Generalized Hyper-Ramsey Resonance

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(Dated: March 12, 2015)

We derive an exact generalization of the Ramsey transition probability to improve ultra-high precision measurement and quantum state engineering when a particle is subjected to controllable separated oscillating fields. The phase-shift accumulated at the end of the interrogation scheme and associated to the particle wave-function is offering a very high-level control of quantum states in various laser parameters conditions. The Generalized Hyper-Ramsey Resonance based on independent manipulation of interaction time, field amplitude and frequency detuning is presented to increase performances in the next generation of atomic, molecular and nuclear clocks, to upgrade high resolution frequency measurement in Penning trap mass spectrometry, for a better control of light induced frequency shifts in matter wave interferometer and quantum information processing.

PACS numbers:

Nowadays, very precise measurements are required in many fields of physics such as metrology and fundamental tests of physical theories. Laser pulsed spectroscopy is one technique to achieve such measurements. It has become a universal tool to investigate interaction between light and matter in quantum clocks \cite{1, 2}, in cavity quantum electrodynamics experiments \cite{3, 4} and in atomic, molecular and neutron interferometry \cite{5, 6, 7}.

To improve resolution of frequency measurements in the atomic and molecular beam resonance method initially invented by I.I. Rabi \cite{8}, N.F. Ramsey proposed to replace the single oscillatory field by a double microwave excitation pulse separated by a free evolution without any electromagnetic field perturbation \cite{9, 10}. The low sensitivity of the Ramsey’s spectroscopy to field inhomogeneities associated to a strong reduction of light-shift on the probing atomic transition has drastically impacted the time and frequency metrology \cite{11, 12, 13} leading to microwave standards at the relative 10\textsuperscript{-16} level of accuracy \cite{14, 15}.

Ultra-high resolution of frequency measurement has been achieved with very long storage time of Doppler and recoil free quantum particles using laser cooling techniques in ion traps \cite{16, 17} and optical lattice clocks \cite{18, 19}. The level of 10\textsuperscript{-18} relative accuracy, almost achieved \cite{20}, requires a very precise control of atomic or molecular interactions to cancel systematic frequency shifts whether fermionic or bosonic species are used \cite{21, 22}. With next generation of quantum clocks, stringent tests of general relativity would be accessible as well as new applications in geophysics and hydrology \cite{23, 24}.

Also more recently, very high precision measurement has become relevant for mass spectrometry \cite{25} where the highest precision was reached using ions in a Penning trap. The use of Ramsey’s method of separated oscillating fields has provided a significant reduction in the uncertainty of the cyclotron frequency and thus of the ion mass of interest combined with a faster acquisition rate \cite{26, 27}.

For the next progress in very high precision, the recent Hyper-Ramsey scheme \cite{28} is a very promising evolution of Ramsey pulsed spectroscopy to overcome some issues. Composite pulses inspired by NMR techniques \cite{29, 30} are now used to compensate simultaneously for noise decoherence, pulse area fluctuation and residual frequency offset due to the applied laser field itself. The probing protocol is reminiscent of the spin echo technique where the sequence of pulses was originally applied to suppress inhomogeneous effects causing a spin relaxation \cite{31, 32, 33}. This new pulsed method was successfully implemented on a single trapped \textsuperscript{171}Yb\textsuperscript{+} ion demonstrating efficient reduction of the light-shift by several orders of magnitude \cite{34}.

