Experimental study of two-photon processes induced by a phase-diffusion field

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A phase-diffusion stochastic field with frequency fluctuations obeying Gaussian Markovian statistics is produced at microwave frequency and used to drive a two-level spin system. The system is tuned to the two-photon (TP) resonance and its TP-induced second-order response is measured as a function of the characteristic parameters of the frequency noise. It is found that, when the statistics of the driving field are varied from the fast- to the slow-modulation limit, the power spectrum of the nonlinear response of the system varies in a continuous way from a Lorentzian form toward a Gaussian one, and its relative width (normalized to the input spectral width) varies from 4.0 ± 0.4 to 2.0 ± 0.2. The experimental results are interpreted on the basis of a semiclassical calculation which takes into account the stochastic nature of the field. The effect of the filter action of the cavity modes is discussed. The relationship with previous experiments and theoretical calculations of TP absorption is also examined.

I. INTRODUCTION

The interaction of an atomic system with a strong radiation of finite bandwidth is affected to a large extent by the statistical properties of the radiation, especially when nonlinear processes are effective. It is now well known that a precise characterization of the statistics of the driving field is required for a complete account of the properties of such phenomena as multiphoton absorption and ionization, resonance fluorescence, four-wave mixing, coherent transients, and so on. Among them, the simultaneous absorption of two photons by a two-level (TL) system has been extensively considered in literature since early work by Mollow[1] and by Lambropoulos.[2] It was pointed out in those papers that the two-photon (TP) absorption processes in TL atomic systems driven by nonmonochromatic radiation depend on the second-order correlation function of the field rather than on its spectral content. That is, for instance, different exciting fields, having identical spectral profiles but originating from statistically nonequivalent fluctuations, yield quite different spectral shape and width of the atomic TP response.[3]

Several models have been proposed over the past 20 years to describe real sources and their output radiation. Among them, widespread attention has been given to the phase-diffusion field (PDF). In this model the amplitude of the field is time independent and its frequency noise is an Ornstein-Uhlenbeck (OU) process,[4,5] namely a stationary Gaussian Markovian process. The power spectrum of the PDF depends on the correlation time of its frequency, evolving in a continuous way from a Lorentzian profile to a Gaussian one, when the correlation time increases from zero to infinity.[6,7] Even if the name PDF is strictly appropriate only in the former limit (when the frequency is a white noise and the phase is a purely diffusive Wiener-Levi process), it is commonly used (and adopted herein) to indicate the more general case of finite correlation time as well. The generalized PDF model was originally devised to take into account the sub-Lorentzian fall down of the spectral wings in real sources. Since then it has been extensively used with considerable success to investigate nonlinear processes and coherent transients induced by nonmonochromatic radiation. In particular, as regards TP transitions in TL systems, it has been shown[8-11] that if the bandwidth of the driving field is much larger than the intrinsic width of the atomic transition, the TP absorption spectrum, considered as a function of the radiation detuning, exhibits a relative width (normalized to the bandwidth of the input field) that varies from 4.0 to 2.0 when the statistics of the field is varied from the fast- to the slow-modulation limit.

To test experimentally the above theories on the interactions between atoms and strong stochastic fields, efforts have been made to produce nonmonochromatic radiation with well-characterized and externally controlled statistical properties.[12-18] This may be achieved by deliberately superposing fluctuations onto an originally coherent field. However, little experimental work has been reported in which this kind of radiation is used for investigating the interaction of the stochastic radiation with a TL system. Bonch-Bruevich *et al.*[19] reported experiments of TP absorption induced by rf fields whose mutual correlation could be varied externally. Knight *et al.*[15] investigated the input-output cross correlation of a nuclear-magnetic-resonance system driven by a rf field whose phase is modulated by a binary telegraph noise. Recent results in the optical region have been reported by Elliott *et al.*, who used a well-characterized PDF for Doppler-free TP absorption[11] and double-optical-resonance[20] experiments. In other experiments[21] concerned with coherent transients at millimeter wavelengths, a different approach has been used in which the field is coherent and the atomic frequency is modulated by external noise.

In this paper the effect of the statistics of the driving field on the TP transitions of a TL system is experimentally investigated by measuring the nonlinear response of a dilute system of $S=\frac{1}{2}$ spins to a microwave radiation whose statistical properties are externally controlled. We report here the experiments carried out using a PDF at microwave frequency, obtained by imposing Gaussian fluctuations onto the field frequency; the case of non-Gaussian fluctuations is examined in the following paper.[22] In the experiments described here the TL system is driven by a TP-resonant high-intensity PDF, and its steady-state TP-induced second-order response is investigated by monitoring the radiation that the system emits in a spectral region centered at the second-harmonic (SH) frequency of the driving field.[23] In particular, we measure the spectral shape and width of the emitted SH radiation, and we study their dependence on the relevant parameters of the Gaussian noise modulating the field frequency; these parameters can be adjusted externally over so wide a range that we can examine the whole excursion from the fast-fluctuation limit to the slow one. Our experimental results show the gradual evolution of the SH spectral profile from a Lorentzian to a Gaussian shape and of its relative width from 4.0 to 2.0. A semiclassical calculation of the nonlinear response of the spin system to a PDF indicates that SH emission, likewise TP absorption, involves at least the second-order correlations of the input field. However, we point out that, at variance to TP absorption, SH emission spectra, taken at a fixed detuning of the exciting radiation, are influenced by the frequency fluctuations of the driving field even if its spectral width is much less than the atomic resonance width. A partial account of our results has been given in a preliminary report.[24]

Section II is devoted to the experimental results. After a brief summary of the main properties of the PDF, we describe our experimental method for obtaining a microwave radiation with well-controlled statistical properties, the experimental apparatus, and the results on the spectral properties of the SH radiation. In Sec. III we first derive the relationship between the statistical properties of the SH radiation and those of the driving field; second, we discuss our experimental results also in view of previous theoretical treatments and experiments.

