FREQUENCY METROLOGY VIA QUANTUM INTERFERENCE IN TWO-PHOTON EXCITATION

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ABSTRACT

Quantum interference in atomic two-photon excitation is exploited for optical demodulation, with phase-sensitive detection at ±12 THz having been achieved. By utilizing the atoms as ultrafast nonlinear mixing elements, a new technique for the comparison of essentially arbitrary frequencies over the range 200-2000 nm is presented.

Quantum interference for atomic two-photon excitation due to multiple excitation paths has been observed and exploited in an experiment which demonstrates phase-sensitive demodulation of optical fields separated by an interval of ±12THz. Application of this capability to frequency metrology has been investigated where the atom is utilized as a nonlinear mixing element to compare unknown "target" to known "reference" frequencies. Our analysis indicates that it should be possible with this technique to provide the to measure almost any frequency in the optical and near IR spectrum.

In our scheme, we use three fields of frequencies of ω_a, ω_b and ω_c to illuminate a ladder transition 1→2→3 with corresponding eigenfrequencies ω_{12}, ω_{23} and $\omega_{13}=\omega_{12}+\omega_{23}$ as shown in Fig. 1. Quantum

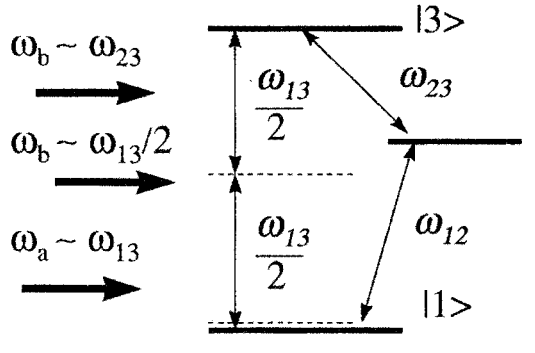

Figure 1 : Two photon ladder scheme for Quantum interference

interference arises due to the existence of two alternative paths of excitation, the "stepwise" dipole absorption of single photons at frequencies ω_a~ω_{12} and ω_b~ω_{23} and the quadrupole two-photon absorption at ω_a~$1/2\omega_{13}$. If the optical phases $\Phi_i=\omega_i t+\varphi$ (i=a,b and c) of the excitation lasers do not sum up according to the "triangle equality" $\Phi_a+\Phi_b=2\Phi_c$, a modulation of the excited state population ρ_{33} at the phase $\Delta=|\Phi_a+\Phi_b-2\Phi_c|$, with the atom acting as a nonlinear mixing element that compares $\Phi_a+\Phi_b$ to $2\Phi_c$.

As a proof-of-principle we have experimentally measured quantum interference of the $6S_{1/2}\rightarrow 6P_{3/2}\rightarrow 6D_{5/2}$ transition in magneto-optically trapped atomic Cesium by exciting the atoms to the $6D_{5/2}$ level using two independent lasers at 852 and 883 nm close to λ_{12} and to $2\lambda_{13}$, and a third laser at 917 nm close to λ_{23}, which is parametrically derived from these first two lasers via a subthreshold optical parametric amplifier. By virtue of the way we generate the 917 beam, we are guaranteed to have that $\omega_a+\omega_b-2\omega_c\equiv 0$, hence $\Delta=|\phi_a+\phi_b-2\phi_c|$. By modulating the phase of the 883 beam by a PZT such that $d\varphi_c/dt=\omega_m$, we find that $\rho_{33}\propto[1+\alpha\cos(2\omega_m t)]$ and hence we expect the excited state population to be modulated due to quantum interference at a frequency $\Omega_m=2\omega_m$. The excited state population is monitored by observing the fluorescence of the decay from the upper to the intermediate level at 917 nm. Figure 2 shows a typical fluorescence signal. Note the reasonable visibility α of the signal that was recorded to be as high as 0.6. Also, note that the data in Fig. 2 represent demodulation of the beatnode between the 852 and 917 fields that have an optical interval separation of $(\omega_a-\omega_b)/2\pi\approx$ 25 THz and where the local oscillator was located at 883 nm.

Control of the quantum interference could be achieved by servoing the phase of the 883 beam with .

