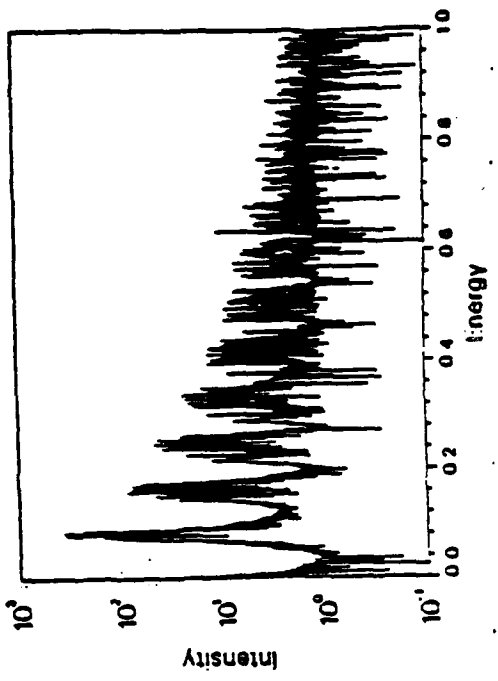
Fig. 4. Uncoupled and coupled spectra for a case having Z=83.
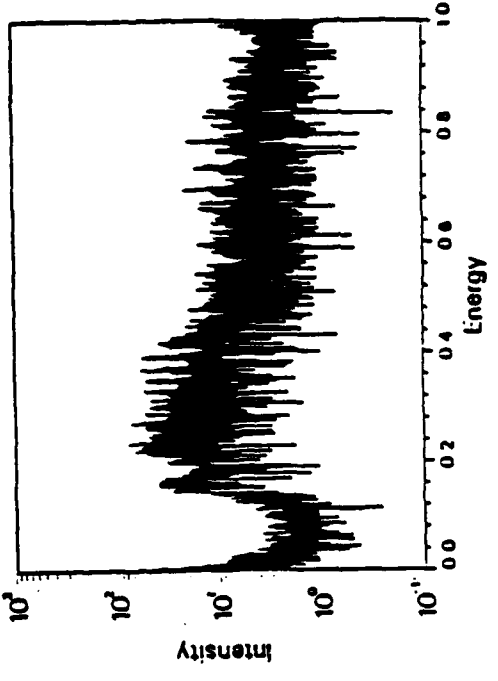
Notes added for clarification:

The parameter a_n has units of length; it physically affects the "sharpness" of the well's slope. A three-dimensional phase space is used to find initial starting conditions. The model must use these starting conditions; we examine only planar trajectories in this version of the model. The values of r in Figs. 1 and 2 depict the spatial extent of trajectories as discussed in the text. We have observed no "locking" of nucleon and electron orbits in this model; the chaotic behavior is indicated by the power spectrum broadening.

31

APPENDIX B

DYNAMICS OF A COUPLED NUCLEAR-ELECTRON MODEL IN AN INTENSE LASER FIELD

A. INTRODUCTION

This appendix documents a presentation on the dynamics of a coupled nuclear-electron model in an intense laser field. The main text of this report is a convenient description of the Presentation, the proceedings is not available as of the time of the preparation of this summary. In particular, the main text describes the information on pages 40 to 48. The first presentation at the Annual Meeting of the Optical Society of America in October 1987, at Rochester, was given in the session on Ultra-High Power Laser Matter Interactions; the Summary and Abstracts are taken from the issue of *Optics News* (September 1987) and Technical Digest (OSA Technical Digest Series, Vol. 22, 1987). The abstract summary from a second talk given at the American Physical Society International Laser Science Conference in November 1987 in Atlantic City in the session on Novel Lasers and Devices is taken from the Bulletin of the American Physical Society as it is printed there.

B. SUMMARY AND ABSTRACT FROM THE OSA MEETING

1. Session Summary

Ultra-High Power Laser Matter Interactions (Chairman: Dr. Reiss)
Dynamics of a Coupled Nuclear-Electron Model in an Intense Laser Field
F.X. Hartmann (IDA), J.K. Munro, Jr., and D.W. Noid (ORNL)
Institute for Defense Analyses/Oak Ridge National Laboratory.

Energy transfer processes in single particle coupled nucleon-electron models interacting with an intense laser field are studied using quantization of the coupled classical Hamiltonian.

2. Abstract

Energy transfer processes in a simple single-particle coupled nuclear-electron model interacting with an intense laser field are studied. In our model, an excited valence proton is bound as an independent particle in a Woods-Saxon potential--its dynamics are characteristic of nuclear motion in the Blatt and Weisskopf single-particle approximation. The electron is bound to the nuclear core in a non-relativistic[1] treatment by a Coulomb potential--its dynamics are characteristic of single-particle electronic transitions. Initial conditions for the classical trajectories are chosen to be states of the separable Hamiltonian, and the Bohr quantization condition is applied. The spectral analysis method[2] is then used to calculate both transition intensities and frequencies for coupled electron-nucleon quantum mechanical transitions. This approach is particularly useful in treating perturbations on the coupled spectra. We have reported[3] instances of strong coupling and chaotic motion in a simple model having extreme ionizations--we report here on dynamics of this model system in the presence of an intense laser field.

C. PRESENTATION

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[1] A misprint appears in the original abstract, "non-relativistic" in the original abstract should be replaced with "relativistic."

[2] D.W. Noid, M.L. Koszykowski and R.A. Marcus, *J. Chem. Phys.* **67**, 404 (1977)

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DYNAMICS OF COUPLED ELECTRON-NUCLEON MOTION IN A LASER FIELD

F. X. HARTMANN, K. K. GARCIA
IDA
J. K. MUNRO AND D. W. NOID
ORNL

35

Summary

Energy transfer processes in single particle coupled nucleon-electron models interacting with an intense laser field are studied using quantization of the coupled classical Hamiltonian.

36

Internal Gamma Conversion and its Inverse

Interactions between electronic transitions of an atom and nucleonic transitions of its nucleus are of interest in the excitation of nuclei from ground states to low-lying excited states or the excitation of short-lived states from long lived isomeric states.