II. EXPERIMENTS

Our experiments can be outlined as follows. A dilute system of N electronic spins $S=\frac{1}{2}$, in a static field $\mathbf{B}=B_0\hat{\mathbf{z}}$, is driven by a nonmonochromatic field $\mathbf{b}(t)$

$$\mathbf{b}(t)=\mathbf{b}_1\exp\{-i[\bar{\omega}t+\phi(t)]\}+\text{c.c.} \tag{1}$$

with constant amplitude and fixed direction, mean frequency $\bar{\omega}$, and stochastic phase $\phi(t)$. The statistical properties of $\phi(t)$ will be specified later on.

B_0 is adjusted to tune the spin frequency ω_0 to the TP resonance condition with the center frequency of the driving field $\omega_0=2\bar{\omega}$; $\mathbf{b}_1$ has both a longitudinal and a transverse component with respect to $\mathbf{B}$,

$$\mathbf{b}_1=b_1(\sin\alpha)\hat{\mathbf{x}}+b_1(\cos\alpha)\hat{\mathbf{z}}\ . \tag{2}$$

When both conditions are fulfilled, the two states of each spin center are coupled by TP transitions, which cause the build up of a resonant transverse second-order magnetization:[23]

$$m_x^{(2)}(t)=2\,\text{Re}[m_{x2}(t)\exp(-2i\bar{\omega}t)]\ , \tag{3}$$

oscillating at the mean frequency $2\bar{\omega}$. In the stationary state, in which we are interested here, the complex amplitude $m_{x2}(t)$ of $m_x^{(2)}(t)$ is expected to fluctuate in time as a consequence of the stochastic nature of $\mathbf{b}(t)$. As will be shown in Sec. III, Eq. (3) generalizes previous results on TP resonance in a $S=\frac{1}{2}$ spin system obtained for a monochromatic driving field.[23]

In our experiments the sample interacts with two electromagnetic (e.m.) modes at $\bar{\omega}$ and at $2\bar{\omega}$. The former (pump mode) creates the field $\mathbf{b}(t)$ over the sample; the latter (detection mode) collects the radiation emitted by the macroscopic dipole $m_x^{(2)}(t)$. So, a microwave signal can be detected that images the nonlinear response of the spin system. By this method we measure the profile and the width of the power spectrum $S_2(\omega-2\bar{\omega})$ of $m_x^{(2)}(t)$ and we investigate their dependence on the statistical properties of the driving field $\mathbf{b}(t)$.

A. The stochastic field

In the PDF model[6,7] the instantaneous frequency deviation $\mu(t)=d\phi/dt$ of the field $\mathbf{b}(t)$ of Eq. (1) is a zero-mean OU process,[4,5] characterized by a Gaussian probability density

$$P(\mu)=(2\pi)^{-1/2}\sigma^{-1}\exp(-\mu^2/2\sigma^2)$$

with a standard deviation σ and by an exponential autocorrelation function

$$R_\mu(\tau)\equiv\langle\mu(t+\tau)\mu(t)\rangle\ ,$$

$$R_\mu(\tau)=\sigma^2\exp(-|\tau|/\tau_c)\ ,$$

with a correlation time τ_c. Due to the Markovian nature of $\mu(t)$, the parameters σ and τ_c completely determine the statistics of $\mathbf{b}(t)$ and, in particular, its power spectrum $S_1(\omega-\bar{\omega})$. As this calculation has appeared in literature repeatedly,[6,7,10,25] we refrain from reproducing it here and we only give the final result:

$$S_1(\omega-\bar{\omega})=4b_1^2\tau_c e^x\text{Re}[x^{-z}\gamma(z,x)]\ , \tag{4}$$

where $z=x-iy$ with $x=(\sigma\tau_c)^2$, $y=(\omega-\bar{\omega})\tau_c$, and $\gamma(z,x)$ is the incomplete γ function.[26] The expression for $S_1(\omega-\bar{\omega})$ is a complicated function of τ_c, σ, and $\omega-\bar{\omega}$. However, as known, two limit situations can be distinguished depending on $\sigma\tau_c$. The former occurs for very fast fluctuations ($\tau_c\to 0$ with finite $\sigma^2\tau_c$); in this limit $S_1(\omega-\bar{\omega})$ narrows down to a Lorentzian:

$$S_1(\omega-\bar{\omega})\simeq 4b_1^2\frac{\sigma^2\tau_c}{(\omega-\bar{\omega})^2+\sigma^4\tau_c^2}\ , \tag{5}$$

with a half width at half maximum (HWHM) $\delta_1=\sigma^2\tau_c$. In the opposite limit of very slow fluctuations ($\tau_c\to\infty$) the field $\mathbf{b}(t)$ can be considered as the static superposition of uncorrelated fields and $S_1(\omega-\bar{\omega})$ duplicates the Gaussian shape of $P(\mu)$:

$$S_1(\omega-\bar{\omega})\simeq 2(2\pi)^{1/2}b_1^2\sigma^{-1}\exp[-(\omega-\bar{\omega})^2/2\sigma^2] \tag{6}$$

with a HWHM $\delta_1=(2\ln 2)^{1/2}\sigma$. Finally, in the intermediate case, the power spectrum $S_1(\omega-\bar{\omega})$ consists of a nearly Lorentzian center part [for $(\omega-\bar{\omega})\tau_c \ll 1$] and of Gaussian wings [for $(\omega-\bar{\omega})\tau_c \gg 1$]. As $\sigma\tau_c$ increases, the Gaussian portion of the spectrum increases until the entire spectrum becomes Gaussian.

