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Two-photon transitions induced by a stochastic microwave field

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Two-photon processes induced by a stochastic field, frequency-modulated by a Gaussian noise, are experimentally investigated in a $S=\frac{1}{2}$ spin system at microwave frequency. The shape and the width of the power spectra of the second-order two-photon–induced response of the spin system are measured for wide ranges of the rms amplitude and of the correlation time of the frequency fluctuations of the driving field.

Multiphoton transitions in atomic systems are affected to a large extent by the statistical properties of the exciting electromagnetic (em) field. In particular, two-photon (TP) resonant absorption and ionization processes are known to depend on the second-order correlation function of the field.[1-7] Among several kinds of models used for describing the stochastic field, the phase-diffusion field model has been given considerable attention. In this model one assumes that the field is amplitude stabilized, and that a random process $\mu(t)$ makes its instantaneous frequency $\omega_i(t)$ fluctuate in time about the mean value $\bar{\omega}$, $\omega_i(t)=\bar{\omega}+\mu(t)$. $\mu(t)$ is usually assumed to be an Ornstein-Uhlenbeck process,[8] namely, to have a Gaussian density function and an exponential autocorrelation function. Two opposite limits are often considered to point out the effects of the field correlations. In the former (fast-modulation or pure phase-diffusion limit) $\mu(t)$ is a white noise and the related phase $\phi(t)=\int_0^t\mu(\alpha)\,d\alpha$ is a Wiener-Lévy process;[8] in this limit the spectral profiles of the driving field, $S_1(\omega-\bar{\omega})$, and of the TP response of the system, $S_2(\omega-2\bar{\omega})$, are expected to be both Lorentzian but to have different widths, Δ_1 and Δ_2, respectively, with $\Delta_2=4\Delta_1$.[1,2,6,7] The opposite (slow-modulation) limit occurs for very long values of the correlation time of $\mu(t)$; it has been calculated that in this limit both $S_1(\omega-\bar{\omega})$ and $S_2(\omega-2\bar{\omega})$ are Gaussian but $S_2(\omega-2\bar{\omega})$ has twice the width of $S_1(\omega-\bar{\omega})$: $\Delta_2=2\Delta_1$.[6] These results were derived within the weak-field approximation, in which the Rabi frequency is assumed to be much less than the atomic resonance linewidth and TP saturation effects are disregarded.

The above theoretical predictions have been recently verified experimentally in both limit cases in a TP Doppler-free optical experiment by superimposing frequency and phase fluctuations onto the driving laser beam.[6]

In this Rapid Communication a two-level electron-spin system and a stochastic microwave field are used (for the first time, to our knowledge) to examine the effect of the statistical properties of the field on the TP processes in the weak-field limit. In particular, we consider here the case of an amplitude-stabilized microwave field, whose frequency is modulated by a Gaussian noise. We report experimental results on the shape and the width of the power spectra of the TP processes induced by this kind of field, as measured at various values of the root-mean-square (rms) amplitude Δ of the frequency fluctuation $\mu(t)$ and of its correlation time τ_c. A reason for interest in the results reported below is that they cover a wide range of the quantity $\Delta\tau_c$ from the fast-modulation limit ($\Delta\tau_c \ll 1$) to the slow one ($\Delta\tau_c \gg 1$). In this regard our experiments are complementary to the optical one.[6]

In the experiments described here an intense microwave field $H_\omega(t)=H_1\exp\{-i[\bar{\omega}t-\phi(t)]\}$, frequency modulated by a zero-mean Gaussian stationary process, $\mu(t)=d\phi(t)/dt$, drives a $S=\frac{1}{2}$ spin system, whose energy-level splitting $\hbar\omega_0$ is tuned by the static field H_0 to the TP resonance $\omega_0=2\bar{\omega}$. The effect of the frequency noise $\mu(t)$ on the TP transitions between the spin levels is investigated here by measuring the power spectra of the radiation that the spins undergoing TP transitions emit in a narrow spectral region centered at the second-harmonic (SH) frequency $2\bar{\omega}=\omega_0$.[9] As known, the emitted SH radiation reproduces the second-order TP-induced response of the spin system; this property was used in previous reports for detecting TP-saturation effects and TP coherent transients.[10,11] Using the SH generation effect to probe TP transitions is here convenient from an experimental point of view, as the frequency separation between the excitation field and the emitted radiation is large enough to allow reliable measurements of their spectral content.

