The Livermore experience: Contributions of J. H. Eberly to laser excitation theory

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Abstract: This article summarizes the developing understanding of coherent atomic excitation, as gained through a collaboration of J. H. Eberly with the Laser Isotope Separation Program of the Lawrence Livermore National Laboratory, particularly aspects of coherence, population trapping, multilevel multiphoton excitation sequences, analytic solutions to multistate excitation chains, the quasicontinuum, pulse propagation, and noise. In addition to the discovery of several curious and unexpected properties of coherent excitation, mentioned here, the collaboration provided an excellent example of unexpected benefits from investment into basic research.

OCIS codes: (270..1670) Coherent optical effects

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1 Introduction

Some three decades ago imaginative scientists at the Lawrence Livermore National Laboratory (LLNL), perceiving a need to supply the nuclear power industry with a cheaper supply of enriched uranium fuel than could be obtained with the aging existing gaseous diffusion plants, sought to apply the remarkable properties of laser light to this problem [1, 2]. Whereas the traditional methods of separating large quantities of isotopes made use of the small mass difference between isotopes, the very narrow spectral bandwidth of lasers offered the opportunity to use the difference in atomic spectra of isotopes (a difference that originates not only in the mass difference and the volume difference of nuclei with differing numbers of neutrons, but also in differences of hyperfine structure due to differences in nuclear spin). So began the program known first as Laser Isotope Separation (LIS) and subsequently as Atomic Vapor Laser Isotope Separation (AVLIS) – as distinguished from schemes to use molecules rather than atoms in the vapor. (It should be noted that the concept of LIS did not originate at Livermore, and was being pursued by numerous other groups [3, 4, 5, 6, 7]. More recent references include [8, 9, 10]).

This isotope separation program at Livermore lasted for some three decades, growing from a few scientists into a major component of support for LLNL [11], before being

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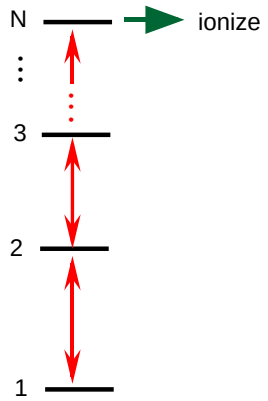

Fig. 1. Schematic diagram of resonant laser excitation of a chain of N energy levels, followed by ionization. Excitation energy increases upwards. Vertical red arrows indicate connections induced by various lasers. Horizontal arrow indicates (ionization) probability loss.

privatized as a part of the United States Enrichment Corporation (USEC). That program ended only after a decision in 1998 by top management of the USEC to cancel totally the support of research and development of this technology. The failure to deploy AVLIS for civilian use was in part a consequence of the very limited market growth in demand for nuclear fuel in the last two decades as well as the very large supply made available (through blending) from weapons grade stockpiles after the Cold War ended; it was not a failure of the science or technology.

The concept pursued at Livermore, with little deviation, was in essence the following simplified process. Start with solid chunks of ordinary uranium, melt and vaporize it under vacuum, form a beam of atomic vapor, and expose the streaming vapor to several coincident beams of laser light. The laser frequencies were carefully chosen to match Bohr transition frequencies along an excitation chain of increasingly more energetic bound states, eventually terminating with an autoionizing state embedded in the photoionization continuum; see Fig. 1. The result of the laser exposure was to photoionize only a chosen isotope (because other isotopes would not be resonant with the lasers). Electrostatic fields would then separate the ions (the desired isotopes) from the background of neutral atoms (the undesired isotopes.)

Very early in the project it was recognized that, in addition to engineering and materials handling challenges, there were many questions of a very fundamental nature that needed to be addressed in order to place the modeling of the separation process on a secure foundation. Indeed, a group of theorists with expertise in chemistry and physics issues (the Theoretical Atomic and Molecular Physics group, or TAMP, headed by Charlie Bender) were assembled, in part to address these. During the startup of the LIS project it was headed by Ben Snavely, who came to Livermore from Eastman Kodak in Rochester. There he had been acquainted with Joe Eberly, and knowing of Joe's ability to ask and answer very fundamental questions about laser excitation, Ben hired Joe as a consultant. Joe's scientific points of contact at LLNL were Bruce Shore and Mike Johnson. Almost from the beginning of this consultantship, which typically involved two visits a year to Livermore, the LIS project made funds available to support basic research at Rochester. You have to understand that even two decades ago the organization of our national laboratories was very different than it is now. Places such as LLNL were able to make funding grants for basic research at the discretion of program leaders, and under the enlightened leadership of Ben Snavely and his successor Jim Davis (1974 to 1986), there was ongoing support for post-docs and students at Rochester. Indeed,

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Joe recently pointed out to BWS that his most widely cited reference [12] was financed with support from LLNL. In that paper they predicted and explained the "collapse and revival" of population oscillations of a two-state atom in a single-mode cavity, one of the few definite ways in which the discreteness of photons is observable. Though unrelated to any application at LLNL, that paper has had a significant impact on all of quantum optics. Sadly, a change of management at Livermore brought an abrupt and unforeseen termination to this work, and the consultantship, in 1987. This document reviews some of the things we learned during this collaboration, doing research that formed the core of a major treatise on coherent excitation, where more details can be found [8]. In the words of our honoree,

> *"Recall those wonderful days in Livermore when we knew only epsilon more than anybody else there, but epsilon was enough."*

Unfortunately it is not possible, in the short space available here, to present a comprehensive discussion of the topics in this article. We aim primarily to summarize work at LLNL on coherent excitation, all of which was influenced by Joe. We have cited all of the joint publications with Joe and LLNL. For further details, and many more references to related and historically significant works, readers should consult the aforementioned book [8].

