Semiclassical Calculations on Multiple-Quantum Transitions

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It is shown that a recent quantum-electrodynamic derivation by Chang and Stehle of double-quantum resonance-frequency shifts is equivalent to a previously known semiclassical approximation, and that neither of these methods is adequate to account for the results of Kusch's experiments. A more-exact semiclassical computation is given, which is in reasonable agreement with Kusch's results in the region where these results are meaningful. It is concluded that the assertion of Chang and Stehle that quantum electrodynamics is necessary for such situations is unfounded.

I. INTRODUCTION

Recently Chang and Stehle[1] used a quantum-electrodynamic approach to analyze the results obtained by Kusch[2] for a two-quantum transition in an atomic-beam experiment. They interpreted their results as agreeing with Kusch's observations very well, and maintained that the use of quantum electrodynamics instead of the semiclassical theory was necessary for the high rf-field strengths involved. Chang and Stehle based their calculations on their previous work,[3] which also purported to show the inadequacy of semiclassical theory in another context. In a comment on this previous work, Pegg and Series[4] maintained that their conclusions were invalid.

In this paper it is shown that the method used by Chang and Stehle to calculate the resonance-frequency shift of the double-quantum transition observed by Kusch is in fact equivalent to the semiclassical approximation[5] that has already been used in such situations.[6,7] Furthermore, the nature of this approximation, together with Kusch's stated limitations to the accuracy of the rectangular-pulse approximation, is such as to indicate that the agreement obtained by Chang and Stehle is merely fortuitous. It is shown that a more-accurate semiclassical computation can give quite reasonable results in the region where the pulse is approximately rectangular.

II. SEMICLASSICAL APPROXIMATION

Salwen's[8] theory, which was originally applied to Kusch's results, uses a second-order perturbation approximation together with the rotating-field approximation to calculate level shifts. As such, it is limited to the accuracy of these two approximations. A semiclassical-approximation method to replace second-order perturbation theory in the region of higher rf field strengths was developed by Pegg.[5] This may be applied either to the time-independent Hamiltonian arising in the rotating-field approximation or, if the effect of the counter-rotating component is to be included, to Shirley's[9] time-independent Floquet Hamiltonian. The procedure to find the resonance frequency is to find the frequency at which the two appropriate roots W of the time-independent secular equation are equal, supposing that the interaction between the given states is zero, after taking account of perturbations due to other states.[5] The procedure can be simplified without greatly affecting the order of the approximation by considering only states which directly interact with the given states,[6,7] that is, which are linked to the given states by off-diagonal matrix elements in the Hamiltonian.

To obtain the results of Chang and Stehle it is sufficient to work in Salwen's rotating reference frame, with the Schrödinger equation

$$\sum_i (\mathcal{H}_{ji} - W\delta_{ji})\langle i|\rangle = 0\,, \tag{1}$$

where $\mathcal{H}_{ji}$ is the time-independent Hamiltonian matrix and $\langle i|\rangle$ its eigenvector coefficients. The diagonal elements are $\mathcal{H}_{jj} = E_j - j\omega$, where E_j are the unperturbed energy levels in the laboratory frame. For the level system under consideration, j and i take the values 2, 1, 0, −1, since we are considering only states which directly interact with $|2\rangle$ and $|0\rangle$, and $\mathcal{H}_{ji} = 0$ for $|i-j|>1$. The second and fourth equations of (1) can be easily substituted into the first and third to give

$$(\tilde{\mathcal{H}}_{22} - W)\langle 2|\rangle + \tilde{\mathcal{H}}_{20}\langle 0|\rangle = 0\,, \tag{2}$$

$$\tilde{\mathcal{H}}_{02}\langle 2|\rangle + (\tilde{\mathcal{H}}_{00} - W)\langle 0|\rangle = 0\,, \tag{3}$$

where

$$\tilde{\mathcal{H}}_{22} = \mathcal{H}_{22} + |\mathcal{H}_{12}|^2/(W - \mathcal{H}_{11})\,, \tag{4}$$

$$\tilde{\mathcal{H}}_{00} = \mathcal{H}_{00} + |\mathcal{H}_{10}|^2/(W - \mathcal{H}_{11}) + |\mathcal{H}_{-10}|^2/(W - \mathcal{H}_{-1-1})\,. \tag{5}$$

