Saturation effects in three-level selective reflection

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We consider nonlinear selective reflection on the interface between dielectric media and dilute vapors of three-level atoms. Assuming a pump-probe scheme with cascade-type excitation, we study the modification ΔR of the reflection coefficient of the probe induced by the presence of the pump. We extend previous calculations of the frequency spectrum of ΔR made for low probe power to the case of arbitrary probe power. One then obtains a strong resonance which has already been observed experimentally. Moreover, for the two-photon resonance present in the low-intensity spectrum, a power splitting can be predicted with increasing intensity [S1050-2947(96)06205-1]

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I. INTRODUCTION

Selective reflection at the interface between dielectric media and dilute atomic vapors has attracted the attention of several authors in recent years. This is because the frequency profile of the resonant reflectivity signal yields information on the behavior of excited-state atoms in the vicinity of a dielectric surface. As has been pointed out in an early paper [1], atoms are in a transient regime after deexcitation at the surface, and this behavior creates spatial dispersion leading to spectral narrowing of the selective reflection profile [2].

Theoretical studies devoted to this effect in two-level atoms have been carried out at both low and saturating intensities for normal incidence, as well as for arbitrary incidence angles [3,4]. An interesting aspect is introduced by considering the case of a pump-probe scheme [5], where one studies the modification of the reflectivity spectrum of a weak probe beam induced by the presence of an intense pump beam, since in this case additional effects such as saturation narrowing and extra resonances are observed. Finally long-range atom-surface interactions can also play a part in selective reflection spectroscopy under special conditions of observation [6,7].

More recently, studies concerning the pump-probe scheme have been extended to the case of three-level atoms where experimental results are also available [8,9]. In a previous paper [10] referred to as I, we have studied nonlinear selective reflection by considering a three-level scheme with two beams in normal incidence and with frequencies tuned respectively to the lower and the upper transition, giving rise to a cascade-type excitation. Following the suggestions made in [1], we have assumed that atoms moving towards the surface are in a stationary regime, whereas those leaving the surface are in a transient one.

In I we assumed that one of the transitions is driven by a strong field with fixed frequency detuning (the pump), and that the other transition is frequency scanned by a weak field (the probe or signal). We have studied two cases with the pump driving either the lower or the upper transition. As the reflection coefficient of the probe is modified by the presence of the pump, the quantity of interest is this modification expressed as a function of the probe frequency detuning. Experimentally this quantity can be separated from ordinary two-level reflectivity by amplitude modulation techniques.

In I we introduced an expansion of the corresponding expression with respect to both the pump and the probe field intensity, and considered only the lowest-order nonvanishing terms. In this paper we extend the calculations to all orders in the intensities of both fields in the case where the pump is tuned to the upper transition, and we consider more specifically probe saturation effects on these spectra.

II. COUPLED DENSITY-MATRIX EQUATIONS

Given a system as shown in Fig. 1, with g, r, and e, respectively, labeling the lower, intermediate and upper levels, we consider the case where the pump is tuned to the vicinity of the transition $r \rightarrow e$, whereas the probe is scanning the transition $g \rightarrow r$. In I the state of an atom was described by a reduced density matrix $\sigma(z,v_z)$ depending on the nor-

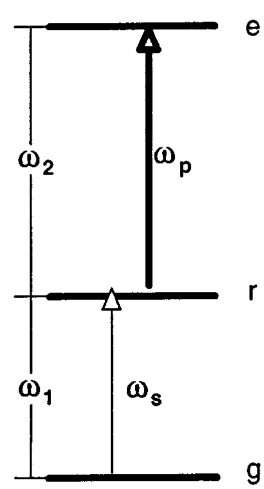

FIG. 1. Cascade three-level system. The pump beam with frequency ω_p is resonant with the upper transition (resonance frequency ω_2), while the probe beam with frequency ω_s is resonant with the lower transition (resonance frequency ω_1).