2 Beginnings

The concept of selective multistep photoionization that formed the basis for the LIS scheme at LLNL was very simple: one chose a set of laser frequencies that would provide a resonant excitation chain from the ground state into the photoionization continuum. The precise wavelengths would be determined by experiment (and would be held in secrecy). The basic challenge for theory was to predict the intensity of the various lasers, given the measured oscillator strengths, such that the ionization would proceed selectively and approach completeness asymptotically - all at least cost.

At that time theorists dealing with radiation effects on vapors gained their understanding from studying textbooks aimed at astrophysicists who sought to model the passage of radiation through stars. The relevant equations expressed the rate of change in atomic populations as being proportional to the energy density (or the flux) of radiation [8]. The proportionality coefficients were the Einstein-Milne B coefficients (or cross sections). It was these radiative rate equations that were used in the first modeling of laser excitation in the LIS program by Rich Davis.

However, even undergraduate physics majors at that time had encountered the time-dependent Schrödinger equation, and knew it as the basic equation governing time evolution at the most fundamental atomic level. This equation differed very significantly from the Einstein rate equations: Rather than deal with linear differential equations for probabilities, it dealt with differential equations for probability amplitudes. Only after squaring these amplitudes did one obtain the observable probabilities. The reliance on amplitudes leads to the possibility of both constructive and destructive interference effects, and so it is possible to obtain very different results from the two approaches.

Surprisingly little had been done with the time-dependent Schrödinger equation at that time. Apart from some special cases mentioned below, it was regarded primarily as a means of deriving rate coefficients by means of time-dependent perturbation theory and Fermi's famous golden rule.

One of the first fundamental questions that had to be addressed when considering laser-induced atomic excitation was: what equations would describe the time evolution of an illuminated vapor, as it would be used in the LIS project? Rate equations or the

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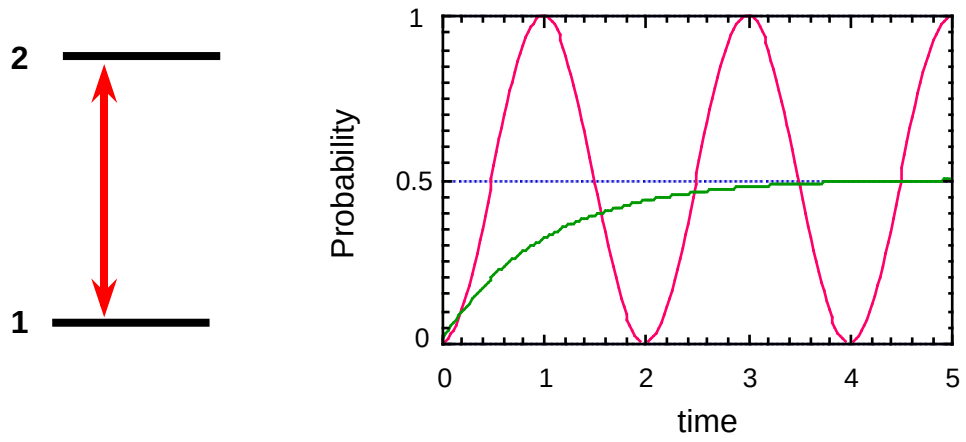

Fig. 2. Time dependence of resonant excitation probability $P_e(t)$ for lossless two-level atom. Monotonic green curve is for rate equations, oscillatory red curve is for the time-dependent Schrödinger equation (oscillation frequency is the Rabi frequency).

Schrödinger equation? Stated somewhat differently, were we to deal with (incoherent) multiple photon absorption or (coherent) multiphoton absorption? As we now understand from numerous textbooks on quantum optics and laser physics, these two types of equations are extreme cases of a formalism that can be dealt with by means of density matrices [8]. When excitation occurs by means of coherent radiation (laser light) then the Schrödinger equation comes close to the correct description. With incoherent light (the astronomical sources or plasma sources) then rate equations are suitable. But this was not so clear in those early days.

In one of his early visits to the LIS project, Joe participated in a lively discussion of the significance of coherence for LIS, organized at the suggestion of Jim Davis, who professed skepticism about the need to consider coherence (and some of the curiosities of the Schrödinger equation) in any practical separation program. The disputants at that time included, besides Joe, Bruce Shore, John Garrison, Mike Johnson, and a few others. Joe gave a masterful lecture on the two-level atom, starting from the most basic ideas of probabilities and the Schrödinger equation, going through what is now very traditional introduction of the rotating wave approximation (RWA), and ending with sinusoidal Rabi oscillations of populations. (These contrast with the monotonic growth of populations illuminated incoherently, as predicted by rate equations; see Fig. 2). All of this was still new and novel at that time. Davis was unconvinced, however, and on the spur of the moment offered a challenge, to be known as The Davis Cup, to anyone who could convince him that coherence was important in his job as leader of the LIS project.

