PHYSICS OF ATOMS AND MOLECULES

Marvin H. Mittleman

Introduction to the Theory of Laser-Atom Interactions

SECOND EDITION

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Preface

The objectives of this edition are slightly modified from those of the First Edition; the modifications are a response to the almost explosive growth of this field that has occurred since the publication of the First Edition. This was most noticeable for both theory and experiment on multiphoton ionization where, I am sorry to say, experimental developments have largely shown the way toward theoretical understanding.

A few sections were deleted from the First Edition, but many more have been added to account for recent work. I have tried to avoid detailed descriptions of calculations which could not be at least outlined here, but there have been so many important numerical experiments that it was essential to include some. There was a time when it would have been essential to discuss the one-dimensional models that have provided understanding for the real problem, but these have been superseded mainly by full three-dimensional calculations of real problems. Some have been described in Sections 7.9 and 8.8.

The new field of atom–atom scattering in an optical trap is dealt with in Section 9.7. I have tried to describe the physics of the problem and, with apologies to many authors, have not described any of the current calculations. I think that the field is sufficiently new that most of these calculations must be considered to be preliminary and, therefore, I cannot determine which are not.

There have been so many colleagues whose insight has helped with the writing of this edition that it would be difficult to name them all. There are some whom I must mention: Ken Kulander, Norm Bardsley, and Abe Szöke at Livermore and Lee Collins and Peter Miloni at Los Alamos. Regarding scattering in traps, Paul Julienne at NIST and Keith Burnett at Oxford helped my education. Again, present and former colleagues at City College were invaluable as was Predrag Krstić as a collaborator.

Contents

Basic Ideas

1.1. Introduction

Since the object of this exercise is the description of interacting atoms and laser fields, the starting point must be the Schrödinger equation describing the time evolution of coupled matter and electromagnetic fields. We shall be interested in relativistic effects in only a few cases so the matter field will be described by the nonrelativistic Schrödinger wave function. However, we must allow for creation and destruction of photons, the particles of the electromagnetic field. This is most conveniently done by resorting to the quantum electrodynamic description of the field.[1] The Schrödinger equation is

$$\left[i\hbar \frac{\partial}{\partial t} - H \right] \Psi = 0 \tag{1.1.1}$$

where

$$H = H_{\text{rad}} + \sum_{j=1}^{j_{\max}} \frac{1}{2m} \left(\mathbf{p}_j - \frac{e_j}{c} \mathbf{A}(\mathbf{r}_j) \right)^2 + V(\mathbf{r}_1 \cdots \mathbf{r}_{j_{\max}}) \tag{1.1.2}$$

The first term is the energy operator of the noninteracting electromagnetic field

$$H_{\text{rad}} = \sum_{k\lambda} \hbar\omega_k \mathbf{n}_{k\lambda} \tag{1.1.3}$$

where the sum k, λ is over a complete set of modes of the electromagnetic field. Usually k is the momentum of the mode and λ numbers the two possible transverse polarizations of the field, $A(r)$. $\hbar\omega_k = \hbar c\,|\mathbf{k}|$ is the energy of the k, λ mode and $\mathbf{n}_{k\lambda}$ is the operator whose eigenvalues are the integer occupation numbers (or the number of photons) of the designated mode. This operator is given by

$$\mathbf{n}_{k\lambda} = a_{k\lambda}^{\dagger} a_{k\lambda} \tag{1.1.4}$$

where $a^\dagger_{k\lambda}(a_{k\lambda})$ is a creation (destruction) operator of photons in the k, λ mode. They obey the commutation relations

$$[a_{k\lambda}, a^\dagger_{k'\lambda'}] = \delta_{k,k'}\delta_{\lambda,\lambda'} \tag{1.1.5}$$

and all other commutators vanish.

The field, $\mathbf{A}(\mathbf{r})$, can be expanded in any complete set, $\mathbf{u}_{k\lambda}(\mathbf{r})$, which satisfies the wave equation

$$[\nabla^2 - \omega^2_{k\lambda}/c^2]\,\mathbf{u}_{k\lambda}(\mathbf{r}) = 0 \tag{1.1.6}$$