Such interactions can proceed via non-radiative energy transfer between the excited electronic state and the initial nuclear state.

isomeric state

lower lying state

Electronic   Nucleonic
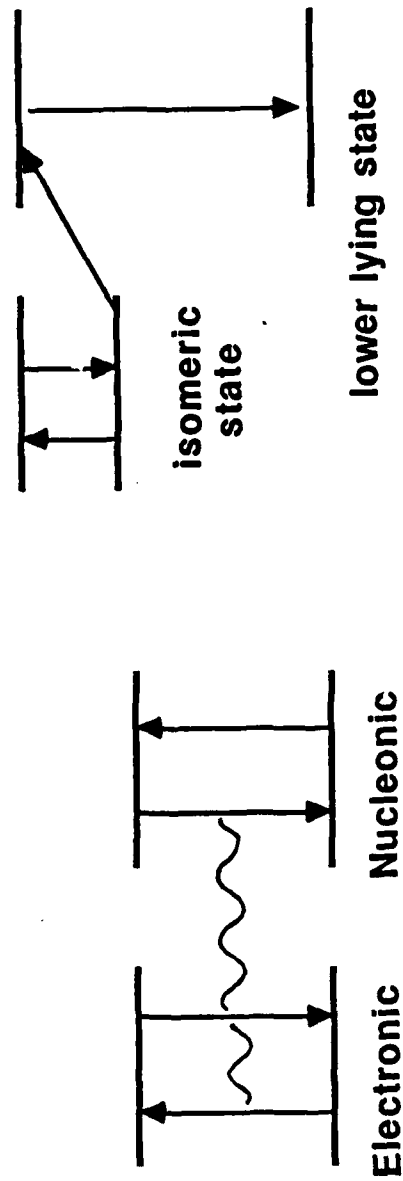
Electronic Excitation of Nuclear Ground States

Experimental

1) 235 U   - Goldanskii and Namiot

2) 235 U   - Izawa and Yamanaka

3) 237 Np - Saito, Shinohara, Otozai

4) 189 Os - Otozai, Arakawa and Morita

Theoretical

1) Outershell Electron Oscillations - Biedenharn, Baldwin, Boyer, Solem

2) Interlevel Transfer - Solem, Rinker

3) 235 U Separation - Morita

4) K shell ionization Bremsstrahlung - Saito, Shinohara, Miura, Otozai

5) Nuclear Excitation - Okamoto

6) Classical Approaches -- with Noid, Koszykowski

7) Strong Laser Approaches -- Berger, Gogny and Weiss

Schemes for Excitations from the Nuclear Ground State
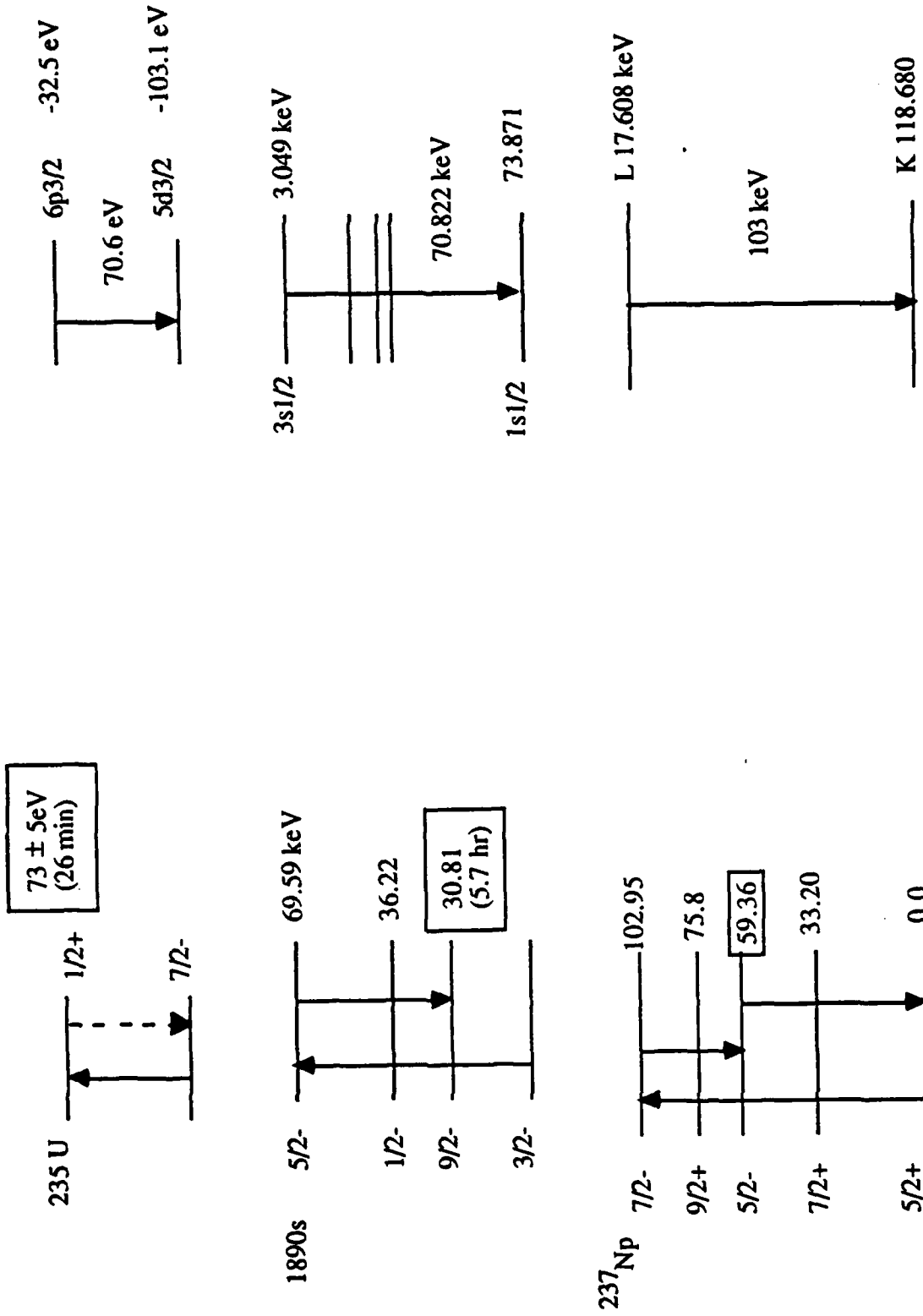
Hamiltonian for the Model

$$H = \frac{\tilde{p}_n^2}{2m_n} + V_0\left\{1 + \exp\left[(r_n - R_0)/a_n\right]\right\}^{-1}$$

$$+ \frac{\tilde{p}_e^2}{2m_e} - \frac{e^2\eta(z-k)}{r_e} - \frac{\eta k e^2}{|r_e - r_n|} + H_{laser}(t)$$

$$m_0c^2\left[\sqrt{\frac{p^2 + m_0^2c^2}{m_0^2c^2}} - 1\right]$$

relativistic kinetic energy

$$\dot{p} = -\frac{\partial H}{\partial x} \qquad \dot{x} = \frac{\partial H}{\partial p} = \frac{pc}{\sqrt{m_0c^2 + p^2}}$$