The experimental setup for generating nonmonochromatic microwave radiation consists of a noise source and two microwave oscillators. The noise source is based on a 32-bit pseudorandom sequence generator, clocked at 16 MHz, with a 168-s repetition interval. After fast digital-to-analog conversion, the noise voltage $v(t)$, having originally a power spectrum of the kind $(\sin x/x)$ with a first null at 16 MHz, is filtered by three-pole low-pass filters with selectable nominal bandwidths of 20 kHz, 200 kHz or 2 MHz, well below the first spectral null. The residual ripple of the noise spectrum after the filter is less than 0.3 dB over the selected bandwidth. The noise amplitude can be adjusted from 1.0 mV to 5.0 V rms. For all the configurations (rms amplitude and bandwidth) used in the experiments, the density function $P(v)$ and the autocorrelation function $R_v(\tau)=\langle v(t+\tau)v(t)\rangle$ of $v(t)$ were preliminarily measured by proper analysis of 10 240 sampling points of $v(t)$, in order to check the Gaussian shape of $P(v)$ and to determine the values of the second moment $\langle v^2\rangle$ and of the correlation time τ_c.

In order to cover a wide range of $\Delta\tau_c$ we used two different low-power ($P \simeq 10$ mW) cw microwave oscillators with built-in frequency-modulation (FM) capability. The former is a cavity-stabilized klystron oscillator, whose FM circuit has a wide bandwidth ($\simeq 5$ MHz) and a low voltage-to-frequency conversion (VFC) ratio $K_{\rm VF} = (52 \pm 1)$ kHz/V; this oscillator was used when the 2-MHz bandwidth of the noise source was selected to reach very low values of $\Delta\tau_c$. The other microwave source is a solid-state sweeper whose FM circuit has a narrower bandwidth and a higher VFC ratio, $K_{\rm VF} = (4.55 \pm 0.05)$ MHz/V. Both microwave sources, when unmodulated, are highly stable, with a residual rms frequency fluctuation less than 1 kHz. The nonlinearity of their VFC was checked to be less than 2% in the used range of the modulating voltage.

In the experiments reported here the selected microwave source was tuned to $\bar{\omega} = 2\times 2.95$ GHz and its FM input was driven by the output signal $v(t)$ of the noise generator. The spectra of the obtained radiation were examined for various values of the quantity $\Delta\tau_c$, with Δ determined from $\langle v^2\rangle$, $\Delta = K_{\rm VF}(\langle v^2\rangle)^{1/2}$. In agreement with the theory of random FM of an oscillator,[12-14] the detected power spectra $S_1(\omega-\bar{\omega})$ were found to be well fitted by Lorentzian and Gaussian shapes down to -60 dB below the maximum for $\Delta\tau_c \lesssim 0.1$ and $\Delta\tau_c \gtrsim 10$, respectively. For intermediate values of $\Delta\tau_c$, $S_1(\omega-\bar{\omega})$ was found to have a mixed shape with Gaussian wings.

The intensity of the obtained radiation was raised to the required power level ($P \simeq 10$ W) by a traveling-wave-tube amplifier, working well below its saturation limit. In the experimental apparatus the spin sample is located in a low-Q bimodal cavity resonating both at $\omega_1 = 2\pi\times 2.95$ GHz (fundamental mode) and at $\omega_2 = 2\omega_1 = 2\pi\times 5.9$ GHz (detection mode), both having widths of 0.8 MHz. When the fundamental mode is excited by the high input power, the detection mode collects the SH radiation emitted by the spin system. The microwave signal picked out from the detection mode is sent to a microwave spectrum analyzer, tuned to $2\bar{\omega}$ and set to a frequency resolution of 1 kHz, where the spectra are visualized, digitized, and stored for further processing. Apart from the noise FM setup, the experimental apparatus is the same as reported in previous papers[10,11] to which we refer for a detailed description.