The simplest of these is the plane-wave set

$$\mathbf{u}_{k\lambda}(\mathbf{r}) = \hat{\varepsilon}_{k\lambda}\frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{V}}, \qquad \lambda = 1, 2 \tag{1.1.7}$$

where

$$\hat{\varepsilon}^2_{k\lambda} = 1, \qquad \hat{\varepsilon}_{k\lambda_1}\cdot\hat{\varepsilon}_{k\lambda_2} = \mathbf{k}\cdot\hat{\varepsilon}_{k\lambda} = 0 \tag{1.1.8}$$

and V is the normalization volume of the field. These are not the most general complete set nor do they satisfy the boundary conditions for any real laser field. A more realistic set of boundary conditions would impose a finite spatial width to the laser field and describe the properties of its focus.[2] However, the realistic description of the beam can always be expanded locally in the set (1.1.7). That is, the beam can never change its amplitude or direction on a scale that is smaller than a wavelength and usually does not vary on a scale that is a good deal larger than a wavelength. Therefore, over a domain set by the scale of the variation of the amplitude, the expansion in the plane-wave set is a good one. We shall be interested in the effects of the laser field on matter so that if the process in question (e.g., ionization, scattering) takes place in a domain small relative to a wavelength, then the laser field is well described as a (local) plane wave. This will almost always be true for ionization but will have to be examined more carefully for scattering processes.

The transverse $(\mathbf{k}\cdot\mathbf{A} = 0)$ vector potential can be expanded in the complete set (1.1.7) or a more general one as

$$\mathbf{A}(\mathbf{r}) = \sum_{k\lambda}\left(\frac{2\pi\hbar c^2}{\omega_k}\right)^{1/2}[a_{k\lambda}\mathbf{u}_{k\lambda}(\mathbf{r}) + a^\dagger_{k\lambda}\mathbf{u}^*_{k\lambda}(\mathbf{r})] \tag{1.1.9}$$

Returning to (1.1.2), the j sum runs over the particles of the system and V represents the Coulomb interactions between all of the (nonrelativistic) charged particles. The other quantities have their conventional meanings. The wave function in (1.1.1) is therefore a function in the configuration space of all of the particles and the Hilbert space of all of the modes of the electromagnetic field. This is the usual starting point for the treatment of the interaction of radiation and matter, which is a much broader problem than the one of interest here.

1.2. Transition to a Classical Description of the Laser Field

We are interested in the interaction of lasers, a very special kind of radiation, with matter. The crucial points that distinguish laser fields from other radiation, for our purposes, are their high intensity and their coherence properties. More specifically, it is the large number of photons in a laser mode. For example, a laser with photons such that $\hbar\omega = 1$ eV with a single-mode flux of 1 mW/cm^2 in a typical coherence volume of 1 cm^3 has about 2×10^5 photons in the field:

$$N = \frac{\text{energy flux}}{\hbar\omega}\frac{V}{c} = \frac{10^{-3}\ \text{W/cm}^2}{1\ \text{eV}}\frac{1\ \text{cm}^3}{3 \times 10^{10}\ \text{cm/s}} \approx 2 \times 10^5$$

This high quantum number makes it likely that the lasers will be accurately described as a classical electromagnetic field. This will now be demonstrated.

We first transform to an interaction representation in which the time evolution due to the field energy, H_{rad}, is absorbed into the wave function

$$\Psi = \exp(-iH_{\text{rad}}\,t/\hbar)\ \Psi_I \tag{1.2.1}$$

which yields

$$\left[i\hbar\frac{\partial}{\partial t} - H_I(t)\right]\Psi_I = 0 \tag{1.2.2}$$

where

$$H_I(t) = \sum_{j=1}^{j_{\max}} \frac{1}{2m_j}\left(\mathbf{p}_j - \frac{e_j}{c}\mathbf{A}(\mathbf{r}_j, t)\right)^2 + V(\mathbf{r}_1 \cdots \mathbf{r}_{j_{\max}}) \tag{1.2.3}$$

and the time dependence introduced into the vector potential by this transformation is given by

$$\begin{aligned}\mathbf{A}(\mathbf{r}, t) &= \exp(iH_{\text{rad}}\,t/\hbar)\ \mathbf{A}(\mathbf{r})\exp(-iH_{\text{rad}}\,t/\hbar) \\ &= \sum_{k\lambda}\left(\frac{2\pi\hbar c^2}{\omega_k}\right)^{1/2}\ [a_{k\lambda}\mathbf{u}_{k\lambda}(\mathbf{r})\,e^{-i\omega_k t} + a^\dagger_{k\lambda}\mathbf{u}^*_{k\lambda}(\mathbf{r})\,e^{i\omega_k t}]\end{aligned} \tag{1.2.4}$$

where (1.1.5) has been used in the form

$$e^{i\omega_k t \mathbf{n}_{k\lambda}} a_{k\lambda} e^{-i\omega_k t \mathbf{n}_{k\lambda}} = a_{k\lambda} e^{-i\omega_k t} \tag{1.2.5}$$

and the vanishing of all commutators other than (1.1.5) has also been used.

