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Comment on "Quantum-Electrodynamical Theory of Atoms Interacting with High-Intensity Radiation Fields"

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It is maintained that in a recent comparison of quantum-electrodynamical with semiclassical treatments of the interaction between atoms and radiation by Chang and Stehle the semiclassical work was misrepresented and that the conclusions reached by these authors are invalid.

Chang and Stehle, in a recent quantum-electrodynamical study of the interaction between atoms and radiation,[1] state "our major purpose is to compare our results with those of the semiclassical treatment." In a subsequent paper[2] they show that some of their results on multiple-quantum transitions in magnetic resonance are in better agreement with the experimental work of Kusch[3] than are the semiclassical calculations of Salwen,[4,5] which were used for comparison with the experiments at the time when they were carried out. Chang and Stehle conclude that semiclassical theory is inferior to their own work in interpreting these and other experiments. We assert that their papers do not constitute a proper comparison with semiclassical theory because Salwen's calculations and other results cited by Chang and Stehle are admittedly only approximate solutions to the semiclassical equations of motion and have been superseded by more recent work in this field to which the authors make no reference. Further, at some points of comparison, Chang and Stehle do not specify their electromagnetic field in sufficient detail.

Salwen gave a general theory of the response of an atom, whose angular momentum was made up of several components, to the fields normally used in magnetic-resonance experiments—a strong static field which may partially decouple the angular momenta and a rotating field to induce resonance transitions. He used a method of approximation to obtain formulas, valid to the lowest nonvanishing order in the rf amplitude, for resonance line shapes and shifts of resonance frequency in single- and multiple-quantum transitions. In Ref. 5 he applied these formulas to the special case of ^{39}K in the ground state. It is our contention that Chang and Stehle have not made proper allowance for the approximate nature of Salwen's formulas.

A different approach to the solution of the semiclassical equation of motion was offered a few years after the publication of Salwen's paper by Shirley,[6] who studied the problem of a two-level atom in an *oscillating* (as distinct from a *rotating*) magnetic field. Shirley showed how to replace the time-dependent Hamiltonian by a time-independent Hamiltonian having an infinite Hermitian matrix, whose diagonalization gives the solution of the problem to any desired order in the field strength. Further, he showed the equivalence of this to a quantized field analysis. He described multiple-quantum transitions in detail, showing how to calculate the transition probabilities and the resonance line shapes.

Shirley's treatment was applied to the many-level atom (Salwen's problem) by Pegg,[7] whose solution differed from Salwen's in essentially the same way as does Chang and Stehle's; namely, by the modification of energy denominators which appear in correction terms. Salwen's energy denominators are the intervals between the states of the atom unperturbed by the rf field. Chang and Stehle's denominators are the same intervals "renormalized by forward-scattering processes." "It seems impossible," say these authors, "to incorporate this into semiclassical theory in any simple way." Pegg's denominators are again the same intervals, but corrected to take account of perturbations of the atom induced according to the equations of semiclassical quantum theory by the rf field. These perturbations are calculated in a time-independent representation, which, for a rotating field, is simply the energy representation of atomic wave functions in a frame rotating with the field. The "renormalization" appears in this treatment simply as the displacement of energy levels by the rf field, which is seen as static in the rotating frame. A full justification of this remark requires, of course, a demonstration that the formulas of Chang and Stehle are substantially identical with those of Pegg. This work is being undertaken.

The two-level atom studied by Shirley was treated at length in Chang and Stehle's first paper.

They compared their expressions for the Bloch–Siegert shift—the displacement of a resonance produced by the antiresonant component of an oscillating field—with semiclassical results quoted only to the first nonvanishing term, where the shifts are proportional to the square of the rf field amplitude. But Shirley's paper quotes the results explicitly up to the sixth power of the field. Shirley's results have been reproduced by Pegg[8] using a different method of solution of the semiclassical equations, and are in agreement also with the recent work of Stenholm,[9] whose method of solution uses continued fractions. These results disagree with that of Chang and Stehle [Eq. (88) of Ref. 1], but their result is based on the approximation $\omega \approx \omega_0$, whereas it is represented in Fig. 7 of Ref. 1 as being valid up to $\delta\omega/2\omega_0 = 0.5$. Here the shift of the resonance is equal to the resonance frequency itself, and the approximation is clearly unjustified.

Shirley's methods were originally applied to the very practical problem of calculating corrections for atomic beam frequency standards.[10] The extension by Pegg was applied in an experimental situation where strong oscillating magnetic fields were used to study excited atoms having complicated hyperfine structure.[11]

There are two particular situations where an exact semiclassical solution can be obtained: One[12] is the well-known "rotating-wave" situation, which can be realized experimentally by subjecting an assembly of magnetic dipoles to the combination of a static magnetic field and a field rotating in the perpendicular plane; the other[13] is the situation where the fields consist of a linearly oscillating component and a parallel static component (which may be zero). Chang and Stehle do not specify the polarization of the fields they discuss; it would appear that they are dealing exclusively with linearly oscillating fields (which would exclude the first case cited above) and that interactions having diagonal matrix elements (the second case above) are of no interest to them. Their strictures against the exact semiclassical solutions are therefore misplaced.

It might have been expected that a study of the interactions of atoms with strong electromagnetic fields (particularly where rf magnetic fields are cited as examples) would contain some reference to the work of Cohen-Tannoudji and his colleagues on "dressed atoms."[14] Many of the results which these authors and others have obtained by a fully quantum-mechanical treatment (for example, the calculation of anomalous *g* factors, of modified resonance line shapes, and of frequency shifts) have been derived also by Pegg and Series[15] and by Pegg[16] using a semiclassical theory.

Chang and Stehle's conclusion is that the region of agreement between results derived from semiclassical and quantum-electrodynamical treatments is in the low-frequency low-field region, "in those systems where transitions involve the emission or absorption of a single photon." This remark is misleading on account of its incompleteness. The agreement holds also for strong fields where the photon number is large compared with unity—a conclusion which can be derived from a correspondence argument. The significant difference between semiclassical and quantized field theories (apart from virtual processes, which Chang and Stehle explicitly neglect) lies in the treatment of spontaneous emission.[17] The interaction matrix elements which determine the evolution of the atomic state vector under stimulated emission and absorption are identical in the two theories; hence, the predictions of the theories must coincide for those situations in which spontaneous emission plays no significant role.

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