Amplification without inversion: Understanding probability amplitudes, quantum interference, and Feynman rules in a strongly driven system

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Density-matrix calculations provide the steady-state conditions for probe amplification or lasing between atomic levels with an uninverted population, but additional insight into the underlying physics is given by a probability amplitude approach. In this paper we derive the gain coefficient from the Feynman diagrams for a probe-laser incident on a resonantly pumped, *V*-type system using time-dependent perturbation theory in a dressed basis. The connection is made to density-matrix calculations for this model, which have been used recently to describe experiments in Rb [A. S. Zibrov *et al.*, Laser Phys. **5**, 553 (1995); Phys. Rev. Lett. **75**, 1499 (1995)]. In the density-matrix calculation the overall gain is possible because the pump-induced coherence of a strongly driven transition leads to probe amplification, despite the lack of inversion on the probe transition. In our amplitude approach we associate a specific physical process with each of the scattering channels for the probe and show how amplification without inversion can be achieved. The amplitude calculation reveals a distinction between stepwise and two-quantum processes. Interference is shown to result from the two-quantum processes, constructive for the amplification channels and destructive for the absorption. Terms appearing in the gain coefficient are traced to different sources in the amplitude and density-matrix approaches. The physical origin of each term is discussed and compared for both approaches. Terms that arise from coherences in the density-matrix approach are shown to correspond to noninterfering stepwise contributions in the amplitude approach. In deriving these results, we find that the Feynman rules that we construct for forming the probability amplitude for an arbitrary scattering process of the electromagnetic field from the coupled atom-strong pump system are consistent with Rayleigh-Schrödinger perturbation theory in the quantum dressed basis. In addition, the spontaneously emitted photons become entangled with the probe field, correlating the emission spectrum with specific scattering channels. [S1050-2947(97)06405-6]

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I. INTRODUCTION

The recent interest in quantum coherence effects in laser-atom systems has prompted some debate on the subject of amplification without inversion (AWI) [1–4]. We define AWI as the observation of laser probe amplification in a system which exhibits no population inversion on the probe transition in any bare or dressed state basis and is a precursor to lasing without inversion (LWI). The overall gain can be traced to a quantum coherence between the levels of the system [5], a fact that is evident in density-matrix calculations. (By convention, we refer to off-diagonal density-matrix elements as *coherences*.) However, additional information related to the physical origin of the gain can be found in the probability amplitudes associated with the different Feynman paths contributing to the *S* matrix for the process. Lasing without inversion schemes have been reported in two-, three-, and four-level atomic models. The experimental atoms, alkali-metal vapors for the most part, have more complicated level structures and are made to interact with laser fields of various polarizations and static magnetic fields [1,3,4].

A simple example of AWI occurs in the weak probe or Mollow spectrum [6] of the two-level atom, pumped off resonance by a strong laser field. A steady-state density-matrix calculation for this system reveals gain near $\Delta'\approx\Delta-\Gamma$, where $\Delta=\Omega-\omega$ and $\Delta'=\Omega'-\omega'$ are the pump and probe detunings from the atomic resonances, respectively, and Γ is the natural linewidth of the transition. (For the two-level atom the pump and probe drive the same transition, so that $\omega=\omega'$.) This small-signal gain feature, often called a ''stimulated Rayleigh resonance,'' is attributed by a density-matrix analysis to the coherence between the atomic levels in both the bare and dressed bases; no population inversion exists in the system. However, the density-matrix approach makes it impossible to keep track of possible interference terms in the wave function which lead to a reduction of the absorption cross section and, therefore, AWI.

Several years ago, Grynberg and Cohen-Tannoudji [7] showed this interference explicitly for the first time by deriving the gain using a perturbation theory for the quantum probability amplitudes. Assuming an off-resonant pump and probe, their calculations were performed in the dressed basis, using the dressed level which adiabatically evolves from the ground state as a proper asymptotic (stable) state of the system. The authors showed that an asymmetry occurs, whereby the two absorption channels interfere destructively, while the amplification process has only one diagram. The reduced absorption allows for overall gain in the absence of a population inversion. A similar prescription was used recently by Grynberg, Pinard, and Mandel [8] to reveal the quantum interference in an off resonant, weak pump AWI scheme for the *V*-type three-level atom. These authors suggested that a resonant, strong pump field theory, needed for an under-

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standing of many experiments but beyond the validity of their perturbation approach, seemed possible in light of recent density-matrix calculations with two-level atoms by Szymanowski and co-workers [9].

Intrigued by these results, we have studied AWI in the strongly pumped, four-level *V*-type system that was used by Zibrov and co-workers [1,10] to explain their AWI and LWI experiments in rubidium. Because of calculations like those in Refs. [7,8], it was believed that AWI occurs in this four-level atom when a quantum interference cancellation of absorption in the amplitudes can be associated with the coherence contribution to the gain in the density-matrix analysis. To our knowledge, this paper details the first amplitude calculation of AWI which is valid for strong, resonant pump fields and shows that probability amplitudes describe the precise mechanisms which allow gain to dominate absorption. We find that both gain and absorption processes are crucial in understanding this system. Our calculations are done using time-dependent perturbation theory to lowest order in a probe field in a semiclassical dressed state basis [11,12]. This basis automatically accounts for the strong, coherent pumping to all orders. A fourth state in the atom is coupled incoherently to the *V* system and acts as both an incoherent pumping reservoir and as a stationary, final state in perturbation theory. Seen as a scattering process, the probe laser scatters off of the incoherently pumped, atom–strong-field system, leading to different Feynman diagrams for gain and for absorption. In the main body of the paper, we have not quantized the pump and probe fields and note that previous quantized field calculations by Grynberg and co-workers [7,8] were independent of the quantum statistics of the field. Our technique automatically includes all multiphoton (nonlinear) pump processes combined with the absorption or emission of a single probe photon and is formally equivalent to using time-independent perturbation theory in the fully quantized dressed basis, as will be demonstrated.

The details of the amplitude approach are spelled out below. One interesting feature of the calculation is to show the role of the vacuum field in selecting the resonances of the dressed atom-field system. A more striking feature of the amplitude approach is that the various terms contributing to the absorption and amplification involve *entangled states* of the probe and vacuum field. As such, the radiation emitted into various modes of the vacuum field can be correlated with specific contributions to probe amplification or absorption. Correlations of this type have been discussed, for example, by Dubetsky and Berman in their amplitude approach to the analysis of recoil- and pressure-induced extra resonances in four-wave mixing signals [13].

In Sec. II we present the model system and review the density-matrix results for AWI. In particular, for $\Delta=\Delta'$, no population inversion on the probe transition is possible when the bare, upper probe level decays faster than the bare, upper pump level [see Fig. 1(a)]. Nevertheless, probe amplification can occur under these conditions. In Sec. III the Hamiltonian, including the necessary couplings to the vacuum field, is transformed into the semiclassical dressed basis. Then, the perturbation diagrams and cross sections for amplification and absorption are developed. Finally, the steady-state conditions are reproduced by the amplitude method. In Sec. IV we show how our results match the strong-field

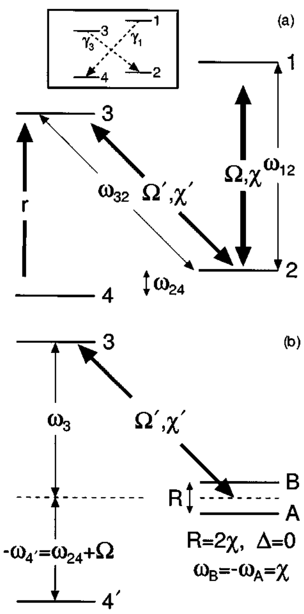

FIG. 1. (a) Energy level and pumping scheme for the four-level atom, in which r is the incoherent pumping rate of level 3. The coherent pump drives the 1-2 transition strongly with a Rabi frequency χ. Amplification without inversion occurs for the probe laser on the 2-3 transition. State 4 is asymptotically stable. (inset) Population decay rates. (b) Dressed energy levels of the atom-strong pump field system for $\Delta=\Omega-\omega_{12}=0$ in the frame rotating at the pump frequency Ω. The probe field at frequency Ω' and the quantum electromagnetic field (not shown) cause transitions between state 3 and the dressed states A and B, which are split in energy by the generalized Rabi frequency $\hbar\mathcal{R}=2\hbar\chi$. Interference results when a superposition of dressed states are formed as an intermediate state during probe field scattering. The dashed line marks the zero of energy.

density-matrix expressions for the gain coefficient and provide the correct conditions for AWI. We then conclude by discussing the basic physics of AWI, as elucidated by the amplitude calculation. In Appendix A the density-matrix master equations for this system are written out and solved in steady state. In Appendix B we describe the Feynman paths for amplification and absorption in the quantum dressed state picture.

The main result of this paper is as follows: for $\Delta=\Delta'=0$, the net probe gain coefficient is proportional to a quantity $\mathcal{G}$ defined as

$$\mathcal{G}=1-\left(\frac{\gamma_3}{\gamma_1}\right)^2+\left(\frac{\gamma_3}{\gamma_1}\right), \tag{1}$$

where γ_3 and γ_1 are the state 3 and state 1 decay rates, respectively. In a *density-matrix approach* to the calculation, the term $1-(\gamma_3/\gamma_1)^2$ can be associated with a population difference of dressed or bare states in the absence of the probe field. There is no population inversion in either the dressed or bare picture if $\gamma_3/\gamma_1>1$. As a consequence of this condition, the terms arising from the population difference can result in probe absorption only. On the other hand, the term (γ_3/γ_1) in $\mathcal{G}$ can be associated with a coherence of dressed or bare states in the absence of the probe. This extra term is responsible for AWI for $1<\gamma_3/\gamma_1<(1+\sqrt{5})/2$. The interpretation of AWI differs significantly in the *amplitude approach* to the calculation. In the amplitude approach, we find that the terms $(\gamma_3/\gamma_1)^2$ and (γ_3/γ_1) in $\mathcal{G}$ arise from *stepwise* contributions to the probe absorption and amplification, respectively. For $\gamma_3/\gamma_1>1$, the stepwise absorption always dominates the stepwise amplification, so that AWI is not possible without an additional contribution. This additional contribution comes from the ''1'' term in Eq. (1) for $\mathcal{G}$, which results from a *two-quantum* amplification process involving constructive interference between different dressed state channels to the same final state. A corresponding contribution to the probe absorption vanishes owing to destructive interference between the dressed channels. (The terms, stepwise and two quantum, are defined in Ref. [14] and will become clear in the text below.) Thus, while destructive quantum interference is crucial for overall probe gain in this system, we do not see its effect directly in the expression for the gain coefficient, viz., Eq. (1) for $\mathcal{G}$. Instead, noninterfering stepwise contributions to the amplification in the amplitude approach are associated with the coherence of bare or dressed states in the density-matrix approach, leading to the term (γ_3/γ_1) in $\mathcal{G}$.

II. AMPLIFICATION WITHOUT INVERSION IN A FOUR-LEVEL ATOM

A. The system

An energy-level diagram and pumping scheme for the four-level atom are displayed in Fig. 1. Figure 1(a) shows the bare energy-level separations, $\hbar\omega_{12}$ and $\hbar\omega_{32}$, and laser field frequencies, Ω and Ω', for the pump and probe transitions, respectively. The strong pump transition between states 1 and 2 has an associated Rabi frequency χ. In the bare picture these levels are ac Stark split by the laser. However, the system is most elegantly understood in a dressed basis, taking into account the strong laser-atom interaction. After a unitary transformation of levels 1 and 2 into the reference frame rotating at the field frequency Ω, the Hamiltonian is transformed again to form the dressed states, A and B. The resulting basis, shown schematically in Fig. 1(b), has been called the semiclassical dressed representation because it takes into account the coupling between the atom and the classical pump field nonperturbatively [11,12].

Though the off-resonant AWI analyses of Refs. [7] and [8] employed a quantized field approach, assuming the pump and probe lasers were in the N- and N'-photon states, respectively, the resulting probe gain turned out to be independent of the field statistics for $N,N'\gg 0$. Quantum dressed states are useful because the fundamental scattering processes can be well defined within the N-photon manifold. While our states, A and B, could be thought of as the N-photon pair of states in the dressed ladder, we prefer to invoke the correspondence between a classical cw field with no photons in the vacuum and a single mode coherent state of the quantum field [15]. In this way we can develop a semiclassical perturbation theory which reproduces the exact quantum results while allowing for arbitrary detunings, field strengths, and decay rates. For completeness, we will define the probe emission and absorption processes for AWI in terms of the quantum dressed states in Appendix B.

The probe field on the 2-3 transition is assumed to be weak and, therefore, treated to lowest order in its Rabi frequency χ'. In the dressed basis, as seen in Fig. 1(b), the probe simultaneously interacts with both dressed states, A and B, which are split in frequency units by the generalized pump Rabi frequency,

$$\mathcal{R}=(\Delta^2+4\chi^2)^{1/2}. \tag{2}$$

When $|\Delta|,|\Delta'|\ll 2\chi$, the probe is seen to be detuned from both dressed states. In addition, an incoherent source pumps population from the auxiliary, ground state 4 into state 3 at the rate r. State 4 is a stable state of the system and can therefore serve as an initial and final state in amplitude perturbation theory.

The inset of Fig. 1(a) shows the radiative decay scheme required to understand the physics of AWI in this system. The population decay rates are γ_1 for the $1\to4$ transition and γ_3 for the $3\to2$ transition. Note that there is no coherent excitation into or out of level 4, justifying its use as an asymptotic state in perturbation theory when r is made arbitrarily small. A steady-state density-matrix calculation can also include spontaneous emission from state 3 to 4 and from state 1 to 2. However, these decay terms turn out to have no influence on the basic physical processes by which AWI occurs and make amplitude calculations all but impossible. This is a subtle point. Decay from state 1 to 2 during coherent, resonant excitation will cause transitions between the semiclassical dressed states, corresponding to cascades between pairs of quantum dressed states from the N-photon manifold to the $(N-1)$-photon manifold and so on. This is the two-level resonance fluorescence problem, which has yet to be solved by probability amplitude methods [12]. While the spontaneous emission between the quantum dressed states has been essential in understanding previous AWI mechanisms, it plays no role in AWI in this scheme, as seen in the Feynman-type diagrams for amplification and absorption in Figs. 2 and 3.

