Phase correlation of laser waves with arbitrary frequency spacing

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The theoretically predicted correlation of laser phase fluctuations in Λ type interaction schemes is experimentally demonstrated. We show, that the mechanism of correlation in a Λ scheme is restricted to high frequency noise components, whereas in a double-Λ scheme, due to the laser phase locking in closed-loop interaction, it extends to all noise frequencies. In this case the correlation is weakly sensitive to coherence losses. Thus the double-Λ scheme can be used to correlate e.m. fields with carrier frequency differences beyond the GHz regime.

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The study of quantum interference effects in optical dense media, such as Electromagnetically Induced Transparency (EIT) [8], is one of the most challenging fields in modern quantum optics research. In ideal EIT atoms are decoupled from the resonant light fields and trapped into a dark state, which depends on the radiation amplitudes and phases. Perfectly phase correlated laser fields, i.e. fields with matched Fourier components, even though resonant, are not absorbed. In this paper we show, that in Λ-type excitation under the terms of EIT phase noise of one laser field is transferred to the other one in a way, that perfect phase correlation, i.e. $\omega_2 - \omega_1 = \text{const.}$, is given for the two laser fields $\omega_1$ and $\omega_2$. Such a perfect correlation is essential for high resolution in quantum interference applications. Among the basic field parameters’ correlation processes in coherently prepared media there is pulse matching [1], amplitude and phase matching [2,3], matched photon statistics [4], and intensity- [5] and phase noise correlation [6,7], which extends also to quantized fields, including squeezing [8] and quantum entanglement [9]. In experiments phase correlated laser waves are usually produced via sideband modulation techniques (electrooptical-, acoustooptical modulators, VCSELs [10]) or optical phase locking [11]. Hence the accessible frequency differences of phase correlated laser fields are restricted to the electronically available frequency limits, presently of the order of GHz. We show, that any pair of laser frequencies, even with frequency spacing far beyond the GHz range, can be correlated in phase in the EIT regime. It can be done easily with a simple experimental setup, provided there is a suitable atomic or molecular medium, whose energy level system allows to combine resonantly the two frequencies in form of a Λ-type excitation scheme.

Experiments are performed on the Na D1 line (590nm) by use of frequency stabilized cw-dye lasers (Fig. 1). The two hyperfine sublevels $F = 1, 2$ of Na ground state $^3S_{1/2}$, spaced by $\omega_{12} = 1771.6$ MHz, are connected to $3^2P_{1/2}, F = 2$ by $\sigma^+$-polarized laser beams (frequencies $\omega_1, \omega_2$) and $\sigma^-$-polarized laser beams (frequencies $\omega_3, \omega_4$), thus forming the Λ- and closed double-Λ transition schemes. Laser 1 emitting frequency $\omega_1$ is stabilized to transition $3^2S_{1/2}, F = 1 - 3^2P_{1/2}, F = 2$. An acousto-optical modulator (AM1) driven at 1771.6 MHz produces the first negative order modulation sideband $\omega_2$, resonant to transition $3^2S_{1/2}, F = 2 - 3^2P_{1/2}, F = 2$. An electrooptical phase modulator (PM), which causes a phase shift of $16.3$ mrad/V, is used to modulate the phase of a part of $\omega_1$ by a 100 MHz band limited white frequency noise. For investigations in double-Λ configuration a second pair of frequency components ($\omega_3, \omega_4$) is generated by means of an electrooptical modulator (EM) driven at 885.8 MHz. The two first order modulation sidebands of carrier $\omega_0$ (Laser 2 is stabilized to the Λ-crossover resonance $3^2S_{1/2}, F = 1, 2 - 3^2P_{1/2}, F = 2$) match the Na ground state hyperfine splitting. To avoid an additional absorption background the carrier frequency $\omega_0$ is suppressed by an electronically stabilized Fabry-Perot etalon. It is combined with a polarizing beamsplitter and a quarter wave plate to circumvent intense retroreflection into the dye laser. A defined phase relation between frequency pairs ($\omega_1, \omega_2$) and ($\omega_3, \omega_4$) is ensured by an internal frequency synchronization of the two frequency generators driving AM1 and EM [12]. The frequency pairs are prepared in circular polarization and transmitted collinearly through the absorption cell at typical incident intensities of 200 mW/cm$^2$ per frequency component. After the cell they are separated by a quarter wave plate and a polarizer and observed separately with a 15 GHz InGaAs Schottky photodiode using heterodyne spectroscopy. To observe the spectral noise...
distribution $S_j(\tau, \omega)$, i.e. the intensity spectrum of phase noise of a single laser component with carrier frequency $\omega_1$, a part of $\omega_1$ is shifted by 260 MHz using acoustooptical modulator AM2 ($\omega_0 = \omega_1 + 260MHz$) and superposed with the transmitted $\omega_1$ laser beam. As $\omega_0$ is free of noise, the spectrum $S_j(\tau, \omega)$ can be observed directly by taking the beat signal $S_{1_5}$ at frequency $|\omega_1 - \omega_5|$ via a 2.8 GHz electronic spectrum analyzer. The optical density $\tau(T)$ was calibrated via an absorption measurement on transition $3^2S_{1/2}, F = 2 - 3^2P_{1/2}, F = 2$. The vapor cell is a 1 cm$^3$ cube with a sidearm containing the sodium reservoir. Vapor density (and $\tau$) is controlled via this reservoir temperature, which is stabilized with an accuracy of 1°C. The windows are kept at higher temperature to avoid darkening. The cell is placed inside an arrangement of three mutually orthogonal Helmholz coils to compensate stray magnetic fields.