In this letter, we have elaborated an exact generalization of the Ramsey transition probability as a function of frequency which is of considerable importance in the design, interpretation and very high accuracy of future laser pulses experiments in fundamental and applied physics. It is producing an accurate control of energy levels at the end of the interrogation sequence as in quantum information processing \cite{35}. To achieve this main goal, the generalized transition probability is derived for independent particles interacting with separated and controllable oscillating fields. Field amplitudes, frequency detunings and pulse durations can be manipulated individually as shown in Fig. 1. Adopting a multi-zone interaction wave-
The phase-shift accumulated after the entire interrogation scheme is

\[
\tan \Phi = \frac{\frac{\delta_j \tan \theta_j + \frac{\Omega_{jk}}{\omega_{jk}} \tan \theta_k}{\Omega_j \omega_k \tan \theta_j + \Omega_k \omega_j \tan \theta_k} + \frac{\Omega_k}{\omega_{jk}} \tan \theta_j \tan \theta_k}{1 - \frac{\delta_j \tan \theta_j + \frac{\Omega_k}{\omega_{jk}} \tan \theta_k}{\Omega_j \omega_k \tan \theta_j + \Omega_k \omega_j \tan \theta_k}} + \frac{\delta_j \tan \theta_j + \frac{\Omega_{jk}}{\omega_{jk}} \tan \theta_k}{\Omega_j \omega_k \tan \theta_j + \Omega_k \omega_j \tan \theta_k} \tan \theta_j \tan \theta_k}
\]

(5)

From Eqs. [1] to [3] lineshape, population transfer efficiency and frequency-shift affecting the resonance can be accu-

To investigate very precisely a modified version of the Ramsey or Hyper-Ramsey spectroscopy, the generalized transition probability has to be dependent on pulse area \( \theta_i = \omega_i \tau_i / 2 \) \((i = i, j, k)\) with different driving Rabi amplitudes \( \Omega_i \) and frequency detunings \( \delta_i \) via the effective frequency \( \omega_i = \sqrt{\delta_i^2 + \Omega_i^2} \). During pulses, some light-shift from off-resonant states may be present and a laser step frequency activated to rectify any anticipated shift thus requiring a redefinition of frequency detunings as \( \delta_i \equiv \delta_i \pm \Delta_i \). Additional phase reversion of laser fields and nonuniform pulsed excitation conditions modifying the entire spectral lineshape are included into the computation of the transition \[31, 45\]. The exact expression of the generalized Ramsey probability for a particle starting from initial state \(|g\rangle\) to final state \(|e\rangle\) is given in a compact form as:

\[
P_{g \rightarrow e} = \alpha \left[ 1 + \beta(\Phi)^2 \right] \left[ 1 + \frac{2\beta(\Phi)}{1 + \beta(\Phi)^2} \cos(\delta T + \Phi) \right],
\]

(1)

with a clock frequency detuning \( \delta \) during free evolution without light. We have introduced for convenience the notation

\[
\beta(\Phi) = \beta \sqrt{1 + \tan^2 \Phi},
\]

(2)

where envelopes \( \alpha, \beta \) driving the resonance amplitude are respectively defined by

\[
\alpha = \left( 1 + \frac{\delta_i^2}{\omega_i^2} \tan^2 \theta_i \right) \left( 1 + \frac{\delta_j^2}{\omega_j^2} \tan^2 \theta_j \right) \left( \frac{\Omega_j}{\omega_j} \tan \theta_j + \frac{\Omega_k}{\omega_k} \tan \theta_k \right)^2 \cos^2 \theta_j \cos^2 \theta_j \cos^2 \theta_k,
\]

(3a)

\[
\beta = \frac{\Omega_{jk}}{\omega_{jk}} \tan \theta_j \left( 1 - \frac{\delta_j \delta_k + \Omega_{jk}}{\omega_{jk}} \tan \theta_j \tan \theta_k \right) \left( \frac{\Omega_j}{\omega_j} \tan \theta_j + \frac{\Omega_k}{\omega_k} \tan \theta_k \right)^2 \left[ 1 - \frac{\delta_j \tan \theta_j + \frac{\Omega_{jk}}{\omega_{jk}} \tan \theta_k}{\Omega_j \omega_k \tan \theta_j + \Omega_k \omega_j \tan \theta_k} \right] + \frac{\delta_j \tan \theta_j + \frac{\Omega_{jk}}{\omega_{jk}} \tan \theta_k}{\Omega_j \omega_k \tan \theta_j + \Omega_k \omega_j \tan \theta_k} \tan \theta_j \tan \theta_k,
\]