This is done in the manner shown by Salwen, except that here, for more accuracy, we have not

replaced W by its unperturbed values, as was done by Salwen. Equations (2) and (3) are of the form of the Schrödinger equation for a two-level system, with $\tilde{\mathcal{K}}_{20}$ the effective off-diagonal Hamiltonian element, and $\tilde{\mathcal{K}}_{22}$, $\tilde{\mathcal{K}}_{00}$ the perturbed states 2 and 0. Now set the approximation procedure to $\tilde{\mathcal{K}}_{20} = \tilde{\mathcal{K}}_{02} = 0$, and then find where the two appropriate values of W are equal, which [from (2) and (3)] is $\tilde{\mathcal{K}}_{22} = \tilde{\mathcal{K}}_{00}$. Substituting the values $W = \tilde{\mathcal{K}}_{22}$ from (2) into (4) and $W = \tilde{\mathcal{K}}_{00}$ from (3) into (5), we find, using the notation of Chang and Stehle, that $\tilde{\mathcal{K}}_{22} = \tilde{\mathcal{K}}_{00}$, when

$$\omega = \tfrac{1}{2}\omega_{2,0} + \delta\omega_{2,0} ,$$

with the resonance frequency shift

$$\delta\omega_{2,0} = \tfrac{1}{2}(\Delta E_2 - \Delta E_0) ,$$

where $\tilde{\mathcal{K}}_{ii} = \mathcal{K}_{ii} + \Delta E_i$; from (4) and (5)

$$\Delta E_2 + |\alpha(2;1)|^2/(-\Delta E_2 + \omega - \omega_{2,1}) = 0$$

and

$$\Delta E_0 + |\alpha(1;0)|^2/(-\Delta E_0 - \omega + \omega_{1,0}) + |\alpha(0;-1)|^2/(-\Delta E_0 + \omega - \omega_{0,-1}) = 0.$$

We note that Salwen's result is obtained by substituting the unperturbed values $\mathcal{K}_{22}$ and $\mathcal{K}_{00}$ for W in (4) and (5).

Apart from obvious misprints and a discrepancy in sign, the above results are identical to the expressions (27) and (23) of Chang and Stehle. The difference in sign is most likely a misprint as well, since the graphical results of Chang and Stehle are obtainable from the above equations but not from their own equations.

At first sight, in view of Fig. 2 of Chang and Stehle, it would appear that the results of this semiclassical approximation and the approach of Chang and Stehle are confirmed by Kusch's measurements. It would be very surprising if this were true, however, for the following reasons.

(i) Kusch's frequencies are measured to an accuracy of about 0.1%, that is, about 1 kHz. The approximation adopted is not this accurate.[6,7]

(ii) No account has been taken of the counter-rotating component, which can be shown by a straightforward calculation to induce a Bloch-Siegert shift of the order of 1 kHz at about 1 G.

(iii) Kusch repeated his experiments and showed that the geometry of the rf circuit had a critical effect on widths and shifts. With a geometry that produced a better approximation to a rectangular pulse, he found that the agreement between the observed and semiclassical widths persisted to much higher rf amplitudes than shown in his Fig. 3 (and Fig. 3 of Chang and Stehle). This is a very strong indication that any theory, such as that of Salwen or of Chang and Stehle, based on the rectangular-pulse approximation is not applicable for rf amplitudes greater than the voltage for which the measured widths depart significantly from the semiclassical widths, in this case about 10 V.

In the light of the above reasons [particularly (iii)], it appears that the agreement obtained by Chang and Stehle is most likely fortuitous.

III. SEMICLASSICAL COMPUTATION

Difficulties (i) and (ii) above can be overcome by replacing the rotating-frame Hamiltonian by Shirley's Floquet Hamiltonian[9] and diagonalizing this by computer, rather than by approximation methods. Because of the unknown shape of the pulse experienced by the atoms, however, it is pointless to attempt to compute the frequency shift or width for an rf voltage of above 10 V or so. The agreement in width is reasonable up to this voltage, and we limit our computations to that of the shift.