B. The Hamiltonian and density-matrix results for the gain

The rotating-wave Hamiltonian in the dipole approximation, including the atom-vacuum interaction, is

$$\begin{aligned}\mathcal{H}=&\sum_{i=1}^{4}\hbar\omega_i\sigma_{ii}+\sum_{\mathbf{k},\lambda}\hbar\Omega_{\mathbf{k}}a^{\dagger}_{\mathbf{k},\lambda}a_{\mathbf{k},\lambda}+\hbar\sum_{\mathbf{k},\lambda}[(g_{41}\sigma^{+}_{41}\\&+g_{23}\sigma^{+}_{23})a_{\mathbf{k},\lambda}+\text{H.c.}]+\hbar\chi(\sigma^{+}_{21}e^{-i\Omega t}+\sigma^{-}_{21}e^{i\Omega t})\\&+\hbar[\chi'\sigma^{+}_{23}e^{-i\Omega' t}+(\chi')^{*}\sigma^{-}_{23}e^{i\Omega' t}].\end{aligned} \tag{3}$$

The energy of the atomic state i is given by $\hbar\omega_i$, and the frequency difference between states i and j is $\omega_{ij}=\omega_i-\omega_j$. The atomic state projection operators are represented by $\sigma_{ii}=|i\rangle\langle i|$, and the atomic raising (lowering) operator, causing a transition between states i and j, is $\sigma^{+}_{ij}=|j\rangle\langle i|$ $(\sigma^{-}_{ij}=|i\rangle\langle j|)$. The second term in Eq. (3) is the free-field Hamiltonian, where $a_{\mathbf{k},\lambda}$ $(a^{\dagger}_{\mathbf{k},\lambda})$ annihilates (creates) a photon of polarization $\hat{\mathbf{e}}_\lambda$ and frequency $\Omega_{\mathbf{k}}$ with wave vector $|\mathbf{k}|=c\Omega_{\mathbf{k}}$. The third term accounts for spontaneous emission by the atom-vacuum interaction with coupling

$$g_{ij} = -i\langle j|\mathbf{d}\cdot\hat{\mathbf{e}}_\lambda|i\rangle\left(\frac{\Omega_{\mathbf{k}}}{2\epsilon_0\hbar V}\right)^{1/2}, \tag{4}$$

where $\langle j|\mathbf{d}\cdot\hat{\mathbf{e}}_\lambda|i\rangle$ is the dipole moment along $\hat{\mathbf{e}}_\lambda$ of the i-j transition, and V is the box quantization volume of the electromagnetic field. The remaining terms of Eq. (3) show the semiclassical, dipole interactions between the applied laser fields and the atom, where the coherent pump acts only on the 1-2 transition and the probe only on the 2-3 transition. We take the pump Rabi frequency χ to be real and positive. Assuming the pump field is strong, the secular limit is defined as

$$\chi \gg \gamma_3, \gamma_1. \tag{5}$$

The equations of motion for the reduced density matrix are derived by tracing over the vacuum field and adding in the incoherent pumping term. These equations and their steady-state solutions are shown in Appendix A. The conclusions drawn from them, whether in a bare or dressed representation, are the same. In the bare basis the presence of AWI results from a coherence between levels 1 and 2 when dressed by a strong, resonant pump. This coherence leads to a contribution to the probe amplification on the 2-3 transition. The density-matrix solution shows that AWI can occur most easily if the pump and probe are both tuned to the atomic resonances. In the dressed basis, this implies that the probe is tuned directly between the dressed states A and B [see Fig. 1(b)]. Probe gain occurs on the transitions between state 3 and each of the dressed states. The overall gain in AWI is then interpreted as arising from a coherence between dressed states A and B.

The gain coefficient in the secular approximation for $\Delta=\Delta'=0$ and weak incoherent pumping is

$$G = \frac{k' n d^2}{2\epsilon_0\hbar}\,\frac{r}{\gamma_3}\,\frac{\gamma_1}{\chi^2}\left[1-\left(\frac{\gamma_3}{\gamma_1}\right)^2+\frac{\gamma_3}{\gamma_1}\right] \tag{6}$$

from Eq. (A12) in Appendix A, where $k'=\Omega'/c$ is the probe wave vector, d is the 2-3 dipole matrix element, and n is the atomic density. The square bracket is the quantity defined as $\mathcal{G}$ in Eq. (1). We set

$$\gamma_3 > \gamma_1, \tag{7}$$

so that no population inversion exists in either the dressed or bare basis. Still, amplification occurs for $G>0$. Again, the density-matrix approach does not show explicitly how quantum scattering processes lead to this AWI. That is the intended purpose of this paper. In Sec. III the amplitude equations for arbitrary parameters in the weak incoherent pump limit are developed. By then specializing to the resonant (coherent) pump case in the secular limit, the physics of the AWI becomes clear in perturbation theory for the probe.

III. DRESSED STATE PERTURBATION THEORY

A. Equations of motion for the amplitudes, incoherent pumping, and cross sections

We write the state vector in a mixed interaction representation of the bare basis,

$$|\psi(t)\rangle = (a_{1,\{\mathbf{k}\}}e^{-i\Omega t}|1\rangle + a_{2,\{\mathbf{k}\}}|2\rangle + a_{3,\{\mathbf{k}\}}e^{-i\omega_{32}t}|3\rangle + a_{4,\{\mathbf{k}\}}e^{i\omega_{24}t}|4\rangle)\otimes e^{-i\Omega_{\{\mathbf{k}\}}t}|\{\mathbf{k}\}\rangle. \tag{8}$$

For each probability amplitude, $a_{i,\{\mathbf{k}\}}$, the index i labels the atomic state $|i\rangle$. The symbol $\{\mathbf{k}\}$ labels a product of j single photon states, $|\mathbf{k}_1,\lambda_1;\mathbf{k}_2,\lambda_2;\ldots;\mathbf{k}_j,\lambda_j\rangle \equiv |\{\mathbf{k}\}\rangle$, created from the vacuum state $|\mathbf{0}\rangle$, which is denoted by $\{\mathbf{0}\}$. The energy of state $\{\mathbf{k}\}$ is written as $\hbar\Omega_{\{\mathbf{k}\}}=\hbar\Sigma_j\Omega_{\mathbf{k}_j}$. The normal interaction representation is used for states 3 and 4 in Eq. (8), while a field interaction representation is used for the strongly driven 1-2 transition. Without loss of generality, the zero of energy is assumed to be $|2,\{\mathbf{0}\}\rangle$, implying $\omega_2=0$. Forming the Schrödinger equation, we perform a Weisskopf-Wigner derivation of the amplitude decay rates by formally integrating the equations for $\dot{a}_{4,\{\mathbf{k}_0\},\mathbf{k}'}$ and $\dot{a}_{2,\{\mathbf{k}_0\},\mathbf{k}'}$, substituting into the equations for $\dot{a}_{1,\{\mathbf{k}_0\}}$ and $\dot{a}_{3,\{\mathbf{k}_0\}}$, respectively, and summing over the $\mathbf{k}'$ mode of the vacuum. The resulting equations of motion, which can be written in the Schrödinger form $i\hbar\dot{\mathbf{a}}=\tilde{H}\mathbf{a}$ for the state vector $\mathbf{a}$ and Hamiltonian matrix $\tilde{H}$, are

$$i\dot{a}_{1,\{\mathbf{k}_0\}} = -\left(\Delta + i\,\frac{\gamma_1}{2}\right)a_{1,\{\mathbf{k}_0\}} + \chi a_{2,\{\mathbf{k}_0\}}, \tag{9a}$$

$$i\dot{a}_{2,\{\mathbf{k}_0\}} = \chi a_{1,\{\mathbf{k}_0\}} + (\chi')^* e^{i\Delta' t}a_{3,\{\mathbf{k}_0\}}, \tag{9b}$$

$$i\dot{a}_{2,\{\mathbf{k}_0\},\mathbf{k}} = \chi a_{1,\{\mathbf{k}_0\},\mathbf{k}} + g_{23}^* e^{i(\Omega_{\mathbf{k}}-\omega_{32})t}a_{3,\{\mathbf{k}_0\}}, \tag{9c}$$

$$i\dot{a}_{3,\{\mathbf{k}_0\}} = \chi' e^{-i\Delta' t}a_{2,\{\mathbf{k}_0\}} - i\,\frac{\gamma_3}{2}\,a_{3,\{\mathbf{k}_0\}}, \tag{9d}$$

$$i\dot{a}_{4,\{\mathbf{k}_0\},\mathbf{k}} = g_{41}^* e^{i(\Omega_{\mathbf{k}}-\omega_{24}-\Omega)t}a_{1,\{\mathbf{k}_0\}}, \tag{9e}$$

where $\Delta=\Omega-\omega_{12}$ and $\Delta'=\Omega'-\omega_{32}$ are the pump and probe detunings, respectively. The quantum field is initially in the vacuum state, and emitted photons cannot act back on the system, so we have dropped interaction terms related to the absorption of photons out of states 2 and 4 into states 3 and 1, respectively. The in terms for states 2 and 4, via spontaneous emission, will be treated perturbatively using Eqs. (9c) and (9e) in the calculation of specific transition amplitudes below. The distinction is clear in these equations between the quantum field transitions, which change the field state from $|\{\mathbf{k}_0\}\rangle$ to $|\{\mathbf{k}_0\},\mathbf{k}\rangle$, and the classical laser transitions, which do not change the state of the vacuum field.

Semiclassical dressed states for the 1-2 transition are introduced via the transformation

$$\mathbf{a}_d = (\tilde{a}_{B,\{\mathbf{k}_0\}}\;\;\tilde{a}_{A,\{\mathbf{k}_0\}}\;\;a_{3,\{\mathbf{k}_0\}}\;\;a_{4,\{\mathbf{k}_0\}})^T = \underset{\sim}{T}\mathbf{a},\quad H_d = \underset{\sim}{T}\tilde{H}\underset{\sim}{T}^\dagger, \tag{10}$$

using the unitary matrix

$$\underset{\sim}{T} = \begin{pmatrix} c & s & 0 & 0\\ -s & c & 0 & 0\\ 0 & 0 & 1 & 0\\ 0 & 0 & 0 & 1\end{pmatrix}, \tag{11}$$

where

$$c \equiv \cos\theta = \left[\frac{1}{2}\left(1-\frac{\Delta}{\mathcal{R}}\right)\right]^{1/2}, \tag{12a}$$

$$s \equiv \sin\theta = \left[\frac{1}{2}\left(1+\frac{\Delta}{\mathcal{R}}\right)\right]^{1/2}, \tag{12b}$$

for the mixing angle $0 \leqslant \theta \leqslant \pi/2$, and $\tilde{a}_{B,\{\mathbf{k}_0\}}$ and $\tilde{a}_{A,\{\mathbf{k}_0\}}$ are dressed state probability amplitudes. This transformation is discussed in more detail in Ref. [12], where the semiclassical dressed states were used to find the pump-probe and resonance fluorescence spectrum of a two-level atom. For our purposes the dressed states,

$$|B\rangle = ce^{-i\Omega t}|1\rangle + s|2\rangle = c|\tilde{1}\rangle + s|\tilde{2}\rangle, \tag{13a}$$

$$|A\rangle = -se^{-i\Omega t}|1\rangle + c|2\rangle = -s|\tilde{1}\rangle + c|\tilde{2}\rangle, \tag{13b}$$

are time dependent in the laboratory frame but look like stationary eigenstates in the rotating frame of the pump, denoted by the tildes.

The amplitude equations become

$$i\dot{\tilde{a}}_{B,\{\mathbf{k}_0\}} = \tfrac{1}{2}(-\Delta+\mathcal{R})\tilde{a}_{B,\{\mathbf{k}_0\}} - ic\,\frac{\gamma_1}{2}\,(c\tilde{a}_{B,\{\mathbf{k}_0\}} - s\tilde{a}_{A,\{\mathbf{k}_0\}}) + s(\chi')^* e^{i\Delta' t} a_{3,\{\mathbf{k}_0\}}, \tag{14a}$$

$$i\dot{\tilde{a}}_{B,\{\mathbf{k}_0\},\mathbf{k}} = \tfrac{1}{2}(-\Delta+\mathcal{R})\tilde{a}_{B,\{\mathbf{k}_0\},\mathbf{k}} - ic\,\frac{\gamma_1}{2}\,(c\tilde{a}_{B,\{\mathbf{k}_0\},\mathbf{k}} - s\tilde{a}_{A,\{\mathbf{k}_0\},\mathbf{k}}) + sg_{23}^* e^{i(\Omega_{\mathbf{k}}-\omega_{32})t} a_{3,\{\mathbf{k}_0\}}, \tag{14b}$$

$$i\dot{\tilde{a}}_{A,\{\mathbf{k}_0\}} = \tfrac{1}{2}(-\Delta-\mathcal{R})\tilde{a}_{A,\{\mathbf{k}_0\}} + is\,\frac{\gamma_1}{2}\,(c\tilde{a}_{B,\{\mathbf{k}_0\}} - s\tilde{a}_{A,\{\mathbf{k}_0\}}) + c(\chi')^* e^{i\Delta' t} a_{3,\{\mathbf{k}_0\}}, \tag{14c}$$

$$i\dot{\tilde{a}}_{A,\{\mathbf{k}_0\},\mathbf{k}} = \tfrac{1}{2}(-\Delta-\mathcal{R})\tilde{a}_{A,\{\mathbf{k}_0\},\mathbf{k}} + is\,\frac{\gamma_1}{2}\,(c\tilde{a}_{B,\{\mathbf{k}_0\},\mathbf{k}} - s\tilde{a}_{A,\{\mathbf{k}_0\},\mathbf{k}}) + cg_{23}^* e^{i(\Omega_{\mathbf{k}}-\omega_{32})t} a_{3,\{\mathbf{k}_0\}}, \tag{14d}$$

$$i\dot{a}_{3,\{\mathbf{k}_0\}} = \chi' e^{-i\Delta' t}(s\tilde{a}_{B,\{\mathbf{k}_0\}} + c\tilde{a}_{A,\{\mathbf{k}_0\}}) - i\,\frac{\gamma_3}{2}\,a_{3,\{\mathbf{k}_0\}}, \tag{14e}$$

$$i\dot{a}_{4,\{\mathbf{k}_0\},\mathbf{k}} = g_{41}^* e^{i(\Omega_{\mathbf{k}}-\omega_{24}-\Omega)t}(c\tilde{a}_{B,\{\mathbf{k}_0\}} - s\tilde{a}_{A,\{\mathbf{k}_0\}}). \tag{14f}$$

The vacuum interactions between states A and B with strength $ics\gamma_1/2$ in Eqs. (14a)–(14d) complicate the dressed representation. These couplings can be dropped in the secular limit [see Eq. (5)] of amplitude perturbation theory.

