Atomic Relaxation in the Presence of Intense Partially Coherent Radiation Fields*

J. H. Eberly
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
(Received 30 August 1976)

I develop a nonperturbative relaxation theory appropriate for an atom subjected to strong finite-bandwidth radiation fields. The partial coherence implied by the finite bandwidth is modeled by simple phase and amplitude correlation functions. Modifications of the Mollow theory of resonance fluorescence are calculated as an example. The finite bandwidth of the applied field does not have a simply additive effect on the predicted fluorescence spectrum.

The use of quasimonochromatic lasers in multiphoton spectroscopy poses a new problem in relaxation theory. In all types of spectroscopy, linewidths are studied for the insights they give into the relaxation interactions of the system under investigation. Thus, it is necessary to have an analysis of atomic relaxation phenomena in the presence of very strong fields that is flexible enough to allow the laser's contribution to be isolated from the other elements making up the whole line. Such an analysis becomes doubly important for those multiphoton effects, that of resonance fluorescence for example, in which a truly fundamental theory such as quantum electrodynamics can be tested experimentally.

Unfortunately, in the cases of the greatest interest, and of the most recent experimental advances, the well-known perturbative relaxation theory of Bloch and Wangsness and of Redfield[1] fails precisely because the laser power is so high. In other words, it has recently become possible to undertake spectroscopic experiments with laser fields of sufficient power that the atomic transition amplitudes change more rapidly in response to the coherent laser than they do in response to the incoherent stochastic (spontaneous radiative and collisional) forces acting on the atom. However, even if all other stochastic forces can be ignored by virtue of the high laser power, it cannot be forgotten that the same laser contributes its own element of stochastic influence on the atom simply because it is not perfectly coherent.

In this Letter I sketch a nonperturbative approach to atomic relaxation processes, an approach that deals directly with the equations of motion of the two-time functions that determine the spectrum. This approach is unaffected by the high strength of the atom's coherent interactions. In further contrast to most previous treatments of relaxation, the present method uses the Heisenberg picture and avoids consideration of the density matrix and its time evolution.

For definiteness we treat the specific problem of resonance fluorescence by a two-level atom. (Other areas of intense-field physics, such as two-photon absorption and multiphoton ionization, provide equally interesting contexts for the application of the present nonperturbative technique.) A strong saturating laser field induces a near-resonant multiphoton "dressing" of a single allowed atomic transition in this case, and observations of the fluorescence spectrum are called for. Stroud and others have recently carried out the first resonance fluorescence experiments.[2] The perfectly coherent zero-bandwidth-laser theory due to Mollow[3] is well known.

The equations obeyed by the Heisenberg operators appropriate to a two-level atom are only three in number once the electromagnetic self-fields have been eliminated[4] in favor of the "natural" longitudinal and transverse relaxation rates appropriate to spontaneous emission, γ_L and γ_T. We work in the instantaneous rotating frame and denote by $\Omega(t)$ and $\varphi(t)$ the instantaneous Rabi rate and laser phase.[5] These are the simplest stochastic elements associated with every realistic laser, and absent from previous theories of the laser-atom interaction. The observed fluorescence spectrum can be shown[6] to be proportional to the real part of a certain definite integral $G(\nu)$:

$$G(\nu) = \int_0^\infty ds \exp(-\Gamma s) \exp[-i(\nu-\omega)s] \langle\langle \exp[i(\varphi_s - \varphi_0)] g_s \rangle\rangle. \quad (1)$$

Here ω is the laser midfrequency; and Γ and ν are the bandwidth and center frequency of the conventional Fabry-Perot interferometer used as a spectrum analyzer. Also, $\varphi_s \equiv \varphi(t+s)$, $\Omega(t+s)$, and $g_s = \rho^\dagger(t+s)\rho(t)$, where ρ and $\rho^\dagger$ are the instantaneous rotating-frame transition operators for the atom, and where t will be assumed to be a time sufficiently long that initial transients have died away. The double angular bracket signifies an average over the stochastic phase and amplitude fluctuations of the laser.