We now transform to the phase representation[3] for the field. Independent coordinates, ϕ, with the range $0 < \phi < 2\pi$, are introduced for each mode and a state with n photons in the k, λ mode is described in this coordinate space by

$$|n_{k\lambda}\rangle = (2\pi)^{-1/2} \exp(in_{k\lambda}\phi_{k\lambda}) \tag{1.2.6}$$

The number operator is then

$$\mathbf{n}_{k\lambda} = -i \frac{\partial}{\partial \phi_{k\lambda}} \tag{1.2.7}$$

and the creation and destruction operators are

$$a_{k\lambda} = e^{-i\phi_{k\lambda}} \left(-i \frac{\partial}{\partial \phi_{k\lambda}}\right)^{1/2}, \qquad a_{k\lambda}^{\dagger} = \left(-i \frac{\partial}{\partial \phi_{k\lambda}}\right)^{1/2} e^{i\phi_{k\lambda}} \tag{1.2.8}$$

which are readily shown to satisfy the commutation relation (1.1.5). This transformation is not a particularly useful one in the general case since the square root of a derivative is difficult to work with, but when the mode occupation numbers are large, as they are for lasers operating well above the lasing threshold,[4] then it can be exploited to good use. For the laser modes only, we let

$$n_{k\lambda} = N_{k\lambda} + \nu_{k\lambda} \tag{1.2.9}$$

where $N_{k\lambda}$ is some average value for this laser mode occupation number during the process and $\nu_{k\lambda}$ is the variation about that number. We shall be interested in $\nu_{k\lambda}$ as large as 10^3 or so, but this is still much smaller than typical values of $N_{k\lambda}$ of interest. If this were not the case, then the dynamics of the laser would be coupled to the atomic processes under study. This is a common phenomenon when many atoms participate in the process. Harmonic generation and its coupling to ionization[5] is a good example. We shall not deal with collective effects of this kind here (except for a brief discussion in Section 7.4, and so we make the unitary transformation which relabels the laser mode states by $\nu_{k\lambda}$ rather than $n_{k\lambda}$

$$|n_{k\lambda}\rangle = e^{iN_{k\lambda}\phi_{k\lambda}} |\nu_{k\lambda}\rangle \tag{1.2.10}$$

The operators, (1.2.8), are changed to

$$a_{k\lambda} = e^{-i\phi_{k\lambda}} \left(N_{k\lambda} - i \frac{\partial}{\partial \phi_{k\lambda}} \right)^{1/2} \tag{1.2.11a}$$

and its Hermitian conjugate

$$a_{k\lambda}^{\dagger} = \left(N_{k\lambda} - i \frac{\partial}{\partial \phi_{k\lambda}} \right)^{1/2} e^{i\phi_{k\lambda}} \tag{1.2.11b}$$

In this representation, $(i\partial/\partial\phi_{k\lambda})$ is of order $\nu_{k\lambda}$ so that an expansion is indicated,

$$a_{k\lambda} = e^{-i\phi_{k\lambda}} \sqrt{N_{k\lambda}} \left(1 - \frac{i}{2N_{k\lambda}} \frac{\partial}{\partial \phi_{k\lambda}} \cdots \right) \tag{1.2.12}$$

with its Hermitian conjugate. If only the leading terms are kept, then

$$\mathbf{A}(\mathbf{r}, t) = \sum_{k\lambda} \left(\frac{2\pi\hbar c^2}{\omega_{k\lambda} V} \right)^{1/2} \begin{bmatrix} \hat{\varepsilon}_{k\lambda} \exp i(\mathbf{k}\cdot\mathbf{r} - \omega_k t - \phi_{k\lambda}) \\ + \hat{\varepsilon}_{k\lambda}^{*} \exp -i(\mathbf{k}\cdot\mathbf{r} - \omega_k t - \phi_{k\lambda}) \end{bmatrix} \tag{1.2.13}$$

where the plane wave representation (1.1.7) is used. This is exactly the form of the classical electromagnetic vector potential with a mode amplitude

$$|A_{k\lambda}| = \frac{c}{\omega_k} |E_{k\lambda}|, \qquad \mathbf{E}_{k\lambda} = \left(\frac{8\pi\hbar\omega_k N_{k\lambda}}{V} \right)^{1/2} \hat{\varepsilon}_{k\lambda} \tag{1.2.14}$$

This relates the amplitude of a classical electromagnetic field to the more fundamental description in terms of the occupation numbers of the field, or the density of photons in the field.