In order to see AWI, we have argued, above and in Appendix A, that the probe must be tuned near its bare line center, $\Delta'=0$, for a strong, resonant pump. In contrast, tuning the probe to a dressed state resonance, $\Delta' \approx \pm\chi$, can lead only to absorption [see Fig. 1(b)]. These conclusions will be reproduced rigorously by our perturbation method. We clarify our understanding of the AWI scattering process at the outset by setting $\Delta=0$ in Eqs. (2), (12a) and (12b), and (14a)–(14d), resulting in the simplifications, $\mathcal{R}=2\chi$ and $c=s=\sqrt{1/2}$. Under these conditions the dressed states B and A are symmetric and antisymmetric superpositions of states $\tilde{1}$ and $\tilde{2}$, respectively, according to Eqs. (13a) and (13b). The amplitude equations reduce to

$$i\dot{\tilde{a}}_{B,\{\mathbf{k}_0\}} = \chi\tilde{a}_{B,\{\mathbf{k}_0\}} - i\,\frac{\gamma_1}{4}\,(\tilde{a}_{B,\{\mathbf{k}_0\}} - \tilde{a}_{A,\{\mathbf{k}_0\}}) + \sqrt{\tfrac{1}{2}}(\chi')^* e^{i\Delta' t} a_{3,\{\mathbf{k}_0\}}, \tag{15a}$$

$$i\dot{\tilde{a}}_{B,\{\mathbf{k}_0\},\mathbf{k}} = \chi\tilde{a}_{B,\{\mathbf{k}_0\},\mathbf{k}} - i\,\frac{\gamma_1}{4}\,(\tilde{a}_{B,\{\mathbf{k}_0\},\mathbf{k}} - \tilde{a}_{A,\{\mathbf{k}_0\},\mathbf{k}}) + \sqrt{\tfrac{1}{2}}\,g_{23}^* e^{i(\Omega_{\mathbf{k}}-\omega_{32})t} a_{3,\{\mathbf{k}_0\}}, \tag{15b}$$

$$i\dot{\tilde{a}}_{A,\{\mathbf{k}_0\}} = -\chi\tilde{a}_{A,\{\mathbf{k}_0\}} + i\,\frac{\gamma_1}{4}\,(\tilde{a}_{B,\{\mathbf{k}_0\}} - \tilde{a}_{A,\{\mathbf{k}_0\}}) + \sqrt{\tfrac{1}{2}}\,(\chi')^* e^{i\Delta' t} a_{3,\{\mathbf{k}_0\}}, \tag{15c}$$

$$i\dot{\tilde{a}}_{A,\{\mathbf{k}_0\},\mathbf{k}} = -\chi\tilde{a}_{A,\{\mathbf{k}_0\},\mathbf{k}} + i\,\frac{\gamma_1}{4}\,(\tilde{a}_{B,\{\mathbf{k}_0\},\mathbf{k}} - \tilde{a}_{A,\{\mathbf{k}_0\},\mathbf{k}}) + \sqrt{\tfrac{1}{2}}\,g_{23}^* e^{i(\Omega_{\mathbf{k}}-\omega_{32})t} a_{3,\{\mathbf{k}_0\}}, \tag{15d}$$

$$i\dot{a}_{3,\{\mathbf{k}_0\}} = \sqrt{\tfrac{1}{2}}\,\chi' e^{-i\Delta' t}(\tilde{a}_{B,\{\mathbf{k}_0\}} + \tilde{a}_{A,\{\mathbf{k}_0\}}) - i\,\frac{\gamma_3}{2}\,a_{3,\{\mathbf{k}_0\}}, \tag{15e}$$

$$i\dot{a}_{4,\{\mathbf{k}_0\},\mathbf{k}} = \sqrt{\tfrac{1}{2}}\,g_{41}^* e^{i(\Omega_{\mathbf{k}}-\omega_{24}-\Omega)t}(\tilde{a}_{B,\{\mathbf{k}_0\}} - \tilde{a}_{A,\{\mathbf{k}_0\}}). \tag{15f}$$

These amplitude equations can be simplified by removing the secular interactions, proportional to $i\gamma_1/4$, that couple the dressed states in Eqs. (15a)–(15d). The eigenvalues of the state A and B subspace are

$$\lambda_{\pm} = -i\,\frac{\gamma_1}{4} \pm \frac{1}{2}\left[4\chi^2 - \left(\frac{\gamma_1}{2}\right)^2\right]^{1/2} \simeq \pm\chi - i\,\frac{\gamma_1}{4}. \tag{16}$$

Thus, the couplings contribute nonsecular results in perturbation theory and are dropped for now. (However, these couplings are crucial in forming the density matrix self-consistently, as will be shown below in Sec. III D, where we derive the steady-state density-matrix elements with our amplitude formalism.) We see that the dressed state amplitudes decay at the rate $\gamma_1/4$, one-half of the rate of the bare state 1. In this limit the system again looks like a four-level atom interacting with a probe and the vacuum simultaneously, where the dressed levels replace levels 1 and 2 of the bare formulation. This is seen in Fig. 1(b), where the dressed states appear as a split doublet between the upper state 3 and the ground state $4'$. States A and B have the energies $\hbar\chi$ and $-\hbar\chi$, respectively.

Accordingly, to simplify the notation, we define ''dressed'' energies or frequencies, remembering that we are in a frame rotating at the pump frequency $\Omega=\omega_{12}$,

$$\omega_B = -\omega_A = \chi, \tag{17}$$

$$\omega_3 = \omega_{32}, \tag{18}$$

$$\omega_{4'} = -\omega_{24} - \Omega = -\omega_{14}. \tag{19}$$

We go to the normal interaction representation of the dressed energy levels by setting $\tilde{a}_{B,\{\mathbf{k}_0\}} = a_{B,\{\mathbf{k}_0\}} e^{-i\omega_B t}$ and $\tilde{a}_{A,\{\mathbf{k}_0\}} = a_{A,\{\mathbf{k}_0\}} e^{-i\omega_A t}$, which is equivalent to writing the state vector of the system as

$$|\psi(t)\rangle = (a_{B,\{\mathbf{k}\}} e^{-i\omega_B t}|B\rangle + a_{A,\{\mathbf{k}\}} e^{-i\omega_A t}|A\rangle + a_{3,\{\mathbf{k}\}} e^{-i\omega_3 t}|3\rangle + a_{4',\{\mathbf{k}\}} e^{-i\omega_{4'} t}|4'\rangle) \otimes e^{-i\Omega_{\{\mathbf{k}\}} t}|\{\mathbf{k}\}\rangle. \tag{20}$$

Comparing this state vector with Eq. (4), the atomic state $4'$ in the rotating frame is written as the ket $|4'\rangle = e^{-i\Omega t}|4\rangle$ and shown to be at an energy $\hbar\Omega$ below the energy of the bare state 4 in Fig. 1(b); the amplitude for this state remains the same, i.e., $a_{4',\{\mathbf{k}\}} \equiv a_{4,\{\mathbf{k}\}}$. The amplitude equations are then

$$i\dot{a}_{B,\{\mathbf{k}_0\}} = -i\,\frac{\gamma_1}{4}\,a_{B,\{\mathbf{k}_0\}} + \sqrt{\tfrac{1}{2}}\,(\chi')^* e^{i(\Omega' - \omega_{3B})t} a_{3,\{\mathbf{k}_0\}}, \tag{21a}$$

$$i\dot{a}_{B,\{\mathbf{k}_0\},\mathbf{k}} = -i\,\frac{\gamma_1}{4}\,a_{B,\{\mathbf{k}_0\},\mathbf{k}} + \sqrt{\tfrac{1}{2}}\,g_{23}^* e^{i(\Omega_{\mathbf{k}} - \omega_{3B})t} a_{3,\{\mathbf{k}_0\}}, \tag{21b}$$

$$i\dot{a}_{A,\{\mathbf{k}_0\}} = -i\,\frac{\gamma_1}{4}\,a_{A,\{\mathbf{k}_0\}} + \sqrt{\tfrac{1}{2}}\,(\chi')^* e^{i(\Omega' - \omega_{3A})t} a_{3,\{\mathbf{k}_0\}}, \tag{21c}$$

$$i\dot{a}_{A,\{\mathbf{k}_0\},\mathbf{k}} = -i\,\frac{\gamma_1}{4}\,a_{A,\{\mathbf{k}_0\},\mathbf{k}} + \sqrt{\tfrac{1}{2}}\,g_{23}^* e^{i(\Omega_{\mathbf{k}} - \omega_{3A})t} a_{3,\{\mathbf{k}_0\}}, \tag{21d}$$

$$i\dot{a}_{3,\{\mathbf{k}_0\}} = \sqrt{\tfrac{1}{2}}\,\chi'(a_{B,\{\mathbf{k}_0\}} e^{-i(\Omega' - \omega_{3B})t} + a_{A,\{\mathbf{k}_0\}} e^{-i(\Omega' - \omega_{3A})t}) - i\,\frac{\gamma_3}{2}\,a_{3,\{\mathbf{k}_0\}}, \tag{21e}$$

$$i\dot{a}_{4',\{\mathbf{k}_0\},\mathbf{k}} = \sqrt{\tfrac{1}{2}}\,g_{41}^*(a_{B,\{\mathbf{k}_0\}} e^{i(\Omega_{\mathbf{k}} - \omega_{B4'})t} - a_{A,\{\mathbf{k}_0\}} e^{i(\Omega_{\mathbf{k}} - \omega_{A4'})t}). \tag{21f}$$

These equations form the foundation of the amplitude analysis.

We are almost ready to calculate the amplification and absorption cross sections. First, however, we must incorporate the incoherent pumping into our formalism. The diagrams for probe emission and absorption in Figs. 2 and 3 each start by the incoherent pumping of level 3 from level 4. The incoherent pumping and spontaneous decay of level 3 allow for the steady state from which the probe scatters. The correct Feynman rules, which account for this pumping, will be apparent later. We start the system at an initial time t_0 with some amplitude to be in the atomic state 3 and the vacuum state of the field and examine the system at time t. In the absence of the probe, the amplitude evolves from Eq. (21e) as

$$a_{3,\{0\}}(t,t_0) = e^{-(\gamma_3/2)(t-t_0)}, \tag{22}$$

where the differential population which is pumped into state 3 between t_0 and $t_0 + dt_0$ must equal $r\,dt_0$. To calculate any observable at time t, we must sum over the ensemble of differential populations. In other words, we integrate over all previous histories of the system such that $-\infty < t_0 \leq t$. As a check, this procedure leads to the correct steady-state population of level 3,

$$|a_{3,\{0\}}(t)|^2 = \int_{-\infty}^{t} r\,dt_0 |a_{3,\{0\}}(t,t_0)|^2 = \frac{r}{\gamma_3}. \tag{23}$$

This method was used by Lamb, for example, in his classic paper on the theory of the laser [16].

The cross sections for any process leading to probe amplification or absorption are proportional to the transition rate into level 4,

$$\sigma = \frac{1}{F} \sum_{\{\mathbf{k}\}} \frac{d}{dt}\left[\int_{-\infty}^{t} r\,dt_0 \left|\sum_{\text{paths}} a_{4',\{\mathbf{k}\}}\right|^2\right], \tag{24}$$

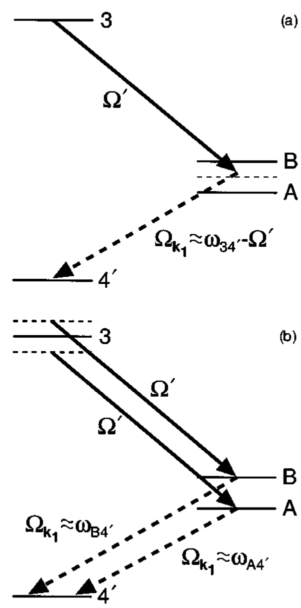

FIG. 2. Scattering channels for probe amplification. Solid arrows indicate probe field transitions to state A or B from state 3 with an interaction energy $\hbar(\chi')^*/\sqrt{2}$, while dashed arrows refer to transitions caused by the quantum field. The arrows are labeled by the frequency of the field. (a) Feynman diagram, suggesting the two-quantum processes. The probe creates a virtual superposition of states A and B, which leads to constructive interference for spontaneous emission at the frequency $\Omega_{\mathbf{k}_1} \approx \omega_{34'} - \Omega' \pm \gamma_3/2$. This interference is maximized when the probe frequency is tuned to the zero of energy, exactly half-way between the dressed states, i.e., $\Omega' = \omega_3$. (b) Stepwise processes. Because state 3 has a finite energy width $\hbar\gamma_3$, we can schematically understand the presence of spontaneous emission at the dressed transition frequencies, $\Omega_{\mathbf{k}_1} \approx \omega_{B4'} \pm \gamma_1/4$ and $\Omega_{\mathbf{k}_1} \approx \omega_{A4'} \pm \gamma_1/4$, as the result of a probe-induced transition from the wings of state 3 to states A and B. No interference occurs since the photon energy difference, $2\hbar\chi$, is well resolved.

where the semiclassical flux factor F [18] is given by

$$\frac{1}{F}=\left(\frac{k'd^2}{2\epsilon_0\hbar|\chi'|^2}\right). \tag{25}$$

The sum over the field states $\{\mathbf{k}\}$ in Eq. (24) includes all single-photon states created during probe scattering, as outlined below. The sum over paths produces a Dyson-like series representation of the amplitude,

$$\sum_{\text{paths}} a_{4',\{\mathbf{k}\}}\equiv a_{4',\{\mathbf{k}\}}(t,t_0)=\int_{t_0}^{t}dt_n\int_{t_0}^{t_n}dt_{n-1}\cdots \times\int_{t_0}^{t_2}dt_1 U(t,\ t_n,\ldots,t_1;t_0)a_{3,\{0\}}(t_1,t_0)$$

for an n-quantum process caused by an evolution operator U. It is U that is implicitly calculated using the amplitude equations (21a)–(21f).

