# Comment on the ultimate single-ion laser-frequency standard

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Dehmelt's proposal for an ultimate laser-frequency standard based on a single trapped ion is examined. Density-matrix calculations are developed for a double-resonance scheme which, under favorable circumstances, amplifies a highly forbidden and narrow electronic transition. The advantages of a pulsed excitation scheme are discussed, and the limitations of cw excitation are exposed.

In a series of provocative papers,[1–4] an ultimate laser-frequency standard, perhaps with a resolution of 1 part in $10^{18}$, has been proposed. The concept is based on bringing a single isolated ion nearly to rest by localizing it in a quadrupole trap and by optically cooling it. Optical line broadening arising from the Doppler effect, collisions, or transit-time broadening are thereby minimized. The clock transition is assumed to be a highly forbidden electronic transition (which we label $2 \rightarrow 3$), but to detect it, a strong transition ($1 \rightarrow 3$) sharing a common lower level is to be monitored in spontaneous emission, each transition being driven by a resonant laser field (Fig. 1). The argument proposed rests on the assumption that a single ion, in contrast to an ensemble, occupies but a single state at any instant; the possibility of a superposition state appears to be excluded. According to this reasoning, the occupation of the metastable state (level 2) or *shelving* of the ion extinguishes the spontaneous emission of the strong transition, signifying that the weak transition has occurred in absorption. The weak-transition signal is thus amplified.

Beyond these ideas, however, the Dehmelt proposal[1–4] does not clearly specify how the measurements are to be performed, for example, whether the two transitions are to be driven at different times in a pulsed sequence or simultaneously with cw laser sources. It is this circumstance which has given rise to a three-level rate equation calculation of Cook and Kimble,[5] where both transitions are driven simultaneously by an *incoherent source.* Although their model addresses the issue of "quantum jumps," rather than an optical-frequency standard, we wish to point out that under cw conditions the two transitions interact in such a way as to produce a large frequency shift and broadening which renders the forbidden transition useless as a frequency standard. Clearly, the ion can exist in a superposition state, and the consequences cannot be ignored.

Additional and perhaps more profound questions are raised by Dehmelt's proposal which have not been discussed. From the density-matrix calculations given below, it follows that the shelving scheme and the anticipated amplification fail in the case of cw excitation, because the weak and strong transitions compete to such an extent that the population of the metastable state is reduced by orders of magnitude. This point has not been appreciated in the earlier literature. In this case, the effect of the weak transition on the strong transition is slight, and partially populating the metastable state does not extinguish the strong transition on the average.

For the Dehmelt proposal to work, the two laser fields must be pulsed so that they alternate in time, not only to avoid dramatic frequency shifts and line broadening, but also to populate the metastable state appreciably. Ideally, the amplitude and duration of the weak-transition pulse should approximate a $\pi$ pulse so that the population is nearly inverted, and then the strong-transition intensity will be greatly diminished, but of course not extinguished.

The intriguing question of how one calculates a single scattering event, rather than an average, is less obvious. This subject invades the area of measurement theory, and such questions as whether the transition occurs abruptly as in a quantum jump, assumed by Dehmelt and by Cook and Kimble, or gradually in time need to be answered in deciding whether the strong transition is extinguished or not, although this may not be a crucial question for a laser-frequency standard.

To explore these questions, we first perform a three-level density-matrix calculation for the case where the weak and strong transitions are driven continuously and simultaneously by two coherent cw fields. We utilize the three-level density-matrix equations of motion,

$$\dot{\tilde{\rho}}_{13} + i\tilde{\rho}_{13}(\Delta - i\gamma_1/2) = i\alpha(\rho_{33} - \rho_{11}) - i\beta\rho_{12} \ , \qquad (1a)$$

$$\dot{\tilde{\rho}}_{23} - i\tilde{\rho}_{23}(\Delta' + i\gamma_2/2) = i\beta(\rho_{33} - \rho_{22}) - i\alpha\rho_{21} \ , \qquad (1b)$$

$$\dot{\rho}_{12} + i\rho_{12}[\Delta + \Delta' - i(\gamma_1 + \gamma_2)/2] = i\alpha\tilde{\rho}_{32} - i\beta\tilde{\rho}_{13} \ , \qquad (1c)$$