40

Summary of the semi-classical approach

① Set up Hamiltonian

② Establish well characteristics for starting points

③ Run trajectories to understand dynamics and frequencies of motion

④ Add time-dependent laser term

⑤ Choose E and ω with 64 phases evenly distributed around 2π for the laser and compute ensemble energy averages
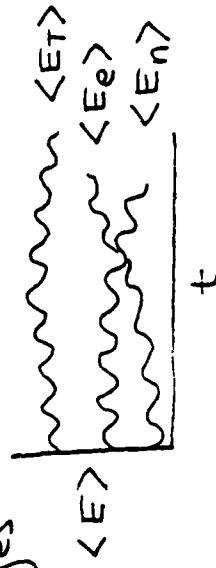
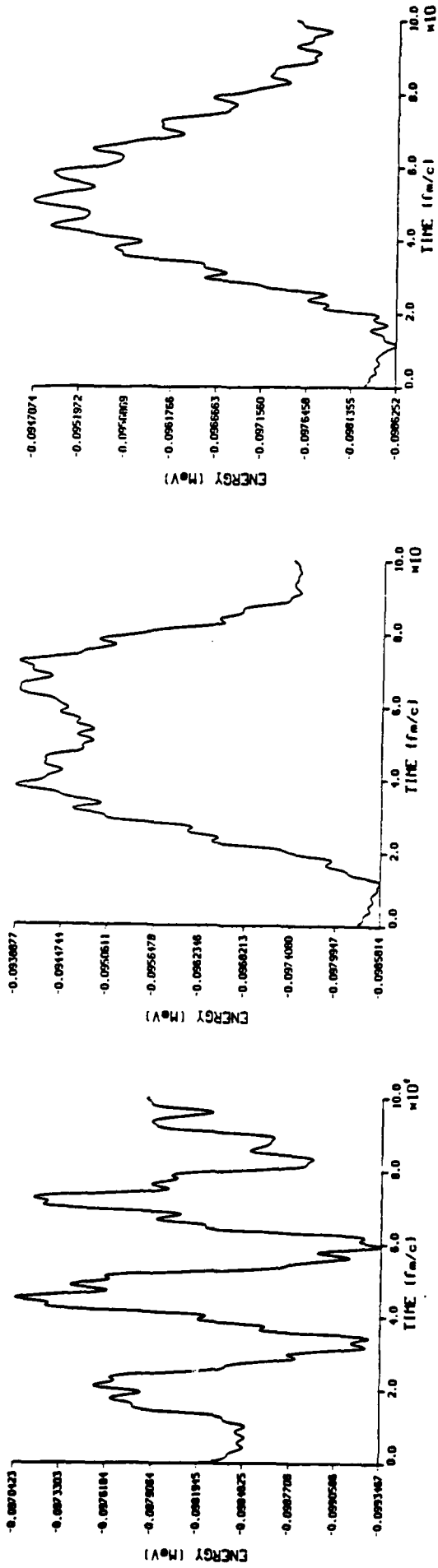⑥ Look at ΔE (energy transfer) as dependent on ω