Even below 10 V, there are still some parameters necessary for calculation that are uncertain. One is the strength of the rf field which Kusch measured as 11.6 V/G noting the possibility of error in this measurement. We shall accept Kusch's measured value here.[10] Another uncertain parameter is n_i, the number of atoms in each beam. This affects the strengths of all the resonances and thus their overlap with the transition $|2\rangle \rightarrow |0\rangle$ under consideration. This is a secondary effect but does make some contribution. From his observed intensities Kusch concluded that an atom in state $|2\rangle$ was in a sufficiently expanded trajectory to collide with the surfaces of the deflecting magnets. As a first trial, therefore, we take $n_2 = 0$ and then, by symmetry, $n_{-2} = 0$, although we find that the value of n_{-2} has negligible effect on the results. Because the transitions $|1\rangle \rightarrow |2\rangle$ and $|0\rangle \rightarrow |2\rangle$ are observed, we must then conclude that the trajectories of atoms in $|1\rangle$ and $|0\rangle$ are such that they allow a reasonable number of atoms in these states to strike the target, possibly with n_1 slightly less than n_0, since the trajectories of atoms in $|1\rangle$ are more expanded.[2] Consequently, we consider the cases $n_1 : n_0 = 1$ and 0.8, to show the effect of variation of the ratio. By symmetry we put $n_{-1} = n_1$.

The number of atoms received at the detector will be

$$N(\omega) = \sum_i n_i P_{i \rightarrow i} , \tag{6}$$

where $P_{i \rightarrow i}$ is the probability that an atom in state $|i\rangle$ remains in state $|i\rangle$. We assume that the

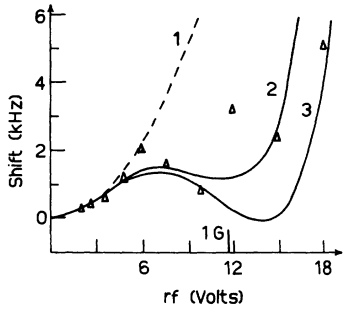

FIG. 1. Shift in resonance frequency as a function of rf amplitude. The curves are based on Salwen's calculation (curve 1) and a more-accurate semiclassical computation for ratios $n_1:n_0=1$ (curve 2) and $n_1:n_0=0.8$ (curve 3).

position of resonance depends very little on the natural width,[5] and adopt Shirley's formula (19) for the time-average probability for atomic-beam experiments. From this we obtain

$$P_{i\to i}=\sum_k \sum_{jl} |\langle i\,k|\lambda_{jl}\rangle\langle\lambda_{jl}|i\,0\rangle|^2, \qquad (7)$$

where $\langle i\,k|\lambda_{jl}\rangle$ is an eigenvector component of the Floquet Hamiltonian. j takes the values -2, -1, 0, $+1$, $+2$, and k and l run from $-\infty$ to $+\infty$. For the calculation here it is sufficient to consider only $k, l=-1$, 0, $+1$, and, for k, $l\neq 0$, to include only Floquet levels which directly interact with the given states $|2,0\rangle$ and $|0,0\rangle$. This leaves an 8×8 Floquet matrix to diagonalize, which is easily done by computer. The resonance frequency is obtained by finding the frequency for which $N(\omega)$ is a minimum. The results are shown in Fig. 1 for both $n_1:n_0=1$ and $n_1:n_0=0.8$.

IV. DISCUSSION

It is to be remembered that experimental points above about 10 V, where the rectangular-pulse approximation fails, are to be disregarded for comparison with the theoretical curves. Because of the uncertainties in the measured parameters mentioned, the results should not be interpreted as good agreement. The semiclassical theory, however, does reasonably reproduce the behavior of the points within the realm of validity of the rectangular-pulse approximation, and we can definitely conclude that the experimental results, while showing up the limits of second-order perturbation theory, do not in any way indicate that semiclassical theory is inadequate. This strongly implies that the assertion made by Chang and Stehle, that semiclassical theory cannot describe Kusch's experimental double-quantum results, is invalid.

Before semiclassical theory can possibly be considered inadequate for such cases, more experiments should be done with controllable and measurable parameters such as the number of atoms in each beam, the rf field strength, and the shape of the rf pulse experienced by the atoms. An optical-pumping or double-resonance-type experiment would circumvent these difficulties. Such experiments in intense rf fields as have been done by Chapman[11] lend support to semiclassical theory.

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[10]We note that Kusch's results and those of Chang and Stehle are not plotted using the measured value of the rf field, which Kusch calls H'. Instead, a value of about 17 V/G is used, which is the adjusted value needed to make the peak of the experimental intensity curve match the theoretical peak. However, in view of the uncertainties in the parameters affecting the beam intensities, together with the likely deviations from the Maxwellian velocity distribution which Kusch estimates should make the experimental intensity peak occur at a somewhat greater value of rf field than the calculated value, it is doubtful if overmuch reliance can be placed on this adjusted value. Furthermore, when we adopt the measured value we find that at low rf fields the experimental frequencies are asymptotic to Salwen's theoretical curve, which is independent of the beam intensities. This is in accord with the predictions of all the theories.
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