B. Amplification cross section

Amplification follows the diagram in Fig. 2(a). The probe stimulates a transition from state 3 to both dressed states simultaneously, creating an intermediate superposition state which decays to level 4′ via spontaneous emission. The perturbation chains for Eqs. (21a)–(21f) are represented by

$$a_{3,\{0\}}(t_1,t_0)\xrightarrow{\sqrt{(1/2)}(\chi')^*}a_{B,\{0\}}(t_2,t_0)\xrightarrow{\sqrt{(1/2)}g_{41}^*}a^{(B)}_{4',\mathbf{k}_1}(t,t_0), \tag{26a}$$

$$a_{3,\{0\}}(t_1,t_0)\xrightarrow{\sqrt{(1/2)}(\chi')^*}a_{A,\{0\}}(t_2,t_0)\xrightarrow{-\sqrt{(1/2)}g_{41}^*}a^{(A)}_{4',\mathbf{k}_1}(t,t_0), \tag{26b}$$

for a total amplitude

$$a_{4',\mathbf{k}_1}(t,t_0)=a^{(B)}_{4',\mathbf{k}_1}(t,t_0)+a^{(A)}_{4',\mathbf{k}_1}(t,t_0). \tag{27}$$

Each arrow represents a transition with the perturbative interaction energy given above the arrow. The wave vector $\mathbf{k}_1$ labels the single-photon field state created from the vacuum. Now, one might think that we can easily see what happens schematically when $\Delta'\approx 0$ from the perturbation chains and Fig. 2(a). The first transition coupling constants are the same, $\sqrt{\frac{1}{2}}(\chi')^*$, but we arrive in intermediate states which have the opposite dressed energies, $\pm\hbar\chi$, producing the virtual superposition state,

$$|\psi\rangle\sim\frac{\sqrt{\frac{1}{2}}(\chi')^*}{\chi}(|A\rangle-|B\rangle). \tag{28}$$

Spontaneous emission, however, causes decay from each dressed state to state 4′ with the opposite sign of the coupling strength, $\pm\sqrt{\frac{1}{2}}g_{41}^*$, showing that the total interference is constructive. This kind of basic argument leads directly to the two-quantum contribution to the gain, the first term of Eq. (6). *However*, this argument *cannot* reproduce the stepwise amplification, shown schematically in Fig. 2(b) [14].

The formal calculation commences by substituting Eq. (22) for $a_{3,\{0\}}(t,t_0)$ into Eqs. (21a) and (21c) and integrating to find the inhomogeneous solutions. The expression for the upper dressed state amplitude is

$$a_{B,\{0\}}(t_2,t_0)=-i\int_{t_0}^{t_2}dt_1 e^{-(\gamma_1/4)(t_2-t_1)}\left(\sqrt{\tfrac{1}{2}}(\chi')^* e^{i(\Omega'-\omega_{3B})t_1}e^{-(\gamma_3/2)(t_1-t_0)}\right)$$
$$=\sqrt{\tfrac{1}{2}}(\chi')^*\,\frac{e^{i(\Omega'-\omega_{3B})t_2}e^{-(\gamma_3/2)(t_2-t_0)}-e^{-(\gamma_1/4)(t_2-t_0)}e^{i(\Omega'-\omega_{3B})t_0}}{(\omega_{3B}-\Omega')+i\left(\frac{\gamma_1}{4}-\frac{\gamma_3}{2}\right)}, \tag{29}$$

with $a_A,\{0\}(t_2,t_0)$ given by the substitution $\omega_{3B}\rightarrow\omega_{3A}$.

We now find $a^{(B)}_{4',\mathbf{k}_1}(t,t_0)$ by direct integration of Eq. (21f) using Eq. (29),

$$a^{(B)}_{4',\mathbf{k}_1}(t,t_0)=\frac{1}{2}\,g_{41}^*(\chi')^*\,\frac{1}{(\omega_{3B}-\Omega')+i\left(\frac{\gamma_1}{4}-\frac{\gamma_3}{2}\right)}\left[\frac{e^{i[\Omega_{k_1}-(\omega_{34'}-\Omega')]t}e^{-(\gamma_3/2)(t-t_0)}-e^{i[\Omega_{k_1}-(\omega_{34'}-\Omega')]t_0}}{[(\omega_{34'}-\Omega')-\Omega_{\mathbf{k}_1}]-i\,\frac{\gamma_3}{2}}\right.$$
$$\left.-e^{i(\Omega'-\omega_{3B})t_0}\,\frac{e^{i(\Omega_{k_1}-\omega_{B4'})t}e^{-(\gamma_1/4)(t-t_0)}-e^{i(\Omega_{k_1}-\omega_{B4'})t_0}}{(\omega_{B4'}-\Omega_{\mathbf{k}_1})-i\frac{\gamma_1}{4}}\right]. \tag{30}$$

Several observations at this point will help to simplify the algebra. First, any terms in the state 4′ amplitudes which vary as $e^{-\gamma_3(t-t_0)/2}$ or $e^{-\gamma_1(t-t_0)/4}$ will not contribute to the cross section after summing and squaring. This is easy to see from Eq. (24)

$$\sigma \sim \frac{d}{dt}\left[\int_{-\infty}^{t} r\, dt_0 e^{-\gamma(t-t_0)}\right]=\frac{d}{dt}\left[\frac{r}{\gamma}\right]=0. \tag{31}$$

We drop these terms to obtain a simpler form for the contributing amplitudes,

$$a^{(B)}_{4',\mathbf{k}_1}(t,t_0)=\frac{1}{2}\, g^*_{41}(\chi')^* \frac{e^{i[\Omega_{k_1}-(\omega_{34'}-\Omega')]t_0}}{(\omega_{3B}-\Omega')+i\left(\frac{\gamma_1}{4}-\frac{\gamma_3}{2}\right)}\left[\frac{-1}{[(\omega_{34'}-\Omega')-\Omega_{\mathbf{k}_1}]-i\,\frac{\gamma_3}{2}}-\frac{-1}{(\omega_{B4'}-\Omega_{\mathbf{k}_1})-i\,\frac{\gamma_1}{4}}\right]. \tag{32}$$

The two terms in the large square bracket from the integration over t_2 combine to cancel the denominator from the first integration. This turns out to be a general feature of any n-step scattering process derived in this manner: all frequency denominators cancel except for those appearing from the integration over t_n. In addition, the common phase factor for both contributing amplitudes here, $e^{i(\Omega_{k_1}-(\omega_{34'}-\Omega'))t_0}$, reflects the energy conservation of the scattering process and is also a general feature of these calculations. Since it does not depend on χ, it is the same for $a^{(B)}_{4',\mathbf{k}_1}(t,t_0)$ and $a^{(A)}_{4',\mathbf{k}_1}(t,t_0)$ and will disappear when the amplitudes are squared; therefore, we also drop this factor. The contributing amplitude in its final form is then

$$a^{(B)}_{4',\mathbf{k}_1}(t,t_0)=\frac{1}{2}\, g^*_{41}(\chi')^*\left[\frac{1}{[\Omega_{\mathbf{k}_1}-(\omega_{34'}-\Omega')]+i\,\frac{\gamma_3}{2}}\right]\times\left[\frac{1}{(\Omega_{\mathbf{k}_1}-\omega_{B4'})+i\,\frac{\gamma_1}{4}}\right], \tag{33}$$

with $-a^{(A)}_{4',\mathbf{k}_1}(t,t_0)$ given by the substitution $\omega_{B4'}\rightarrow\omega_{A4'}$.

Thus the two amplitudes contain two resonances each, corresponding to different spontaneous emission frequencies. The first resonance is for the two-quantum process from state 3 to state 4′, shown in Fig. 2(a), with spontaneous emission peaked at $\Omega_{\mathbf{k}_1}=\omega_{34'}-\Omega'$ with the full width at half maximum (FWHM) γ_3. First, the probe field drives a transition between state 3 and a virtual state somewhere between states A and B, which then decays to state 4′ via spontaneous emission. The resonance is common to $a^{(B)}_{4',\mathbf{k}_1}$ and $a^{(A)}_{4',\mathbf{k}_1}$ and therefore leads to interference when the amplitudes are summed and squared, a feature which is discussed in detail below. The second resonance is for spontaneous decay directly from each dressed state to the final state and is peaked at $\Omega_{\mathbf{k}_1}=\omega_{B4'}$ or $\Omega_{\mathbf{k}_1}=\omega_{A4'}$ with FWHM $\gamma_1/2$, independent of the probe frequency. These resonances suggest stepwise processes that can be seen as an initial, probe-induced transition from the wings of state 3 to the dressed states, followed by decay to state 4′ from that dressed state, as in Fig. 2(b). These paths do not interfere in the final state, as they lead to emission at distinct frequencies that indicate that the decay was from state A or state B unambiguously. We see that both types of processes are automatically included in each of the two amplitudes, $a^{(B)}_{4',\mathbf{k}_1}$ and $a^{(A)}_{4',\mathbf{k}_1}$. In fact, these are precisely the amplitudes of time-independent perturbation theory in a quantum dressed basis [19], as outlined in Appendix B. This point is emphasized further in the absorption calculation below.

To find the amplification cross section, we use the definition of the cross section, Eqs. (24) and (27), together with

$$\frac{d}{dt}\left[\int_{-\infty}^{t} r\, dt_0\right]=r$$

and

$$\sum_{\mathbf{k}} \rightarrow \frac{V}{(2\pi c)^3}\int_0^{2\pi}\int_{-1}^{1}\int_0^{\infty}\Omega^2_{\mathbf{k}} d\Omega_{\mathbf{k}} d(\cos\,\theta_{\mathbf{k}})d\varphi_{\mathbf{k}}. \tag{34}$$

Under the Weisskopf-Wigner approximation of spontaneous emission, the $\Omega_{\mathbf{k}_1}$ integration extends from $-\infty$ to ∞, and the function $\Omega^2_{\mathbf{k}_1}|g_{41}|^2$ is evaluated at the complex poles in an $\Omega_{\mathbf{k}_1}$ contour integration, removing the ultraviolet divergence. Using the residue theorem by closing the contour in the upper-half plane,

$$\begin{aligned}\sigma_{\text{amp}}&=\frac{1}{F}\sum_{\mathbf{k}_1,\lambda_1}\frac{d}{dt}\left[\int_{-\infty}^{t} r\, dt_0 |a^{(B)}_{4',\mathbf{k}_1}(t,t_0)+a^{(A)}_{4',\mathbf{k}_1}(t,t_0)|^2\right]\\&=\frac{r}{4F}\,|\chi'|^2\,\frac{\gamma_1}{2\pi}\,2\pi i\\&\quad\times\sum_{\text{residues}}\left|\frac{1}{[\Omega_{\mathbf{k}_1}-(\omega_{34'}-\Omega')]+i\,\frac{\gamma_3}{2}}\right|^2\\&\quad\times\left|\frac{1}{(\Omega_{\mathbf{k}_1}-\omega_{B4'})+i\,\frac{\gamma_1}{4}}-\frac{1}{(\Omega_{\mathbf{k}_1}-\omega_{A4'})+i\,\frac{\gamma_1}{4}}\right|^2.\end{aligned} \tag{35}$$

The pole at $\Omega_{\mathbf{k}_1}=(\omega_{34'}-\Omega')+i\gamma_3/2$ maximizes the constructive interference between the two, dressed channels when $\Delta'=\Omega'-\omega_3=0$. On the other hand, tuning the probe directly to a dressed resonance, $\Omega'=\omega_{3B}$ or $\Omega'=\omega_{3A}$, removes the interference and maximizes the amplification cross section. We will see in Sec. III C that the tuning, $\Omega'=\omega_{3B}$ or $\Omega'=\omega_{3A}$, also maximizes the absorption cross section, preventing AWI. Hence, we have the result that a probe tuned to the bare resonance, $\Delta'=0$, produces a beneficial interference for AWI in the strongly dressed system. In the secular limit it is easy to see the interference by evaluating the residues of Eq. (35) for $\Delta'=0$,

$$\sum_{\text{residues}} \cdots = \frac{1}{i\gamma_3}\left|\frac{1}{-\omega_B}-\frac{1}{-\omega_A}\right|^2+\frac{1}{\omega_B^2}\frac{2}{i\gamma_1}+\frac{1}{\omega_A^2}\frac{2}{i\gamma_1}$$
$$=\frac{1}{i\gamma_3}\left|\frac{1}{-\chi}-\frac{1}{\chi}\right|^2+\frac{1}{\chi^2}\frac{2}{i\gamma_1}+\frac{1}{\chi^2}\frac{2}{i\gamma_1}, \qquad (36)$$

yielding the amplification cross section,

$$\sigma_{\text{amp}}=\frac{|\chi'|^2}{F}\frac{r}{\gamma_3}\frac{\gamma_1}{\chi^2}\left[1+\frac{\gamma_3}{\gamma_1}\right]. \qquad (37)$$

We clearly identify the first term in the square brackets as the contribution arising from constructive interference of the two-photon process [Fig. 2(a)] through both dressed channels simultaneously, correlating with spontaneous emission at $\Omega_{\mathbf{k}_1}=|\omega_{4'}|$. In comparison with the density-matrix results, this term corresponds to the emission owing to the steady-state population of level 3. In contrast, the second term in the square brackets is the gain arising from the dressed state coherence in the density matrix calculation. We have found the physical origin of this term to be the stepwise process [Fig. 2(b)], first to each dressed state individually and then to state 4′ via spontaneous emission. As a result, this term is associated with spontaneous emission at the dressed transition frequencies, $\Omega_{\mathbf{k}_1}=\omega_{B4'}=|\omega_{4'}|+\chi$ and $\Omega_{\mathbf{k}_1}=\omega_{A4'}=|\omega_{4'}|-\chi$. This type of stepwise process is only possible when the initial state has a width, like γ_3, so that a probe can pick out the detuning from the initial state that will give a resonance at the dressed state. Notice that our ability to correlate the two amplification processes with distinct spontaneous emission frequencies rests on Eq. (35), which is essentially the differential spectrum of the scattered radiation at $\Omega_{\mathbf{k}_1}$ before the residues are evaluated.

