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Degenerate four-wave-mixing intensity noise fluctuations as a spectroscopic tool

H. Failache, M. Oriá,* and A. Lezama

Instituto de Física, Facultad de Ingeniería, Casilla de Correo 30, 11300 Montevideo, Uruguay

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We explore the use of degenerate four-wave-mixing (DFWM) intensity noise fluctuations as a spectroscopic tool for the study of atomic dynamics. Experimental results obtained from the spectral analysis of the intensity noise of the DFWM signal produced by a free running diode laser in a rubidium vapor cell are presented. For low exciting field intensities, the spectra are single peaked with a linewidth comparable to the atomic natural linewidth while at higher intensities a structure devleops revealing the existence of a dynamic Stark effect. The results are compared with numerical simulations of the power spectrum of the DFWM signal produced by a phase-diffusing field incident on two-level atoms.

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I. INTRODUCTION

The observation of the temporal behavior of the response of atomic samples to time varying light-field excitations can provide useful information on the atomic dynamics. Different techniques relying on the exposure of atoms to time-dependent radiation allow the determination of several atomic parameters, such as energy-level structures, relaxation times, collisional rates, etc. A particular class of time-dependent atom-light interaction phenomena are those produced by the illumination of atoms with stationary randomly fluctuating electromagnetic waves [1–7]. The main motivation for the study of these phenomena arises from the fact that light fluctuations are unavoidable in actual experiments, and consequently, the study of the effects of light fluctuations on the observed signals is essential to their accurate interpretation. A second motivation comes from the fact that the study of the atomic response to randomly fluctuating fields can be used for spectroscopic purposes. An example of a physical observable that can be monitored for the study of the atomic response to stochastic fields is the emitted fluorescence. Most of the research in this field is indeed concerned with this phenomenon [1,2,5,8–14].

The interest in the study of fluctuating light fields interacting with atoms has recently been reinforced due to the generalized use of single-mode diode lasers in atomic physics experiments. These devices are well known for allowing the possibility of generating light fields characterized by very small amplitude fluctuations and large phase fluctuations. Recently, Yabuzaki, Mitsui, and Tanaka [15] have shown in a very simple experiment that useful spectroscopic information can be extracted from the spectral analysis of the fluctuation (noise) in the intensity of a diode laser beam after traversing a resonant atomic gas cell. The observed signal, also studied in further detail by McIntyre *et al.* [16], can be interpreted as being due to the heterodyning of the incident laser field with the coherent radiation emitted by the laser-excited atomic polarization. Due to its phase fluctuations, the laser light randomly explores a wide frequency range within the inhomogeneously (Doppler) broadened atomic absorption line. Consequently, the observed noise-power spectrum extends over a frequency range comparable to the Doppler width. Within this range, small narrow features can be observed at frequencies that correspond to hyperfine-level splittings of the excited state. In a recent paper, Walser and Zoller [17] have theoretically studied the use of laser-noise-induced polarization signals as spectroscopic tools. They have analyzed the fluctuations in the transmitted intensity of different kinds of stochastic fields after their passage through an atomic medium. Their results show that quantities such as the mean transmitted intensity, the intensity variance, and its correlation spectrum reveal interesting information on the stochastic nature of the exciting field and on the atomic dynamics.

The purpose of this paper is to demonstrate that degenerate four-wave mixing (DFWM) [18] can also be used as a spectroscopic tool, in addition to fluorescence and transmission, for the study of the response of atomic samples to randomly fluctuating fields [6,7,19–27]. The choice of four-wave mixing is based on the following specific properties: (a) Unlike fluorescence, four-wave mixing is a coherent process allowing the generation of a well-defined beam in the phase-matching direction. Also, due to the triple resonance nature of DFWM, the produced signal can be quite intense, even with low-power lasers. (b) While single-wave transmitted light has to be considered as the result of the joint contributions of the incident laser field and the coherent field produced by the atomic polarization, DFWM can be considered as a pure atomic signal in the sense that it is fully generated by the (third-order, nonlinear) atomic polarization and no light is present at the detector in the absence of the atomic medium. (c) DFWM is a better velocity-selective mechanism than single-wave transmission [28].