Microwave PDF's can be obtained by modulating the frequency of a microwave oscillator by a Gaussian noise voltage.[13,14,17,21] Our noise source is based on a 32-bit Gaussian digital generator of pseudorandom sequences clocked at 16 MHz with a repetition interval of 168 sec. After digital-to-analog conversion, the noise voltage, having originally a power spectrum of the kind $(\sin\omega)/\omega$ with a first null at 16 MHz, is filtered by a low-pass filter with adjustable bandwidth BW: 20 kHz, 200 kHz, or 2 MHz. To characterize the resulting noise voltage $v(t)$ we measured its probability density $P(v)$ and its autocorrelation function $R_v(\tau)\equiv\langle v(t+\tau)v(t)\rangle$ by proper analysis of

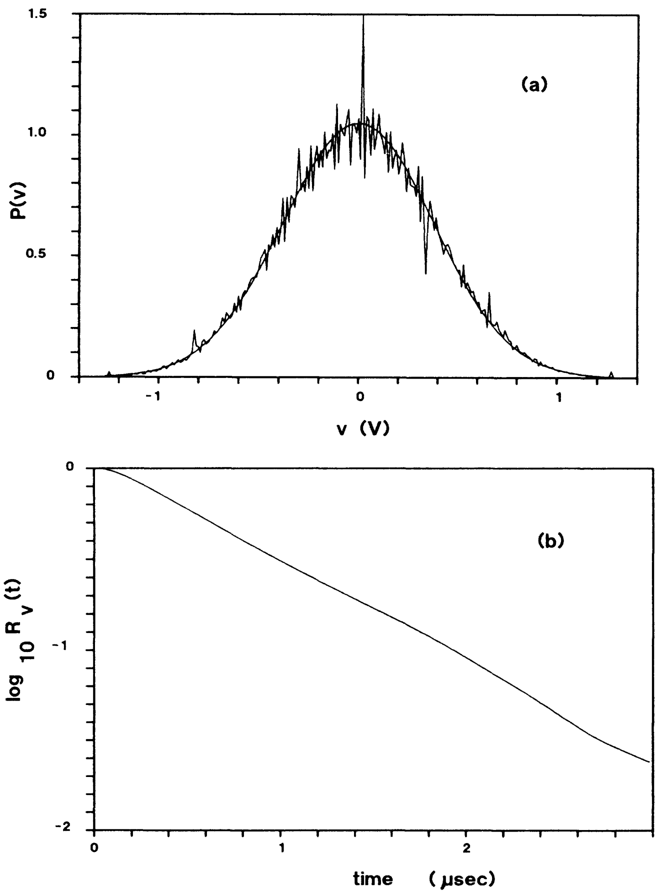

FIG. 1. Statistical analysis of the noise voltage $v(t)$ (a) Probability density $P(v)$; (b) autocorrelation function $R_v(\tau)$. Both curves refer to the voltage signal $v(t)$ obtained for a nominal BW of 200 kHz and a rms amplitude of 0.38 V. The smooth curve (a) is the Gaussian least-squares fit; its standard deviation is in perfect agreement with the nominal rms amplitude. The characteristic time τ_c of the exponential law (not shown) that best fits the experimental $R_v(\tau)$ is $\tau_c=0.87$ μsec.

nearly 10^6 sampling points of $v(t)$. Typical results, obtained for a noise voltage with nominal BW of 200 kHz and rms amplitude $\bar{v}$ of 0.38 V, are reported in Fig. 1. As shown, the experimental distribution $P(v)$ in Fig. 1(a) is well fitted by a Gaussian law up to the maximum value of v examined (nearly $4\bar{v}$). On the other hand, $R_v(\tau)$ in Fig. 1(b), apart from a short delay time ($\simeq 50$ nsec) is well described by a single exponential law, with maximum deviation of $\pm 3\%$ for $\tau \lesssim 2\tau_c$ and $\pm 10\%$ for $\tau \lesssim 4\tau_c$. Similar results were obtained when the BW of 20 kHz was selected, as well as for other rms amplitudes. When the 2-MHz BW was selected, small deviations both of $P(v)$ from the Gaussian distribution in the wings ($v \gtrsim 3\bar{v}$) and of $R_v(\tau)$ from the single-exponential form could be noticed. The extent to which these deviations influence the experimental results reported below will be discussed later on. For all the configurations used in the experiments described here, $P(v)$ and $R_v(\tau)$ were preliminarily measured and analyzed to get the values of $\bar{v}$ and of $\tau_c=\bar{v}^{-2}\int_0^\infty R_v(\tau)d\tau$. The rms amplitude $\bar{v}$ of $v(t)$ could be varied from 1.0 mV to 5.0 V rms. τ_c was found to be $\tau_c=7.6\pm0.4$ μsec, $\tau_c=0.87\pm0.05$ μsec, and $\tau_c=0.11\pm0.01$ μsec, for the three nominal values (20 kHz, 200 kHz, and 2 MHz, respectively) of the noise BW.

The noise voltage $v(t)$ is used to drive the frequency-modulation (FM) input of a microwave oscillator, preliminarily tuned to $\bar{\omega}$. The resulting frequency fluctuation $\mu(t)$ is expected to have the same statistics as $v(t)$, provided that the fm network is linear and its frequency response is flat over the whole noise BW. In particular, the standard deviation σ of $\mu(t)$ is $\sigma=K_{\mathrm{VFC}}\bar{v}$, where K_{VFC} is the voltage-to-frequency conversion (VFC) ratio of the fm circuit. In order to cover a wide range of the quantity $\sigma\tau_c$, we used two different low-power cw microwave oscillators with built-in fm capabilities. The former is a cavity-stabilized klystron oscillator whose fm circuit has a wide BW ($\simeq$5MHz) and a relatively low VFC ratio, $K_{\mathrm{VFC}}=50.5\pm0.5$ kHz/V; this was used when the noise BW of 200 kHz and 2 MHz were selected to get low values of $\sigma\tau_c$ ($\lesssim 0.5$). The latter oscillator is a solid-state sweeper, whose fm circuit has a narrower BW ($\simeq$50 kHz) and a higher VFC ratio, $K_{\mathrm{VFC}}=4.8\pm0.1$ MHz/V. Both oscillators, when not modulated, are highly stable with a residual BW of their output signal less than 1 kHz; the nonlinearity of their fm circuit is less than 2%.