$$\dot{\rho}_{11} = i\alpha(\tilde{\rho}_{31} - \tilde{\rho}_{13}) - \rho_{11}\gamma_1 \ , \qquad (1d)$$

$$\dot{\rho}_{22} = i\beta(\tilde{\rho}_{32} - \tilde{\rho}_{23}) - \rho_{22}\gamma_2 \ , \qquad (1e)$$

$$\dot{\rho}_{33} = i\alpha(\tilde{\rho}_{13} - \tilde{\rho}_{31}) + i\beta(\tilde{\rho}_{23} - \tilde{\rho}_{32}) + \rho_{11}\gamma_1 + \rho_{22}\gamma_2 \ , \qquad (1f)$$

developed by Brewer and Hahn[6] but modified for the case of an isolated atom having a "V"-level structure, level 3 being the common lower level. It is assumed that a strong cw optical field drives the (1-3) transition at the Rabi frequency $\alpha = \mu_{13}E_0/2\hbar$ and a weak optical field the (2-3) transition at a rate $\beta = \mu_{23}E_0'/2\hbar$, where $\alpha >> \beta$ (Fig. 1). States

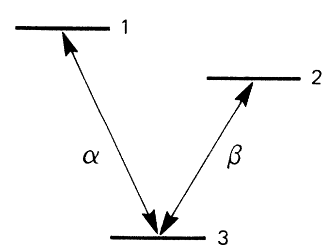


FIG. 1. Energy-level diagram where the Rabi frequencies satisfy $\alpha >> \beta$.

 

$|1\rangle$ and $|2\rangle$ decay to $|3\rangle$ by spontaneous emission at rates $\gamma_1$ and $\gamma_2$, respectively, where $\gamma_1 >> \gamma_2$. The tuning parameters of the strong and weak transitions are defined by $\Delta = -\Omega_1 - \omega_{31}$ and $\Delta' = \Omega_2 + \omega_{32}$, respectively, $\Omega_{1,2}$ being an angular optical frequency, and $\omega_{ij} = (E_i - E_j)/\hbar$ is a level splitting. The tilde denotes the transformations $\rho_{13} = \tilde{\rho}_{13} e^{-i\Omega_1 t}$ and $\rho_{23} = \tilde{\rho}_{23} e^{-i\Omega_2 t}$ to a double rotating frame.

To obtain perturbative solutions of (1) under steady-state conditions, we first derive to zeroth order in $\beta$ the well-known two-level solutions,

$$\tilde{\rho}_{13}^{(0)} = \alpha(\Delta + i\gamma_1/2)/(\Delta^2 + \gamma_1^2/4 + 2\alpha^2) \ ,$$
$$\rho_{33}^{(0)} = (\Delta^2 + \gamma_1^2/4 + \alpha^2)/(\Delta^2 + \gamma_1^2/4 + 2\alpha^2) \ , \tag{2}$$

for the strong transition, where $\rho_{33}^{(0)} + \rho_{11}^{(0)} = 1$.

We then require that the steady-state population of the metastable state

$$\rho_{22}^{(2)} = \frac{i\beta}{\gamma_2}(\tilde{\rho}_{32}^{(2)} - \tilde{\rho}_{23}^{(2)}) \ , \tag{3}$$

which follows from (1e), be evaluated perturbatively to second order in $\beta$, as this quantity signifies whether the weak transition has occurred or not. By means of (1b) and (1c) and the zeroth-order terms (2), we find that

$$\rho_{22}^{(2)} = 4\frac{\gamma_1}{\gamma_2}\frac{\beta^2}{(\gamma_1^2 + 8\alpha^2)} \frac{\left[\frac{\gamma_1}{4}(\gamma_1+\gamma_2) + \frac{\gamma_2}{\gamma_1}\alpha^2\right]\left[\frac{\gamma_2}{4}(\gamma_1+\gamma_2) + \alpha^2\right] + \Delta'^2\left[\frac{\gamma_1\gamma_2}{4} + \left(1 + \frac{\gamma_2}{\gamma_1}\right)\alpha^2\right]}{\left[\Delta'^2 - \alpha^2 - \frac{\gamma_2}{4}(\gamma_1+\gamma_2)\right]^2 + \Delta'^2\left[\gamma_2 + \frac{\gamma_1}{2}\right]^2} \ . \tag{4}$$

In simplifying the above equation, we have assumed without significant loss of generality that the strong transition is driven on resonance ($\Delta = 0$).