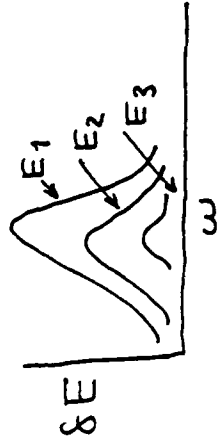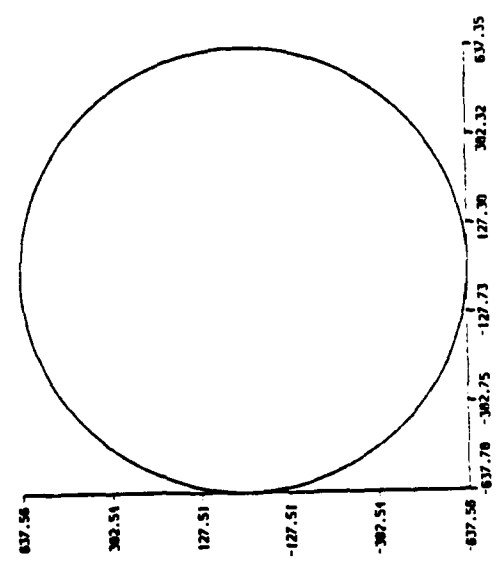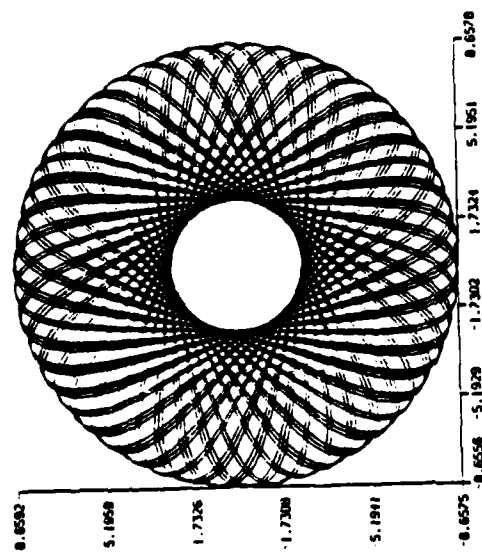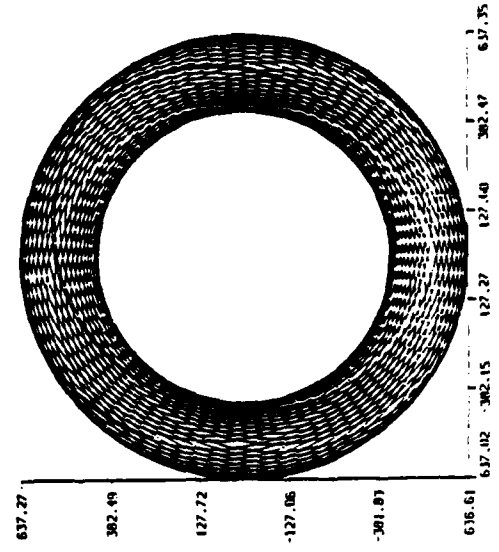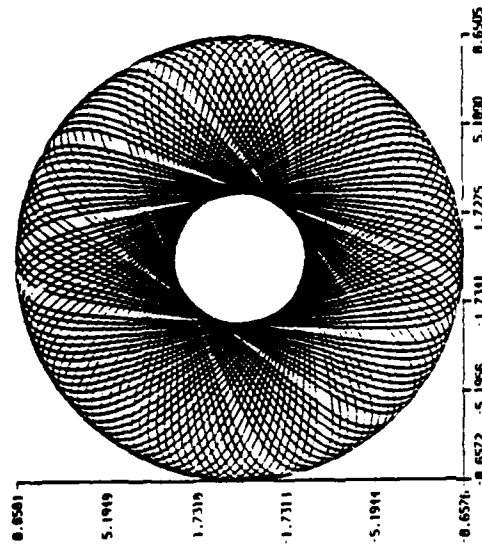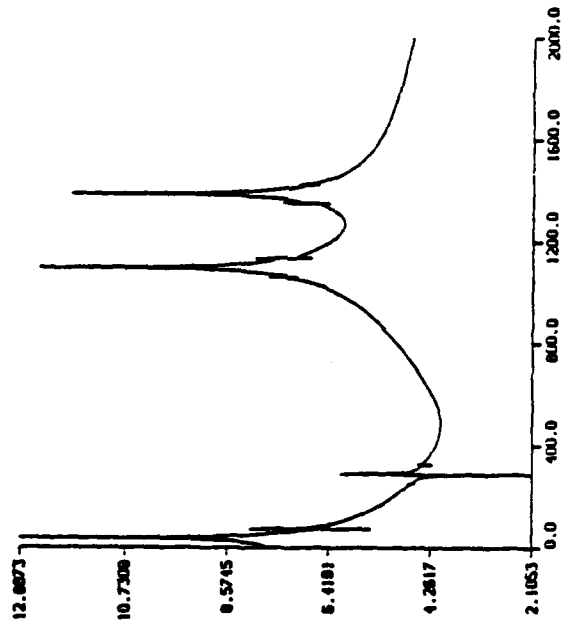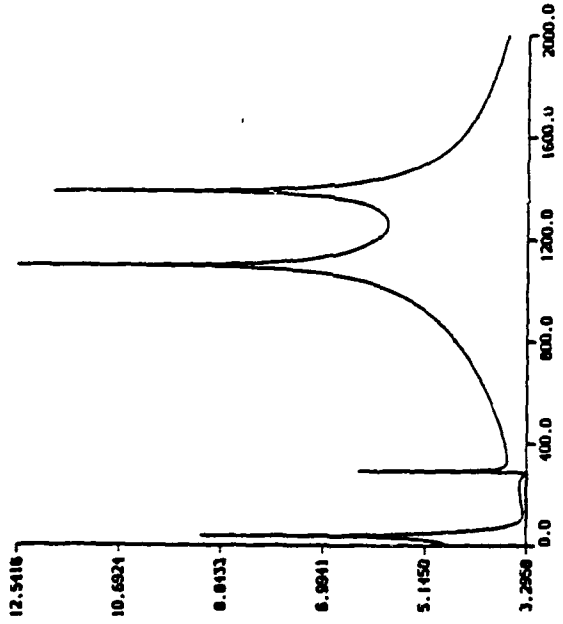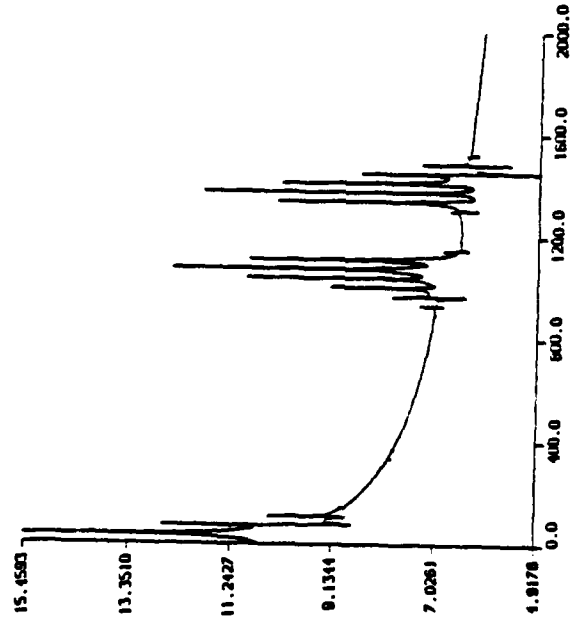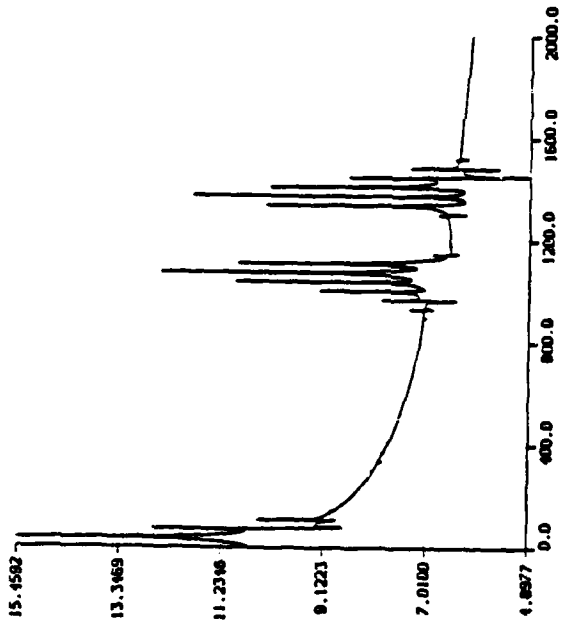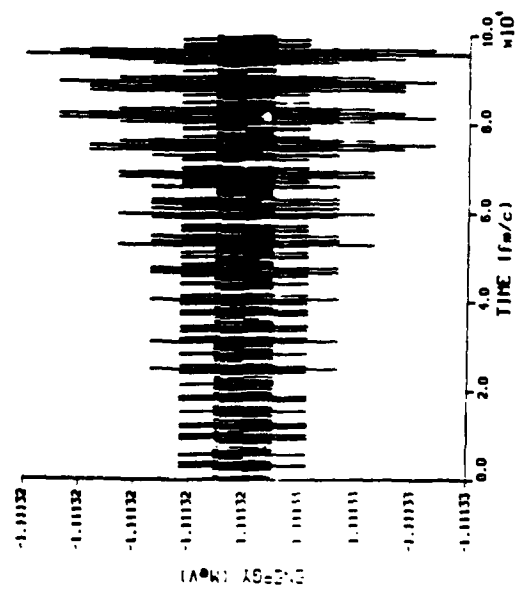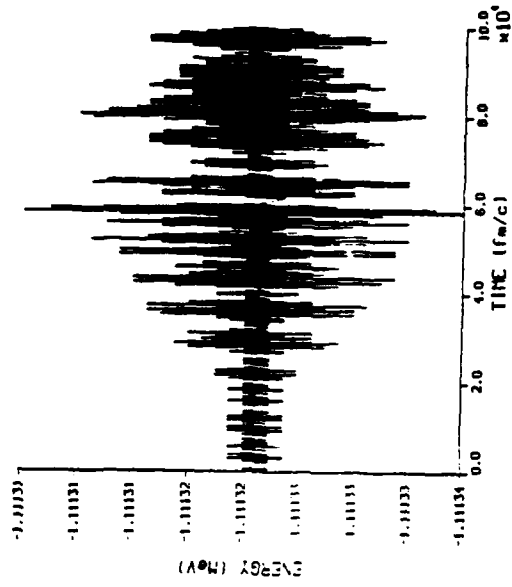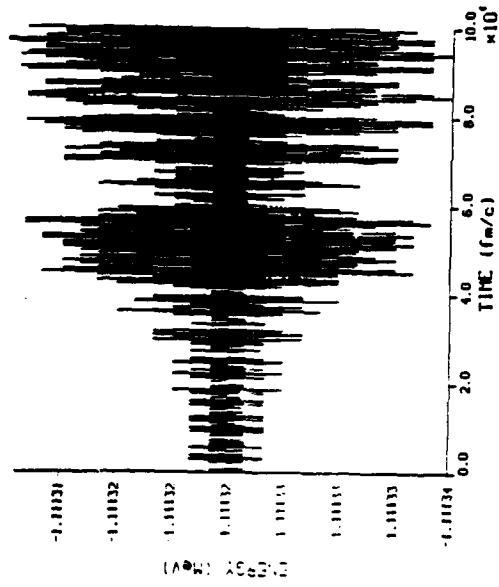