The noise-modulated microwave radiation was examined by means of a microwave spectrum analyzer, tuned at $\bar{\omega}$ and set to its maximum frequency resolution (1 kHz). Representative power spectra are reported in Fig. 2 for three different values of $\sigma\tau_c$: 2(a) $\sigma\tau_c=0.058\pm0.005$; 2(b) $\sigma\tau_c=0.53\pm0.05$; (2c) $\sigma\tau_c=4.5\pm0.4$. The smooth curves in Fig. 2 are the theoretical spectra calculated from Eq. (4) by numerical evaluation of the incomplete γ function with the experimental values of σ and τ_c. The agreement is remarkable for the spectrum in Fig. 2(a), down to -55 dB below the maximum, namely up to nearly 400 widths ($\delta_1/2\pi=5.2$ kHz); note that, for this value of $\sigma\tau_c$, the spectrum $S_1(\omega-\bar{\omega})$ is well described by a Lorentzian form down to

-45 dB. For $\sigma\tau_c=0.53$, the experimental spectrum in Fig. 2(b) (with $\delta_1/2\pi=49$ kHz) is well fitted by the theoretical one down to nearly -45 dB, its very far wings tending to fall down faster than expected. Finally, for the largest value of $\sigma\tau_c$ examined [Fig. 2(c)] the agreement is limited to the range 0 to -20 dB. In fact, for $\omega-\overline{\omega}>5\sigma$, the wings of the experimental spectrum fall down faster than expected. This discrepancy in the wings is perhaps caused by imperfections of the distribution $P(v)$ at high values of v. We recall that the Gaussian nature of $P(v)$ could be tested only for $v<4\overline{v}$; it is possible that, due to limitations of the digital noise generator, higher values of v occur less frequently than for a true Gaussian variable. This manifests mainly in the wings of the spectra taken for high values of $\sigma\tau_c$, as the narrowing action of the fluctuations is only of minor importance and the spectrum tends to reproduce $P(v)$.

We measured the HWHM δ_1 of $S_1(\omega-\overline{\omega})$ as a function of $\sigma\tau_c$. The experimental data are reported in Fig. 3, grouped in three branches according to the value of τ_c. Solid lines plot the theoretical δ_1 versus $\sigma\tau_c$ obtained from Eq. (4). A fair agreement is found in the whole investigated range, except for $\sigma\tau_c\lesssim 0.03$. However we note that, for these points, the measured values of δ_1 ($\delta_1/2\pi\lesssim 3$ kHz) are probably altered by the frequency resolution (1 kHz) of the spectrum analyzer.

By the way, we use data in Fig. 3 to make clear a point of our experimental procedure. In our experiments the stochastic radiation is used to feed a resonant cavity whose mode has a half width of nearly 400 kHz. To avoid severe distortions of the field statistics we are confined to use driving fields with $\delta_1/2\pi\lesssim 150$ kHz; on the opposite side, a minimum value od δ_1 ($\delta_1/2\pi\gtrsim 3$

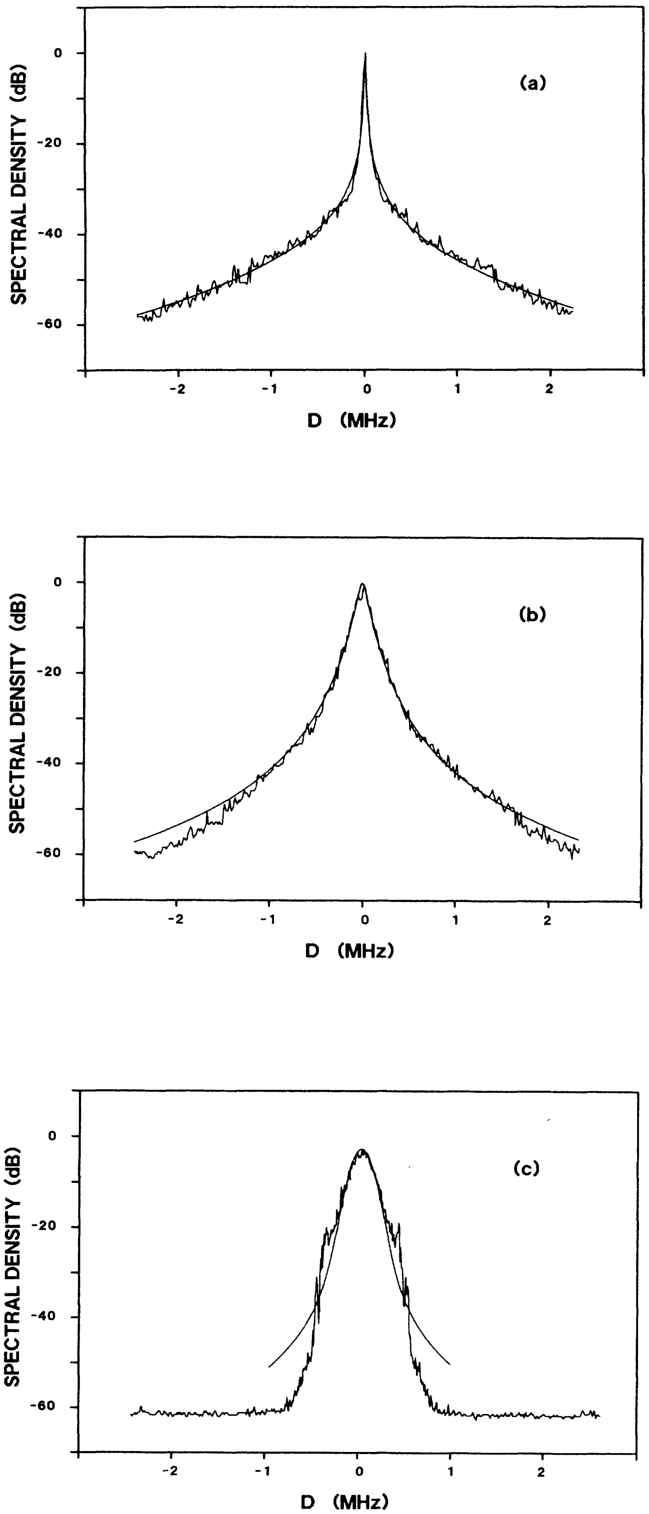

FIG. 2. Representative power spectra of the microwave radiation obtained for $\sigma=5.42\times 10^5$ rad/sec, BW of 2 MHz, $\sigma\tau_c=0.058\pm 0.005$ (a); $\sigma=6.03\times 10^5$ rad/sec, BW of 200 kHz, $\sigma\tau_c=0.53\pm 0.05$ (b); $\sigma=5.91\times 10^5$ rad/sec, BW of 20 kHz, $\sigma\tau_c=4.5\pm 0.4$ (c) The HWHM δ_1 are $\delta_1/2\pi=5.2\pm 0.1$ kHz (a); $\delta_1/2\pi=49\pm 2$ kHz (b); $\delta_1/2\pi=101\pm 5$ kHz (c). Smooth curves are theoretical spectra.