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mal distance z and on the normal velocity component v_z of the atom with respect to the dielectric surface. We have introduced the Laplace-transformed quantity

$$\hat{\sigma}(p,v_z)=\int_0^{\infty}dz\ e^{-pz}\sigma(z,v_z), \tag{1}$$

together with its stationary limit defined as

$$\overline{\sigma(v_z)}=\lim_{p\to 0^+}p\hat{\sigma}(p,v_z), \tag{2}$$

Let σ_{rg} be the matrix element representing the coherence between levels r and g. Then the selective reflection spectrum of the probe beam is shown to be proportional to the integral expression

$$\mathfrak{I}=\mathrm{Re}\int_{-\infty}^{+\infty}dv_zW(v_z)[\Theta(-v_z)\overline{\sigma_{rg}}(v_z)-2ik_s\Theta(v_z)\times\hat{\sigma}_{rg}(-2ik_s,v_z)], \tag{3}$$

where k_s is the wave vector of the probe beam. $W(v_z)$ is a normalized Maxwell-Boltzmann distribution function and $\Theta(-v_z)$ and $\Theta(v_z)$ are Heaviside functions selecting atoms with negative and positive velocity components, respectively. Thus for atoms moving toward the surface only the stationary value of σ_{rg} is relevant, whereas for atoms leaving the surface its Laplace-transformed quantity (1) with $p=-2ik_s$ is needed.

In I, for matrix elements of $\hat{\sigma}$ the following set of coupled equations were derived:

$$\begin{aligned}
v_zp\hat{\sigma}_{ee}&=\frac{i\Omega_p}{2}(\hat{\sigma}_{re}-\hat{\sigma}_{er})-A_2\hat{\sigma}_{ee},\\
v_zp\hat{\sigma}_{rr}&=\frac{i\Omega_p}{2}(\hat{\sigma}_{er}-\hat{\sigma}_{re})+\frac{i\Omega_s}{2}(\hat{\sigma}_{gr}-\hat{\sigma}_{rg})-A_1\hat{\sigma}_{rr}+A_2\hat{\sigma}_{ee},\\
v_zp\hat{\sigma}_{gg}&=\frac{i\Omega_s}{2}(\hat{\sigma}_{rg}-\hat{\sigma}_{gr})+A_1\hat{\sigma}_{rr}+v_z,\\
v_zp\hat{\sigma}_{eg}&=i\tilde{\Delta}_{eg}\hat{\sigma}_{eg}+\frac{i\Omega_p}{2}\hat{\sigma}_{rg}-\frac{i\Omega_s}{2}\hat{\sigma}_{er}-\tfrac{1}{2}A_2\hat{\sigma}_{eg},\\
v_zp\hat{\sigma}_{er}&=i\tilde{\Delta}_{er}\hat{\sigma}_{er}+\frac{i\Omega_p}{2}(\hat{\sigma}_{rr}-\hat{\sigma}_{ee})-\frac{i\Omega_s}{2}\hat{\sigma}_{eg}-\tfrac{1}{2}(A_1+A_2)\hat{\sigma}_{er},\\
v_zp\hat{\sigma}_{rg}&=i\tilde{\Delta}_{rg}\hat{\sigma}_{rg}+\frac{i\Omega_p}{2}\hat{\sigma}_{eg}+\frac{i\Omega_s}{2}(\hat{\sigma}_{gg}-\hat{\sigma}_{rr})-\tfrac{1}{2}A_1\hat{\sigma}_{rg},
\end{aligned} \tag{4}$$

which has to be solved for $\hat{\sigma}_{rg}$. Here Ω_p and Ω_s designate Rabi frequencies associated with the pump and the probe (signal) field, respectively, and defined in the usual way as the scalar products of these fields with the corresponding dipole transition elements. (For convenience we set $\hbar=1$.) A_1 and A_2 are constants of natural decay referring to the transitions $r\to g$ and $e\to r$, respectively. Furthermore we introduced Doppler-shifted frequency detunings defined in terms of the incident frequencies ω_p and ω_s and of the transition frequencies ω_{er} and ω_{rg} by the relations

$$\begin{aligned}
\tilde{\Delta}_{rg}&=\omega_s-\omega_{rg}-k_sv_z=\Delta_s-k_sv_z,\\
\tilde{\Delta}_{er}&=\omega_p-\omega_{er}-k_pv_z=\Delta_p-k_pv_z\\
\tilde{\Delta}_{eg}&=\tilde{\Delta}_{rg}+\tilde{\Delta}_{er}=\Delta_p+\Delta_s-(k_p+k_s)v_z,
\end{aligned} \tag{5}$$

Assuming copropagating geometry, we set $k_s,k_p>0$.