Eventually, largely as the result of several years of collaboration between Joe and Bruce, Davis acknowledged that it was indeed important to base modeling on the Schrödinger equation rather than rate equations, and he graciously made an award of The Davis Cup (to BWS). The original cup was simply a styrofoam coffee cup (probably the one used by Davis himself that day), but eventually it became a heavy vessel of machined brass, mounted on a mahogany base.

3 The Excitation Chain

Already in 1976 Joe had wondered about a very basic issue concerning a chain of excitations, such as those indicated in Fig. 1. It was known that, in a two-state excitation

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followed by ionization, a sufficiently high ionization rate would damp out the Rabi oscillations and give results that were predictable from rate equations. What would happen if there were a chain of excitations, leading to a final ionization? Would an increase of the ionization rate cause the entire chain to lose characteristics of coherent excitation? Or would the incoherence be confined to the final step?

With his then graduate student Jay Ackerhalt, Joe answered these questions as follows: incoherence affects only the final stage at first, but as that stage becomes incoherent, then it can affect the preceding stage [13]. Ultimately one can have a completely incoherent sequence, in which the excitation rates proceed faster and faster as the population rises along the excitation sequence. Interesting though this regime is, it turned out not to be an optimum for purposes of isotope separation.

4 Jay Ackerhalt

One of the early benefits to Livermore from the collaboration with Joe was the arrival at LLNL of Jay Ackerhalt in 1976, fresh from his PhD work at Rochester where he had been Joe's first graduate student, and had devised an elegant way of treating spontaneous emission by means of a source field and Heisenberg equations of motion. Jay was only briefly at LLNL, before moving to his career at Los Alamos, but his work at Livermore, including his code BICENT, helped elucidate the connections between rate equations and the Schrödinger equation [13, 14, 15, 16]. Though his stay at LLNL was brief, he participated in many enjoyable discussions at Livermore. Joe and Bruce were amongst the speakers at the special memorial session for Jay held in September 1992 at the Institute of Laser Science Convention.

5 The Three State Atom

Although our first theoretical concerns were with two-level atoms (Joe was, after all, renown for co- authoring with Les Allen the classic textbook on two-level atoms [17]), very soon we began considering the next logical extension, the three-level atom. It will seem quite curious to readers today, but at that time the three-level atom had not been subject to very much scrutiny (examples of other work include [18, 19], see [8]), and we published a paper in which we described some of the most elementary properties of the three-state system, subject to steady radiation fields [15]. In particular, we presented analytic solutions for the probability amplitudes, something that may seem obvious in retrospect but was, at the time, still publishable. This analysis based on the Schrödinger equation, and its implied complete coherence, was followed by an elucidation of the changes in excitation that would be produced by incoherence, as described by a density matrix [14].

Although this step, from two states to three, was a significant advance at that time, we had no idea of the remarkable effects that would much later be discovered when sequential pulses act on a three-level atom. The effects of counter-intuitive pulse sequences came to be recognized only after work at Rochester involving Fuk Hioe, Yossi Oreg and Joe [20].

6 The Lambda System: Dark States

One of the most remarkable novelties of the three state atom became obvious during our numerical modeling of three-state excitation. Suppose you have a two state atom, resonantly excited by a steady beam of radiation. Suppose further that the excited state can ionize, perhaps by an additional steady photoionizing field. Then a long steady pulse will eventually completely deplete the initial state, converting all the atoms into ions. This is pretty obvious, though there are some subtleties that may not be obvious at first.

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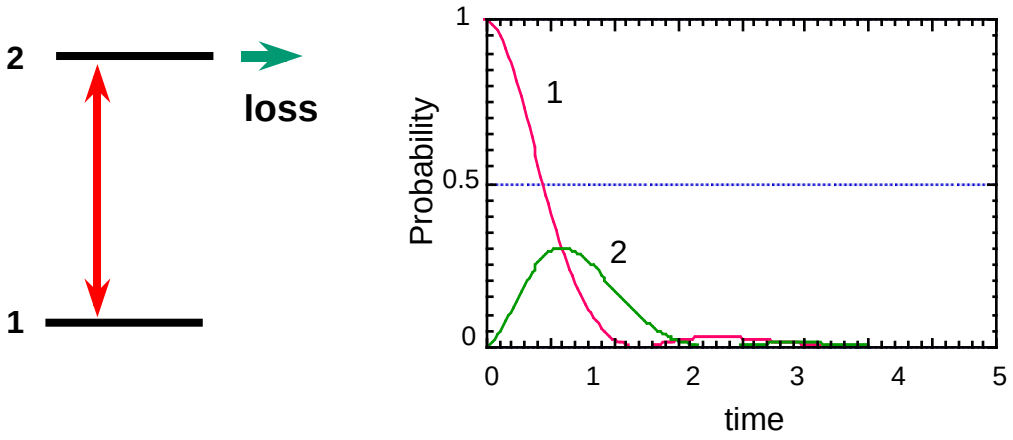

Fig. 3. Time dependence of resonant excitation probabilities $P_n(t)$ for two-level atom with loss from upper level. Curves are marked with level number n. Times are in units of the loss rate; the Rabi frequency is twice this rate. The diagram at the left shows the laser-induced excitation linkage between the two levels. All population is eventually lost.