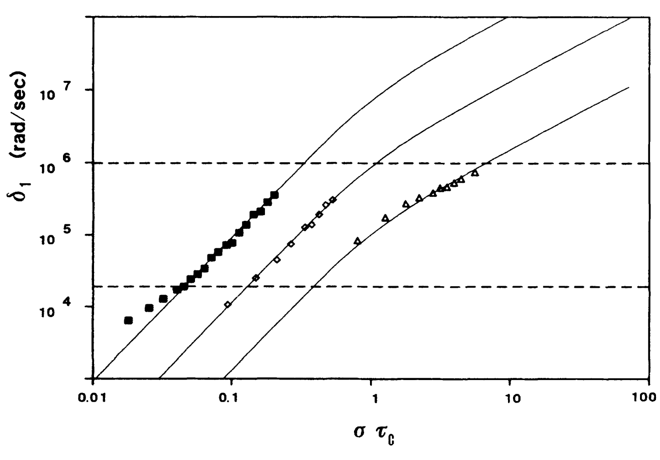

FIG. 3. Experimental values of the HWHM δ_1 of the spectra $S_1(\omega-\overline{\omega})$ vs $\sigma\tau_c$ obtained for three different values of the nominal BW of the noise signal: BW=2 MHz (■); BW=200 kHz (◇); BW=20 kHz (△). Solid lines are the theoretical curves calculated with the appropriate value of τ_c: $\tau_c=0.11$ μsec, $\tau_c=0.87$ μsec, and $\tau_c=7.6$ μsec, respectively. Dashed lines ($\delta_{1\min}=2\pi\times 3$ kHz and $\delta_{1\max}=2\pi\times 150$ kHz) enclose the range of δ_1 usable for the experiments, as explained in the text.

kHz) is fixed by the frequency resolution of the spectrum analyzer. Data in Fig. 3 show how, by varying both the noise BW and its rms amplitude, a wide range of $\sigma\tau_c$ can be covered without violating the above requirements.

B. The SH response

For the measurements reported here, we have chosen a standard spin system, a sample of powdered 1, 1-diphenyl-2-picrylhydrazyl (DPPH), whose ESR properties are well known in literature.[27] The spin centers are free radicals ($S=\frac{1}{2}$) with a concentration $n=10^{21}$ cm^{-3}; their ESR line, homogeneously narrowed by exchange interactions, has a peak-to-peak width of $\simeq 0.25$ mT. In our experimental conditions ($B=0.21$ T, $T=4.2$ K) the longitudinal (T_1) and the transverse (T_2) relaxation times are $T_1=T_2\simeq 0.07$ μsec.

In our setup the sample is located in a bimodal cavity resonating both at $\omega_{c1}=2\pi\times 2.95$ GHz (pump mode) in a partially coaxial mode and at $\omega_{c2}=2\pi\times 5.9$ GHz (detection mode) in its rectangular TE_{102} mode. Fine tuning of the harmonic mode to $\omega_{c2}=2\omega_{c1}$ is achieved by inserting a quartz rod. Both modes have a relatively low quality factor Q, with a half width of nearly 400 kHz. The radiation obtained as described above, with its mean frequency tuned to the pump mode ($\bar{\omega}=\omega_{c1}$), is first raised to the required power level ($\simeq 0.5$ W) by means of a travelling-wave tube amplifier (TWTA), working well below its saturation level ($P_{sat}\simeq 50$ W). The TWTA output, filtered to eliminate residual harmonics, is used to feed the pump mode. When the TP resonance condition ($\omega_0=2\bar{\omega}$) is established, the detection mode of the cavity collects the SH radiation emitted by the spin sample. A microwave signal, centered at ω_{c2}, is picked out from the detection mode filtered by a high-pass filter and amplified by a low-noise linear amplifier (the gain is ~ 50 dB, and the band is 5.7–6.2 GHz). Finally, the signal is detected by a microwave spectrum analyzer, tuned to ω_{c2} and set to a frequency resolution of 1 kHz, where the spectra can be visualized, digitized, and stored for further off-line processing. All the measurements described here were taken at the exact TP resonance and with $\alpha=54.6°$ to maximize SH emission.[23]

By this method we measured the power spectra of the SH response of the spin system at various values of the rms amplitude $\bar{v}$ and of the correlation time τ_c of the noise $v(t)$ modulating the driving field frequency. Representative spectra $S_2(\omega-2\bar{\omega})$ of the SH radiation, for three different values of $\sigma\tau_c$, are shown in Fig. 4. These spectra were taken at the same values of $\sigma\tau_c$ as those in Fig. 2. A detailed discussion of the properties of these spectra is postponed to the next section, where they will be compared to theoretical profiles. Here we only point out a qualitative feature, that is, their gradual evolution from a nearly Lorentzian form toward a nearly Gaussian one, on increasing $\sigma\tau_c$. In this respect the power spectra $S_2(\omega-2\bar{\omega})$ seem to exhibit a similar behavior as those of the driving field. However, we anticipate that a quantitative account of the experimental spectra will be possible only for their center parts; in fact the