The line shape of this expression reveals that the weak transition has not one but two resonances located at

$$\Delta' = \pm[\alpha^2 + (\gamma_2/4)(\gamma_1+\gamma_2)]^{1/2} \ . \tag{5}$$

In other words, the Rabi sidebands of the strong transition are implanted on the weak transition through the common level 3, which is a dressed state. Since we assume that $\alpha > \gamma_1 >> \gamma_2$, the frequency shift (5) is essentially $\Delta' \simeq \pm\alpha$. With the above inequalities, Eq. (4) can be simplified in the high-field limit to yield

$$\lim_{\alpha \to \infty} \rho_{22}^{(2)} = \frac{(\gamma_1/2\gamma_2)\beta^2[\Delta'^2 + (\gamma_2/\gamma_1)\alpha^2]}{(\Delta'^2 - \alpha^2)^2 + \Delta'^2\gamma_1^2/4} \ , \tag{6}$$

which shows rather remarkably that the weak-transition linewidth (full width at half maximum) is

$$\Delta\omega_{1/2} = \gamma_1/2 \ , \tag{7}$$

and not $\gamma_2$. This result may be counterintuitive, since one might expect, naively, that the strong transition would power broaden the weak transition. In any event, since $\gamma_1 >> \gamma_2$, the weak-transition linewidth is totally dominated by the strong transition.

According to Eq. (6), the magnitude of the level-2 population at resonance $\Delta' = \pm\alpha$ is

$$\rho_{22}^{(2)}(\Delta' = \pm\alpha) = 2\beta^2/\gamma_1\gamma_2 \ , \tag{8a}$$

and also the corresponding change in the population of state 1 is

$$\rho_{11}^{(2)}(\Delta' = \pm\alpha) = -\tfrac{1}{2}\rho_{22}^{(2)}(\Delta' = \pm\alpha) \ . \tag{8b}$$

Equation (8a) is to be compared with the corresponding Cook-Kimble result[5] $\sigma_1^2/m_1^2$, which in our notation takes the form

$$[\rho_{22}^{(2)}]_{CK} \sim 2\beta^2/\gamma_2^2 \ . \tag{9}$$

Assuming $1/\gamma_2 = 50$ msec and $1/\gamma_1 = 20$ nsec, numbers used by Dehmelt[1-4] to illustrate the shelving principle for $Tl^+$, we see that the Cook-Kimble result disagrees with (8a) by the large factor $\gamma_1/\gamma_2 = 2.5 \times 10^6$. Equation (9) disagrees, because the three-level problem has been reduced to a two-level problem, which cannot include the competition of the two transitions. According to (8b), the change in intensity of the strong transition due to the weak transition generates the signal $\Delta I_1 = \gamma_1 \Delta\rho_{11} = \beta^2/\gamma_2$, whereas in the two-level problem the weak transition generates the signal $I_2 = \gamma_2\rho_{22} = 2\beta^2/\gamma_2$. Therefore, in this case there is no amplification.

Now consider the low-field limit of (4). By comparing terms in the numerator, we see that when $\alpha << \frac{1}{2}\sqrt{\gamma_1\gamma_2}$, (4) reduces to

$$\rho_{22}^{(2)} = \frac{\beta^2}{(\Delta')^2 + \gamma_2^2/4} \ . \tag{10}$$

This result displays the expected linewidth of $\gamma_2/2$ when both transitions are driven weakly. Note that the crossover point $\alpha \sim \frac{1}{2}\sqrt{\gamma_1\gamma_2}$ between high- and low-field limits is more restrictive than the crude approximation $\alpha \sim \gamma_1/2$.

To summarize, the large frequency shift (5) and line broadening (7) and the smallness of the population change in state 1, Eq. (8b), makes the cw excitation scheme unattractive.