In addition to g_s it is useful[7] to define $d_s = \rho(t+s)\rho(t)$. The equations for g_s and d_s, as functions of s, follow from the three Heisenberg equations for ρ, $\rho^\dagger$, and ρ_3 (the atomic inversion). By formal integration ρ_3 can be eliminated exactly, leaving two coupled linear integrodifferential equations with stochastic coefficients:

$$\dot{g}_s = -(\gamma_T + i\dot{\varphi}_s)g_s - i\Omega_s\rho - \tfrac{1}{2}\int_0^s dt' \exp[-\gamma_L(s-t')]\,\Omega_s\Omega_{t'}(g_{t'} - d_{t'}); \tag{2a}$$

$$\dot{d}_s = -(\gamma_T - i\dot{\varphi}_s)d_s + i\Omega_s\rho + \tfrac{1}{2}\int_0^s dt' \exp[-\gamma_L(s-t')]\,\Omega_s\Omega_{t'}(g_{t'} - d_{t'}). \tag{2b}$$

A formal integration of Eqs. (2a) and (2b) may also be made, but no further real progress is possible without knowledge of the statistical properties of the laser. In theoretical treatments of strong resonant laser-atom interactions to date, only the simplest possible assumption has been made, namely that the laser is perfectly coherent and both φ and Ω are constant.[8] In the present example—that of resonance fluorescence—such an assumption leads from Eqs. (2) directly to Mollow's results[3]; however, a more realistic view is taken of the laser in the present theory. We take the laser phase and amplitude fluctuations to be characterized by the following correlation properties:

$$\langle\langle \exp(i\varphi_s)\exp(-i\varphi_{s'})\rangle\rangle = \exp(-\gamma_\varphi|s-s'|); \tag{3a}$$

$$\langle\langle \Omega_s\Omega_{s'}\rangle\rangle = \Omega^2\exp(-\gamma_A|s-s'|). \tag{3b}$$

Clearly, γ_φ and γ_A will play the role of phase and amplitude bandwidths in the theory; and clearly, when they are set equal to zero we recover the no-fluctuation theory.[9] For simplicity I make an assumption that can only be partially justified, that atom-field variable products can be decorrelated into atom-atom and field-field variable pairs: $\langle\langle \Omega_r\Omega_s\rho_s^\dagger\rho_0\rangle\rangle = \langle\langle \Omega_r\Omega_s\rangle\rangle\langle\langle \rho_s^\dagger\rho_0\rangle\rangle$, etc. Such a decorrelation of the second-order atom and field variable products is in general only approximately valid. It is, however, clearly superior to the decorrelation of a mean-field theory (such as the Bloch-Wangsness-Redfield theory) where one would further decompose $\langle\langle \rho_s^\dagger\rho_0\rangle\rangle$ into $\langle\langle \rho_s^\dagger\rangle\rangle\langle\langle \rho_0\rangle\rangle$, etc.

The simple assumption above allows the most important sources of laser incoherence to be incorporated into the theory in a realistic way. The formal integration of Eqs. (2) leads now to a pair of coupled second-order Volterra integral equations. With no perturbative approximations these can be Laplace-transformed into a pair of algebraic equations for the Laplace transforms $\langle\langle g\rangle\rangle$ and $\langle\langle d\rangle\rangle$. For example, the Laplace transform of $\langle\langle g_s\rangle\rangle$ denoted by $\hat{g}(z)$ is

$$\hat{g}(z) = \frac{F(z)}{(z+\tilde{\gamma}_T)[(z+\tilde{\gamma}_T)(z+\tilde{\gamma}_L)+\Omega^2]}, \tag{4}$$

where F is a simple polynomial, Ω is the usual no-fluctuation Rabi frequency, and $\tilde{\gamma}_T$ and $\tilde{\gamma}_L$ are given by

$$\tilde{\gamma}_T = \gamma_T + \gamma_\varphi, \tag{5a}$$

$$\tilde{\gamma}_L = \gamma_L + \gamma_A. \tag{5b}$$

The form of $\hat{g}(z)$ in (4), but not the parameters $\tilde{\gamma}_L$ and $\tilde{\gamma}_T$, are familiar from the Mollow theory.

The Mollow spectrum is triple-peaked, and in the high-power limit the peaks are all Lorentzians, with two identical side peaks located at a frequency separation Ω above and below the central peak. The curious feature of the spectrum is that neither the widths nor the heights of the side peaks are the same as that of the central peak. Given the fundamental ratio 2:1 of the area under the central peak to the area under either side peak, it is sufficient to state the ratio of central to side peak heights. In the Mollow case this ratio is[10]

$$H_0 : H_\pm = (\gamma_L + \gamma_T) : \gamma_T, \tag{6}$$

and under the conditions encountered in the experiments to date $\gamma_L = 2\gamma_T$, so that the ratio associated with the Mollow spectrum is exactly 3:1.