For reference the amplification cross section for a probe tuned to either dressed resonance, $\Omega'=\omega_{3B}$ or $\Omega'=\omega_{3A}$, is

$$\sigma'_{\text{amp}}=\frac{|\chi'|^2}{F}\frac{r}{\gamma_3}\frac{4}{2\gamma_3+\gamma_1}, \qquad (38)$$

which can be verified by using Eq. (35). This cross section, as previously promised, is larger (by a factor of $\sim\chi^2/\gamma^2$) than the cross section [Eq. (37)] for the tuning of interest for AWI, $\Omega'=\omega_3$.

We conclude the calculation of the amplification cross section with an interesting and unexpected observation. Recently, some authors have suggested that if the physical origin of AWI in this system was revealed, the coherence contribution to the gain in the density-matrix approach would be shown to result from a quantum interference cancellation of absorption in the amplitude calculation [1]. We expected to find this result. On the contrary our analysis has proved that constructive quantum interference plays an important role for the gain but does not correspond to the coherence term in the density-matrix calculation. Rather, the noninterfering, stepwise amplification corresponds to the coherence feature. As we shall see below, these amplification processes occur in the presence of absorption suppression by destructive quantum interference.

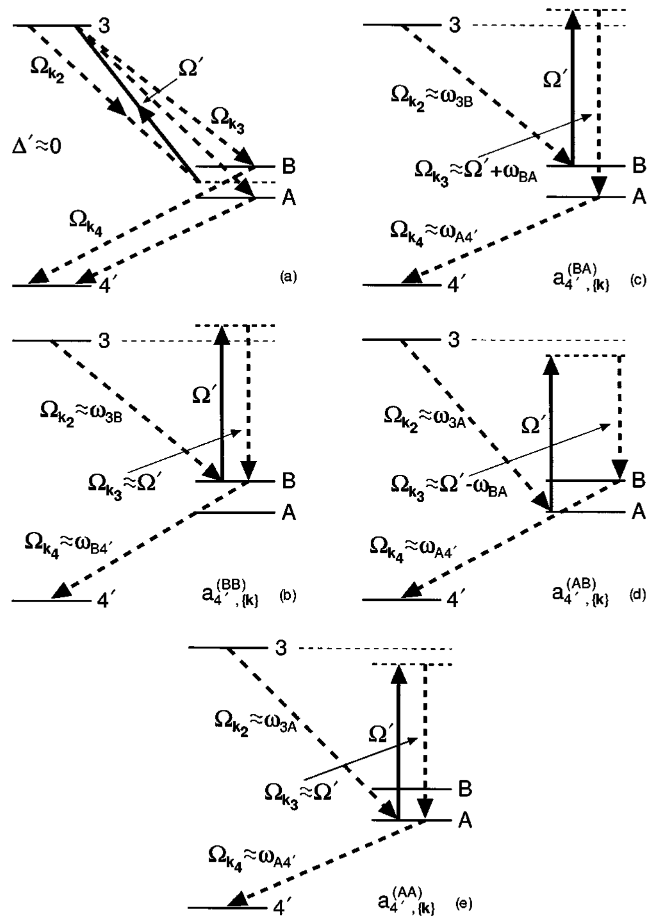

FIG. 3. Feynman diagrams for probe absorption. Solid arrows indicate probe field transitions to state 3 from state A or B with an interaction energy $\hbar\chi'/\sqrt{2}$, while dashed arrows refer to transitions caused by the quantum field. The arrows are labeled by the frequency of the field. (a) Possible scattering channel, suggesting an initial, spontaneous emission at $\Omega_{\mathbf{k}_2}\approx\Omega'$. After then absorbing a probe photon, destructive interference at state 3 between the two channels, states A and B, causes this contribution to vanish. (b) Feynman diagram for the amplitude $a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, where the probe field undergoes Rayleigh scattering from state B after an initial emission at $\Omega_{\mathbf{k}_2}\approx\omega_{3B}$. (c) Feynman diagram for $a^{(BA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, where the probe field undergoes anti-Stokes Raman scattering from state B to state A. (d) Feynman diagram for $a^{(AB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, where the probe field undergoes Stokes Raman scattering from state A to state B. (e) Feynman diagram for $a^{(AA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, where the probe field undergoes Rayleigh scattering from state A.

C. Absorption cross section

The basic absorption process is diagrammed schematically in Fig. 3(a). We write the four possible Feynman paths for the amplitude equations using Eqs. (21a)–(21f). The first two steps are spontaneous emission at frequency $\Omega_{\mathbf{k}_2}$ from state 3 to either state A or B, followed by immediate absorption of a probe photon back to state 3. Subsequent decay through the dressed states to state 4′ occurs by the spontaneous emission of two more photons, $\Omega_{\mathbf{k}_3}$ and $\Omega_{\mathbf{k}_4}$. The four perturbation chains are

$$a_{3,\{0\}}(t_1,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a_{B,\mathbf{k}_2}(t_2,t_0)\xrightarrow{\sqrt{1/2}\chi'}a^{(B)}_{3,\mathbf{k}_2}(t_3,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a^{(B)}_{B,\mathbf{k}_2,\mathbf{k}_3}(t_4,t_0)\xrightarrow{\sqrt{1/2}g^*_{41}}a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}(t,t_0), \tag{39a}$$

$$a_{3,\{0\}}(t_1,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a_{B,\mathbf{k}_2}(t_2,t_0)\xrightarrow{\sqrt{1/2}\chi'}a^{(B)}_{3,\mathbf{k}_2}(t_3,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a^{(B)}_{A,\mathbf{k}_2,\mathbf{k}_3}(t_4,t_0)\xrightarrow{-\sqrt{1/2}g^*_{41}}a^{(BA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}(t,t_0), \tag{39b}$$

$$a_{3,\{0\}}(t_1,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a_{A,\mathbf{k}_2}(t_2,t_0)\xrightarrow{\sqrt{1/2}\chi'}a^{(A)}_{3,\mathbf{k}_2}(t_3,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a^{(A)}_{B,\mathbf{k}_2,\mathbf{k}_3}(t_4,t_0)\xrightarrow{\sqrt{1/2}g^*_{41}}a^{(AB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}(t,t_0), \tag{39c}$$

$$a_{3,\{0\}}(t_1,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a_{A,\mathbf{k}_2}(t_2,t_0)\xrightarrow{\sqrt{1/2}\chi'}a^{(A)}_{3,\mathbf{k}_2}(t_3,t_0)\xrightarrow{\sqrt{1/2}g^*_{23}}a^{(A)}_{A,\mathbf{k}_2,\mathbf{k}_3}(t_4,t_0)\xrightarrow{-\sqrt{1/2}g^*_{41}}a^{(AA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}(t,t_0), \tag{39d}$$

where

$$a_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}=a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}+a^{(BA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}+a^{(AB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}+a^{(AA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} \tag{40}$$

is the total amplitude, and the dependence on t and t_0 is implicit.

Before we perform the calculation, we can get a picture of what is happening for $\Delta'\approx 0$. First, consider an initial emission at $\Omega_{\mathbf{k}_2}\approx\Omega'$, as suggested by Fig. 3(a). The source of interference is then evident: we have started in and then revisited the atomic state 3 through two different channels. The vacuum appears to create a virtual superposition state at time t_2 which is antisymmetric,

$$|\psi\rangle\sim\frac{\sqrt{1/2}g^*_{23}}{\chi}(|A\rangle-|B\rangle), \tag{41}$$

and, therefore, looks like a dark state for absorption. But, this is only true for the two-quantum process of an initial emission at $\Omega_{\mathbf{k}_2}\approx\Omega'$ followed by absorption of a probe photon, just as constructive interference for the amplification process only occurred when a downward transition followed from an antisymmetric superposition of states A and B.

The vacuum field can also pick out the dressed frequencies of the system, so that spontaneous emission is possible at $\Omega_{\mathbf{k}_2}\approx\omega_{3B}$ and $\Omega_{\mathbf{k}_2}\approx\omega_{3A}$, allowing for a detuned absorption of the probe back to level 3. This leads to the four distinct absorption paths from the initial state 3 to the ground state 4′, as shown in Figs. 3(b)–3(e). The physical meaning of each path is clear, suggesting stepwise absorption processes. The paths of Eqs. (39a) and (39b), Figs. 3(b) and 3(c), respectively, each start by spontaneous emission at the $3-B$ resonance frequency, $\Omega_{\mathbf{k}_2}\approx\omega_{3B}$, in making the transition to level B. The first path then corresponds to Rayleigh scattering of the probe off of state B through state 3, leading to $\Omega_{\mathbf{k}_3}\approx\Omega'$, while the second path indicates spontaneous anti-Stokes Raman scattering from state B to state A through state 3 by absorbing a probe photon and emitting a spontaneous photon at $\Omega_{\mathbf{k}_3}\approx\Omega'+\omega_{BA}$. The final emission to state 4′ from these paths can only occur at the dressed resonance frequencies, $\Omega_{\mathbf{k}_4}\approx\omega_{B4'}$ and $\Omega_{\mathbf{k}_4}\approx\omega_{A4'}$, respectively. Similarly, we can consider the paths of Eqs. (39c) and (39d), Figs. 3(d) and 3(e), respectively, which start by spontaneous emission at the $3-A$ resonance frequency, $\Omega_{\mathbf{k}_2}\approx\omega_{3A}$. The path of Fig. 3(d) corresponds to Stokes Raman scattering from state A to state B through state 3 with $\Omega_{\mathbf{k}_3}\approx\Omega'-\omega_{BA}$ and decay to state 4′ with $\Omega_{\mathbf{k}_4}\approx\omega_{B4'}$. Finally, the path of Fig. 3(e) considers the Rayleigh scattering of the probe from state A through state 3 with $\Omega_{\mathbf{k}_3}\approx\Omega'$, followed by decay to state 4′ with $\Omega_{\mathbf{k}_4}\approx\omega_{A4'}$. These processes, each creating a distinct set of spontaneous photons, do not interfere and give us a clear picture of how absorption occurs in this system.

Consider the diagram of Fig. 3(b) and Eq. (39a), which defines the amplitude $a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$. We integrate the amplitude equation (21b) for $a_{B,\mathbf{k}_2}(t_2,t_0)$ using the initial condition of Eq. (22) for $a_{3,\{0\}}(t,t_0)$, creating a photon in state $|\mathbf{k}_2\rangle$:

$$a_{B,\mathbf{k}_2}(t_2,t_0)=-i\int_{t_0}^{t_2}dt_1e^{-(\gamma_1/4)(t_2-t_1)}\times\left(\sqrt{\tfrac{1}{2}}g^*_{23,\mathbf{k}_2}e^{i(\Omega_{\mathbf{k}_2}-\omega_{3B})t_1}e^{-(\gamma_3/2)(t_1-t_0)}\right). \tag{42}$$

This amplitude, in turn, provides the driving term for the $a^{(B)}_{3,\mathbf{k}_2}(t_3,t_0)$ equation of motion, Eq. (21e), describing the absorption of a probe photon. Thus, formally integrating Eq. (21e),

$$a^{(B)}_{3,\mathbf{k}_2}(t_3,t_0)=-i\sqrt{\tfrac{1}{2}}\chi'\int_{t_0}^{t_3}dt_2 e^{-(\gamma_3/2)(t_3-t_2)}\times a_{B,\mathbf{k}_2}(t_2,t_0)e^{-i(\Omega'-\omega_{3B})t}, \tag{43}$$

where $a_{B,\mathbf{k}_2}(t_2,t_0)$ is given by Eq. (42). And so on for the final two vacuum interactions to derive $a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$.

The explicit form of this amplitude is lengthy before we make the same algebraic simplifications which led to a simpler form for the amplification amplitudes. In general for an n-step process, there are 2^n different terms in each amplitude after n time integrations when derived this way, as in Eq. (30) for the amplification calculation. But, half of the terms are always proportional to $e^{-\gamma(t-t_0)}$, so we throw them out, as we did for the amplification calculation to form Eq. (32). This leaves 2^{n-1} different terms, each with a common phase reflecting energy conservation. For example, the phase for the absorption calculation which is common to each of the four amplitudes, including the eight terms of $a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, is $e^{i(\Omega_{\mathbf{k}_2}+\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\Omega'-\omega_{34'})t_0}$. Dropping this common phase, the 2^{n-1} terms of each amplitude always combine to eliminate all of the denominators produced by the first $n-1$ time integrations and form a single term with n denominators. These are the n multiphoton resonances that can be formed by the energy differences between the *final state* and *the initial and each intermediate state*, including a negative imaginary energy referring to the width of a state. This is exactly the result of time-independent perturbation theory, where the nth-order amplitude to be in some final state is of this form for a system with a single initial state, such as $|3,\{\mathbf{0}\}\rangle$ [20].

For example, the eight terms combine in our case to give four frequency denominators in the amplitude,

$$a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}=\left(\sqrt{\tfrac{1}{2}}g^*_{41,\mathbf{k}_4}\right)\left(\sqrt{\tfrac{1}{2}}g^*_{23,\mathbf{k}_3}\right)\left(\sqrt{\tfrac{1}{2}}\chi'\right)\times\left(\sqrt{\tfrac{1}{2}}g^*_{23,\mathbf{k}_2}\right)\times\left[\frac{1}{(\Omega_{\mathbf{k}_2}+\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\Omega'-\omega_{34'})+i\,\frac{\gamma_3}{2}}\right]\times\left[\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{B4'}-\Omega')+i\,\frac{\gamma_1}{4}}\right]\times\left[\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{34'})+i\,\frac{\gamma_3}{2}}\right]\times\left[\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{B4'})+i\,\frac{\gamma_1}{4}}\right]. \tag{44}$$

The first line of vertex factors are simply the interaction energies of each single quantum transition in the Feynman diagram. The four complex Lorentzians are formed, in order here, by the energy difference between the final and initial state of the system and then between the final state and each of the three intermediate states, including the probe photon energy for each state. We can see this from Fig. 3(b). (These transition energies are also easy to see in a fully quantized treatment, as in Appendix B). If we go back to the absorption calculation, the same explanation holds for Eq. (33).