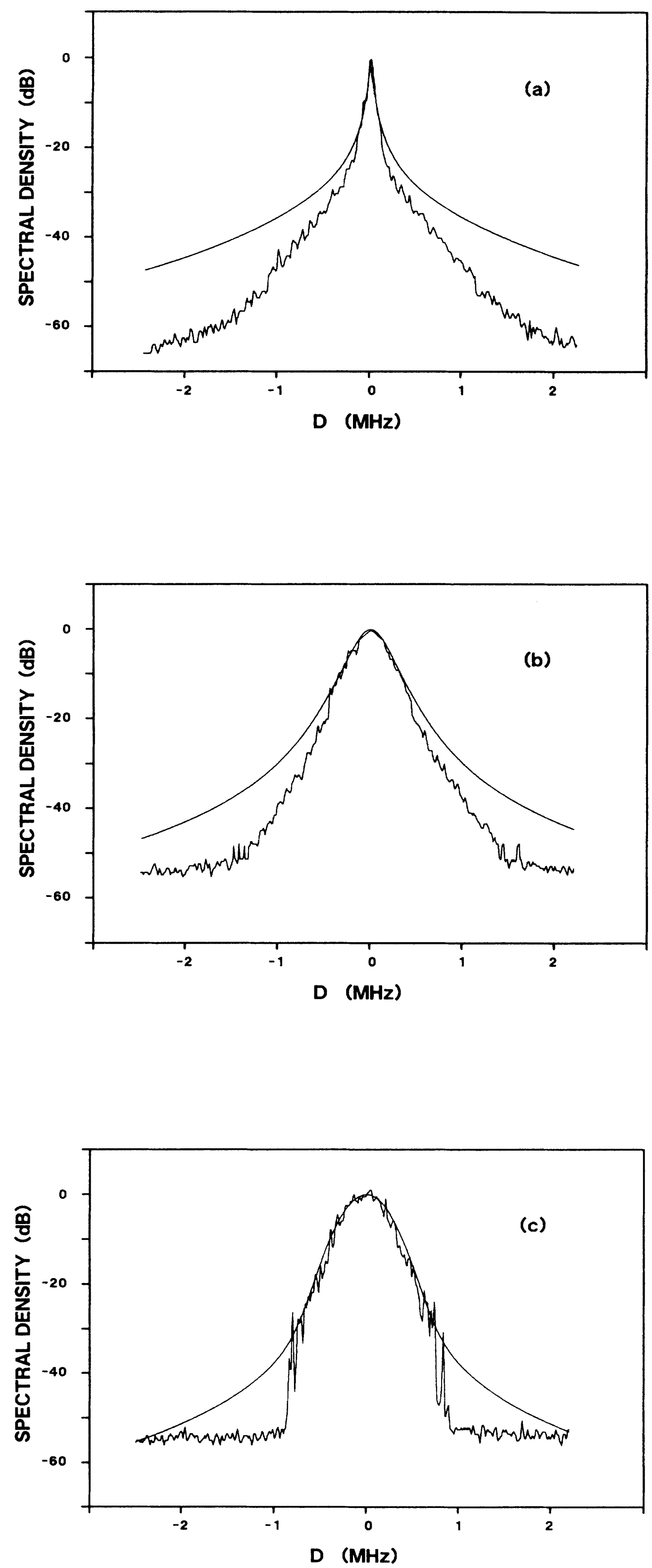

FIG. 4. Experimental power spectra of the SH radiation emitted by the sample at the TP resonance, corresponding parts [(a)–(c)] to the driving fields as described in Fig. 2. Solid lines are theoretical spectra.

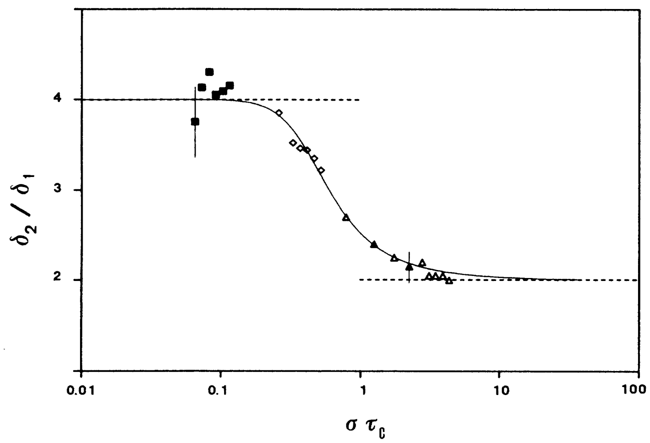

FIG. 5. Experimental values of δ_2/δ_1 vs $\sigma\tau_c$. The range of $\sigma\tau_c$ was covered by varying the rms amplitude of the noise voltage for each available BW of the noise source: 20 kHz (△), 200 kHz (◇), and 2 MHz (■). The solid line plots the theoretical dependence.

far wings of $S_2(\omega-2\bar{\omega})$ will be found to fall down faster than expected. The reason is that the emission of radiation far from the center is limited by the frequency response of the detection mode, whose finite width cuts down the wings of $S_2(\omega-2\bar{\omega})$. We verified that if the detection mode is slightly mistuned from the harmonic condition ($\omega_{c2}\neq 2\omega_{c1}$), one of the wings is greatly enhanced even if at the expense of the opposite one and of the center part.

The comparison between input radiation and second-order response of the system can be carried out in a more reliable way in terms of their spectral widths. Actually, the effect of varying the statistical properties of the driving field on the SH response of the system becomes evident if the HWHM δ_2 of the spectra $S_2(\omega-2\bar{\omega})$ are considered. Experimental results are reported in Fig. 5, where we plot the values of the ratio δ_2/δ_1 measured at various values of $\sigma\tau_c$. As shown, on increasing $\sigma\tau_c$, from the Lorentzian to the Gaussian limit, the output-to-input width ratio δ_2/δ_1 gradually decreases from 4.0 ± 0.4 to 2.0 ± 0.2.

To conclude this experimental section we note that in our experimental conditions the TP Rabi frequency[28] is of the order of 10^4 rad/s and the TP saturation factor less than 10^{-6}, as estimated on the basis of the values of driving field amplitude and of the relaxation times, thus fulfilling the weak-field condition.