(3b)

including a reduced variable

\[
\frac{\delta_{jk} \tan \theta_{jk}}{\omega_{jk}} = \left( \frac{\delta_j \Omega_k - \Omega_j \delta_k}{\Omega_j \omega_k \tan \theta_j + \Omega_k \omega_j \tan \theta_k} \right) \tan \theta_j \tan \theta_k.
\]

(4)

The phase-shift accumulated after the entire interrogation scheme is

\[
\delta_{jk} \tan \theta_{jk} = \left( \frac{\delta_j \Omega_k - \Omega_j \delta_k}{\Omega_j \omega_k \tan \theta_j + \Omega_k \omega_j \tan \theta_k} \right) \tan \theta_j \tan \theta_k.
\]

(5)

function model \[33\], we have derived the analytical form of the generalized transition probability and the phase-shift driving the resonance frequency position around the extremum of the central fringe. Such a resonance can be coherently excited by stimulated two-photon Raman transitions \[38-40\], by magnetically induced transitions \[41\] or by quadrupolar radio-frequency field interaction in mass spectrometry \[42\]. The sequence of composite pulses summarized in Fig. 1 and described by the analytical expression of the transition probability is investigated to improve control of external perturbations such as light-shift or residual magnetic fields on the lineshapes \[33, 42\]. It can be operated in trapped atom clocks and extended to ion trap mass spectrometers and molecular beams or fountain devices where moving particles are interacting with multiple oscillating fields during a free fly.

FIG. 1: (color online). Quantum system with a narrow \(|g\rangle \leftrightarrow |e\rangle\) clock transition probed by a laser pulse excitation. The sequence of composite pulses with specified laser parameters includes detuning \(\delta\), field amplitude \(\Omega\), pulse duration \(\tau\) where \(l = i, j, k\) and a free evolution time \(T\) between pulses.
The phase-shift given by Eq. 5 is the main result needed in precise control of quantum states to suppress any frequency-shift induced by the laser excitation itself. By tailoring phase-shift parameters and engineering the resonance amplitude, it is possible to generate quantum spectroscopy signals for increased resolution or better stability. Following [16], Eq. 5 can be decomposed into three terms as:

$$\Phi = \arctan \left[ \frac{\delta_i \tan \theta_j + \delta_j \tan \theta_k}{1 - \frac{\delta_i \delta_j + \Omega_i \Omega_j}{\omega_j \omega_k} \tan \theta_j \tan \theta_k} \right] + \arctan \left[ \frac{\delta_{jk} \tan \theta_{jk}}{\omega_{jk}} \right] + \arctan \left[ \frac{\delta_i \tan \theta_i}{\omega_i} \right].$$

(6)

Some combinations of these phases related to each pulse area may interfere to eliminate the central fringe frequency-shift by particular choices of laser step frequency, pulse duration and phase reversion of the light field. Composite pulses are thus realized where the key point is to establish efficient quantum control protocols which compensate for frequency-shift and are robust to small change in pulse area while achieving a highly contrasted population transfer between the targeted states [38]. Such quantum engineering of phase-shift has been recently proposed in the Hyper Raman-Ramsey spectroscopy of a stimulated two-photon forbidden clock transition eliminating the detrimental light-shift contribution [38].

The generalized Hyper-Ramsey transition probability has been computed with some pulse protocols reported in Table I. The standard sequence referring to the Ramsey (R) protocol is compared with two others sequences based on nonstandard G-H-R protocols. The panels of Fig. 2 display resonance fringes corresponding to selected protocols in Table I. In Fig. 2(a), Ramsey fringes have been simulated using parameters following the R protocol with two \( \pi/2 \) pulse areas. Hyper-Ramsey fringes have also been simulated with \( \pi/2 \) and \( \pi - \pi/2 \) pulse areas according to G-H-R protocols leading to the same resonance line shown in Fig. 2(b).