From these considerations, the total, contributing amplitude $a_{4,\{\mathbf{k}\}}$ is time independent for any process, and the cross section of Eq. (24) simplifies to [21]

$$\sigma_{\text{abs}}=\frac{r}{F}\sum_{\{\mathbf{k},\lambda\}}|a_{4',\{\mathbf{k}\}}|^2. \tag{45}$$

The cross section is simply the probability to be in the final system–quantum-field state, summed over the field modes, multiplied by the pumping rate of the initial state, and divided by the flux.

These arguments then lead directly to simple Feynman rules for the formation of any multiphoton scattering amplitude and cross section. In particular, we get the other three amplitudes we need by permuting $\omega_{B4'}$ and $\omega_{A4'}$ in the second and fourth frequency denominators of Eq. (44) and multiplying by -1 for $a^{(BA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$ and $a^{(AA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$. Factoring the common denominators, the sum of amplitudes for the four absorption pathways can be written

$$a_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}=a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}+a^{(BA)}_{4'\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}+a^{(AB)}_{4'\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}+a^{(AA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}=\left(\sqrt{\tfrac{1}{2}}g^*_{41,\mathbf{k}_4}\right)\left(\sqrt{\tfrac{1}{2}}g^*_{23,\mathbf{k}_3}\right)\left(\sqrt{\tfrac{1}{2}}\chi'\right)\left(\sqrt{\tfrac{1}{2}}g^*_{23,\mathbf{k}_2}\right)\times\left[\frac{1}{(\Omega_{\mathbf{k}_2}+\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\Omega'-\omega_{34'})+i\,\frac{\gamma_3}{2}}\right]\left[\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{34'})+i\,\frac{\gamma_3}{2}}\right]\left[\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{B4'}-\Omega')+i\,\frac{\gamma_1}{4}}\right.$$
$$\left.+\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{A4'}-\Omega')+i\,\frac{\gamma_1}{4}}\right]\left[\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{B4'})+i\,\frac{\gamma_1}{4}}-\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{A4'})+i\,\frac{\gamma_1}{4}}\right]. \tag{46}$$

Forming the cross section and summing over $\mathbf{k}_2,\lambda_2$ in the Weisskopf-Wigner approximation,

$$\sigma_{\rm abs}=\frac{r}{F}\sum_{\mathbf{k}_2,\lambda_2;\mathbf{k}_3,\lambda_3;\mathbf{k}_4,\lambda_4}|a_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}|^2$$
$$=\frac{r}{16F}|\chi'|^2\sum_{\mathbf{k}_3,\lambda_3;\mathbf{k}_4,\lambda_4}|g_{23,\mathbf{k}_3}|^2|g_{41,\mathbf{k}_4}|^2\left|\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{34'})+i\,\frac{\gamma_3}{2}}\right|^2\left|\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{B4'}-\Omega')+i\,\frac{\gamma_1}{4}}\right.$$
$$\left.+\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{A4'}-\Omega')+i\,\frac{\gamma_1}{4}}\right|^2\left|\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{B4'})+i\,\frac{\gamma_1}{4}}-\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{A4'})+i\,\frac{\gamma_1}{4}}\right|^2. \tag{47}$$

The single pole for $\Omega_{\mathbf{k}_2}$ in Eq. (46) defines its peak emission frequency in terms of $\Omega_{\mathbf{k}_3}$ and $\Omega_{\mathbf{k}_4}$,

$$\Omega_{\mathbf{k}_2}\approx\Omega'+\omega_{34'}-\Omega_{\mathbf{k}_3}-\Omega_{\mathbf{k}_4}, \tag{48}$$

allowing us to correlate the $\Omega_{\mathbf{k}_2}$ photon with the others even after we have summed over it. The resulting form in Eq. (47) does not depend directly on $\Omega_{\mathbf{k}_3}$, but only on $\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}$. This implies that the spectrum associated with spontaneous emission at $\Omega_{\mathbf{k}_4}$ to state $4'$ will be peaked at the frequencies $\omega_{A4'}$ and $\omega_{B4'}$. We now perform the sum over $\Omega_{\mathbf{k}_3}$ explicitly to see the quantum interference in the absorption.

The first Lorentzian with $\Omega_{\mathbf{k}_3}$ in Eq. (47) has a pole in the upper-half plane at $\Omega_{\mathbf{k}_3}=\omega_{34'}-\Omega_{\mathbf{k}_4}+i\gamma_3/2$. Using Eq. (48), this correlates with $\Omega_{\mathbf{k}_2}\approx\Omega'$, an initial spontaneous emission at the probe frequency. By contour integration over $\Omega_{\mathbf{k}_3}$ in the Weisskopf-Wigner approximation, this pole contribution in the secular limit gives

$$\sum_{\mathbf{k}_3,\lambda_3}|g_{23},\mathbf{k}_3|^2\cdots=\frac{\gamma_3}{2\pi}\frac{2\pi i}{i\gamma_3}\left|\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{B4}-\Omega')+i\,\frac{\gamma_1}{4}}+\frac{1}{(\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_{A4}-\Omega')+i\,\frac{\gamma_1}{4}}\right|^2_{\Omega_{\mathbf{k}_3}=\omega_{34'}-\Omega_{\mathbf{k}_4}}$$
$$=\left|\frac{1}{(\omega_{3B}-\Omega')+i\,\frac{\gamma_1}{4}}+\frac{1}{(\omega_{3A}-\Omega')+i\,\frac{\gamma_1}{4}}\right|^2\xrightarrow[\Delta'=0]{}O\left(\left[\frac{\gamma_1}{\chi^2}\right]^2\right), \tag{49}$$

where we have pulled out the $\Omega_{\mathbf{k}_3}$-dependent parts of Eq. (47). For $\Omega'-\omega_3=\Delta'=0$ we have completely destructive interference in the secular limit. The two terms which cancel here are precisely from the antisymmetric superposition of states A and B which would form after an initial emission at $\Omega_{\mathbf{k}_2}\approx\Omega'$. In terms of the four amplitudes, as implied by Fig. 3(a) and by Eqs. (46) and (47), $a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$ interferes destructively with $a^{(AB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$ for a final emission on the $B-4'$ transition at $\Omega_{\mathbf{k}_4}=\omega_{B4'}$, and $a^{(BA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$ interferes destructively with $a^{(AA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$ for a final emission on the $A-4'$ transition at $\Omega_{\mathbf{k}_4}=\omega_{A4'}$. Thus, we have discovered the origin of destructive interference in this AWI scheme.

We still have two contributing poles in the $\Omega_{\mathbf{k}_3}$ contour integration of Eq. (47),

$$\Omega_{\mathbf{k}_3}=\omega_{B4'}+\Omega'-\Omega_{\mathbf{k}_4}+i\gamma_1/4,\quad \Omega_{\mathbf{k}_3}=\omega_{A4'}+\Omega'-\Omega_{\mathbf{k}_4}+i\gamma_1/4. \tag{50}$$

As in the $\Omega_{\mathbf{k}_2}$ integration, these poles determine the emission peak at $\Omega_{\mathbf{k}_3}$ as a function of $\Omega_{\mathbf{k}_4}$. The first pole is common to the amplitudes $a^{(BB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$ and $a^{(BA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, while the second is common to $a^{(AB)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$ and $a^{(AA)}_{4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$. The first resonance in Eq. (47) gives a factor $1/\chi^2$ for the residue of either pole (50) in the secular approximation. For simplicity, we first look at the pole $\Omega_{\mathbf{k}_3}=\omega_{B4'}+\Omega'-\Omega_{\mathbf{k}_4}+i\gamma_1/4$. The other pole will not interfere in the secular limit, i.e., it gives corrections to the absorption of the order γ_1^2/χ^2 smaller than the leading term. Evaluating the pole contribution at $\Omega_{\mathbf{k}_3}=\omega_{B4'}+\Omega'-\Omega_{\mathbf{k}_4}+i\gamma_1/4$ from Eq. (47),

$$\sum_{\mathbf{k}_3,\lambda_3;\mathbf{k}_4,\lambda_4}|g_{23,\mathbf{k}_3}|^2|g_{41,\mathbf{k}_4}|^2\cdots=\frac{1}{\chi^2}\frac{\gamma_3}{2\pi}2\pi i\,\frac{2}{i\gamma_1}\sum_{\mathbf{k}_4,\lambda_4}|g_{41,\mathbf{k}_4}|^2\left|\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{B4'})+i\,\frac{\gamma_1}{4}}-\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{A4'})+i\,\frac{\gamma_1}{4}}\right|^2, \tag{51}$$

where the sum over $\mathbf{k}_3,\lambda_3$ reduces to the coefficients of the right-hand side. The same expression results from evaluating the pole contribution at $\Omega_{\mathbf{k}_3}=\omega_{A4'}+\Omega'-\Omega_{\mathbf{k}_4}+i\gamma_1/4$, and the two residues add. This gives the total cross section,

$$\sigma_{\rm abs}=\frac{r}{4F}|\chi'|^2\frac{1}{\chi^2}\frac{\gamma_3}{\gamma_1}\sum_{\mathbf{k}_4,\lambda_4}|g_{41,\mathbf{k}_4}|^2\left|\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{B4'})+i\frac{\gamma_1}{4}}-\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{A4'})+i\frac{\gamma_1}{4}}\right|^2$$

$$\simeq\frac{r}{4F}|\chi'|^2\frac{1}{\chi^2}\frac{\gamma_3}{\gamma_1}\sum_{\mathbf{k}_4,\lambda_4}|g_{41,\mathbf{k}_4}|^2\left[\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{B4'})^2+\left(\frac{\gamma_1}{4}\right)^2}+\frac{1}{(\Omega_{\mathbf{k}_4}-\omega_{A4'})^2+\left(\frac{\gamma_1}{4}\right)^2}\right]=\frac{|\chi'|^2}{F}\frac{r}{\gamma_3}\frac{\gamma_1}{\chi^2}\left(\frac{\gamma_3}{\gamma_1}\right)^2. \quad (52)$$

The $\Omega_{\mathbf{k}_4}$ resonances do not interfere in the secular limit, hence we have dropped the cross term in the second equality. The sum over $\mathbf{k}_4,\lambda_4$ gives a ''2'' for each term in square brackets, showing that each of the four amplitudes contributes one-fourth of the final value of the absorption cross section. Using Eqs. (48), (50), and (52), the spontaneous emission frequencies that correlate with each of the four amplitudes are those pictured in Figs. 3(b)–3(e). To tie this expression back to the steady-state density-matrix result of Eq. (A14), the absorption here is fully accounted for by the stimulated transition to state 3 from the steady-state populations, $\rho_{AA}^{(0)}$ and $\rho_{BB}^{(0)}$, which each contribute half of the absorption.

Finally, we can show from Eqs. (47) and (48) that by tuning to the dressed resonances, $\Omega'=\omega_{3B}$ or $\Omega'=\omega_{3A}$,

$$\sigma'_{\rm abs}=\frac{|\chi'|^2}{F}\frac{r}{\gamma_1}\frac{4}{2\gamma_3+\gamma_1}. \quad (53)$$

Again, this tuning produces an absorption cross section $\sim\chi^2/\gamma^2$ larger than the cross section for $\Delta'=0$, Eq. (52). Furthermore, for $\Omega'=\omega_{3B}$ or $\Omega'=\omega_{3A}$, the absorption is larger than the amplification, Eq. (38), by γ_3/γ_1, showing by our amplitude method that a lack of inversion leads to overall absorption. Therefore, AWI is impossible for direct tuning of the probe to the dressed resonances.

D. Steady-state density-matrix elements from the amplitudes

As a final exercise, we can calculate the steady state established in the absence of the probe. We know that $\rho_{33}^{(0)}$ equals r/γ_3, as derived in Eq. (23) above and corroborated by Eq. (A10) in Appendix A. Now, we need to find $\rho_{AA}^{(0)}$, $\rho_{BB}^{(0)}$, and $\rho_{AB}^{(0)}$. These quantities will confirm that our formalism reproduces all of the relevant, strong-pump results of the density-matrix approach, Eqs. (A13a) and (A13b).

Returning to the amplitude equations for $a_{A,\mathbf{k}_2}(t,t_0)$ and $a_{B,\mathbf{k}_2}(t,t_0)$, Eqs. (21b) and (21d), with the initial condition given by Eq. (22), we conjecture that spontaneous transitions from state 3 to states A and B *in the absence of the probe* allow the steady-state dressed populations and coherences to form. Therefore, we have

$$\rho_{AA}^{(0)}\equiv\sum_{\mathbf{k}_2,\lambda_2}\int_{-\infty}^{t}r\,dt_0|a_{A,\mathbf{k}_2}(t,t_0)|^2, \quad (54a)$$

$$\rho_{BB}^{(0)}\equiv\sum_{\mathbf{k}_2,\lambda_2}\int_{-\infty}^{t}r\,dt_0|a_{B,\mathbf{k}_2}(t,t_0)|^2, \quad (54b)$$

$$\rho_{AB}^{(0)}\equiv\sum_{\mathbf{k}_2,\lambda_2}\int_{-\infty}^{t}r\,dt_0\tilde{a}_{A,\mathbf{k}_2}(t,t_0)\tilde{a}^*_{B,\mathbf{k}_2}(t,t_0). \quad (54c)$$

For the definition of $\rho_{AB}^{(0)}$, we use the full equations of motion, Eqs. (15b) and (15d), including the previously unimportant coupling terms between states A and B. From Eq. (42) for $a_{B,\mathbf{k}_2}(t,t_0)$, we readily verify that $\rho_{AA}^{(0)}=\rho_{BB}^{(0)}=r/\gamma_1$, where we have summed over the vacuum in the Weisskopf-Wigner approximation. Clearly, no inversion exists in the dressed basis when $\rho_{AA}^{(0)},\rho_{BB}^{(0)}>\rho_{33}^{(0)}$, which again shows that $\gamma_3>\gamma_1$ must hold.