Note that the second equation in (4) implicitly contains the normalization condition

$$\hat{\sigma}_{gg}+\hat{\sigma}_{ee}+\hat{\sigma}_{rr}=\frac{1}{p}. \tag{6}$$

III. OUTLINE OF THE RESOLUTION METHOD

The method consists of eliminating all coherences from the set of Eqs. (4). For the two populations $\hat{\sigma}_{gg}$ and $\hat{\sigma}_{ee}$ one then obtains a set of two equations which constitutes virtually an analytical solution of our coupled equations. In this way the numerical evaluation of the unknown quantities is considerably simplified as compared with a numerical resolution of the initial set of six equations.

Consider first the last three of Eqs. (4). From these we eliminate $\hat{\sigma}_{eg}$ and, after a somewhat lengthy but straightforward calculation obtain the following expressions:

$$\begin{aligned}
\hat{\sigma}_{er}&=i\frac{\Omega_p\Omega_s^2}{8DD_{rg}D_{er}}\frac{1}{p}+\frac{i\Omega_p}{2D_{er}}\left[1-\frac{\Omega_s^2}{4D}\left(\frac{2}{D_{rg}}+\frac{1}{D_{er}}\right)\right]\hat{\sigma}_{rr}\\
&\quad-\frac{i\Omega_p}{2D_{er}}\left[1+\frac{\Omega_s^2}{4D}\left(\frac{1}{D_{rg}}-\frac{1}{D_{er}}\right)\right]\hat{\sigma}_{ee},\\
\hat{\sigma}_{rg}&=\frac{i\Omega_s}{2D_{rg}}\left(1-\frac{\Omega_p^2}{4D_{rg}D}\right)\frac{1}{p}\\
&\quad+\frac{i\Omega_s}{2D_{rg}}\left[-2+\frac{\Omega_p^2}{4D}\left(\frac{2}{D_{rg}}+\frac{1}{D_{er}}\right)\right]\hat{\sigma}_{rr}\\
&\quad+\frac{i\Omega_s}{2D_{rg}}\left[-1+\frac{\Omega_p^2}{4D}\left(\frac{1}{D_{rg}}-\frac{1}{D_{er}}\right)\right]\hat{\sigma}_{ee},
\end{aligned} \tag{7}$$

where we have set

$$\begin{aligned}
D_{er}&=v_zp-i\tilde{\Delta}_{er}+\tfrac{1}{2}(A_1+A_2),\\
D_{rg}&=v_zp-i\tilde{\Delta}_{rg}+\tfrac{1}{2}A_1,\\
D_{eg}&=v_zp-i\tilde{\Delta}_{eg}+\tfrac{1}{2}A_2\\
D&=D_{eg}+\frac{\Omega_p^2}{4D_{rg}}+\frac{\Omega_s^2}{4D_{er}}.
\end{aligned} \tag{8}$$

Note that $\hat{\sigma}_{gg}$ has also been eliminated by means of the normalization condition (6).

To complete the process of eliminating the coherences, the first and the third of Eqs. (4) can now be used. For this purpose, in addition to the quantities $\hat{\sigma}_{er}$ and $\hat{\sigma}_{rg}$ given by