Non-linear behaviors of the central fringe frequency-shift versus a small frequency perturbation in the clock detuning have been investigated and presented in Fig. 3(a) and 3(b). The curves are compared to the linear frequency shift resulting from a single Rabi pulse excitation for guiding-eye. Fig. 3(a) shows the effect of a small external frequency offset on the central fringe position. The Ramsey resonance is more sensitive to the induced shift than the Generalized Hyper-Ramsey resonance which is still locked to the correct frequency but with a small asymmetry in the profile. The major advantage of the G-H-R pulsed technique is a very low frequency shift of the signal with respect to some variation in the frequency offset. We can see on Fig. 3(b) that for small perturbations as a light-shift correction or a residual magnetic field applied on the clock detuning during pulses, the dependence on the central fringe position is still very flat with a strong non-linear behavior.
linear response to the perturbation. The combination of $\pi/2$ and $\pi - \pi/2$ pulse areas in Eq. (4) compensates all terms over a large residual error in frequency as well as their first and second order sensitivity to frequency fluctuation. The suppression of the central fringe shift is also insensitive to small pulse area variation by inserting an intermediate "echo" pulse with a phase inversion of the light field during the intermediate $\pi$ pulse or by reversing phase during initial and final $\pi/2$ pulses (see Table I). In parallel, a laser frequency step may be ultimately applied during pulses to compensate any frequency offset larger than the width of the resonance.

In conclusion, the generalized Hyper-Ramsey resonance provides a new performing tool deeply required for the next generation of quantum interferometers sensitive to very small laser parameters fluctuations. Although the initial Ramsey’s method of separated oscillating fields has proven to be very useful in fundamental and applied physics based on laser pulsed spectroscopy, it still has some fundamental limitations. To overcome these issues, a non standard generalization of the Ramsey protocol has been considered and demonstrated to be even more successful sixty-five years after the original scheme was proposed. The Hyper-Ramsey spectral resonance has been fully extended in this letter to include potential biases, higher-order light-shift corrections on detunings and various modifications of laser parameters exploring non linear frequency responses of quantum particles for ultra precise frequency measurement.

The application of the generalized Hyper-Ramsey resonance should be able to strongly improve frequency uncertainty measurements in next tests of fundamental physics based on atomic or molecular fountains 17-49, in charged ions 50, 51, for small changes in molecular vibrational frequencies based on clocks sensitive to potential variation in the electron-to-proton mass ratio 52, 53 and in searching for a weak parity violation in chiral molecules by laser spectroscopy 54. Quantum phase-shift engineering should impact high resolution mass spectrometry based on the application of the Ramsey method to short-lived ions stored in Penning traps 20 and laser pulsed spectroscopy in cold molecule chemistry 55, 56. An improvement of fitting the generalized Hyper-Ramsey resonance with spectroscopic data may tighten uncertainties in evaluation of frequency shifts when tracking time-variation of the fine structure constant in Stark decelerated cold molecules 57, in measuring gravitationally induced quantum phase shifts for neutrons 58 and for observing spin dependent nuclear scattering lengths of neutrons in Ramsey interferometers 59. In the future, a pair of stretched hyperfine states from a $^{229}$Th nuclear transition may provide a large suppression of several external field shifts 60, 61, where an ultra-narrow clock transition will offer an exquisite test of nuclear quantum engineering spectroscopy at the next level of $10^{-19}$ relative accuracy.

I deeply acknowledge C. Janssen for checking all calculations, E. Arimondo, J. Dalibard, B. Darquié, M. Glass-Maujean, M. Minissale, A.D. Ludlow, E. de Clercq, M-L. Dubernet-Tuckey and Y. Té for discussions and a careful reading of the manuscript.

V.I.Yu. and A.V.T. were supported by the RFBR (grants 14-02-00712, 14-02-00939, 14-02-00806), by the Russian Academy of Sciences, by Presidium of the Siberian Branch of the Russian Academy of Sciences, by the RF Ministry of Education and Science (state assignment No. 2014/139 project No. 825).

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