41

Trajectories

42

Basic Frequencies

43

Ensemble-averaged energy as a function of time in the laser field.
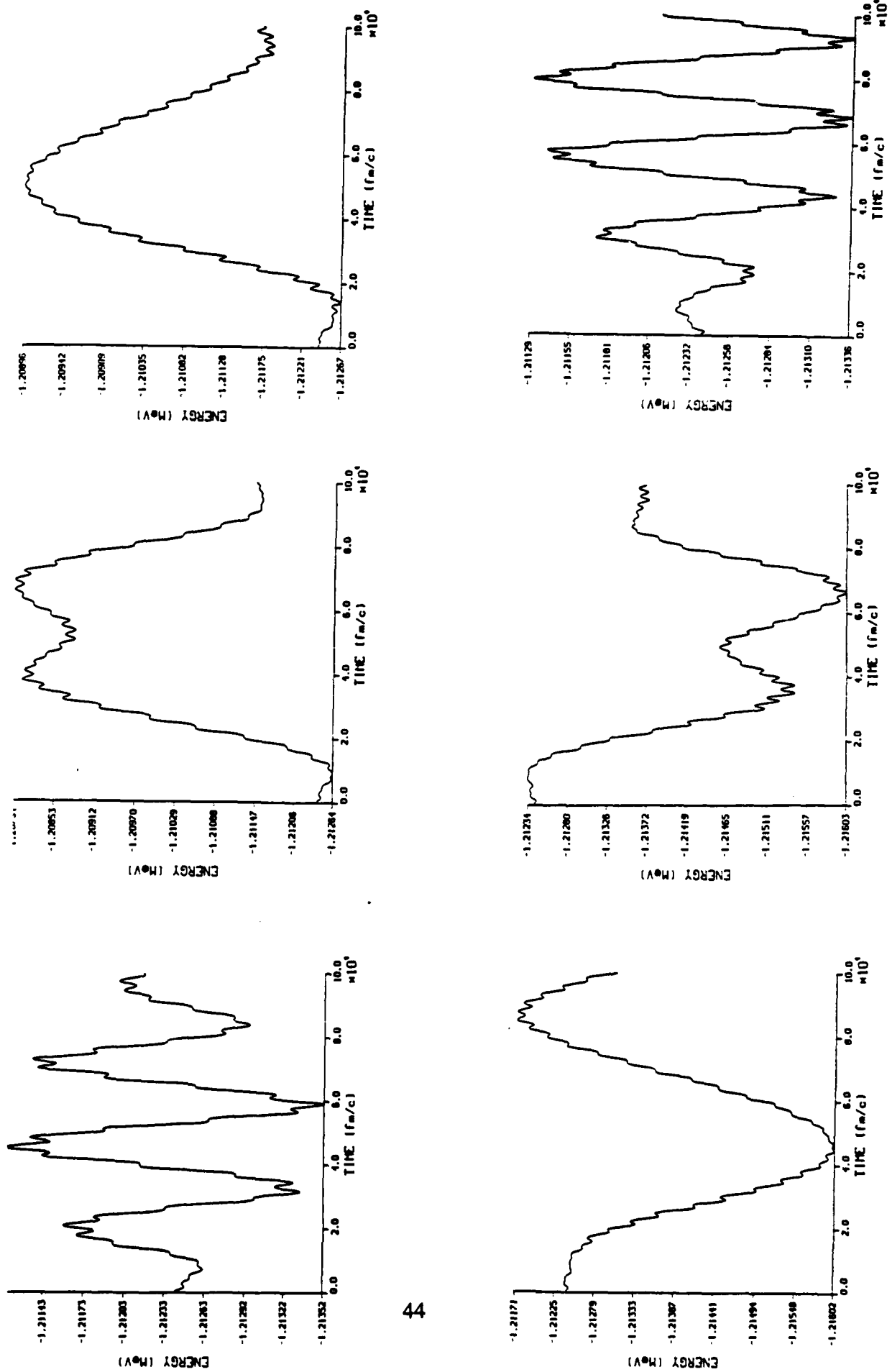
Ensemble-averaged energy as a function of time in the laser field.
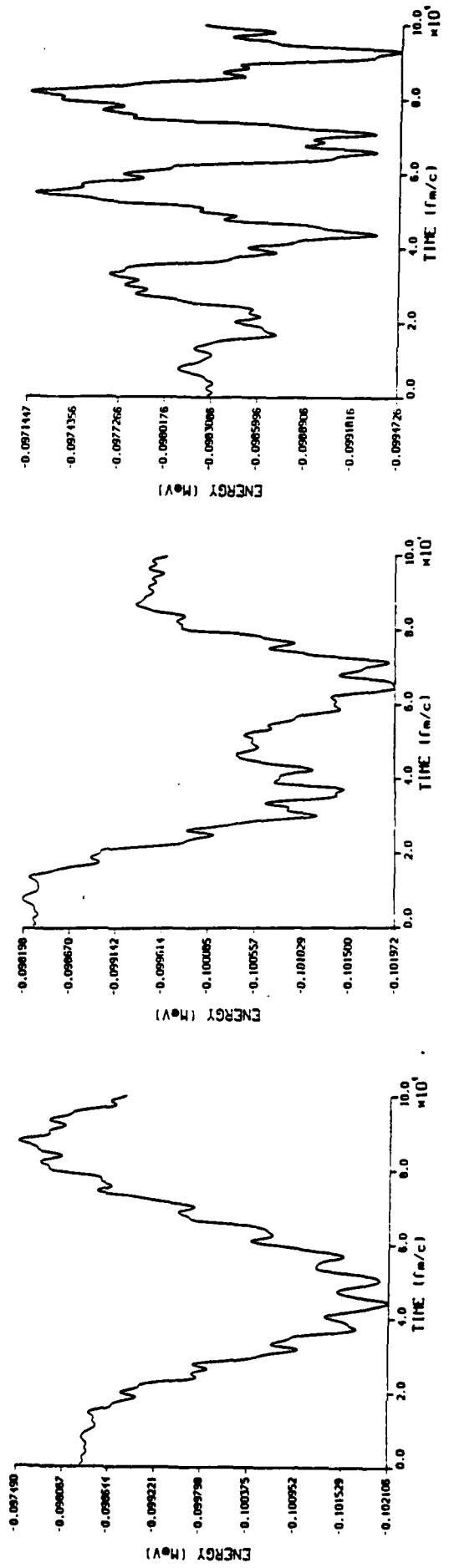