A theoretical analysis of the correlation of phase fluctuations for EIT in $\Delta$ systems is performed in refs. [4, 5]. It is shown, that the spectrum $W_\phi$ of phase-difference fluctuations $\delta \varphi = \delta \varphi_1 - \delta \varphi_2$ of two laser fields $\omega_1$ and $\omega_2$ decays with the propagation distance: $W_\phi(z, \omega) = W_\phi(0, \omega) \exp\left(-\int_0^z 2\kappa(z', \omega) dz'\right)$. Here $z$ is the propagation distance, and $\omega$ is the noise frequency. Contributions from the atomic noise are neglected, which is justified under the conditions of EIT. Obviously the laser phase fluctuations $\delta \varphi_1$ and $\delta \varphi_2$ become more and more correlated with the propagation distance. Slight extension of the theory in [4, 5], assuming equal dipole moments, decay rates and Rabi frequencies of the involved atomic transitions and no phase correlation before interaction, gives for the individual spectra of phase fluctuations $W_{11}, W_{22}$ and for the cross-correlation $W_{12}$ the following approximate dependencies on the propagation path

$$W_{11}(z, \omega) = \frac{1}{4}(W_{11}(0, \omega) + W_{22}(0, \omega))(1 + e^{-2x}) +$$
$$\frac{1}{2}(W_{11}(0, \omega) - W_{22}(0, \omega))$$

(1)

$$W_{22}(z, \omega) = \frac{1}{4}(W_{11}(0, \omega) + W_{22}(0, \omega))(1 + e^{-2x}) -$$
$$\frac{1}{2}(W_{11}(0, \omega) - W_{22}(0, \omega))$$

(2)

$$ReW_{12}(z, \omega) = \frac{1}{4}(W_{11}(0, \omega) + W_{22}(0, \omega))(1 - e^{-2x})$$

(3)

$$ImW_{12}(z, \omega) = \frac{1}{2}(W_{11}(0, \omega) - W_{22}(0, \omega))e^{-x}$$

(4)

with $x = \int_0^z \kappa(z', \omega) dz'$. These eqs. show, that two laser fields transfer and exchange their noise properties in the course of propagation. After a sufficient long propagation path, the noise spectra of both fields are identical and the fields are perfectly correlated: $|W_{12}|^2 = \sqrt{(W_{11}W_{22})^2}$. If initially only one of the fields has fluctuations above shot noise ($W_{11}(0, \omega) \neq 0, W_{22}(0, \omega) = 0$), then we observe from the above eqs. the noise transfer from component $\omega_1$ to the initially noise free component $\omega_2$: $W_{22}(z, \omega) = \frac{1}{4}W_{11}(0, \omega)(1 - e^{-x})^2$. Simultaneously the cross-correlations $ReW_{12}(z, \omega)$ grow exponentially at comparable rate.