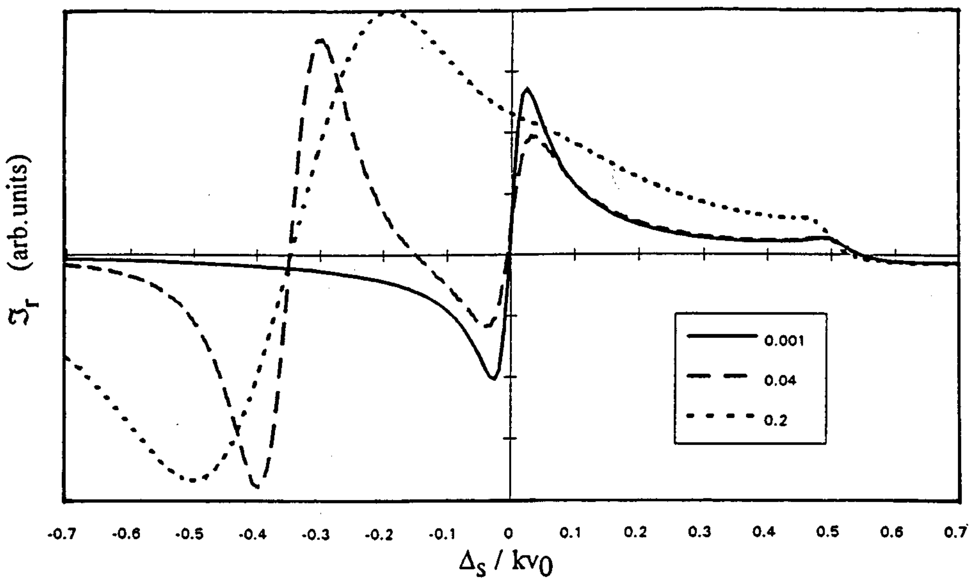

FIG. 2. Selective reflection relative amplitude $\Im_r$ vs probe detuning for negative pump detuning and for different values of probe Rabi frequency. The following parameters have been taken: ρ=0.7, $A_1=A_2=0.05kv_0$, $\Delta p=-0.5kv_0$, and $\Omega p=0.01kv_0$, where $k=(k_k+k_p)/2$.

Eqs. (7) we also need the quantities $\hat{\sigma}_{re}$ and $\hat{\sigma}_{gr}$, which in the case of nonreal p are not the complex conjugates of the former. Neither are $\hat{\sigma}_{ee}$ and $\hat{\sigma}_{rr}$ real quantities in this case, i.e., in contrast to σ, $\hat{\sigma}$ is not Hermitian. However, the required expressions for $\hat{\sigma}_{er}$ and $\hat{\sigma}_{rg}$ are just the complex conjugates of Eqs. (7) established formally by treating p as if it were a real quantity. This is because of the analyticity of these expressions, which implies that if they are valid for real p then they are also valid for any p.

Thus we write Eq. (7) in the form

$$\hat{\sigma}_{er}=\alpha_1\frac{1}{p}+\alpha_2\hat{\sigma}_{rr}+\alpha_3\hat{\sigma}_{ee},$$
$$\hat{\sigma}_{rg}=\beta_1\frac{1}{p}+\beta_2\hat{\sigma}_{rr}+\beta_3\hat{\sigma}_{ee}; \tag{9}$$

then we have

$$\hat{\sigma}_{re}=\alpha_1^\dagger\frac{1}{p}+\alpha_2^\dagger\hat{\sigma}_{rr}+\alpha_3^\dagger\hat{\sigma}_{ee},$$
$$\hat{\sigma}_{gr}=\beta_1^\dagger\frac{1}{p}+\beta_2^\dagger\hat{\sigma}_{rr}+\beta_3^\dagger\hat{\sigma}_{ee}, \tag{10}$$

where the quantities $\alpha_1^\dagger$, etc. are obtained from α_1, etc. by taking complex conjugates with p real, although in the end p is allowed to be nonreal so that in fact $\alpha_1^\dagger$ is different from α_1^*. By substituting expressions (9) and (10) into the first and the third of Eqs. (4), with $\hat{\sigma}_{gg}$ replaced again by means of (6), we obtain the following set of two equations for the two diagonal elements $\hat{\sigma}_{rr}$ and $\hat{\sigma}_{ee}$:

$$\frac{i\Omega_p}{2}(\alpha_2-\alpha_2^\dagger)\hat{\sigma}_{rr}+\left[\frac{i\Omega_p}{2}(\alpha_3-\alpha_3^\dagger)+v_zp+A_2\right]\hat{\sigma}_{ee}=-\frac{i\Omega_p}{2}(\alpha_1-\alpha_1^\dagger)\frac{1}{p},$$
$$\left[\frac{i\Omega_s}{2}(\beta_2-\beta_2^\dagger)+v_zp+A_1\right]\hat{\sigma}_{rr}+\left[\frac{i\Omega_s}{2}(\beta_3-\beta_3^\dagger)+v_zp\right]\hat{\sigma}_{ee}=-\frac{i\Omega_s}{2}(\beta_1-\beta_1^\dagger)\frac{1}{p}, \tag{11}$$

where the coefficients α_1, etc. are obtained by identifying Eqs. (9) and (7) with $p=-2ik_s$, which is the argument of $\hat{\sigma}_{rg}$ in the general expression (3).

From these equations one can obtain explicit expressions for the quantities $\hat{\sigma}_{gg}$ and $\hat{\sigma}_{ee}$ which could be evaluated directly. However, since these quantities are complex, in Eq. (11) we chose to sort out real and imaginary parts. Solving the resulting set of four real equations is equivalent, as far as computing is concerned, with evaluating the corresponding complex quantities from their analytic expressions numerically.

The results for $\hat{\sigma}_{rr}$ and $\hat{\sigma}_{ee}$ are then substituted into the second of Eqs. (9) to yield the required matrix element $\hat{\sigma}_{rg}$ which enters expression (3) with its imaginary part. This general expression (3) also involves the stationary value $\overline{\sigma_{rg}}$. According to the general definition (2), we introduce the quantities

$$\overline{\sigma_{rr}}=\lim_{p\to0^+}p\hat{\sigma}_{rr},\quad \overline{\sigma_{ee}}=\lim_{p\to0^+}p\hat{\sigma}_{ee},$$

which can be determined by multiplying Eqs. (11) by p, and taking the limit $p\to0^+$. Since p is now real, α_1 and $\alpha_1^\dagger$ are complex conjugate, so that we have $\alpha_1-\alpha_1^\dagger=2\alpha_1^I(0)$ with the superscript I designating the imaginary part, and with p set equal to 0.

Our Eqs. (11) then reduce to the form

$$-\Omega_p\alpha_2^I(0)\overline{\sigma_{rr}}+[-\Omega_p\alpha_3^I(0)+A_2]\overline{\sigma_{ee}}=\Omega_p\alpha_1^I(0),$$
$$[-\Omega_s\beta_2^I(0)+A_1]\overline{\sigma_{rr}}-\Omega_s\beta_3^I(0)\overline{\sigma_{ee}}=\Omega_s\beta_1^I(0). \tag{12}$$

Because of the hermiticity of $\overline{\sigma}$ these equations contain only real quantities. After having solved the equations for $\overline{\sigma_{rr}}$ and