On the other hand, to find the steady-state coherence $\rho_{AB}^{(0)}$, we must formally solve the coupled amplitude equations, (15b) and (15d), for $\tilde{a}_{A,\mathbf{k}_2}$ and $\tilde{a}_{B,\mathbf{k}_2}$ because the off-diagonal couplings between the dressed amplitudes are crucial to get the correct, off-diagonal density-matrix element in a secular expansion. To first order in γ_1/χ, the probability amplitudes are

$$\tilde{a}_{B,\mathbf{k}_2}(t,t_0)=-i\frac{g_{23}^*}{\sqrt{2}}\int_{t_0}^{t}dt_1\left[e^{-(i\chi+\gamma_1/4)(t-t_1)}+\left(\frac{-i\gamma_1}{8\chi}\right)\times e^{(i\chi-\gamma_1/4)(t-t_1)}\right]e^{i(\Omega_{\mathbf{k}_2}-\omega_3)t_1}e^{-(\gamma_3/2)(t_1-t_0)}, \quad (55a)$$

$$\tilde{a}_{A,\mathbf{k}_2}(t,t_0)=-i\frac{g_{23}^*}{\sqrt{2}}\int_{t_0}^{t}dt_1\left[e^{(i\chi-\gamma_1/4)(t-t_1)}+\left(\frac{i\gamma_1}{8\chi}\right)\times e^{-(i\chi+\gamma_1/4)(t-t_1)}\right]e^{i(\Omega_{\mathbf{k}_2}-\omega_3)t_1}e^{-(\gamma_3/2)(t_1-t_0)}. \quad (55b)$$

Formally, to first order in γ_1/χ, the off-diagonal terms in the Hamiltonian affect only the eigenvectors of the state A and B subspace, not the dressed eigenvalues, $\pm\chi-i\gamma_1/4$, as seen here. The second term in each square bracket represents this change in the eigenvector, which mixes in the other dressed state. Substituting into Eq. (54c), we find that $\rho_{AB}^{(0)}\simeq ir/2\chi$. This is the correct value for the dressed coherence, and we have now confirmed that our method produces the steady-state density-matrix elements.

IV. DISCUSSION AND CONCLUSION

Gathering the results of the amplitude calculation, Eqs. (37) and (52), we arrive at the overall gain coefficient for $\Delta=\Delta'=0$,

$$G=(\sigma_{\mathrm{amp}}-\sigma_{\mathrm{abs}})n=\frac{k'nd^2}{2\epsilon_0\hbar}\frac{r}{\gamma_3}\frac{\gamma_1}{\chi^2}\left[1+\left(\frac{\gamma_3}{\gamma_1}\right)-\left(\frac{\gamma_3}{\gamma_1}\right)^2\right], \tag{56}$$

where n is the atomic density. This is identical to the density-matrix result, Eq. (6) or (A12). Moreover, our derivation identified the physical origin in an amplitude approach of each of the three terms in the square brackets and thereby allowed us to associate different amplitude perturbation processes with the contributing elements of the density matrix. We see that the condition that $\gamma_3>\gamma_1$ not only assures that our system is uninverted, but also assures that the two-quantum contribution, (γ_3/γ_1), to the amplification process is larger than the stepwise contribution ''1.'' When the ratio of decay rates becomes too large, the stepwise absorption, $(\gamma_3/\gamma_1)^2$, will swamp both amplification contributions. The boundary between these cases is given by

$$1+\frac{\gamma_3}{\gamma_1}=\left(\frac{\gamma_3}{\gamma_1}\right)^2\Leftrightarrow\frac{\gamma_3}{\gamma_1}=\tfrac{1}{2}(1+\sqrt{5})\cong 1.62. \tag{57}$$

(That the upper limit on the ratio of the decay rates forms the golden mean is an interesting side note.) The range of decay ratios for which AWI is possible is then

$$1<\frac{\gamma_3}{\gamma_1}<\tfrac{1}{2}(1+\sqrt{5}). \tag{58}$$

Our strong, resonant pump results lead to different conclusions than previous analyses of off-resonant, weak pump AWI. In those schemes, interferences between *different orders* of perturbation theory led to reduced absorption [7,8]. Here, interferences occur in the *same order* of perturbation theory because we probe between two intermediate levels, the dressed states. These interferences are present for both amplification and absorption diagrams. With the destructive interference eliminating the possibility of two-quantum absorption, only stepwise absorption directly from the dressed states is allowed. This stepwise absorption is larger than the stepwise amplification. The additional, two-quantum amplification which arises from constructive interference can, therefore, be interpreted as the reason behind AWI from an amplitude approach.

A significant new finding is our ability to correlate the spontaneous emission spectrum with distinct amplification and absorption pathways, owing to the entanglement of the probe and vacuum fields. The two-quantum amplification process is the only path which accompanies emission at the frequency $|\omega_{4'}|$. However, both the stepwise amplification and absorption processes correlate with emission at the sidebands, $|\omega_{4'}|\pm\chi$. In order to distinguish these two processes, the absorption pathways lead to the additional spontaneous emission of two photons near the probe frequency Ω'. These two frequencies, $\Omega_{\mathbf{k}_2}$ and $\Omega_{\mathbf{k}_3}$, are a distinct pair for each of the four absorption diagrams, as seen in Figs. 3(b)–3(e). Therefore, for each scattered probe photon we can track exactly which Feynman pathway was taken using photon coincidence spectroscopy of the fluorescence. The exception is the precise path of the two-quantum amplification, where quantum interference does not allow us to know which intermediate channel, state A or B, was used for emission at $|\omega_{4'}|$.

What about our *a priori* omission of spontaneous decay from state 1 to 2 (and therefore between states A and B)? Considering that the steady state is established by the two pump fields such that the dressed states have a negligible population difference in the secular limit for $\Delta=0$, it is reasonable that spontaneous emission from A to B and vice versa plays an unimportant role in defining the physical mechanisms for probe amplification and absorption. In the quantum dressed picture a dynamic equilibrium is established where the decay out of the N-photon manifold of dressed states is compensated by decay into this manifold. Therefore, the populations reach the same steady state in each manifold for $N\gg 1$. As for the dressed state coherence, which provides the important gain contribution for AWI to take place in the density-matrix approach, it would decay at a faster rate than $\gamma_1/2$ without changing the basic physics [17].

Throughout this paper we have described our subject as AWI in the secular limit. More precisely, the probe gain coefficient (56) specifies the lowest-order amplification in a χ^{-1} expansion for resonant pump and probe fields. To produce results which are correct beyond the secular approximation, but still restricted to a weak probe, we would have to diagonialize the state 1-2 subspace exactly in Eqs. (9a)–(9e). Such a unitary transformation is also restricted to the decay scheme we have chosen in Fig. 1(a) and is characterized by eigenvectors and eigenvalues which form a true basis of the coupled atom, pump field, and vacuum. A perturbation calculation in this new basis would be valid for arbitrary decay rates, γ_1 and γ_3. In the secular limit for either a semiclassical or quantum pump field, this representation would reduce to the dressed basis.

The assumptions of weak incoherent pumping and no decay from levels 3 to 4 were necessary to avoid the infinite number of quantum pathways associated with multistep excitation and saturation of the 3-4 transition by the incoherent pump. In this context we have constructed a perturbation theory which allows for a treatment of strong atom-pump field coupling in the presence of the vacuum and which leads to clear Feynman rules for any probe or vacuum scattering process. The key is to use the dressed basis. We have applied that theory to analyze AWI in a system used to model recent experimental results [1] by developing Feynman diagrams which clearly define the amplification and absorption of a probe laser. These Feynman diagrams identify the role of quantum interference when transitions are induced simultaneously to each dressed state. The interferences were shown to be constructive for amplification and destructive for absorption. In this way we have discovered the physical origin of AWI in such a system.

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APPENDIX A: DENSITY-MATRIX MASTER EQUATIONS

The master equations for the bare atomic populations and coherences for the closed system outlined in Sec. II A can be written by inspection since we already know the relevant decay rates [see Fig. 1(a)] in the Weisskopf-Wigner approximation. These equations are

$$\dot{\rho}_{11} = -\gamma_1 \rho_{11} - (i\chi\tilde{\rho}_{21} + \text{c.c.}), \tag{A1a}$$

$$\dot{\rho}_{22} = \gamma_3 \rho_{33} + (i\chi\tilde{\rho}_{21} + \text{c.c.}) + (i\chi'\tilde{\rho}_{23} + \text{c.c.}), \tag{A1b}$$

$$\dot{\rho}_{33} = -r(\rho_{33} - \rho_{44}) - \gamma_3\rho_{33} - (i\chi'\tilde{\rho}_{23} + \text{c.c.}), \tag{A1c}$$

$$\dot{\rho}_{44} = \gamma_1\rho_{11} + r(\rho_{33} - \rho_{44}), \tag{A1d}$$

$$\dot{\tilde{\rho}}_{21} = -i\chi(\rho_{11} - \rho_{22}) - \left(i\Delta + \frac{\gamma_1}{2}\right)\tilde{\rho}_{21} - i(\chi')^*\tilde{\rho}_{31}, \tag{A1e}$$

$$\dot{\tilde{\rho}}_{23} = -i(\chi')^*(\rho_{33} - \rho_{22}) - \left(i\Delta' + \frac{\gamma_3}{2}\right)\tilde{\rho}_{23} - i\chi\tilde{\rho}_{13}, \tag{A1f}$$

$$\dot{\tilde{\rho}}_{13} = i(\chi')^*\tilde{\rho}_{12} - i\chi\tilde{\rho}_{23} + \left(i\delta - \frac{\gamma_1 + \gamma_3}{2}\right)\tilde{\rho}_{13}, \tag{A1g}$$

$$\rho_{14} = \rho_{34} = 0, \tag{A1h}$$

with the supplementary definitions for coherences in a field interaction representation,

$$\tilde{\rho}_{12} = \rho_{12}e^{i\Omega t} = (\tilde{\rho}_{21})^*, \quad \tilde{\rho}_{32} = \rho_{32}e^{i\Omega' t} = (\tilde{\rho}_{23})^*,$$
$$\tilde{\rho}_{31} = \rho_{31}e^{i(\Omega' - \Omega)t} = (\tilde{\rho}_{13})^*. \tag{A2}$$

The closure condition is $\Sigma_{i=1}^{4} \rho_{ii} = 1$. In Eq. (A1g) the relative pump-probe detuning is defined as

$$\delta = \Delta - \Delta'. \tag{A3}$$

To first order in the probe field, the off-diagonal element is

$$\tilde{\rho}_{23}^{(1)} = -i(\chi')^* \left[(\rho_{33}^{(0)} - \rho_{22}^{(0)}) + \frac{i\chi\tilde{\rho}_{12}^{(0)}}{\frac{\gamma_1 + \gamma_3}{2} - i\delta} \right] \times \left[\frac{\gamma_3}{2} + i\Delta' + \frac{\chi^2}{\left(\frac{\gamma_1 + \gamma_3}{2} - i\delta\right)} \right]^{-1}, \tag{A4}$$

where the steady-state density-matrix elements in the absence of the probe are

$$\rho_{33}^{(0)} - \rho_{22}^{(0)} = \frac{r\left[\chi^2 \frac{\gamma_1}{\gamma_3}\left(1 - \frac{\gamma_3}{\gamma_1}\right) - \left(\frac{\gamma_1}{2}\right)^2 - \Delta^2\right]}{r\left[\left(\frac{\gamma_1}{2}\right)^2 + \Delta^2\right] + \beta\chi^2 \frac{\gamma_1}{\gamma_3}} \tag{A5}$$

and

$$\tilde{\rho}_{12}^{(0)} = \frac{i\chi}{\frac{\gamma_1}{2} - i\Delta}(\rho_{11}^{(0)} - \rho_{22}^{(0)})$$
$$= \frac{i\chi}{\frac{\gamma_1}{2} - i\Delta}\left(\frac{-r\left[\left(\frac{\gamma_1}{2}\right)^2 + \Delta^2\right]}{r\left[\left(\frac{\gamma_1}{2}\right)^2 + \Delta^2\right] + \beta\chi^2\frac{\gamma_1}{\gamma_3}} \right) \tag{A6}$$

for

$$\beta = \gamma_3 + 2r\left(1 + \frac{\gamma_3}{\gamma_1}\right). \tag{A7}$$

In Eq. (A4), $\tilde{\rho}_{23}^{(1)}$ has two distinct parts. The first term in the first set of square brackets reflects the expected absorption which results from having more population in state 2 than in state 3, which is always the case for $\gamma_3 > \gamma_1$ by Eq. (A5). The second term, however, can be positive real, demonstrating that any overall gain is due to a coherence established by the pump. This coherence of the pump transition $\tilde{\rho}_{12}^{(0)}$ is coupled to the probe through the off-diagonal element $\tilde{\rho}_{13}^{1}$ [see Eq. (A1g)], demonstrating that coherence between the bare upper levels 1 and 3 in the presence of the pump is crucial for AWI [1]. When this gain mechanism overcomes the population contribution, AWI occurs. In order to maximize this term, we require $\delta = 0$ in Eq. (A4). This vanishing pump-probe detuning simultaneously maximizes the coherent pump's saturation of the probe polarization, as seen in the denominator of Eq. (A4), indicating a strong-pump phenomenon. On the other hand, if the probe is tuned to one of the Rabi sidebands ($\Delta' \approx \pm\chi$) for $\Delta = 0$, the gain term proportional to $\tilde{\rho}_{12}^{(0)}$ will be down by $\sim(\gamma_1 + \gamma_3)/2\chi$ with respect to the population term and will not contribute in the secular limit.

For $\delta = 0$ the small-signal gain coefficient per unit length is given by

$$G = -\frac{k' n d^2}{\epsilon_0 \hbar}\,\text{Im}\left(\frac{\tilde{\rho}_{23}^{(1)}}{(\chi')^*}\right) = \frac{k' n d^2}{\epsilon_0 \hbar}\frac{2\chi^4\gamma_1 r}{(\gamma_1 + \gamma_3)^2}\left[\frac{\gamma_1 + \gamma_3}{\gamma_3} \times \left(1 - \frac{\gamma_3}{\gamma_1}\right) - \left(\frac{\gamma_1}{2\chi}\right)^2\left(\frac{\gamma_3}{2\chi} + \frac{2}{\gamma_1 + \gamma_3}\right) + 1\right]\frac{1}{D_1 D_2}, \tag{A8}$$

where $k' = \Omega'/c$ is the probe wave vector, d is the 2-3 dipole matrix element, n is the atomic density,

$$D_1=\left(\frac{\gamma_3}{2}+\frac{2\chi^2}{\gamma_1+\gamma_3}\right)^2+\Delta^2,$$

$$D_2=r\left[\left(\frac{\gamma_1}{2}\right)^2+\Delta^2\right]+\beta\chi^2\frac{\gamma_1}{\gamma_3}. \tag{A9}$$

The gain coefficient is seen to be strongly peaked around the detuning $\Delta=\Delta'=0$.