(For example, if the photoionizing radiation is made very intense, it will actually slow the rate of ionization.) What is quite unexpected is that if you have a second low-lying state, initially populated, and you link this state with the same ionizing excited state by means of a second resonantly tuned laser field, you will not obtain complete ionization. No matter how intense the two excitation fields, and how long you wait, some population will remain in the two low-lying states. Figures 3 and 4 illustrates the dramatic change produced by adding a second leg to the excitation linkage, in the so-called "lambda" configuration.

Nowadays it is understood that this un-ionized population is trapped in a coherent superposition state, a so-called "dark state" or "population trapping state" [21]. But our first encounter of this phenomena was quite unexpected [22]. Carlos Stroud subsequently pointed out to BWS that this coherence had been discovered not only during work with his students Rich Whitley and Bob Gray [23, 24] but some years before, by Arimondo and Orriols [25] who nowadays get the credit for observing this population trapping effect in optical transitions. A rather simple example of exactly this coherent effect is to be found in the example of coordinate choices for treating excitation involving degenerate magnetic sublevels, identified by magnetic quantum number M, of transitions between states of well defined angular momentum J. Figure 5 illustrates this.

Population trapping states are an essential prerequisite for the success of various schemes for transferring population adiabatically, as in the Stimulated Raman Adiabatic Passage (STIRAP) process [26, 27]. The foundation for this line of work was laid at Rochester, and described in a paper by Eberly, Hioe and Oreg [20] who pointed out how adiabatic states (i.e. instantaneous eigenstates of the Hamiltonian), in multilevel systems, can be used to carry population between specified physical states by means of suitably crafted laser pulses. The significance of this theoretical work became evident with the experimental work of Klaas Bergmann and his co-workers; for a review see [28, 29]. Here too, significant questioning by Joe brought new insights into this process [30].

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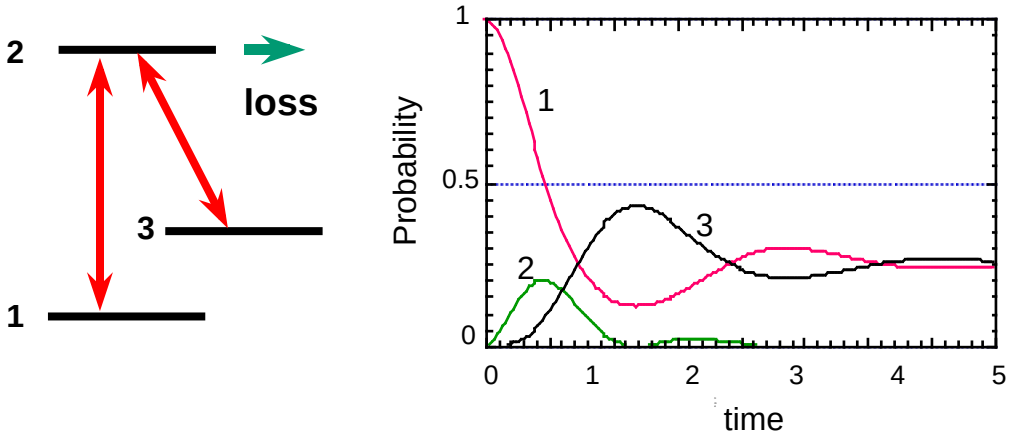

Fig. 4. Time dependence of resonant excitation probabilities $P_n(t)$ for three-level lambda system, with loss from level 2, for population initially all in level 1. Curves are marked with level number n. Times are in units of the loss rate; the Rabi frequencies are each twice this rate. The diagram at the left shows the laser-induced excitation linkages between the three levels. After a long time one fourth the population resides in level 1 and another fourth in level 3; only half has been lost.

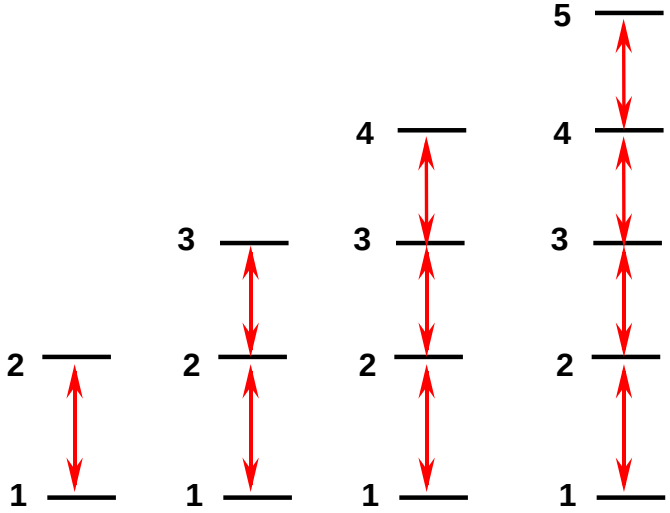

Fig. 5. Example of linkages of linearly polarized light between magnetic sublevels of a transition between angular momentum $J = 1$ and $J = 0$. (a) Using a coordinate system in which the z (quantization) axis lies along the electric field. (b) Using a coordinate system in which the electric field direction is taken as the x axis, and the light is considered a coherent superposition of right- and left-circular polarization. This is an example of the lambda system of Fig. 4; it is equivalent to the linkage of (a).