In order to test this prediction experimentally, we observe the intensity noise spectra $S_1(\tau, \omega)$ and $S_2(\tau, \omega)$ by taking the beat signals $S_{15}$ and $S_{25}$ respectively (the centers of the noise spectra are normalized to zero frequency by subtracting the frequency difference of the two carriers). Further we observe the dependence of the FWHM $\Delta S_{12}$ of the beat signal $S_{12}$ on $\tau$. An increasing optical path length $z$ is simulated by changing the optical density $\tau$ via cell heating. Usually the random frequency jitter of light emitted by two different lasers is uncorrelated, and the width $\Delta S_{12}$ of the beat signal at frequency difference $\omega_2 - \omega_1$ is the sum of the linewidths of the two lasers. Phase correlation due to EIT improves with $\tau$, thus $\Delta S_{12}(\tau)$, similar to $W_\phi(z)$, reflects the degree of correlation between different frequency sidebands of $\omega_1$ and $\omega_2$. $\Delta S_{12} = 0$ corresponds to perfect correlation. Fig. 2(a) shows a measurement of the beat signal $S_{15}$, that represents the spectrum of phase fluctuations $S_1(\tau, \omega)$ modulated onto carrier $\omega_1$.

![Figure 2: Phase noise transfer at $\tau = 7.3$: Intensity spectra $S_{15}(\omega)$ (a) and $S_{25}(\omega)$ (b), representing the spectra of phase fluctuations $S_1(\tau, \omega)$ and $S_2(\tau, \omega)$ respectively; $\Delta R = 0$ (black curve) and $\Delta R = 20MHz$ (gray curve).](image)

The waveguide structure is caused by the HF noise amplifier characteristic, the central peak occurs at the exact carrier frequency difference. The measurement of beat signal $S_{25}$ (Fig. 2(b) - black curve) exactly represents the spectrum of phase fluctuations $S_2(\tau, \omega)$: A noise transfer from frequency component $\omega_1$ to the initially noise free frequency component $\omega_2$, as predicted by eq. 2, is obvious.

Under the same assumptions as for eqs. 1-4 we derived the noise transfer rate

$$\kappa = \kappa_0 \frac{\Gamma_0^2}{\Gamma_g^2 + (\omega - \Delta R)^2} \frac{\omega^2}{\Gamma_g^2 + \omega^2}.$$  

(5)
where \( \kappa_0 = 4\mu |\Omega|^2 N/\gamma^2 T_q \) is the maximum rate, with transition coupling elements \( \mu_j = \omega_j d_j^2 / \hbar c \), dipole transition moments \( d_j \), spontaneous decay rates \( \gamma_j \), Rabi frequencies \( \Omega_j = d_j E_j / \hbar \) (all assumed equal), atom density \( N \), Raman detuning \( \Delta_R = \omega_i - \omega_2 - \omega_1 \), and the transparency window width \( \Gamma_g = \Gamma + 2 |\Omega|^2 / \gamma \). \( \Gamma \) is the dark state coherence decay rate. The noise transfer rate \( \kappa \) shows a typical Lorentzian profile with respect to the Raman detuning \( \Delta_R \), with the width \( \Gamma_g \). Only for a small band of Raman detuning \( \Delta_R \) around the noise frequency \( \omega \) the transfer rate is of considerable magnitude - thus the efficient noise transfer is due to EIT. This fact is supported by our measurements: In Fig.2(b), the second (gray) curve is obtained for \( \Delta_R \) larger than the transparency window width. We see, that no efficient noise transfer occurs, except for the beat frequencies \( \omega \approx \Delta_R = 20 MHz \), in correspondence with Fig.3. We have performed such measurements for series of different Raman detunings and observed, that the noise transfer efficiency depends on \( \Delta_R \) as a Lorentzian function, in very good agreement with eq. 6.