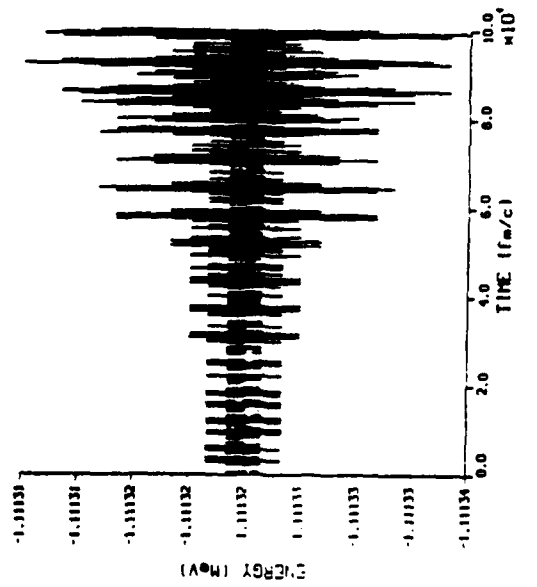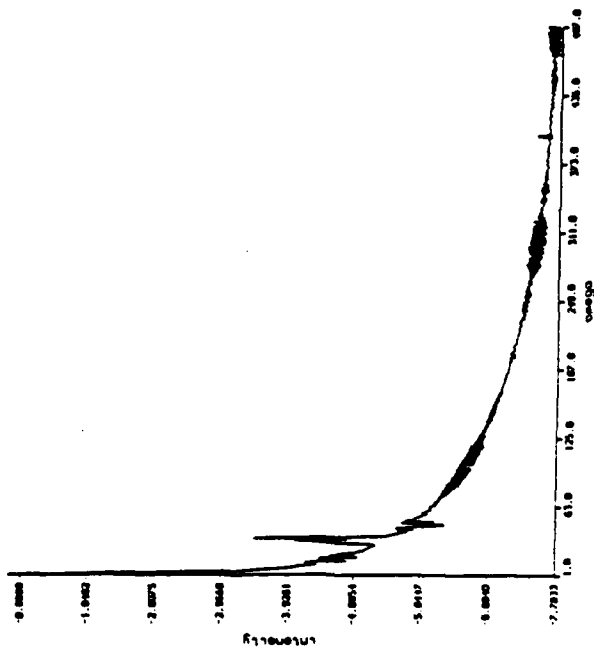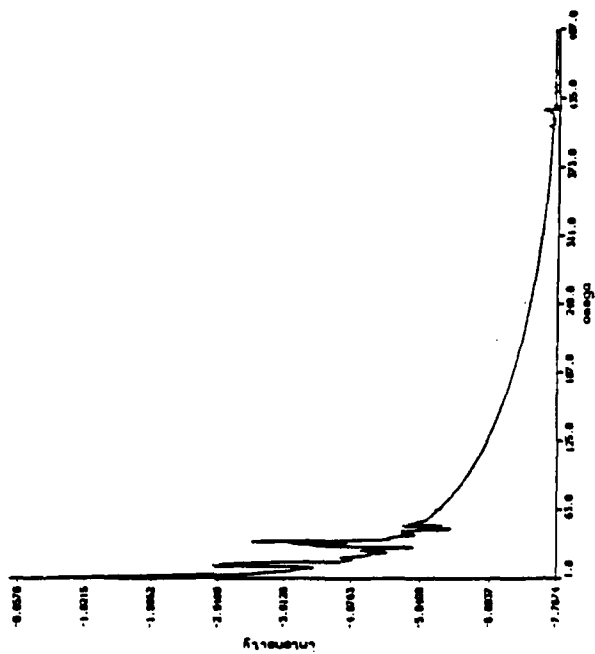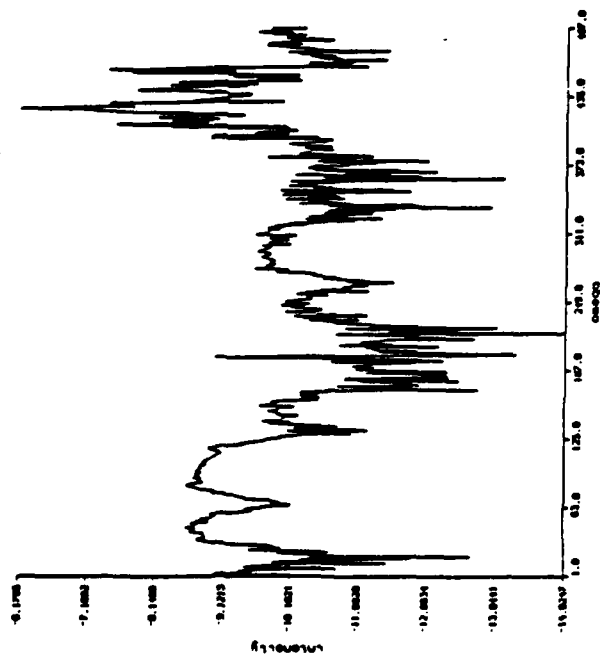# Ensemble-averaged energy as a function of time in the laser field.