The above-mentioned features of DFWM suggest the following physical picture: The random-phase fluctua-

*Permanent address: Departamento de Física,Universidade Federal da Paraíba, Joao Pessoa, Paraíba, Brazil.

tions of the laser light correspond to instantaneous frequency variations of the exciting fields. One can thus imagine the exciting process as one in which the input beams interact at any given time with a single-velocity class of atoms, producing in this particular class a spatial modulation of the atomic polarization, which is in turn responsible for the nonlinear light generation. As the laser frequency randomly fluctuates, different velocity classes are sequentially excited. Since the correlation time of the laser fluctuations is generally short compared with the atomic relaxation times, this process approaches a sequence of almost impulsional excitation of different velocity classes. Each excited-velocity class of atoms is then responsible for the emission of a DFWM train evolving in a time scale determined by the atomic relaxation times. One can thus assume that the DFWM light arising from the atomic polarization generated through nonlinear interaction with the exciting beams should have a typical correlation time related to the characteristic time scale of the atomic motion. We have checked this hypothesis through the spectral analysis of the intensity fluctuations of the DFWM signal.

This paper is organized as follows. In Sec. II we present experimental observations of the power spectrum of the intensity noise of a DFWM light beam. In Sec. III we develop numerical simulations of the DFWM signal produced by a two-level system illuminated with phase-diffusing fields. Both results are compared and discussed in Sec. IV.

II. EXPERIMENTAL OBSERVATIONS

We present in this section the experimental results obtained from the analysis of the noise carried by the DFWM signal, produced in a Rb vapor cell excited by three beams arising from the same diode laser. The geometry of the experiment corresponds to the boxcar configuration [29], with no delay introduced among the three beams.

A. Experimental setup

We use a commercial diode laser (Sharp LT021) that is current and temperature stabilized and that is tuned to the resonance frequencies of the two natural rubidium isotopes. No external line-narrowing setup is used for this laser, and special care is taken to avoid laser feedback using a Faraday optical isolator (Isowave Mod. I-80-U4). After traversing the optical isolator the laser spectral profile is monitored with a confocal Fabry-Pérot analyzer. Using this device, we measure the laser linewidth (30±5 MHz) and check for the absence of optical laser feedback that manifests itself through instabilities in the laser frequency and/or intensity and produces a characteristic linewidth reduction.

The collimated laser beam is split into three different paths of equal length. The three beams are circularly polarized and made to cross inside a 5-mm-long cell containing Rb vapor at a temperature of 90 °C. They are aligned in order to pass at the three corners of a 1-cm side square target placed 1 m after the vapor cell. The relative intensities of the three laser beams were 1:0.9:0.7. The total available power in the interaction region is 2.5 mW, and the beam's diameter is 0.5 mm. Neutral density filters were used to vary the light beam's intensities. The DFWM signal arises in the direction defined by the interaction point and the fourth square target corner. When the total available power is sent to the cell the DFWM signal power is typically 10 μW. The DFWM signal is detected with an avalanche photodiode-amplifier module (APAM) (Hamamatsu C5331), with a 10–100-MHz bandpath. The amplified signal from the APAM is sent to a 100-MHz digital oscilloscope, where the temporal evolution of the DFWM signal intensity is recorded. The oscilloscope traces, typically consisting of 4096 sample points corresponding to a 40.96-μs total scan time, were transferred to a computer for subsequent analysis.

B. Results

Figure 1 shows the intensity of the DFWM signal fluctuations as a function of the laser frequency using full available excitation power. Since our detection electronics is high-pass (cutoff frequency, 10 kHz) and the laser is slowly scanned through the atomic resonances, only the rapidly varying ac part of the DFWM intensity contributes to this spectrum. Due to optical pumping, only the transitions arising from the upper hyperfine ground-state levels participate in the process. The noise presented in Fig. 1 was recorded using the "peakdet" acquisition mode of the scope showing the extreme values reached by the signal during each temporal channel. An interesting feature of this signal is the observed asymmetry of the fluctuations with respect to the baseline, indicating large relative amplitude variations. This asymmetry decreases as the excitation power is reduced. With the maximum available laser power the rms of the DFWM signal intensity over the detection bandwidth is roughly equal to the average signal intensity. This is to be compared with the excitation beams that have negligible intensity fluctuations.