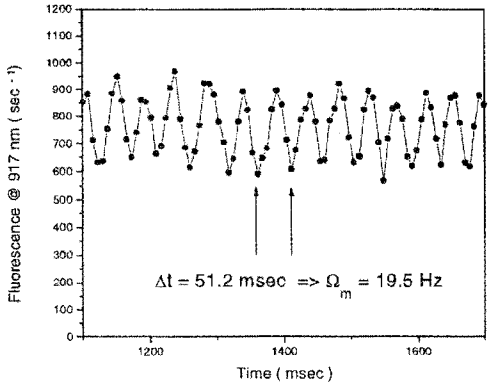

Figure 2: Typical modulation of the fluorescence at 917 nm and hence of the excited state population ρ_{33}. Here the modulation frequency is $\Omega_m/2\pi$~20 Hz .

respect to that of the 852 and 917 beams. In addition, although in our case $\omega_a+\omega_b-2\omega_c \equiv 0$ one can have three independent lasers for which the above condition is not a-priori satisfied. In that case control the quantum interference beatnote could be utilized to phase lock the three independent lasers.

Our scheme is easily generalized to other two-photon transitions as for example in the alkali elements, which can be conveniently laser cooled and trapped. By constraining consideration to transitions where each of the three wavelengths involved is less than 2 μm, we find that there are about 6800 possible ladder transitions for obtaining phase-sensitive detection of $\omega_a+\omega_b-2\omega_c$, where in general the three frequencies are separated by 10's-100's of THz . Note that each of these transitions may by viewed as a nonlinear mixer characterized by its "local oscillator" (LO) frequency $\Omega_{LO}=\omega_c$ and its "sideband separation" defined by $\Delta f=|\omega_a-\omega_c|=|\omega_b-\omega_c|$. Figure 3 shows these characteristics of the 6800 "nonlinear mixers" of our database .

Turning next to applications in frequency metrology, we note that the task of comparing two lasers at different frequencies has so far been addressed by means of multiple stages of conventional non-linear mixing in crystals and electrooptic modulation[1-3]. By contrast our proposal utilizes atoms as ultra-high frequency demodulators in conjunction with 1-3 stages of nonlinear mixing. With our extensive data base of two-photon transitions, we find that we can connect a dense set of "target" frequencies over the range 200 - 2000 nm with the "reference" frequencies (standards) quoted in Ref[3]. Here we assume that offsets below 300GHz can be compensated via electrooptic modulation. To demonstrate the "density" of coverage of our method, we have identifed the appropriate alkali atom and transition that would allow for measurement and phase-stabilization of frequencies over the range 1300 - 1500 nm (relevant for fiber optics communications [4]) in increments of 1nm. Here we employ as frequency standards a subset of the reference frequencies quoted in Ref. [3] (namely 514, 633, 657 and 778 nm). Note that the density of coverage can readily be extended below 1 nm and that target frequencies over the interval 200-2000 nm can be accessed. For example we also have developed a measurement strategy for the Ly-α line in Hydrogen (121.6 cm) by measuring its 4th subharmonic at 972.8 nm.

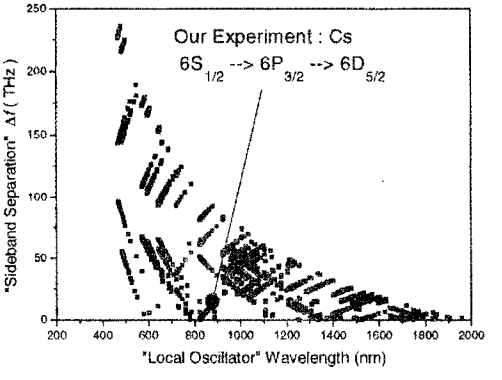

Figure 3: Characteristics of the "atomic nonlinear mixers" of our data base. The one which we implemented experimentally has $\Omega_{LO}/2\pi= 3\times10^{14}$ Hz corresponding to the wavelength of 883 nm and sideband separation $\Delta f/2\pi=12.5$ *THz*.

Perturbative as well as master equation theoretical analyses of the quantum interference have been performed. In addition we have characterized the efficiency of the mixing process with reference to our experiment and the theoretical model.

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