Basic Frequencies
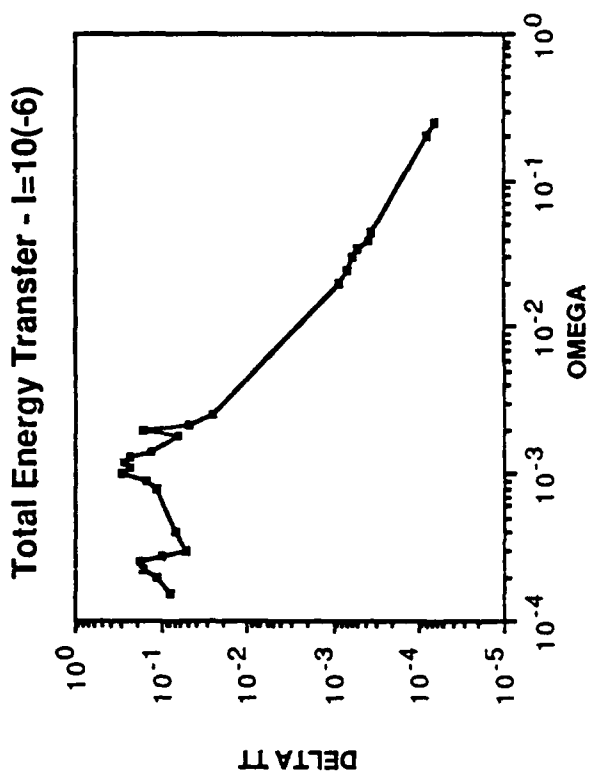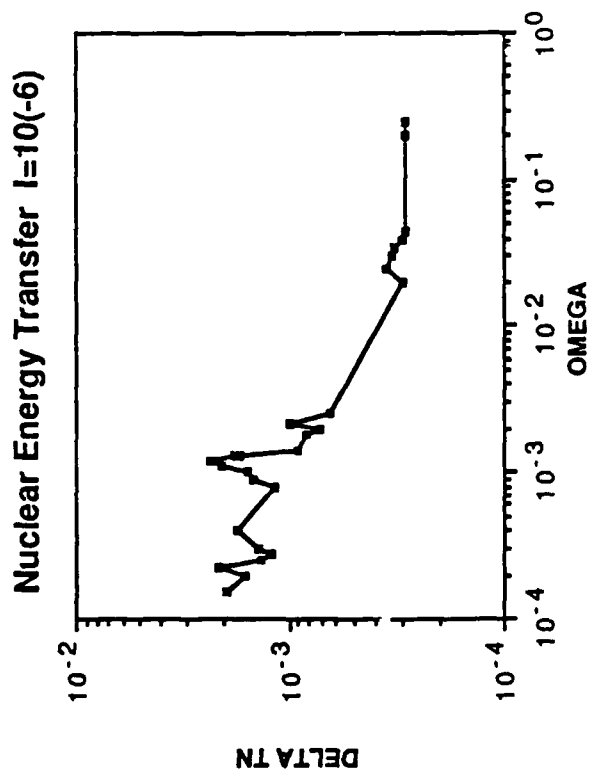

47

Frequency and Intensity Dependence of the Energy Transfer

Nuclear Energy Transfer I=10(-6)

Electron Energy Transfer I=10(-6)

Total Energy Transfer - I=10(-6)

Frequency and Intensity Dependence of the Energy Transfer

Intensity Dependence of Energy Transfer

Energy (x64)