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7 Solving the Schrödinger Equation

In the earliest days of our involvement with the theory of coherent excitation, there was some effort to apply the very elegant and elaborate machinery of Green's functions and resolvent operators to find formal expressions for time dependent probability amplitudes. In retrospect it is clear that such an approach would have been of little use in the LIS project. A remark by Bernie Lippmann (of the Lippmann-Schwinger equation, then a consultant at LLNL) started us in a new and ultimately much more promising direction: he asked "Why not just solve numerically the set of coupled ordinary differential equations?"

The numerical approach, supplemented by analytical insights whenever possible, became the basis for our theoretical work. It was a simple matter to integrate the time-dependent Schrödinger equation (even on the mainframe batch-process computers that preceded the personal computers of today) and observe the population changes. We became computational "experimenters" [31], an approach that Joe has often used since then to discover new physics.

8 Analytic Solutions for the NState Atom

When one treats an excitation sequence in which a set of laser fields link a succession of nondegenerate energy levels in a ladder-like arrangement (an excitation chain), then each energy state is coupled to at most two other states, and the Hamiltonian forms a tri-diagonal matrix with elements only along the diagonal and the two adjacent bands. In the commonly used generalized rotating wave approximation [8], the elements of this matrix vary in time only with the slow change of pulse envelopes; the diagonal elements are differences between summed photon energies and excitation energies (in frequency units these are cumulative detunings), whereas the off diagonal elements are interaction energies (in frequency units these are half Rabi frequencies). In the simplest idealization, the Hamiltonian remains constant in time. Flushed with success at finding analytic solutions to the three-state atom, Joe posed a question: For what number of levels N could one find analytic solutions (for the populations of an excitation chain excited by constant intensities)?

We recognized almost as soon as the question had been posed that there is a special case in which solutions exist for an infinite number of levels, namely the harmonic oscillator. The analytic solution for this model system, not just for constant intensity but for arbitrary pulses, was already well known as one of the soluble problems of quantum mechanics [32].

About this time Iwo and Sophie Bialynicki-Birula came as visitors to Rochester – part of what became a very active Polish Connection that Joe established with scientists at the Polish Academy of Sciences in Warsaw. They immediately recognized how to map the Schrödinger equation for the N-state atom onto equations of 19th century special function theory, thereby obtaining exact analytic solutions [33].

The key to this work was realizing the importance of eigenstates of the Hamiltonian. Although the ultimate concern is with transferring population between the free-field "physical states" (or "bare states") of the atom, the mathematical description of time evolution is greatly simplified by introducing eigenstates of the Hamiltonian ("dressed states" or, for time-varying fields, "adiabatic states"). In the special case of constant laser fields and a chain of excitation linkages, the Hamiltonian is a tridiagonal matrix. For particular choices of laser intensities the elements of such matrices are identical with the three terms of recursion relationships for the classical polynomials of Hermite, Laguerre, Chebyshev and others [33]. Each family of polynomials is associated with a specific set of Rabi frequencies and detunings. Solutions exist both for arbitrary finite

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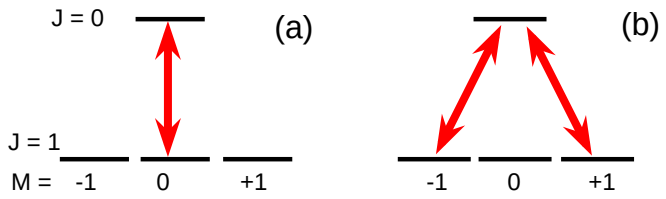

Fig. 6. Examples of resonant N-level linkages in a lossless excitation chain. From left to right $N = 2$, 3, 4 and 5.

N and for the limit of an infinite number of levels.

Although the analytic solutions do offer interesting insights [34] they apply only to steady excitation, and so their usefulness was limited.

9 Periodicities

The most obvious novelty of two-state excitation by a steady coherent field is the exact periodicity of the probabilities, which vary sinusoidally. When the radiation frequency exactly coincides with the Bohr transition frequency, there occurs complete population transfer; the oscillations occur at the Rabi frequency. When the laser is detuned from resonance, the oscillations become more rapid and the excitation becomes less complete, but it remains purely sinusoidal. This behavior contrasts markedly with the monotonic approach to a steady-state value predicted by rate equations for similar steady excitation conditions. In particular, at most half of the population can be excited according to rate equations, whereas the Schrödinger equation predicts periodic complete population transfer. Our work with three-state atoms showed that here too the populations varied periodically with time, though the changes were not described by a single pure sinusoid. Quite naturally there arose the question: is all steady coherent excitation, of an arbitrary N-state chain, periodic?