An important feature follows from the factor \( \omega^2 / (\Gamma^2 + \omega^2) \) in eq. 6: There is no fluctuations correlation for frequencies inside the transparency window \( \omega < \Gamma_g \), independent on the values of \( \Delta_R \) ! This is due to the adiabatic regime in this noise frequency range, where small variations in the laser phase are so slow, that the atom follows the evolution of the fields and remains in dark state. Only high-frequency noise components \( \omega > \Gamma_g \) are correlated. As the laser intensity itself exponentially decreases with optical density, and \( \Gamma_g \sim \Omega^2 \), the ultimate low-frequency threshold for phase correlation is determined by \( \Gamma \). This fact is clearly demonstrated in our measurements: We observed the \( S_{12} \) beat signal at different optical densities \( \tau \) for zero Raman detuning. Since noise transfer happens, the spectra \( S_1 (\tau, \omega) \) and \( S_2 (\tau, \omega) \) are almost identical, and the corresponding beat signal \( S_{12} \) shows a Lorentzian profile with a half width \( \Delta S_{12} \) limited by \( \Gamma_g \). The FWHM \( \Delta S_{12} (\tau) \) was evaluated and depicted in Fig.3(a): As \( \omega_1 \) and \( \omega_2 \) propagate, more and more lower noise frequencies \( \omega \) become correlated, and \( \Delta S_{12} (\tau) \) exponentially decreases and asymptotically approaches the limit set by \( \Gamma \). The curve \( \Delta S_{12} (\tau) \) was fitted by an exponential decay function yielding a dark state coherence decay rate \( \Gamma = 0.3 (1 MHz) \), which is in good agreement with measurements of \( \Gamma \) made independently of the present experiment. Additionally measurements of \( \Delta S_{12} (\tau) \) at 8MHz Raman detuning show, that outside the EIT regime there is no correlation effect at all!

In principle, the rate \( \Gamma \) can be made very small if the low lying states \( |1 \rangle \) and \( |2 \rangle \) of a \( \Lambda \) system are close in energy. However, for a considerable energy difference of \( \omega_1 \) and \( \omega_2 \) (e.g., in the optical range), \( \Gamma \) will be quite large, of the order of \( \gamma \). Moreover, the correlation mechanism requires EIT, which in turn requires sufficient high intensities \( |\Omega|^2 \gg \Gamma \gamma \), so that the transparency window gets even wider. Thus, in such realistic cases, only small parts of the phase noise spectrum can be correlated. This problem can be avoided if one uses atom excitation in the double-\( \Lambda \) scheme: Here the phase noise spectra have approximately the same dependence on the propagation length as above - eqs. 11-12, but similar relations are valid for any pair of the four participating frequency components. Thus noise transfer and correlation of phase fluctuations occur between all four radiation fields. Essential for the double-\( \Lambda \) system is the different noise transfer coefficient, which for the stationary situation is given by

\[
\kappa \simeq \kappa_0 \frac{\Gamma_g^2 \cos \varphi_0 + (\omega - \Delta_R)^2 (1 + \cos \varphi_0)}{\Gamma_g^2 + (\omega - \Delta_R)^2},
\]