The results reported here were obtained in a standard spin system, a powder diphenylpicrylhydrazyl (DPPH) sample, whose electron-spin resonance properties are well known:[15] $S = \frac{1}{2}$, spin concentration $n \simeq 10^{21}$ cm^{-3}, longitudinal and transverse relaxation times $T_1 = T_2 = 7\times 10^{-8}$ s in our working conditions ($H_0 = 2.1$ kG, $T = 4.2$ K). At our maximum available power level, the TP-induced Rabi frequency in this system is of the order of 10^5 rad/s,[11] so that the weak-field conditions, $\chi T_2 \ll 1$ and $\chi^2 T_1 T_2 \ll 1$ are fulfilled in our experiments.

In this sample we measured the SH emission spectra at various values of the noise voltage rms amplitude and bandwidth. When $\langle v^2\rangle$ and τ_c were so regulated that $\Delta\tau_c \lesssim 0.1$ or $\Delta\tau_c \gtrsim 3.0$ the measured spectral profiles of the SH radiation were found to have the same shape as the spectrum of the driving field (Lorentzian or Gaussian, respectively). At intermediate values of $\Delta\tau_c$, where the power spectra of the input radiation were observed to have a mixed shape (Lorentzian in the center and Gaussian in the wings) the SH radiation was observed to have a similarly mixed shape but with a little more pronounced Gaussian part. Typical SH spectra obtained for representative values of $\Delta\tau_c$ are reported in Fig. 1. The observed behavior is in agreement with theoretical predictions.[1,2,6,7]

The widths (half-widths at half-maximum) of the SH output (Δ_2) and of the input radiation (Δ_1) spectra were measured and compared with each other at various values of the quantity $\Delta\tau_c$ that characterizes the input field statistics. The results are reported in Fig. 2, where the measured values of the ratio Δ_2/Δ_1 are plotted against $\Delta\tau_c$. As shown, the experimental Δ_2/Δ_1 varies from 4.2 ± 0.4 to 2.0 ± 0.2 when $\Delta\tau_c$ is varied from 0.036 to 4.4. Both limit values of Δ_2/Δ_1 are in agreement with theoretical values.[1,2,6,7]

In the measurements described above the investigated range of $\Delta\tau_c$ was restricted to $0.036 < \Delta\tau_c < 4.4$, less than the one actually available, in order to ensure that experimental spectra could not be affected by the residual FM of the microwave sources nor by the finite bandwidth of the cavity modes. By properly selecting the bandwidth of the noise source, we could scan this range of $\Delta\tau_c$ while keeping the width Δ_1 within the safety limits 3 kHz $< \Delta_1/2\pi <$ 100 kHz.

The experimental results reported above show the evolution of the TP-induced SH emission spectra from the Lorentzian to the Gaussian shape and the corresponding decrease of Δ_2/Δ_1 from 4 to 2 when $\Delta\tau_c$ is increased from the fast-modulation limit toward the slow one. As noted above, our results complement those obtained in the optical region,[6] which were restricted to the ranges $\Delta\tau_c \lesssim 0.2$ and $\Delta\tau_c \gtrsim 4$. It is worth remarking that the noise voltage used here for performing the random FM of the microwave source is a Gaussian process but not an Ornstein-Uhlenbeck process. In fact we measured nonexponential autocorrelation functions of $v(t)$. No attempt was made to modify the bandshape of $v(t)$ to come within the at noise model. The nonexponential shape of the experimental $R_v(\tau)$ does not invalidate the comparison with the theoretical limit values of Δ_2/Δ_1 and with the limit shapes of $S_2(\omega-2\bar{\omega})$. In fact, on the hand, the Lorentzian shape of the power spectrum of the second-order response of the system and the value 4 of the ratio Δ_2/Δ_1 (fast-modulation limit) are peculiar of a Wiener-Lévy process, to which any Gaussian frequency noise is expected to tend for τ_c tending to zero.[7] On the other hand, in the slow-modulation limit the input field spectrum reproduces the density function of $\mu(t)$ with complete decorrelation of its spectral components and the properties of the TP processes are expected to depend only on the Gaussian nature of $\mu(t)$. Obviously, the behavior in the intermediate range of $\Delta\tau_c$ depends on the particular form of $R_\mu(\tau)$.