We proceeded by computing numerical solutions to the Schrödinger equation for an excitation chain having constant intensity fields. More specifically, we examined an N-state excitation ladder (with no ionization loss), resonant at each step and for which all Rabi frequencies were the same or increased like those of harmonic-oscillator dipole moments. Figure 6 shows the linkage patterns for some of these chains. Trial after trial, we increased the number of states in the chain. The four-state chain was not periodic, even for very long times. But the five-state chain was periodic. Perhaps only odd-integer numbers of levels would show periodicities. Or perhaps only when N was a prime number would populations vary periodically. A bit of good-natured wagering took place as we awaited the display of population histories from the computer. If the periodicities were not immediately obvious, then perhaps the computations had not been carried to sufficiently long times.

In the end, theory [33] provided a simple answer to the puzzles presented by the numerical experiments: because the eigenvalues of the Hamiltonian were, in the cases we were studying, not multiples of any common factor, the population histories were sums of incommensurable sinusoids. The result was not periodic. (The five-state atom was something of an exception to this general rule.) The question of periodicities had no immediate application to the isotope separation project, because all excitation would there take place in the presence of population loss to the ionization continuum. Nevertheless the question entertained theorists. Some years later Dick Cook, then a graduate student at Livermore, revisited this question, and pointed out that it would not be difficult to arrange laser intensities so that the Hamiltonian eigenvalues were evenly spaced, and therefore the populations would undergo periodic change. The Hamiltonian was, in

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fact, the familiar one of an angular momentum system – a magnetic moment in a static field [35].

10 Time-averaged populations; multiphoton resonances

In the RWA, the diagonal elements of the Hamiltonian matrix express the (cumulative) detuning of laser frequencies from relevant Bohr frequencies. If you use a separate laser for each transition, then it is possible to select the frequencies such that the diagonal elements all vanish. This is a very useful situation for efficient excitation along the chain. On the other hand, if you have only a single laser, then the detunings are not zero. In particular, for an anharmonic oscillator (such as occurs with a Morse potential) then the detunings form a regular sequence.

With our computational tools for treating arbitrary (but constant) Hamiltonians, we had begun to examine the long-term average populations found in such lossless chains, starting from an initially populated ground state. Studies of anharmonic oscillators showed some very interesting results. The numerical simulations of averaged populations, when plotted on a logarithmic scale to reveal finer details of small population changes, showed remarkable structure as a function of the laser frequency. One could see the anticipated multiphoton resonances as Lorentz profiles whose very narrow widths originated in the very long times required to achieve the multiphoton population transfer from the ground state. But, rather surprisingly, each of these was superposed on a succession of broader profiles attributable to lower-order transitions between pairs of excited states. Although this work benefitted from discussions with Joe, it was published only later, as a part of a textbook [8].

11 Degeneracies

The simple excitation chain, represented by a tri-diagonal Hamiltonian, is an idealization of use to theorists, but of only limited direct application for modeling real atoms, particularly real uranium atoms. In the real world atomic energy levels possess hyperfine structure as a result of the weak interaction between magnetic moments of spinning atomic nuclei and the magnetic fields generated by the moving electrons. An atom with nonzero overall angular momentum J has $2J + 1$ discrete orientations with respect to an arbitrary quantization axis, and each of these magnetic sublevels has equal energy in free space. Although selection rules provide some limitation on possible linkages between atomic basis states, the combination of hyperfine structure and magnetic sublevels greatly increases the complexity of the Hamiltonian matrix [36]. Figure 7 (a) illustrates a simple example of such complexity.

One might think that there would be little hope for any simplification of a general Hamiltonian of this sort. Interestingly, if the various sublevels are degenerate (meaning that one neglects external static magnetic and electric fields and neglects also the small hyperfine splitting) then, as Jim Morris discovered [37], it is possible to introduce a new set of (theoretical) atomic basis states in which the Hamiltonian describes a set of independent ladders. Instead of two degenerate levels, in which the Hamiltonian exhibits a complicated pattern of linkages, one deals instead with the mathematical problem of several independent two-state atoms, and the modeling reduces to application of the well-known two-level atom of Allen and Eberly [17]. Significantly, if there are more lower-energy sublevels than upper-level ones, the independent set of two-state excitations will be supplemented by a number of ground states that have no excitation; they are generalizations of the dark states (population trapping states) of the three-state lambda linkage. Figure 7(b) illustrates this. This interesting Morris-Shore transformation has application whenever one deals with degeneracies. Though Joe was not a co-author on

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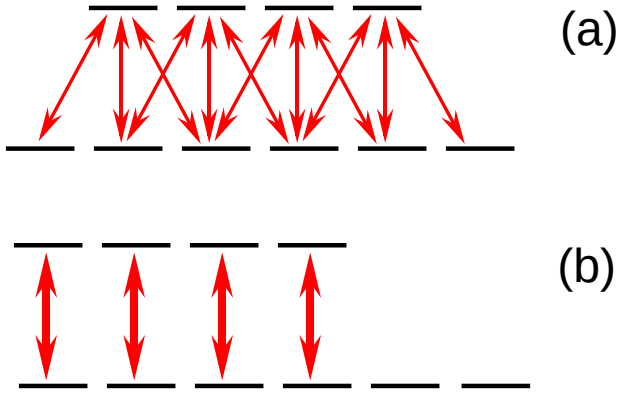

Fig. 7. (a) Example of linkage pattern such as would be found with degenerate hyperfine interaction, $F = 5/2$ to $F = 3/2$. (b) Equivalent system is four independent two-state systems, and two unconnected ("dark") states.

these works, they appeared at the time of his regular LLNL visits, and they contributed notably to our present understanding of coherent atomic excitation.