To study the temporal evolution of the DFWM signal fluctuations the laser was manually tuned to the center of the Doppler-broadened absorption line of one of the two

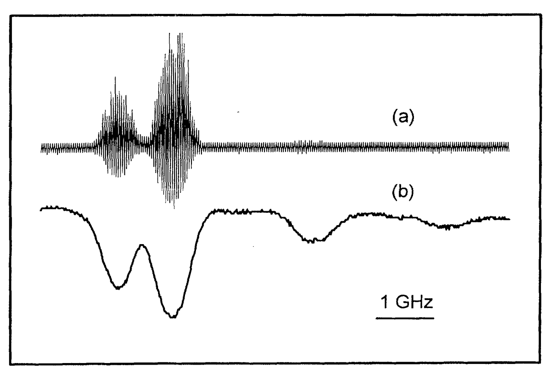

FIG. 1. (a) Observed DFWM noise as a function of the laser frequency. (b) Single-beam absorption under the same experimental conditions as (a).

relevant transitions by monitoring the transmission through an auxiliary low-density cell. In the interaction cell the beam attenuation at the line center was kept below 50%.

The temporal variations of the DFWM intensity signals were analyzed as follows. While keeping the laser frequency constant, we used the digital oscilloscope in the $Y\times t$ mode and registered a series of samples of the temporal evolution of the DFWM signal during 40.96 μs. Each temporal recording consisted of 4096 digitized sampling points. The digitized temporal evolutions were transferred to a computer for spectral analysis. From each temporal recording, we subtracted the mean value and performed a digital Fourier transform. The final spectra correspond to the averaging of the square modulus of the Fourier transforms of 64 oscilloscope traces. Large and narrow spurious peaks corresponding to unavoidable electromagnetic ambient noise were systematically present in the spectra. These peaks were removed from the final spectra using a numerical procedure that replaces abnormally large deviations of the spectrum by the average values of the neighboring points. The resulting DFWM fluctuations power spectra are shown in Fig. 2 for different values of the total laser power in the interaction region. As the excitation power is decreased, the noise spectrum width approaches a constant value of around 8 MHz, close to the homogeneous atomic linewidth (5.9 MHz) and substantially smaller than the laser linewidth. For increasing power, the fluctuation spectrum broadens and develops a local minimum around zero frequency and a maximum shifted to higher values. Similar results were observed with the two rubidium isotopes. The above observations did not depend on the precise position of the laser frequency as long as it remained within the Doppler absorption line.

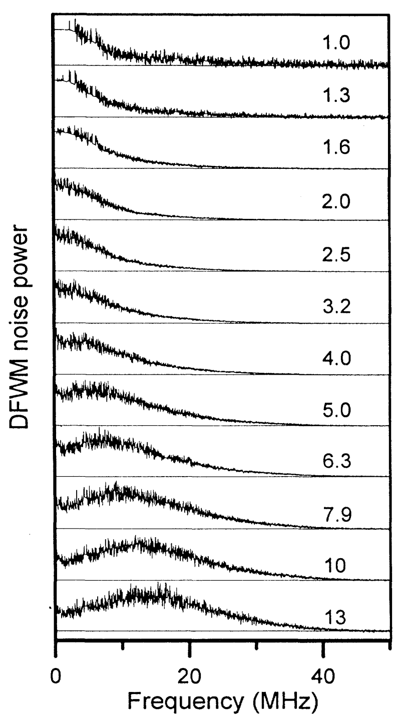

FIG. 2. Observed DFWM noise spectra for different values of the total exciting-field power. The numbers indicate relative values of the total laser power in the atomic cell for constant intensity ratios between the beams. All spectra have been normalized to the same maximum value.

III. NUMERICAL SIMULATIONS

In order to analyze the experimental results we have developed a numerical simulation of the DFWM signal produced in an ideal two-level system by phase-fluctuating laser fields. It is well known that an amplitude-stabilized laser such as a diode laser produces a field that approaches a phase-diffusing field (PDF). Such a field has a temporal dependence of the form $E(t)=E_0\exp\{-i[\omega t+\varphi(t)]\}+\text{c.c.}$, where E_0 is a constant-field amplitude and $\varphi(t)$ is a Gaussian random phase with the following stochastic properties:

$$\langle\dot{\varphi}(t)\rangle=0\ , \tag{1}$$

$$\langle\dot{\varphi}(t),\dot{\varphi}(t')\rangle=2b\delta(t-t')\ . \tag{2}$$

The corresponding field has a Lorentzian line shape with a full width at half maximum (FWHM) given by b. This field can be easily simulated numerically. We consider the generation of DFWM by a PDF incident upon a homogeneous sample of two-level atoms with states $|1\rangle$ and $|2\rangle$. The total incident field is

$$E(\mathbf{r},t)=E_0e^{-i[\omega t+\varphi(t)]}[e^{i\mathbf{k}_1\mathbf{r}}+ge^{i\mathbf{k}_2\mathbf{r}}]+\text{c.c.}\ , \tag{3}$$

where g is an adimensional expansion parameter assumed to be small. The DFWM signal arises in the direction $\mathbf{k}_s=2\mathbf{k}_1-\mathbf{k}_2$. To simulate this signal we solve numerically the optical Bloch equations for the two-level system according to the procedure described in Refs. [25] and [27]. This method allows the calculation within the rotating-wave approximation of the slow varying envelope of the third-order nonlinear density-matrix coherence coefficient $\tilde{\rho}_{12}^{(3)}(t)$ that accounts for the coherent light generation along $\mathbf{k}_s$. The calculation is valid to all orders for the field propagating along $\mathbf{k}_1$ and to first order in the field propagating along $\mathbf{k}_2$. We assume an optically thin medium and take the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$ to be negligibly small. Apart from inessential propagation corrections, the DFWM light field intensity is given by

$$I(t)\propto|\tilde{\rho}_{12}^{(3)}(t)|^2\ . \tag{4}$$

We use in the computations a time unit equal to the transverse atomic relaxation time T_2. We assume that the relaxation is purely radiative ($T_2=2T_1$). For the numerical integration of the optical Bloch equations, we use Euler's procedure with a time increment equal to $0.001\times T_2$. This increment has been tested to provide numerical errors no larger than 10%. The numerical integration extends over a total time interval of $70T_2$. The atomic system is assumed to be initially in the ground state. To eliminate spurious effects due to the initial tran-

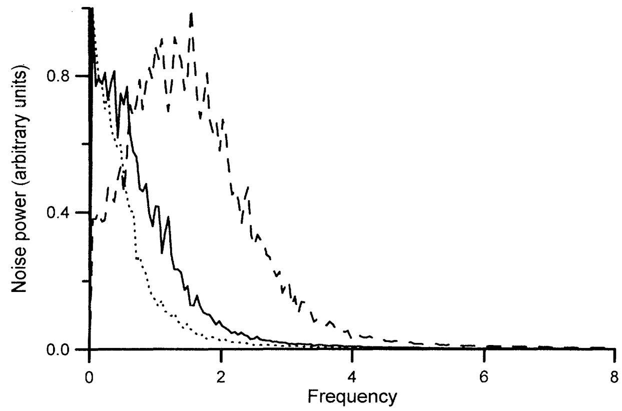

FIG. 3. Simulated DFWM intensity noise power spectra for different values of the Rabi frequency Ω. The frequency scale is taken in units of the natural atomic linewidth (FWHM) Γ. The laser linewidth is $b=5\Gamma$. All spectra have been normalized to the same maximum value. $\Omega=0.01\Gamma$, dotted; $\Omega=\Gamma$, continuous; $\Omega=2\Gamma$, dashed.

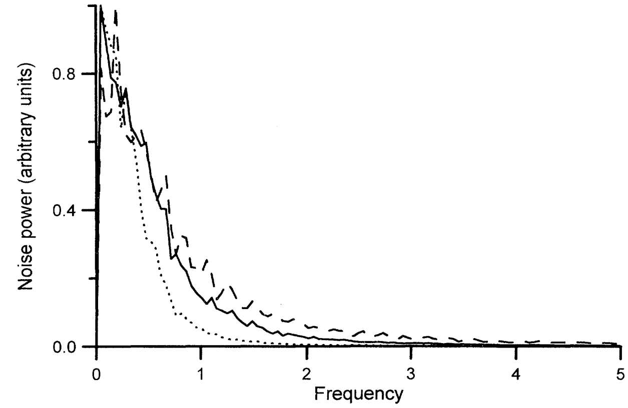

FIG. 5. Simulated DFWM intensity noise power spectra for different values of the laser linewidth b with $\Omega=0.01\Gamma$. The frequency scale is taken in units of the natural atomic linewidth (FWHM) Γ. $b=0.5\Gamma$, dotted; $b=5\Gamma$, continuous; $b=50\Gamma$, dashed. All spectra have been normalized to the same maximum value.

sient, only the results corresponding to $t>4.5T_2$ are taken under consideration in the analysis. The power spectrum of the fluctuations of the calculation DFWM is taken as the square of the Fourier transform of the difference between this signal and its mean value. The results are averaged over 100 runs for a given choice of parameters.