In considering the time-dependent solutions, we treat a case more appropriate to Dehmelt's suggestion,[1-4] where the two fields are applied alternately in time, and the effects of superposition are eliminated, as in recent microwave experiments.[7] Assume that the 2-3 transition is driven by an initial pulse of duration $T$, and then a second pulse beginning at time $T$ excites the 1-3 transition. Ideally, the first pulse is a $\pi$ pulse so that the population of the three-level system resides totally in level 2 at time $T$ and thereafter feeds state 3 by spontaneous emission. The problem then reduces to a two-level calculation which can be treated exactly using a Laplace transform technique.[8] At short times, the nutation oscillation damps out in a time $\sim 1/\gamma_1$, whereas the long-time behavior is given by

$$\rho_{11}(t) \cong \frac{2\alpha^2\rho_{22}(T)}{4\alpha^2 + (1/2)\gamma_1^2}(1 - e^{-\gamma_2(t-T)}) \ . \tag{11}$$

For $\alpha^2 >> \gamma_1^2$, the population will be distributed equally between states 1 and 3, and the fluorescence signal will have grown in a time $\sim 1/\gamma_2$ from 0 to 50% of the maximum value. This case represents the maximum amplification possible or $\frac{1}{2}\gamma_1/\gamma_2$. A relevant point is that if the strong and weak transitions are excited by pulses of duration $1/\gamma_1$ and $1/\gamma_2$, respectively, then the intensities required to achieve the $\pi$-pulse condition are related by $I_2 = I_1\gamma_2/\gamma_1$, i.e., rather interestingly the weak transition requires a far weaker pulse intensity than the strong transition.

As an alternative, assume that the initial or preparative pulse is sufficiently long ($\gtrsim 1/\gamma_2$) such that the steady-state population

$$\rho_{22}(T) = \frac{\beta^2}{2\beta^2 + \Delta'^2 + (1/4)\gamma_2^2} \tag{12}$$

is essentially reached. Application of the second or probing pulse then yields the long-term behavior

$$\rho_{11}(t) = \frac{2\alpha^2}{4\alpha^2 + (1/4)\gamma_1^2}[1 - \rho_{22}(T)e^{-\gamma_2(t-T)}] \ . \tag{13}$$

Thus, if the two-pulse sequence is repeated many times, Eq. (13) predicts that the strong transition faithfully monitors the spontaneous decay of the weak transition. For each photon emitted in the 2-3 transition, $\frac{1}{2}\gamma_1/\gamma_2$ photons are radiated in the 1-3 transition, and in the relevant case of $Tl^+$, the amplification[2] is about $10^6$. From (12), we also see that the clock transition can exhibit power broadening.

It should be stressed that the density-matrix solutions as presented here pertain to averaged quantities, for example, to the average fluorescence intensity over many observations of a single ion or equivalently to a single observation of an ensemble of independent ions. Furthermore, the time dependence is smoothly varying rather than discontinuous. If the preparative and monitoring pulses are repeated, there will be a statistical distribution in the measurement of the fluorescence intensity from one pulse sequence to the next because of the fluctuations associated with spontaneous emission. On the average, as demonstrated above, it is possible to observe a significant reduction in the 1-3 fluorescence rate when the weak transition 2-3 is partially saturated. This is in contrast with the steady-state scheme, where on the average only a negligible change in the fluorescence signal can be obtained, which is expected to escape experimental observation because of the statistical nature of the emission process.

Compare now the proposal of Dehmelt and the subsequent work of Cook and Kimble who resort to an intuitive picture where the individual quantum event of spontaneous emission exhibits a discontinuous jump between a strongly emitting and a nonemitting state. Whether these fluctuations switch the strong transition completely on or off is a fundamental question itself and will be reported subsequently in a detailed quantum statistical calculation.[9,10] Moreover, if these discontinuities do occur, it can be shown in a simple calculation that the time-averaged emission over the "on" period $1/\gamma_2$ and the "off" period $(1/\gamma_2)(1-\rho_{22})/\rho_{22}$ is $I = \gamma_1(1-\rho_{22})/2$, in agreement with the density-matrix calculation. Of course, for an ultimate laser-frequency standard, it is the average quantity that is of importance.

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