12 The Continuum

The termination of the laser-induced excitation process underlying LIS was the photoionization continuum. By this one means that the active electron no longer is confined to a finite region around the atomic nucleus; it is not restricted to a discrete (quantized) kinetic energy but may have any energy exceeding the binding energy. The wavefunction of such an electron extends to infinity, meaning that the electron will travel away from the ion as time advances. For many years the photoionization continuum had been regarded as a sort of one-way exit into a probability sink, as was the case in our modeling of the excitation chain. In recent years, however, it has been recognized that the continuum can exhibit coherence properties [38].

One of the traditional ways of treating a continuum has been by "box quantization": place the particle into a large box, compute whatever properties are of interest, and then, in the resulting formulas, allow the box size to become indefinitely large while retaining total unit probability. As a step along this path one deals with a situation in which the RWA Hamiltonian has a large number of discrete detunings around the resonance detuning. The structure is often referred to as a quasicontinuum, and when truncated to a finite set of states, offers a tractable means of numerical modeling either the large number of molecular bound states [39] or else true contnuum properties. Although some of the properties of such a quasicontinuum do mimic a continuum (e.g. one can deduce the Golden-Rule transition probabilities), the presence of discrete energies leads to interesting recurrence effects that do not occur with a true continuum [40, 41].

On one of his visits Joe told us about some of the very surprising results that he and his colleagues had discovered [40], and this led us to examine such structures. Although most of the Livermore work appeared only in a textbook [8], one collaborative paper made explicit use of this model [42]. Old records show that Jay Ackerhalt was also a participant in this work.

13 Propagation

The excitation of atoms by a given radiation pulse is an idealization suited to treatment of very thin samples of gas. As the vapor path of the light becomes longer, the atoms will inevitably leave an imprint on the light, which in turn will alter the excitation of

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atoms deeper in the sample. To correctly model a thick sample of matter one must deal not only with the time-dependent Schrödinger equation at each position, one must also use Maxwell's equations to describe the propagation of the field through the excitable medium.

Already in his book with Les Allen [17], Joe had discussed some of the simple effects that occur with optically thick media, such as self-induced transparency and the formation of solitons. Such effects were obviously of relevance to our LIS project: we would like to use up as many photons as possible while creating as many ions as possible. Thus it was essential to treat optically thick material, in which the fields acting on the downstream atoms were significantly different from the initial laser beams. Joe was instrumental in helping us to formulate the equations properly, and in understanding some of the novel physics that occurs when one has several pulses, of different colors, passing through a medium of multilevel atoms. Amongst the interesting discoveries he shared were "simultons", two different colored pulses that acting on a three-level atom (an example of Cook-Shore periodicities) travel together unchanged [43, 44].

As with simple coherent excitation of atoms, intuition based upon incoherent excitation often leads to incorrect results. Here too, the collaboration with Joe helped clarify some of the unexpected physics of multiple simultaneous pulses coupled to coherently excitable atoms. We found that intuition based on incoherent excitation was simply wrong when it attempted to treat multiple pulses passing through material that had been coherently excited. The publications from Livermore that involved Joe were [39, 45, 46].

14 Noise

Radiation from a real laser is not the idealized monochromatic train of waves used in simple treatments of multilevel atomic excitation. Inevitable fluctuations of the radiation give a finite bandwidth to the light. In the late 1970's many people were studying the effects of noise on laser-excited atoms, for example Eberly [47], Agarwal [48], Cohen-Tannoudji [18] and Zoller [49]. The book [8] cites many others; see also the books by Gardiner [50]. In the 1980's Krzysztof Wodkiewicz came to Rochester for a visit, bringing with him an interest in stochastic processes and their effects on radiation. About this time we had begun thinking about modeling random fluctuations of fields, using Monte-Carlo type integration techniques. During one of Joe's visits to Livermore, we recognized that work by Anatoly Burshtein offered much better methods for describing the random variations of laser radiation [51, 52]. Over a period of several months Joe, Krzysztof and Bruce worked out a number of interesting applications of these, and other techniques for treating noisy pulses[53, 54, 55]. These papers dealt with forms of Markovian jump processes. Most other papers on noise have been devoted to continuous noise, usally Gaussian -Markovian. Such processes can be viewed as limiting cases of Markovian jumps, as was shown by Burshtein and his coworkers.

One thing became very clear from this work: if you can find some alternative to treating fluctuations by means of straightforward averaging of many stochastic time histories (integration of the Schrödinger equation with random changes) you will be able to see much more clearly the properties of the solutions - for example, shapes of peaks in fluorescence spectra.