In the extended relaxation theory sketched here, there are three additional relaxation rates, in addition to γ_L and γ_T, namely the phase and amplitude bandwidths of the laser, γ_φ and γ_A, and the bandwidth of the interferometer, Γ. We can discuss the application of the present theory to the resonance-fluorescence problem by noting that our spectrum, proportional to $G(\nu)$ in Eq. (1), is itself the Laplace transform of g_s with the variable $z = i(\nu-\omega) + \Gamma + \gamma_\varphi$. It is then only a matter of comparing $\hat{g}(z)$ in Eq. (4), evaluated at this value of z, with the corresponding expression in the

Mollow theory, in order to see that the replacements

$$\gamma_L \to \gamma_L + \gamma_A + \gamma_\varphi + \Gamma \qquad (7a)$$

and

$$\gamma_T \to \gamma_T + 2\gamma_\varphi + \Gamma \qquad (7b)$$

have the following effects on the height ratio $r = H_0/H_+$: (a) $r = 3$, if the fundamental spontaneous radiation rate γ_L dominates all laser-associated relaxation rates (the Mollow case); (b) $r > 3$, if laser phase fluctuations are completely negligible[11]; (c) $r = \frac{3}{2}$, if laser amplitude fluctuations are negligible, and the phase relaxation rate γ_φ dominates the spontaneous rate γ_L; and (d) $r = 2$, if the instrumental bandwidth Γ of the interferometer dominates the other widths.[12]

Several remarks may be made in summary. First, I have shown that it is possible to incorporate an arbitrary laser linewidth into the theory in a simple, nonperturbative, and not unrealistic way. The laser can have either phase or amplitude flucturations or both, and can be arbitrarily intense. Second, as the replacement rules (7a) and (7b) show, the inclusion of a laser linewidth does not in general modify a spectral line in a simple additive way. Thus, laser phase and amplitude fluctuations give rise to unexpectedly complex and dissimilar effects in the quantum theory of atom-field interactions. In the resonance-fluorescence case, for example, I have shown that amplitude fluctuations tend to raise and the phase fluctuations tend to lower the value of the basic peak-height ratio. Third, the basic method sketched here may be expected to be applicable to a number of currently interesting problems. In the correlation theory of resonant multiphoton ionization,[12] for example, the basic equations are, not accidentally, practically identical to Eqs. (2). Finally, I must point out that I have used a particular pairwise decorrelation following Eqs. (3); while this decorrelation is the most natural, others are possible. For example, a similar pairwise decorrelation may be carried out with the average, rather than the instantaneous, rotating-frame variables. As might be expected, the replacement rules are then somewhat more complicated, and the ratio in case (c) above changes slightly. Further discussion of this point and a semiclassical interpretation of the replacement rules, as well as a treatment of the nonstationary and multilaser problems, will be undertaken in separate publications.

During the course of the research leading to this paper, I have had the benefit of helpful conversations and suggestions from J. R. Ackerhalt, G. S. Agarwal, C. M. Bowden, B. W. Shore, and K. Wódkiewicz. I am pleased to thank H. Walther for his hospitality at the University of Munich, where part of the work was done.

---

*Research partially supported by the U. S. Energy Research and Development Administration.

[1]See, for example, F. Bloch, Phys. Rev. 105, 1206 (1957), and earlier papers referred to there. An excellent description of these relaxation theories is provided by C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963).

[2]Strong-field resonance fluorescence has been observed for the first time by F. Schuda, C. R. Stroud, Jr., and M. Hercher, J. Phys. B 7, L198 (1974); and more recently by F. Y. Wu, R. E. Grove, and S. Ezekiel, Phys. Rev. Lett. 35, 1426 (1975); and H. Walther, in *Laser Spectroscopy*, edited by S. Haroche *et al.* (Springer, Berlin, 1975), p. 363. Observations in the weak-field limit have also been reported; H. M. Gibbs and T. N. C. Venkatesan, in Proceedings of the Symposium on Resonant Light Scattering, Cambridge, Massachusetts, April 1976 (unpublished).

[3]B. R. Mollow, Phys. Rev. 188, 1969 (1969). See also more recent discussions by B. R. Mollow, Phys. Rev. A 12, 1919 (1975); G. Oliver, E. Ressayre, and A. Tallet, Lett. Nuovo Cimento 2, 777 (1971); S. Hassan and R. K. Bullough, J. Phys. B 8, L147 (1975); D. F. Walls and H. J. Carmichael, J. Phys. B 8, L77 (1975), and 9, 1199 (1976); and H. J. Kimble and L. Mandel, Phys. Rev. A 13, 2123 (1976).

[4]J. R. Ackerhalt and J. H. Eberly, Phys. Rev. A 10, 3350 (1974). See also L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975), Chap. 7.

[5]In writing $\Omega(t)$ and $\varphi(t)$ as numerical functions, not operators, we anticipate a slight extension of Mollow's demonstration (Ref. 3, second citation) that prescribed external fields can be treated essentially in the quantum theory.