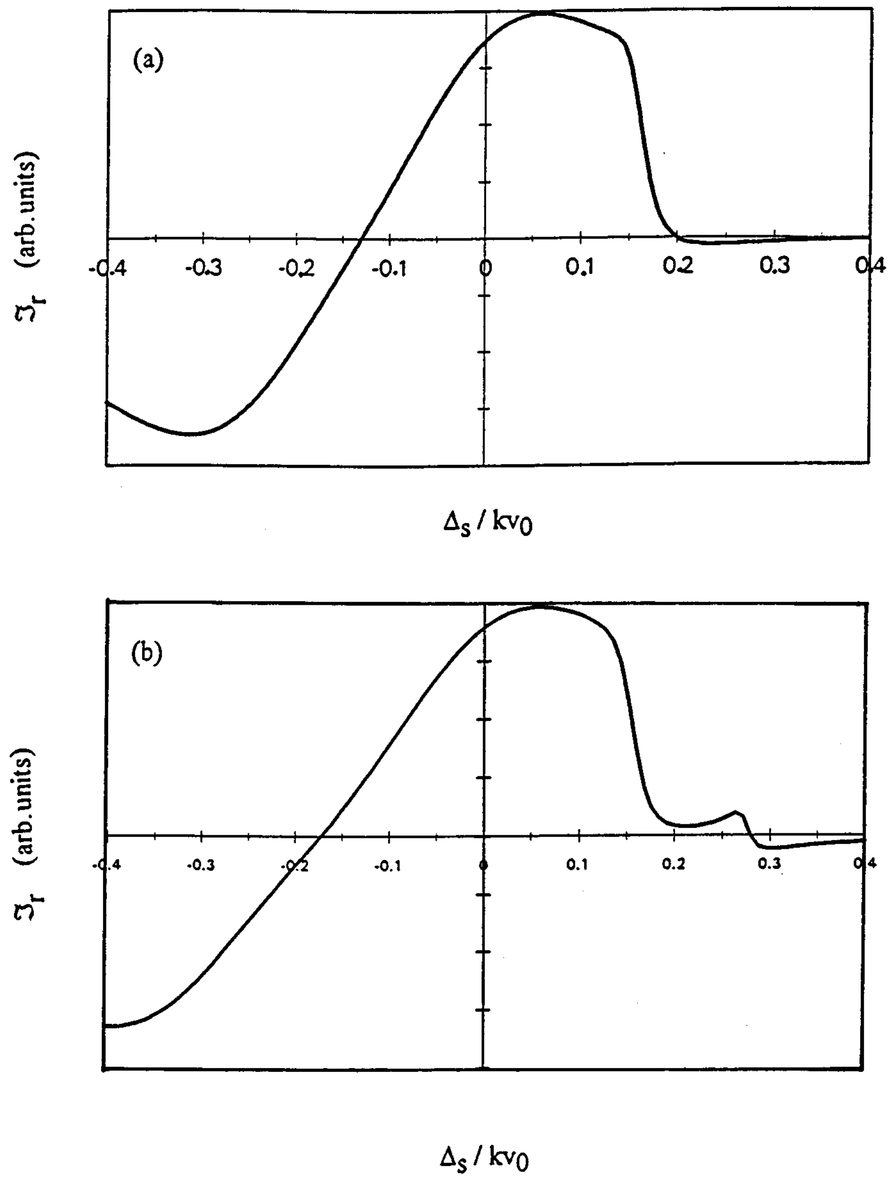

FIG. 3. Selective reflection relative amplitude $\Im_r$ vs probe detuning for negative pump detuning. The following parameters have been taken: $A_1=0.1kv_0$, $A_2=0.01kv_0$, $\Delta p=-0.2kv_0$, $\Omega_s=0.2kv_0$, and $\Omega p=0.01kv_0$, where $k=(k_s+k_p)/2$, and ρ=0.7 for (a) and ρ=0.93 for (b).

$\overline{\sigma_{ee}}$, we calculate the required matrix element $\overline{\sigma_{rg}}$ from the second of Eqs. (9), which now takes the form

$$\overline{\sigma_{rg}}=\beta_1(0)+\beta_2(0)\overline{\sigma_{rr}}+\beta_3(0)\overline{\sigma_{ee}}. \tag{13}$$

In fact only the real part of this expression, which is obtained in (13) by replacing the quantities β_1, β_2, and β_3 by their real parts, enters into (3).

Finally we indicate the particular form taken by expressions (8) which determine the values of the coefficients in Eqs. (11), (12), and (13). With the argument $p=-2ik_s$ relevant for the matrix $\hat{\sigma}$, in the transient case we have

$$\begin{aligned} D_{er}&=-i\Delta_p+i(k_p-2k_s)v_z+\tfrac{1}{2}(A_1+A_2),\\ D_{rg}&=-i\Delta_s-ik_sv_z+\tfrac{1}{2}A_1,\\ D_{eg}&=-i(\Delta_p+\Delta_s)+i(k_p-k_s)v_z+\tfrac{1}{2}A_2, \end{aligned} \tag{14}$$

whereas with $p=0$ the values for the stationary case are

$$\begin{aligned} D_{er}&=-i\Delta_p+ik_pv_z+\tfrac{1}{2}(A_1+A_2)\\ D_{rg}&=-i\Delta_s+ik_sv_z+\tfrac{1}{2}A_1,\\ D_{eg}&=-i(\Delta_p+\Delta_s)+i(k_p+k_s)v_z+\tfrac{1}{2}A_2. \end{aligned} \tag{15}$$