15 Intense Field Physics

As the AVLIS program proceeded successfully on course towards deployment as a commercially viable enterprise, emphasis shifted away from physics to engineering designs. A change in management brought an end to all support for basic research, and for Joe

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around 1987. But this did not end his Livermore connection, it only redirected the points of contact.

To place this new connection into context one needs to recall the work by Joe and his student Zhifang Deng (son of the then Premier of China), aimed at providing a simple understanding of some of the properties of a photoionization continuum then being experimentally discovered [56, 57, 58]. For years most physicists had regarded the electronic states above the ionization limit as an incoherent sink of probabilities. The view was that an electron, once ejected from an atom, was forever lost. But experiments demonstrated that an electron, in leaving the atom under the influence of a strong laser field, could absorb more than the minimum number of photons needed to overcome the binding energy. These excess photons produce a succession of peaks in the photoelectron spectrum, a phenomena that became known as above threshold ionization (ATI) [59]. During a visit to Livermore, Peter Knight recognized that a structured continuum offered an opportunity to enhance the production of harmonics of the strong laser field [60], as subsequently was demonstrated experimentally. Prompted in part by the development of laser sources capable of producing brief electric fields that would overwhelm the binding field of the nuclear attraction on electrons, theorists were examining a new regime of atomic and optical physics. Much of this theoretical work made use of techniques for modeling an electron in space and time, an area in which Ken Kulander at Livermore was uniquely qualified to contribute [61, 62, 63]. This collaboration did much to clarify the complicated processes that occur when atoms are exposed to intense radiation fields [64, 65].

16 Closing Remarks

The work of Joe Eberly for Livermore from 1973 to 1987 not only helped establish the basic conditions needed for successful commercial laser-induced isotope separation, but it also revealed many of the interesting properties of coherent atomic excitation [8]. It is the latter aspect of his collaboration, documented in more than a dozen papers, that holds the more lasting legacy for science. The collaboration came at a time when it was still considered desirable that a National Laboratory engage in and support not only applied research directed at finding an immediate solution to some identified engineering problem but also basic research intended only to enlarge the base of knowledge in physics.

This basic research at LLNL underlay the ultimate success of the theoretical modeling effort, based on a computer code written by Bob Nelson (and later extended by Ron White) that combined multiphoton ionization of the atoms (described by the time-dependent Schrödinger equation) and propagation of the laser beams (based on the Maxwell equations). Using only experimentally determined oscillator strengths and wavelengths, the theory was confirmed (without free parameters) for optically thick and thin transitions over very long propagation paths. This was a remarkable accomplishment considering the complexity of the theoretical modeling, which included hyperfine structure, polarization effects (magnetic sublevels), stimulated Raman scattering, etc., and the daunting experimental challenges of measuring absolute photoionization yields.

17 Acknowledgments

We are pleased to honor Joe Eberly by recalling fondly his many contributions to science at the Lawrence Livermore National Laboratory; to him we dedicate this review article.

BWS says: The years of collaboration with Joe Eberly have been particularly memorable and valuable for me personally; the many long enlightening discussions with him, and with Peter Knight, formed the basis for what subsequently became a major two-

volume textbook on coherent excitation [8]. One of the great treats of a visit with Joe at Livermore was his skill in placing our various computational efforts into a larger landscape. He would inevitably point out some greater significance to our work than we had recognized. And he was a great dinner guest. My children Tim and Hilary share fond memories of suppertime conversations about Louis XI, the "universal spider", and demonstrations of strange attractors on a simple pocket calculator.

MAJ says: Those were heady times for a new Ph.D.-spending hours around hallway tables "arguing" with Joe about how atoms and photons really behaved, making the connections from two-level atoms and perfect plane waves to the laboratory reality of uranium atoms with their 400 relevant sublevels and not-so-perfect, not-so-monochromatic lasers. The textbooks may indeed have had in them all the theory we required, but Joe's voice carrying down the hall was the excuse we welcomed to show someone the data and figure out what it really meant.

KCK says: Just knowing Joe has had many benefits. Ann Orel and I were trying to get some financial support from a young DOD contract monitor to study a laser-molecule collision process. I was schmoozing with said fellow regarding our proposal and happened to mention that Joe was visiting us. He said, "You know Joe Eberly!?" I said, "Of course." Our funding was awarded in full shortly thereafter.

JID says: I had only recently joined the Lab from industry when I met Joe in 1974. By then, I was a somewhat hardened industrial physicist, and had already developed a healthy skepticism of excessive theorizing about practical problems. I let Joe and his colleagues at the Lab know that they had my support for developing a comprehensive theory of multiphoton ionization of atoms in the vapor phase providing it would be useful to the furtherance of our understanding and progress in developing AVLIS. Joe and Bruce Shore and others accepted this challenge and within a matter of months established the need for a detailed quantum mechanical theory of the multiphoton processes involved. Joe's professional and personal approach were essential in establishing the initial effort and he continued to be a very positive contributor for more than a decade during the entire time that I was director of the AVLIS program. I personally appreciate Joe, not only for what he did for the Lab and AVLIS but especially for his savoir faire.

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