[6]The expression given in Eq. (1) is the long-time limit of the general nonstationary spectrum appropriate in this case. See K. Wódkiewicz and J. H. Eberly, unpublished, and to be published.

[7]See P. W. Milonni, Ph.D. thesis, University of Rochester, 1974 (unpublished).

[8]However, an important discussion of multiphoton ionization in which the laser is assigned an intensity bandwidth and a square spectral shape is due to P. Lambropoulos, Phys. Rev. 9, 1992 (1974).

[9]Assumptions (3a) and (3b) are suggested by elementary laser theory as well as by their simplicity and flexibility. Obviously they would need to be expected if higher-than-second-order correlations, or if several different lasers, played roles in the problem under consideration. Such extensions do not appear to be difficult to undertake.

[10]For a semiclassical "explanation" of the linewidths that give rise to this height ratio in the Mollow case, see C. Cohen-Tannoudji, in *Laser Spectroscopy*, edited by S. Haroche *et al.* (Springer, Berlin, 1975), p. 336.

[11]Results in this case have already been reported by the author in Proceedings of the Symposium on Resonant Light Scattering, Cambrdige, Massachusetts, April 1976 (unpublished). The subsequent incorporation of phase-fluctuation effects into the resonance-fluorescence theory has also been achieved by G. S. Agarwal (private communication). See also remarks by Cohen-Tannoudji, Ref. 10.

[12]K. Wódkiewicz and J. H. Eberly, in preparation, and Ref. 6 (to be published).

Enhancement of Dielectronic Recombination by Plasma Electric Microfields*

V. L. Jacobs,† J. Davis, and P. C. Kepple
Plasma Physics Division, Naval Research Laboratory, Washington, D. C. 20375
(Received 24 September 1976)

An investigation has been made of the l-redistribution effect of quasistatic ion electric microfields on dielectronic recombination. Using the linear-Stark-effect approximation, a hydrogen ion density of 10^{14} cm^{-3} is found to produce a threefold enhancement in the rate for the dielectronic recombination process $Fe^{+23}(2s)+e^- \rightarrow Fe^{+22}(2p, nl) \rightarrow Fe^{+22}(2s, nl)+\hbar\omega$.

In this Letter we report on the application of techniques from Stark-broadening theory[1] to calculate the dielectronic recombination rates for multiply charged Fe ions under the influence of plasma electric microfields. Results are presented for a dielectronic recombination process where the effects of the surrounding charged particles have been found to be important at surprisingly low densities.

The conventional interpretation of certain spectral line intensities may be distorted, or even invalidated, by the combined effect of dielectronic-recombination satellites[2] whose wavelengths are indistinguishable from that of the associated resonance line from the recombining ion. In addition, dielectronic recombination has been shown[3] to be the dominant recombination process for nonhydrogenic impurity ions in low-density high-temperature plasmas, such as the solar corona and the discharges produced in controlled thermonuclear fusion experiments.

Dielectronic recombination is the result of a radiationless capture into a doubly excited state

$$X^{+Z}(i)+e^-(\epsilon_i)\rightarrow X^{+(Z-1)}(j,nl), \quad (1)$$

followed by a stabilizing radiative transition involving de-excitation of the recombining ion core

$$X^{+(Z-1)}(j,nl)\rightarrow X^{+(Z-1)}(k,nl)+\hbar\omega. \quad (2)$$

At low densities, where the final excited ions cascade to their ground states in times that are short compared with the electron-ion collision time, the overall recombination rate simply equals the total rate for all stabilizing radiative transitions.

Burgess and Summers[4] have pointed out that with increasing density the overall recombination coefficient defined by Bates, Kingston, and McWhirter[5] is reduced by the effects of collisional ionization on the highly excited nl states, which can play the most important role in the dielectronic recombination process. Burgess and Summers[4] were also the first to suggest that the dielectronic recombination rate could be enhanced by the effects of collisionally induced angular-momentum redistribution of the doubly excited states.

The spectral intensity arising from the stabilizing radiative transitions may be calculated by employing the techniques of modern Stark-broadening theory,[1] which must be applied in its most general form in order to describe the effects of perturbing electrons and ions on the nearly dengerate l sublevels of the upper and lower states. The action of the ions is customarily treated in the quasistatic and long-range dipole (Stark effect) approximations.[6] The effects of electron collisions may be taken into account by applying the generalized impact approximation[7-10] to the quasistatic Stark components before performing the average over the ion electric microfield distribution.

In the present investigation, account is taken only of mixing of the nearly degenerate l sublevels due to the action of the quasistatic ion electric microfields. We defer to a future investigation the inclusion of electron collisions, which will become important with increasing density.