III. THEORY AND DISCUSSION

Consider a TL system with a transition frequency ω_0 tuned at TP resonance with the field $\mathbf{b}(t)$ of Eq. (1). The strength of the TP coupling between the ground and the excited state of each spin center, in the absence of other near-resonant intermediate levels, is measured by the (generalized) TP Rabi frequency ω_R,[28] which, for the field $\mathbf{b}(t)$ of Eq. (1), can be written as

$$\omega_R(t)=\sin(2\alpha)\omega_1^2(t)/\omega_0 \;, \tag{7}$$

where $\omega_1(t)=\gamma b_1\exp[i\phi(t)]$ and γ is the gyromagnetic ratio. $\omega_R(t)$ is a stochastic process, whose properties depend on the particular process $\phi(t)$.

We describe the time evolution of the spin system by its density matrix $\rho_{ij}(t)$, where i,j = 1,2. As known,[29] near the TP resonance and making the slowly varying amplitude and the rotating-wave approximations, the equations of motion of the density matrix elements are conveniently written in terms of the population difference $n(t)=\rho_{11}(t)-\rho_{22}(t)$ and the TP coherence $\sigma_{12}(t)=\rho_{12}(t)\exp(-2i\bar{\omega}t)$:

$$\left[\frac{d}{dt}+i\Delta+\Gamma_2\right]\sigma_{12}(t)=-\tfrac{1}{2}i\omega_R(t)n(t)\;, \tag{8a}$$

$$\left[\frac{d}{dt}+\Gamma_1\right]n(t)=\Gamma_1 n_0-i[\omega_R(t)]^*\sigma_{12}(t)+i\omega_R(t)[\sigma_{12}(t)]^*\;. \tag{8b}$$

In these equations Δ is the TP resonance detuning $\Delta=2\bar{\omega}-\omega_0-\Delta_s$ including a small resonance shift $\Delta_s=(\sin^2\alpha)\gamma^2 b_1^2/\omega_0$;[28] $\Gamma_1=T_1^{-1}$, $\Gamma_2=T_2^{-1}$; n_0 is the thermal-equilibrium population difference.

Integrating formally Eq. (8a) we obtain:

$$\sigma_{12}(t)=\tfrac{1}{2}i\int_0^t\exp[(i\Delta+\Gamma_2)(t'-t)]\times\omega_R(t')n(t')dt'\;, \tag{9a}$$

which can be used to derive the formal solution of Eq. (8b),

$$n(t)=n_0-\mathrm{Re}\int_0^t\exp[\Gamma_1(t'-t)]dt'\int_0^{t'}\exp[(i\Delta+\Gamma_2)(t''-t')][\omega_R(t')]^*\omega_R(t'')n(t'')dt''\;. \tag{9b}$$

Now we take advantage of our experimental condition that the time scale of the fluctuations of the driving field and, hence, the observational time scale are much longer than the relaxation times of the system. This allows to approximate the exponential terms in the above integrals by their limits for $\Gamma_1,\Gamma_2\rightarrow\infty$, namely by δ functions. We get the following solutions:

$$\sigma_{12}(t)=-\tfrac{1}{2}(\Delta-i\Gamma_2)^{-1}\omega_R(t)n(t)\;, \tag{10a}$$

$$n(t)/n_0=(\Delta^2+\Gamma_2^2)/\{\Delta^2+\Gamma_2^2+\omega_R(t)[\omega_R(t)]^*\Gamma_2/\Gamma_1\}\;. \tag{10b}$$

As the field amplitude is fixed and only its phase $\phi(t)$ is stochastic, $\omega_R(t)[\omega_R(t)]^*$ and, hence, $n(t)$ are time independent. Within this approximation the solution for $n(t)$ is the same as for a monochromatic field with frequency $\bar{\omega}$ and amplitude b_1. The situation is different as regards the response of the system, $m_{x2}(t) = -\frac{1}{2}N\gamma\hbar\sigma_{12}(t)$. In fact, even in this limit, the TP coherence $\sigma_{12}(t)$ in Eq. (10a) is a stochastic process. In particular, according to Eq. (10a), it has the same statistical properties as the TP Rabi frequency $\omega_R(t)$. We get

$$m_{x2}(t) = -\tfrac{1}{4}(\Delta - i\Gamma_2)^{-1}\hbar\gamma N(n/n_0)\omega_R(t)\ . \tag{11}$$

According to Eq. (11) the power spectrum $S_2(\omega-2\bar{\omega})$ of $m_{x2}(t)$ is proportional to the power spectrum $S_R(\omega)$ of the TP Rabi frequency $\omega_R(t)$:

$$S_2(\omega-2\bar{\omega}) = \tfrac{1}{16}\hbar^2\gamma^2N^2\frac{(\Delta^2+\Gamma_2^2)}{(\Delta^2+\Gamma_2^2+\omega_R\omega_R^*\Gamma_2/\Gamma_1)^2}S_R(\omega)\ . \tag{12}$$

For our experiments, carried out at the TP resonance $\Delta - 0$ and in the weak-field limit, Eq. (12) simplifies to

$$S_2(\omega-2\bar{\omega}) = \tfrac{1}{16}\hbar^2\gamma^2N^2T_2^2S_R(\omega)\ . \tag{13}$$

The power spectrum $S_R(\omega)$ can be calculated in a straightforward way. In fact, from Eq. (7), $\omega_R(t) = \omega_{R0}\exp[-i\phi_R(t)]$, with $\omega_{R0} = (\gamma^2b_1^2/\omega_0)\sin 2\alpha$ and with $\phi_R(t) = 2\phi(t)$, is a stochastic process, whose instantaneous frequency is an OU process with correlation time τ_c and variance $\sigma_R = 2\sigma$. By the same procedure followed for calculating $S_1(\omega-\bar{\omega})$, $S_R(\omega)$ can be expressed in terms of the incomplete γ function, and we obtain for $S_2(\omega-2\bar{\omega})$

$$S_2(\omega-2\bar{\omega}) = S_0e^{x'}\mathrm{Re}[(x')^{-z'}\gamma(z',x')]\ , \tag{14}$$

where $x' = (2\sigma\tau_c)^2$, $y' = (\omega-2\bar{\omega})\tau_c$, $z' = x' - iy'$, and $S_0 = \frac{1}{4}(\hbar\gamma N\omega_{R0}T_2)^2\tau_c$.