IV. RESULTS AND DISCUSSION

According to I the modification of the reflection coefficient R of the probe beam, due to the presence of the pump, is given by the expression (same units as in I)

$$\Delta R=\frac{4n(n-1)}{(n+1)^3}\frac{\pi N\Omega_s}{2|E_s|^2}\Im, \tag{16}$$

where n and N, respectively, are the refraction index of the dielectric and the number density of the atoms. Here we consider relative values of ΔR and therefore disregard the constant factor in the above expression of ΔR, and evaluate only the quantity $\Im$ as defined by Eq. (3). We have computed this quantity by numerical integration and plotted the result as function of the probe frequency for different fixed values of the pump frequency. We recall that in I three cases were considered: (i) the pump exactly on resonance, (ii) blue detuning of the pump by an amount kv_0, and (iii) red detuning of the pump by an amount kv_0 with $k=(k_s+k_p)/2$. The analysis was adapted to the case of the transitions $6S_{1/2}\rightarrow 6P_{3/2}$ (852 nm) and $6P_{3/2}\rightarrow 8D_{5/2}$ (621 nm) in cesium, for which experimental data are available [8]. Accordingly, for the ratio k_s/k_p the value of 0.7 has been taken.

By applying the method presented in this paper, we extended these calculations to the case of high probe power where additional effects can be expected. One then notices that the most interesting case is that of negative frequency detuning $\Delta_p<0$, because in this case extra resonances appear in the spectrum at saturating probe power.

Let us first recall that, as shown in I, at low intensities only two resonances are present for $\Delta_p<0$: one with a dispersion like shape around $\Delta_s=0$, and one narrow asymmetrical peak at positive frequency $\Delta_s=-\Delta_p$.

(i) At high probe intensities an extra resonance appears around the value $\Delta_s=(k_s/k_p)\Delta_p$. Experimental observation of this resonance has been reported in [9] In Fig. 2 we compare the profile obtained in this case with the low-intensity one. In this comparison we eliminate a trivial factor $\Omega_s\Omega_p^2$ by considering instead of $\mathfrak{F}$ the reduced quantity $\mathfrak{I}_r=\mathfrak{I}/(\Omega_s\Omega_p^2)$ which is intensity independent in the low-intensity case. As can be seen, the saturation-induced resonance has a dispersionlike shape, and its width is proportional to the probe Rabi frequency Ω_s. It corresponds to selection by the pump beam of atoms moving towards the surface, i.e., to atoms which are in steady-state interaction with the light beams. The resonance appears when the probe intensity is high enough to make the population of the intermediate level non-negligible.

(ii) Another effect is the modification of the resonance at $\Delta_s=-\Delta_p$, which in I was attributed to two-photon transitions in atoms with low normal velocity. As seen in Fig. 3(a) this resonance is no longer present as an independent peak, but manifests itself as a sudden bend on the high-frequency side of the central structure which now is strongly broadened by the intensity effect. This interpretation implies that the resonance is shifted toward the red with respect to the low-intensity value. Moreover an additional peak appears at a position shifted to the blue with respect to the original resonance as shown in Fig. 3(b). [In Fig. 3(a) this peak is not visible simply because its intensity is to weak.] These features can be viewed as a splitting of the original two-photon resonance under the influence of high probe power.

V. CONCLUSION

We have shown that due to saturating probe power additional resonances appear in the nonlinear reflection spectrum of a three-level cascade system. In particular the dispersion structure at frequency $\Delta_s=-\Delta_p$ with $\Delta_p<0$ observed in the low-intensity spectrum appears to be split into two components at high intensity. This effect, which is partially masked by the broad structure of the central resonance, could be made more clearly visible if instead of the curves presented here their derivatives were considered. Experimentally this can be achieved by using frequency modulation rather than amplitude modulation as assumed in this paper [6]. Preliminary results on the subject of frequency modulation spectra are reported elsewhere [9].

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