Finally we wish to comment on two aspects of our experimental conditions. The former concerns the quantity $\Delta_1 T_2$. In the whole investigated range of $\Delta\tau_c$ the condition $\Delta_1 T_2 \ll 1$ was fulfilled, which implies that all the com-

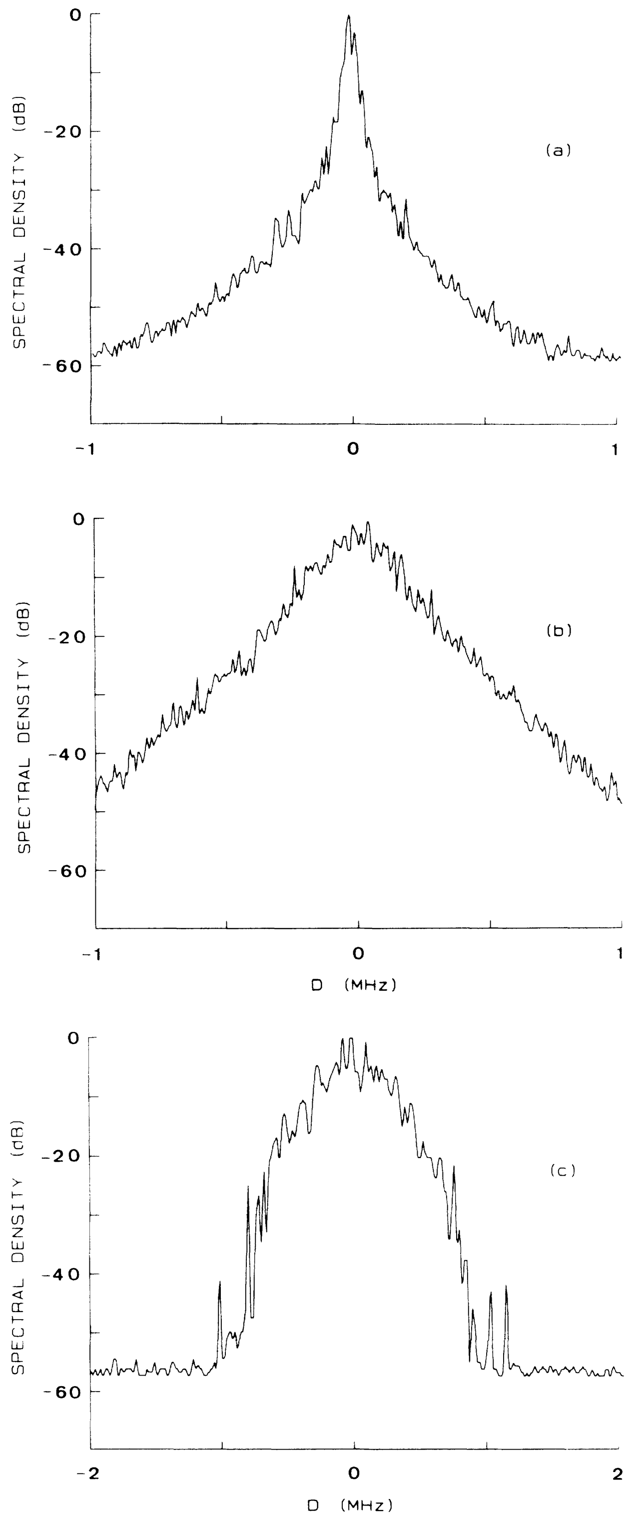

FIG. 1. Experimental power spectra of the second-harmonic radiation emitted by the sample at the TP resonance for three different values of the parameter $\Delta\tau_c$ of the driving field: (a) $\Delta\tau_c = 0.05$; (b) $\Delta\tau_c = 0.3$; (c) $\Delta\tau_c = 4.4$. As explained in the text, the spectra (a) and (c), corresponding to the fast- and to the slow-modulation limit, respectively, are well fitted by a Lorentzian (a) and by a Gaussian (c) shape.

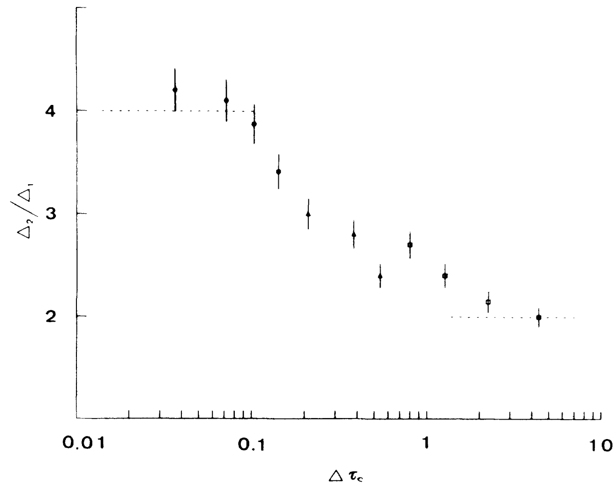

FIG. 2. Experimental values of the width ratio Δ_2/Δ_1 vs the parameter $\Delta\tau_c$ of the driving field. The range of $\Delta\tau_c$ was covered by varying the rms amplitude of the noise voltage for each available bandwidth of the noise source: 20 kHz (□), 200 kHz (Δ), 2 MHz (●). Except for the point at $\Delta\tau_c = 0.036$, which was taken at $\Delta_1 = 2\pi\times3.0$ kHz, all the other values of Δ_2/Δ_1 were measured with Δ_1 within the range: $2\pi\times8 \text{ kHz} < \Delta_1 < 2\pi\times100$ kHz. Dashed lines indicate the asymptotic theoretical values of Δ_2/Δ_1.