Here \( \kappa_0 \) is the same as in eq. 5, and \( \Gamma_g = \Gamma + 4 |\Omega|^2 / \gamma \) for a double-\( \Lambda \) system. \( \varphi_0 = (\varphi_1 - \varphi_2) - (\varphi_3 - \varphi_4) \) is the value of the mean relative phase of the transition excitation loop at a given optical length. The phase \( \varphi_0 \) itself evolves with the propagation distance, and inside the transparency window \( (\omega - \Delta_R) < \Gamma_g \) the phase \( \varphi_0 \) approaches the value \( 2\pi n \), while outside the transparency window \( \gamma \) changes very slowly, and on a scale \( \kappa_0^{-1} \varphi_0 \) is almost constant. For noise frequencies inside the transparency window \( \k \simeq \kappa_0 \cos \varphi_0 \), while outside \( \k \simeq \kappa_0 (1 + \cos \varphi_0) \). Consequently the noise transfer coefficient is not zero in the whole frequency range. In contrast to EIT in the \( \Lambda \) system, the correlation of phase fluctuations takes place for all noise frequencies \( \omega \), including the low-frequency range! This unlimited correlation is demonstrated in the experiment: After transmission in double-\( \Lambda \) excitation

**FIG. 3:** FWHM \( \Delta S_{12} (\tau) \) of the \( \omega_1 / \omega_2 \) beat signal in dependence on \( \tau \) at zero- and 8MHz Raman detuning: (a) In the \( \Lambda \)-scheme the exponential decay is limited by \( \Gamma \); (b) In the double-\( \Lambda \)-scheme the exponential decay asymptotically approaches zero.
the beat signals $S_{12}(\omega)$ and $S_{44}(\omega)$ (the latter is initially free of noise) are observed separately (Fig.1). Due to the noise amplifier’s cut-off frequency of 0.5 MHz the phase noise spectrum $S_\tau(\tau,\omega)$ and accordingly the beat signal $S_{12}(\omega)$ shows a distinct dip around zero peak.

![Intensity spectra $S_{12}(\omega)$ and $S_{44}(\omega)$, taken separately at $\tau = 7.3$ in case of four-frequency excitation in double-Λ-scheme.](image)

Such a pronounced dip is not found in the $S_{44}(\omega)$ intensity spectrum, which confirms a phase noise transfer without frequency limits for the double-Λ regime. Analogous to the measurements in Fig.3(a) we evaluated the FWHM $\Delta S_{12}(\tau)$ (Fig3(b)). In accordance with our model $\Delta S_{12}(\tau)$ exponentially decreases and (within the margin of fitting error) asymptotically approaches zero.

In double-Λ excitation the noise transfer coefficient $\kappa \neq 0$ almost independent on the laser intensity, also for large $\Gamma$ (e.g. for large carrier frequency differences). Inside the transparency window the correlation happens, in general, slower than in the $\Lambda$ system, as can be seen from the different decay rates of $\Delta S_{12}(\tau)$ in Fig 3. This is the price to pay for low-frequency noise correlation. It is interesting that for a singular point $\varphi_0 = \pi$, where no atomic coherence is built up, the transfer rate is negative for all noise frequencies and at all propagation distances (no phase matching occurs at $\varphi_0 = \pi$, see ref 3): The noise grows. This fact has also been mentioned in ref. 3.

The double-Λ system can be applied for a phase correlation of lasers with substantially different wavelengths (e.g. correlation of UV with IR) up to the shot noise (and even beyond - using entanglement 13). In practice a double-Λ excitation scheme is established easily - each of the two fields of an appropriate $\Lambda$ configuration can be shifted in frequency by an equal amount using sideband modulation, and afterwards all four resulting fields are superimposed in the medium. The process also works in the degenerated double-Λ configuration 14. Here the mean relative phase $\varphi_0$ is constant, and can easily be controlled and put to zero 17. Such a setup might be relevant for any EIT application to modern nonlinear optics, where standard phase correlation techniques do not suffice. Besides high precision spectroscopy we expect promising applications in quantum information processing 15, quantum state engineering 12, or long distance quantum communication 20. As to the realization of a quantum repeater, quantum correlated photon pairs have already been generated 21 using the EIT-based technology of light pulse storage 22. Phase correlated excitation of optical materials with high nonlinearities and low loss might well become essential for the controlled generation of entangled states and quantum logic operations in future quantum computer design.

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