We have investigated the power spectrum of the DFWM signal fluctuations as a function of the incident-field amplitude, expressed in terms of the corresponding Rabi frequency $\Omega=2E_0\mu/\hbar$ (μ is the atomic dipole matrix element), and the laser linewidth b. In all the calculations g is taken as 0.01. No detuning between the laser and the atomic system is assumed. Figures 3 and 4 show the obtained noise spectrum for different values of the Rabi frequency and for the realistic value $b=5\Gamma$, where Γ is the atomic natural linewidth ($\Gamma=2/T_2$). The spectra are normalized to the same maximum value. At low field intensities the spectra show a single peak close to $\nu=0$, whose width is of the order of the atomic linewidth. As the Rabi frequency approaches the natural linewidth, the spectrum first broadens and then presents a maximum shifted to higher frequencies. For larger values of Ω (Fig. 4) the peak at $\nu\approx 0$ again becomes dominant, while two sidebands develop at frequencies corresponding to Ω and 2Ω, revealing the existence of dynamic Stark splitting. This spectrum is reminiscent of the resonance fluorescence spectrum first observed by Mollow [30]. While in the fluorescence spectrum only sidebands at $\pm\Omega$ are observed, the DFWM noise spectrum shows a sideband at 2Ω. This is a consequence of the fourth power dependence of the DFWM noise spectrum on the atomic coherence terms. Figures 5 and 6 show the variation in the calculated spectra as a function of the laser linewidth for $\Omega=0.01$ and 10, respectively. For the lowest intensi-

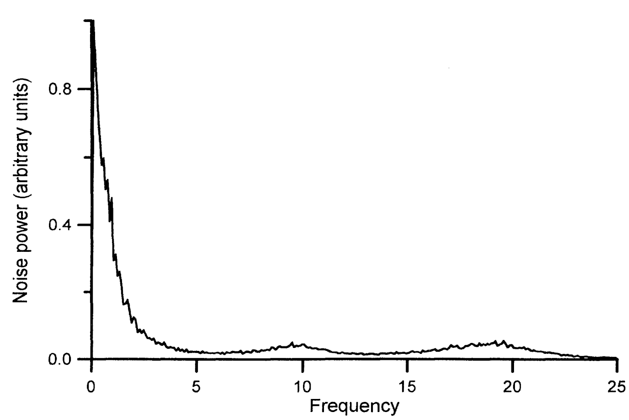

FIG. 4. Simulated DFWM intensity noise power spectrum for $\Omega=10\Gamma$ and $b=5\Gamma$. The frequency scale is taken in units of the natural atomic linewidth (FWHM) Γ.

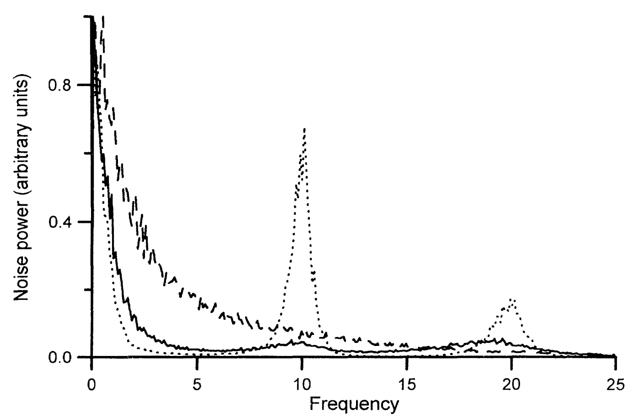

FIG. 6. Simulated DFWM intensity noise power spectra for different values of the laser linewidth b with $\Omega=10\Gamma$. The frequency scale is taken in units of the natural atomic linewidth (FWHM) Γ. $b=0.5\Gamma$, dotted; $b=5\Gamma$, continuous; $b=50\Gamma$, dashed. All spectra have been normalized to the same maximum value.