ponents of the input spectrum $S_1(\omega-\bar{\omega})$ are nearly equally at the TP resonance with the system. We recall that, according to Mollow's theory,[1] just in this limit the width of the spectrum $W_2(\omega_0-2\bar{\omega})$ of the TP-absorption rate is expected to be not affected by the statistical properties of the input field, being determined merely by the intrinsic width of the resonance line. However, the contrast with our experimental results is only superficial. In fact, whereas the spectrum $W_2(\omega_0-2\bar{\omega})$, as calculated by Mollow[1] and measured in the optical experiment,[6] represents the TP-absorption rate as a function of the resonance detuning $(\omega_0-2\bar{\omega})$, in our experiments the spectrum $S_2(\omega-2\bar{\omega})$ of the SH radiation is measured while keeping fixed the TP-resonance condition, $\omega_0=2\bar{\omega}$. In the latter case the width of the spin transition, even if larger than the bandwidth of the driving field, cannot mask the stochastic broadening of the system response. The other condition that characterizes our experiments is $\tau_c/T_2 \gtrsim 1$ and implies that in the observation time scale the system is in a quasiequilibrium state in which the transient regimes excited by the changes of the input field frequency are rapidly smeared out. So, the system can be considered as a zero-memory stochastic system and its stationary second-order response, for a given class of processes, is expected to depend only on the statistical properties of the driving field. Experimental work is in progress to explore the opposite condition $\tau_c \lesssim T_2$.

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