Intensity at w=0.0012

TE
TN
TT
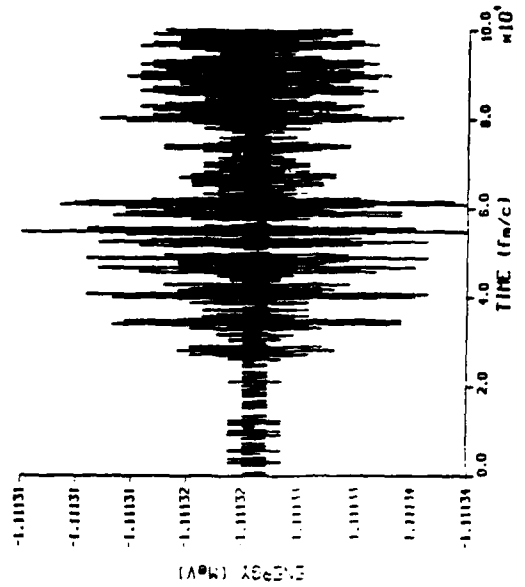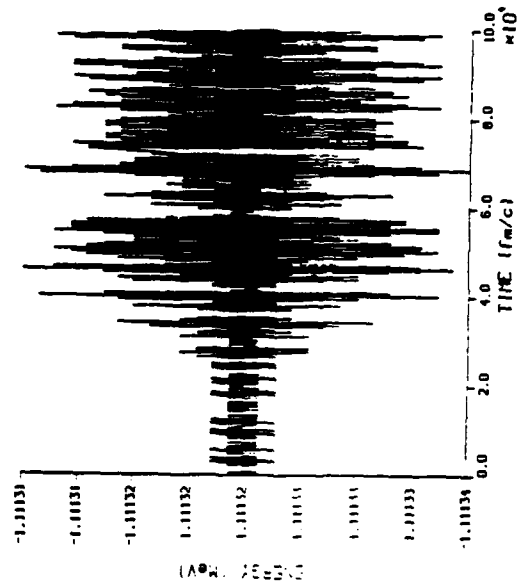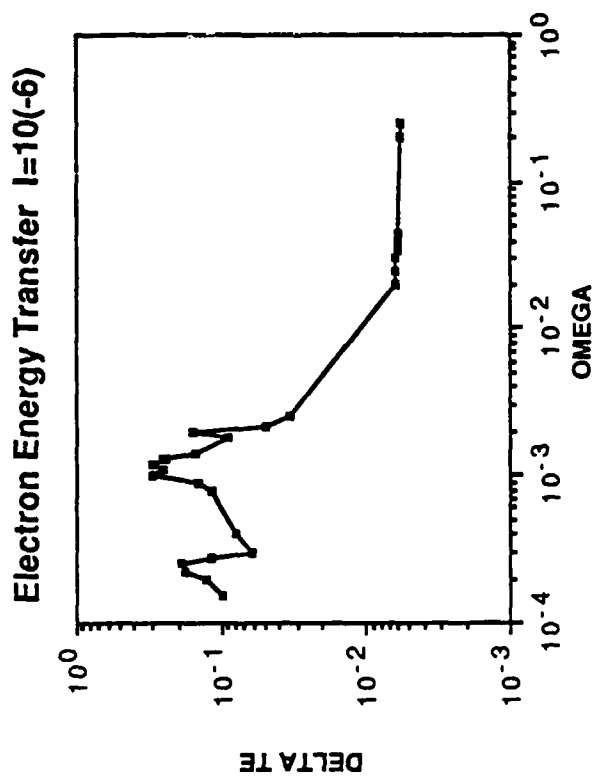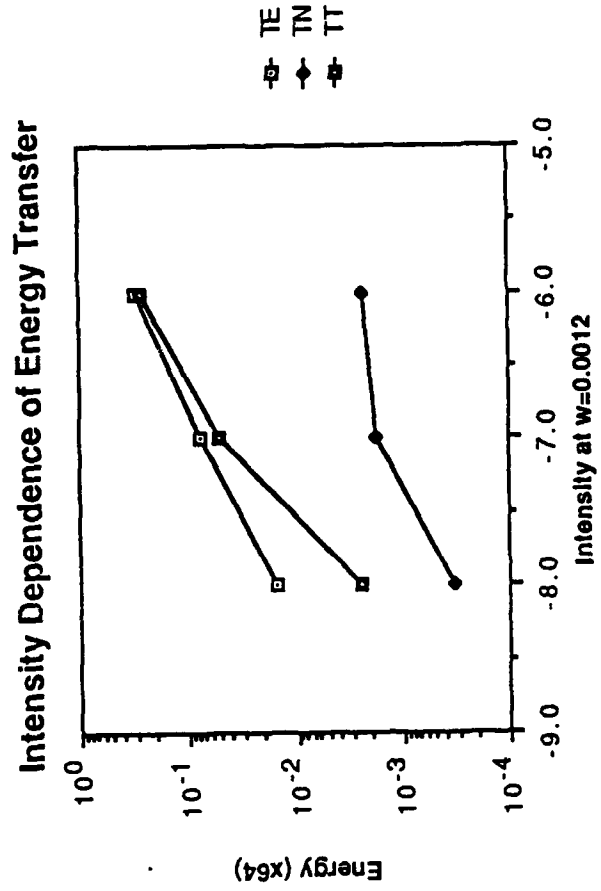
Summary of Approach

	Change in Nucleon Energy ΔNE	Change in Electron Energy (MeV*) ΔEE
No laser non-rel.	0.0000924	0.0000063
no laser rel.	0.00028	0.0059
$\omega = 12 \ (10^{-8})$	0.00034	0.0152
$\omega = 12 \ (10^{-7})$	0.0017	0.075
$\omega = 12 \ (10^{-6})$	0.0023	0.29

• Summed for 64 trajectories.

50

Conclusions

1) An approach to exploring non-radiative energy transfer in coupled systems and in intense laser fields has been set up and used to study energy transfer in a single-particle electron and single particle nucleon models.

2) The nucleon model sets the scale of the frequency response of the coupled system--currently this is too high for practical applications. The nucleon model can be scaled to examine lower frequency transitions and collective motions included.

3) The electron part of the model basically describes a tightly found electron and with suitable changes can be extended to treat outer shell electrons and collective motions (in the laser field).

4) Energy transfer in this system not only depends on ω but also on intensity. Electrons closest to the nucleus have greater coupling, electrons furthest are most affected by the laser. This aspect is interesting to pursue.

D. ABSTRACT FOR PRESENTATION AT THE APS ILS-III MEETING

Dynamics of Coupled Electron - Nucleon Motion in a Laser Field,[4]

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A simple model of coupled electronic and nucleonic independent particle motion is used to study energy transfer processes in the presence of a laser field. The complete classical Hamiltonian of the model includes nuclear, electronic and laser field terms. Initial conditions for the classical trajectories are chosen to be states of the separable Hamiltonian. A method[5] to extract spectral information from the trajectories is then used to calculate both transition intensities and frequencies for the coupled quantum mechanical transitions. Strong coupling and chaotic motions have been reported in the electron-nucleon model for parameters which physically correspond to higher core charges and extreme ionizations.[6] In this paper we report on the dynamics of this system in the presence of a laser field, a problem of interest in nuclear interlevel transfer.[7]

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[4] Work supported by the Dept. of Energy (Martin-Marietta Energy Systems) and the Institute for Defense Analyses.

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