So, the profile of $S_2(\omega-2\bar{\omega})$ is expected to have the same shape as $S_1(\omega-\bar{\omega})$ (Lorentzian and Gaussian) in the two limits of very fast and very slow fluctuations, and a mixed shape (a nearly Lorentzian peak and Gaussian wings) in the intermediate range of $\sigma\tau_c$. The smooth curves reported in Figs. 4(a)–4(c) plot the theoretical spectra $S_2(\omega-2\bar{\omega})$ as calculated from Eq. (14) for the experimental values of σ and τ_c. The agreement with the experimental spectra is good near the center, say, down to -20 dB below the maxima, whereas the wings of the experimental spectra fall down faster than expected. The finite width of the cavity mode is responsible for this disagreement in the wings and compels us to limit our comparison between theory and experiment to the center part of the spectra.

The agreement of Eq. (12) with the experimental results is quantitative as regards the width ratio δ_2/δ_1. The solid curve of Fig. 5 plots the theoretical ratio δ_2/δ_1 versus $\sigma\tau_c$, as obtained numerically from Eqs. (4) and (14). It fits quite well the experimental points in the whole investigated range of $\sigma\tau_c$. The dependence of the relative width δ_2/δ_1 on $\sigma\tau_c$ has an intuitive explanation. In fact the second-order response of the spin system [Eq. (11)] is a stochastic process whose frequency fluctuation is an OU variable having the same correlation time but a standard deviation twice larger than the input field. According to Eqs. (7) and (14), $\delta_2\tau_c$ is related to $2\sigma\tau_c$ by the same functional dependence as $\delta_1\tau_c$ to $\sigma\tau_c$. This dependence is not simply linear and yields a dependence of δ_2/δ_1 on $\sigma\tau_c$. In particular, in the fast-modulation limit, as $\delta_1\tau_c$ tends to $\sigma^2\tau_c^2$ and $\delta_2\tau_c$ tends to $4\sigma^2\tau_c^2$, δ_2/δ_1 tends to 4; on the other hand, in the slow limit $\delta_1\tau_c$ tends to $\sigma\tau_c$ and $\delta_2\tau_c$ tends to $2\sigma\tau_c$, so that δ_2/δ_1 tends to 2.

Two comments on our experiments are in order. The first one concerns the assumption made above that $\mu(t)$ is an OU (Markovian and Gaussian) process. As pointed out in Sec. II, our noise signal can be considered an OU process to a good approximation only when either the 200 kHz or the 20 kHz noise BW is selected, which, according to Fig. 2, means for the measurements taken at $\sigma\tau_c \gtrsim 0.1$. In spite of this, the agreement found between experimental and theoretical results in Fig. 5 seems to regard the whole investigated range of $\sigma\tau_c$. The reason is that for some of the theoretical results mentioned above, the Markovian hypothesis for $\mu(t)$ [namely the assumption of a single exponential $R_\mu(\tau)$] is not necessary. In fact, if only the Gaussian hypothesis is retained for $\mu(t)$, $b(t)$ is still a Kubo oscillator,[30] as it obeys the stochastic multiplicative equation $\dot{b} = ib[\bar{\omega}+\mu(t)]$ with a normal, steady-state, and zero-mean $\mu(t)$. According to Kubo's statistical theory, the Lorentzian and Gaussian limits of $S_1(\omega-\bar{\omega})$, for very fast and very slow fluctuations, are independent of the particular form of $R_\mu(t)$. The same is true for $S_2(\omega-2\bar{\omega})$, since, according to Eqs. (7) and (11), $\omega_R(t)$ and $m_{x2}(t)$ are Kubo oscillators as well, and no hypothesis on the statistics of $\phi(t)$ is involved in our procedure for obtaining Eqs. (10). So, all the properties of the second-order response of the system, in both limits of fast and slow fluctuations, including the asymptotic behavior of the width ratio, are independent of the Markovian hypothesis, whose relevance regards only the intermediate range of $\sigma\tau_c$.

The latter comment is on the relationship between SH emission and TP absorption. Our experimental results show that the second-order response of a TP-resonant two-level system to a PDF is a phase-diffusion process as the driving field, with an output-to-input width ratio varying from 4.0 to 2.0, as the quantity $\sigma\tau_c$ is varied. A semiclassical calculation of the output spectra supports these results. This behavior is reminiscent of experimental[11] and theoretical[1,8–10] results on TP absorption. However, it should be emphasized that our results refer to the case in which the bandwidth δ_1 of the driving field is much less than the intrinsic width of the resonance line $\delta_1T_2 \ll 1$, or that which is the same, the coherence time of the driving field is much longer than the relaxation time; this condition is well satisfied in the experiments reported here and it rests on the basis of the approximations used to derive Eqs. (10). We recall that, in this limit, when the TL system behaves like a zero-memory system, the TP absorption spectral shape is expected to be unaffected by the statistical properties of the field and to reproduce the broad atomic line shape. This is consistent with our Eq. (10b), where $n(t)$ is calculated to be the same as for a monochromatic excitation with a mean TP Rabi frequency ω_{R0}. The different effect of the stochastic driving field on TP absorption and on SH emission can be explained as follows. The TP absorption spectra (either measured or calculated) represent the TP transition rate as a function of the TP detuning Δ. In this kind of spectral analysis of the TP processes, the effect of the field statistics can be observed only when the spectral width of the driving field overcomes the atomic one; in particular, it is completely masked by the atomic line shape in a zero-memory system. On the other hand, the SH emission spectra considered here represent the spectral content of the radiation emitted at a fixed value of the TP detuning, $\Delta=0$ in particular. In this case, as the SH emission process depends on the second-order coherence $\sigma_{12}(t)$ rather than on the population difference, the effect of the input-field statistics manifests itself even if the driving field spectrum is narrower than the resonance line. In this respect, SH emission seems to be more advantageous than TP absorption for evidencing the effects of the statistical properties of the driving field on the TP response.

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