ty the spectra show a unique peak. When the laser width is taken smaller than the atomic linewidth, the peak narrows as expected for a field approaching a classical monochromatic wave. On the other hand, as the laser spectrum is taken broader than the atomic line, the noise spectrum width remains close to the atomic linewidth. This suggests that for weak-field excitation the noise spectrum of the DFWM signal provides an easy way for the determination of the homogeneous atomic linewidth. As the Rabi frequency is increased (Fig. 6), the spectra develop sidebands at Ω and 2Ω. The sidebands can be well resolved, provided that $b \ll \Omega$.

IV. DISCUSSION

To proceed with the discussion, we have to keep in mind that there are substantial differences between the assumptions of the numerical simulations that consider an ideal system and the actual experimental situation. With the first term, there is in practice a velocity distribution of the atoms in the gas that has not been considered in the calculations and cannot in principle be ignored. Another difference between the experiment and the numerical calculation comes from the fact that, in the experiment, three beams are used that intersect at a finite angle of 10 mrad, while in the calculation only two waves are considered and the angle between their wave vectors is taken as zero. However, in the theoretical model the effect of one of the fields is considered to all orders. This allows the use of such a model for the simulation of experimental situations where several (two, in the case of DFWM) undistinguishable beams are combined with a probe beam [25–27]. The theoretical simulation is expected to be better suited for the description of the weak-field limit, where saturation by the probe beam and spatial inhomogeneities due to the finite angle between the beams play a minor role, than for the strong-field limit, where the influence of these parameters should be considerable (see the discussion below).

The above results show that the spectrum of the intensity noise carried by the DFWM signal provides information on the atomic dynamics. Two situations must be distinguished. In the low-field limit the experimental spectra show a width that approaches the atomic natural linewidth and is substantially narrower than the other relevant frequencies: the laser width (30 MHz) and the Doppler width ($\sim$400 MHz). There is a significant similarity between the observed low-field spectra and the corresponding simulation for the ideal two-level system. This similarity suggests that the distribution of the atomic velocities in the atomic gas sample plays a minor role in this limit. This sub-Doppler nature of the low-field DFWM noise spectrum should be attributed to the velocity selective nature of the DFWM mechanism.

As the exciting fields' intensities are increased, the spectra display broadening and a peak shift. Since only the laser amplitude is varied, the change in the spectrum must be attributed to saturation of the sample. However, in this case the observed spectra are significantly different from those simulated for the homogeneous two-level system. This difference may be due to the following factors which have not been considered in the calculations. (i) *Inhomogeneity of the field amplitude in sample.* All atoms within the transverse beam profile contribute to the DFWM signal. To test the influence of this factor, we have made some attempts to select the generating atoms in the beam's cross section by imaging the interaction region and using a pinhole. No evidence for a position dependence of the high-field spectra could be observed. (ii) *Spatial modulation of the exciting light.* In addition to the beam's cross section variation there is a second cause for spatial field intensity distribution in the sample that is due to the finite angle between the beams not considered in the calculation. In this case, the interference between the exciting fields produces at the sample a three-dimensional intensity grating with a small spatial period. Consequently the total DFWM signal arises from atoms with different Rabi frequencies. (iii) *Atom velocity distribution.* Although the velocity distribution plays a minor role at low-field intensities, its influence in the case of saturating fields cannot in principle be ignored. Further study of the high-field spectra is necessary.

V. CONCLUSION

We have studied the power spectrum of the intensity of light generated through DFWM. Although the experiments were carried out with a simple experimental setup using an atomic vapor cell, the spectra show features that can be associated with the atomic dynamics. A comparison of the experimental results with numerical simulations reveals that at low-field intensities the atomic motion plays a minor role in the spectral width, suggesting that the DFWM noise can be readily used for an estimate of the atomic natural linewidth. At higher exciting-field power, the observed spectra differ substantially from the corresponding simulations, indicating that a more realistic calculation, taking into account the atomic motion and the spatial light modulation within the sample, is essential to the interpretation of the observations. Further work in this direction is currently underway.

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