The correction to (1.2.13) is

$$\delta\mathbf{A} = \frac{1}{2} \sum_{k\lambda} \left(\frac{2\pi\hbar c^2}{N_{k\lambda}\omega_k V} \right)^{1/2} \begin{bmatrix} \hat{\varepsilon}_{k\lambda} \exp i(\mathbf{k}\cdot\mathbf{r} - \omega_k t - \phi_{k\lambda}) \left(-i \dfrac{\partial}{\partial \phi_{k\lambda}} \right) \\ + \hat{\varepsilon}_{k\lambda}^{*} \left(-i \dfrac{\partial}{\partial \phi_{k\lambda}} \right) \exp -i(\mathbf{k}\cdot\mathbf{r} - \omega_k t - \phi_{k\lambda}) \end{bmatrix}. \tag{1.2.15}$$

It is small for lasers operating well above their lasing thresholds, $N_{k\lambda} \gg 1$.

Fields of arbitrary polarization are described by the explicit form for the polarization vector

$$\hat{\varepsilon}_{k\lambda} = \hat{x} \cos \frac{\eta_{k\lambda}}{2} + i\hat{y} \sin \frac{\eta_{k\lambda}}{2} \tag{1.2.16}$$

which results in a vector potential for the k, λ mode,

$$\mathbf{A}_{k\lambda}(\mathbf{r}, t) = \frac{|E_{k\lambda}|}{\omega_k}\left(\hat{x}\cos(\omega t - \mathbf{k}\cdot\mathbf{r} + \phi_{k\lambda})\cos\frac{\eta_{k\lambda}}{2} + \hat{y}\sin(\omega t - \mathbf{k}\cdot\mathbf{r} + \phi_{k\lambda})\sin\frac{\eta_{k\lambda}}{2}\right) \tag{1.2.17}$$

The parameter η is the polarization angle for the mode. $\eta = 0$, $(\pi/2)$ describes linear polarization in the x (y) direction, $\eta = \pm\pi/4$ describes opposite circular polarizations, and other values describe elliptic polarizations. In this form the time-averaged power of the laser beam, proportional to A^2, is independent of the polarization angle.

With the prescription (1.2.17) for each mode, the Schrödinger equation describes particles in the field of an operator that looks like a classical prescribed electromagnetic field with phase parameter $\phi_{k\lambda}$ which are still operators. For a single-mode field the one phase parameter can be asborbed into a translation of t, and since physical results will almost always be independent of the origin of t, then these results will no longer depend on $\phi_{k\lambda}$. However, for a multimode laser, only one of the phase parameters can be eliminated in this way and so we can expect that physical results will depend on the remaining phases. When these are not known, ensemble averages over them are necessary. This is discussed briefly in Section 1.6.

The preceding discussion dealt with the transition to a classical description of the field for the case when the number of photons in each mode is large. A complementary derivation of this transition was given by Mollow.[6] It is based on the coherent states of the electromagnetic field. These states have been shown[7] to be the quantum electromagnetic states which most closely approximate the classical description of the field. They can be defined as eigenstates of the photon destruction operator, $a_{k\lambda}$. They can be written as

$$\psi_{k\lambda}(t) = \sum_{n=0}^{\infty} e^{-i\omega_k t}\xi_n\,|n\rangle \tag{1.2.18}$$

The requirement that they be normalized eigenstates of $a_{k\lambda}$ results in

$$\xi_n = \exp[-\langle n_{k\lambda}\rangle/2]\,\frac{(\alpha_{k\lambda})^n}{(n!)^{1/2}} \tag{1.2.19}$$

where

$$\alpha_{k\lambda} = \langle n_{k\lambda}\rangle\, e^{-i\phi_{k\lambda}} \tag{1.2.20}$$

with $\langle n_{k\lambda}\rangle$ being the average occupation number of the mode. Then

$$a_{k\lambda}\psi_{k\lambda}(t) = e^{-i\omega_k t}\alpha_{k\lambda}\psi_{k\lambda}(t) \tag{1.2.21}$$