In the limits $\chi\gg\gamma_1,\gamma_3$ and $\Delta=\Delta'=0$, assuming weak incoherent pumping, $r\ll\gamma_1,\gamma_3$, we find

$$\rho_{33}^{(0)}-\rho_{22}^{(0)}=\rho_{33}^{(0)}-\rho_{11}^{(0)}=\frac{r}{\gamma_3}\left(1-\frac{\gamma_3}{\gamma_1}\right)=\rho_{33}^{(0)}\left(1-\frac{\gamma_3}{\gamma_1}\right), \tag{A10}$$

$$\tilde{\rho}_{12}^{(0)}=\frac{2i\chi}{\gamma_1}(\rho_{11}^{(0)}-\rho_{22}^{(0)})=-\frac{i}{2}\frac{r}{\chi}, \tag{A11}$$

and

$$G=\frac{k'nd^2}{2\epsilon_0\hbar}\frac{r}{\gamma_3}\frac{\gamma_1}{\chi^2}\left[1-\left(\frac{\gamma_3}{\gamma_1}\right)^2+\frac{\gamma_3}{\gamma_1}\right] \tag{A12}$$

for the probe population inversion, the pump coherence, and the gain coefficient, respectively. The gain coefficient clearly shows that the population difference contributes as $1-(\gamma_3/\gamma_1)^2$, while the (γ_3/γ_1) term comes from the coherence contribution to the amplification.

The dressed picture is particularly easy to understand in this limit. The semiclassical dressed states are split in energy by $2\hbar\chi$, each having a FWHM of $\hbar\gamma_1/2$. Probe or vacuum radiation tuned between these states creates an atomic superposition, which can either enhance or detract from further transitions. The steady-state, dressed density-matrix elements are

$$\rho_{AA}^{(0)}=\rho_{BB}^{(0)}=\tfrac{1}{2}(\rho_{11}^{(0)}+\rho_{22}^{(0)}-\tilde{\rho}_{12}^{(0)}-\tilde{\rho}_{21}^{(0)})=\frac{r}{\gamma_1}, \tag{A13a}$$

$$\rho_{AB}^{(0)}=(\rho_{BA}^{(0)})^*=\tfrac{1}{2}(\tilde{\rho}_{21}^{(0)}-\rho_{11}^{(0)}+\rho_{22}^{(0)}-\tilde{\rho}_{12}^{(0)})=\frac{i}{2}\frac{r}{\chi}, \tag{A13b}$$

showing from Eq. (A4) that

$$\tilde{\rho}_{23}^{(1)}\sim-i(\chi')^*\left[\tfrac{1}{2}(\rho_{33}^{(0)}-\rho_{AA}^{(0)})+\tfrac{1}{2}(\rho_{33}^{(0)}-\rho_{BB}^{(0)})+\frac{i\chi\rho_{BA}^{(0)}}{\frac{\gamma_1+\gamma_3}{2}-i\delta}\right]\left[\frac{\chi^2}{\left(\frac{\gamma_1+\gamma_3}{2}\right)}\right]^{-1}. \tag{A14}$$

We now identify absorption with the lack of inversion in the dressed picture and amplification owing to a dressed coherence.

Above, the gain coefficient has been written in a form in Eq. (A12) that emphasizes its proportionality to (i) the steady-state population of level 3, r/γ_3, and (ii) the decay rate γ_1. This form suggests a perturbation path for an amplitude calculation that starts by incoherent pumping out of level 4 into level 3 and ends by decay back into level 4 from the dressed states. The gain coefficient is discussed in Secs. II and IV with respect to our amplitude approach.

APPENDIX B: FEYNMAN DIAGRAMS FOR AWI USING QUANTUM DRESSED STATES

The Feynman diagrams can be defined in a fully quantized dressed basis as well. Instead of trying to draw a picture of transitions between different dressed levels, the perturbation chains that were used in the amplitude calculation above provide the same information. From time-independent perturbation theory, we know how to construct the different amplitudes from these pathways [19,20]. Assuming a resonant pump field, the quantum dressed levels of the N-photon manifold are written as

$$|B,N,N',\{\mathbf{k}\}\rangle=\sqrt{\tfrac{1}{2}}(|1,N-1,N',\{\mathbf{k}\}\rangle+|2,N,N',\{\mathbf{k}\}\rangle), \tag{B1a}$$

$$|A,N,N',\{\mathbf{k}\}\rangle=\sqrt{\tfrac{1}{2}}(-|1,N-1,N',\{\mathbf{k}\}\rangle+|2,N,N',\{\mathbf{k}\}\rangle), \tag{B1b}$$

where $|1\rangle$ and $|2\rangle$ are the upper and lower atomic states, respectively, N and N' are the number of pump and probe photons, respectively, and $\{\mathbf{k}\}$ refers to the set of single photon states, $|\mathbf{k}_1,\lambda_1;\mathbf{k}_2,\lambda_2;\ldots\rangle\equiv|\{\mathbf{k}\}\rangle$, created by spontaneous emission, each with an energy $\hbar\Omega_{\mathbf{k}_i}$. The (complex) energies of the quantum dressed states in frequency units, taking $\omega_{2,0,0,\{0\}}=0$, are

$$\omega_{B,N,N',\{\mathbf{k}\}}=\chi-i\frac{\gamma_1}{4}+N\Omega+N'\Omega'+\sum_i\Omega_{\mathbf{k}_i}, \tag{B2a}$$

$$\omega_{A,N,N',\{\mathbf{k}\}}=-\chi-i\frac{\gamma_1}{4}+N\Omega+N'\Omega'+\sum_i\Omega_{\mathbf{k}_i}. \tag{B2b}$$

Levels 3 and 4 in the N-photon manifold have the energies

$$\omega_{3,N,N',\{\mathbf{k}\}}=\omega_3-i\frac{\gamma_3}{2}+N\Omega+N'\Omega'+\sum_i\Omega_{\mathbf{k}_i}, \tag{B3a}$$

$$\omega_{4,N,N',\{\mathbf{k}\}}=\omega_4+N\Omega+N'\Omega'+\sum_i\Omega_{\mathbf{k}_i}. \tag{B3b}$$

The Hilbert space, expanded in this dressed basis, leads to a Schrödinger equation consistent with the semiclassical amplitude equations of motion, Eqs. (15a)–(15f).

The initial state of the system is $|4,N,N',\{0\}\rangle$. Incoherent pumping at the time t_0 pumps the system into $|3,N,N',\{0\}\rangle$. We stress again that the incoherent pumping process, when accounted for formally, leads to the cross section,

$$\sigma=\frac{r}{F}\sum_{\{\mathbf{k}\}}|a_{4,N-1,n',\{\mathbf{k}\}}|^2, \tag{B4}$$

where F is now the quantum flux factor [18], and $n'=N'+1$ for probe amplification and $n'=N'-1$ for probe absorption. The perturbation chains for amplification are

$$|3,N,N',\{0\}\rangle \xrightarrow{\sqrt{1/2}(\chi')^*} |B,N,N'+1,\{0\}\rangle \xrightarrow{\sqrt{1/2}g_{41}^*} |4,N-1,N'+1,\mathbf{k}_1\rangle, \tag{B5a}$$

$$|3,N,N',\{0\}\rangle \xrightarrow{\sqrt{1/2}(\chi')^*} |A,N,N'+1,\{0\}\rangle \xrightarrow{-\sqrt{1/2}g_{41}^*} |4,N-1,N'+1,\mathbf{k}_1\rangle, \tag{B5b}$$

where paths (B5a) and (B5b) lead to $a^{(B)}_{4,N-1,N'+1,\mathbf{k}_1}$ and $a^{(A)}_{4,N-1,N'+1,\mathbf{k}_1}$, respectively. The condition for energy conservation of the scattering process is

$$E_{4,N-1,N'+1,\mathbf{k}_1}-E_{3,N,N',\{0\}}\approx\Omega_{\mathbf{k}_1}+\omega_4-\Omega+\Omega'-\omega_3=0, \tag{B6}$$

implying spontaneous emission at $\Omega_{\mathbf{k}_1}=\Omega-\omega_4=|\omega_{4'}|$ for the $\Delta'=0$, two-quantum amplification process, where $\omega_{4'}$ was defined for the semiclassical calculation by Eq. (19). (From this, we can identify the semiclassical state $|4'\rangle$ in the rotating frame of the pump with the coupled atom-field state $|4,N-1\rangle$. For example, see Refs. [19, 20].) The final transition energies,

$$E_{4,N-1,N'+1,\mathbf{k}_1}-E_{B,N,N'+1,\{0\}}\approx\Omega_{\mathbf{k}_1}-|\omega_{4'}|-\chi=0, \tag{B7a}$$

$$E_{4,N-1,N'+1,\mathbf{k}_1}-E_{A,N,N'+1,\{0\}}\approx\Omega_{\mathbf{k}_1}-|\omega_{4'}|+\chi=0, \tag{B7b}$$

give the stepwise resonances. Finally, we notice that the full scattering process, including incoherent pumping, started in state 4 in the N-photon manifold of the pump and ends in state 4 in the $(N-1)$-photon manifold with a spontaneous photon emitted; both states are asymptotically stable in the weak incoherent pumping limit, $r\ll\gamma_3,\gamma_1$.

The four Feynman diagrams for absorption are

$$|3,N,N',\{0\}\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |B,N,N',\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}\chi'} |3,N,N'-1,\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |B,N,N'-1,\mathbf{k}_2,\mathbf{k}_3\rangle \xrightarrow{\sqrt{1/2}g_{4}^*} |4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4\rangle, \tag{B8a}$$

$$|3,N,N',\{0\}\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |B,N,N',\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}\chi'} |3,N,N'-1,\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |A,N,N'-1,\mathbf{k}_2,\mathbf{k}_3\rangle \xrightarrow{-\sqrt{1/2}g_{41}^*} |4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4\rangle, \tag{B8b}$$

$$|3,N,N',\{0\}\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |A,N,N',\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}\chi'} |3,N,N'-1,\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |B,N,N'-1,\mathbf{k}_2,\mathbf{k}_3\rangle \xrightarrow{\sqrt{1/2}g_{41}^*} |4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4\rangle, \tag{B8c}$$

$$|3,N,N',\{0\}\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |A,N,N',\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}\chi'} |3,N,N'-1,\mathbf{k}_2\rangle \xrightarrow{\sqrt{1/2}g_{23}^*} |A,N,N'-1,\mathbf{k}_2,\mathbf{k}_3\rangle \xrightarrow{-\sqrt{1/2}g_{41}^*} |4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4\rangle, \tag{B8d}$$

corresponding to $a^{(BB)}_{4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, $a^{(BA)}_{4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, $a^{(AB)}_{4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, and $a^{(AA)}_{4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}$, respectively. Energy conservation of the absorption diagrams requires

$$E_{4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}-E_{3,N,N',\{0\}}\approx\omega_4-\Omega-\Omega'+\Omega_{\mathbf{k}_2}+\Omega_{\mathbf{k}_3}+\Omega_{\mathbf{k}_4}-\omega_3\approx 0, \tag{B9}$$

just as the semiclassical condition in Eq. (46). Furthermore, the three intermediate resonances to the final state are evident from the three final transitions in each of the four pathways.

The final state of the amplification diagrams was shown to be $|4,N-1,N'+1,\mathbf{k}_1\rangle$, distinguishing it clearly from that of the absorption process, $|4,N-1,N'+1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4\rangle$. As a result, these two states do not interfere, allowing us to write the overall quantum cross section for amplification as $\sigma_{\mathrm{amp}}-\sigma_{\mathrm{abs}}$. The definition of these fully quantized Feynman diagrams is satisfying from a fundamental point of view. Yet, the physics of AWI and the conclusions drawn from the calculation remain the same, except in the truly quantum limit, where the number of probe photons N' becomes small. The ramifications of a probe field in this limit will not be discussed here.

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[14] The term *stepwise* refers to probe scattering processes which have resonances for spontaneous emission at the dressed transition frequencies of the system, as shown schematically for the amplification diagrams in Fig. 2(b). On the other hand, the term *two-quantum* denotes a resonance for spontaneous emission at a frequency which is the difference between a two-quantum transition frequency, such as $\omega_{34'}$, and a harmonic perturbation frequency, such as the probe-laser frequency Ω'. The two-quantum process for the amplification diagrams is portrayed in Fig. 2(a), which suggests that quantum interference will occur between the different two-quantum channels. These terms give a physical interpretation to the different multiphoton resonances of the Feynman diagrams but convey a different meaning in the amplitude approach than related terms used to describe density-matrix (Liouville) perturbation chains.

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[18] The quantum flux factor for a scattering process is $F = N'c/V$, where N' is the number of photons in the probe beam, c is the speed of light, and V is the quantization volume. The semiclassical flux factor is derived by equating the classical intensity of the probe, $\frac{1}{2}c\epsilon_0|E'|^2$, with the quantum energy flux density, $(N'c/V)\hbar\Omega'$, and using the definition of the Rabi frequency, $\chi' = -dE'/2\hbar$, to eliminate the field amplitude E'.

[19] For example, see J. J. Sakurai, *Modern Quantum Mechanics*, revised edition (Addison-Wesley, Reading, MA, 1994), p. 292. Sakurai shows the results of time-independent perturbation theory to second order, which is all that we need for the amplification process. In our notation for amplification, we are interested in the amplitude to be in state 4 with one photon in the quantum field for an initial state $|3,\{0\}\rangle$: $a_{4,k_1} = \langle\Psi|3,\{0\}\rangle$, where $|\Psi\rangle$ is the (time-independent part of the) second-order perturbed wave function of the state $|4',\mathbf{k}_1\rangle$ in the semiclassical dressed basis. In the fully quantized dressed basis the equivalent amplitude is $a_{4,N-1,N'+1,\mathbf{k}_1} = \langle\Psi|3,N,N',\{0\}\rangle$, where $|\Psi\rangle$ is the stationary perturbed wave function of the state $|4,N-1,N'+1,\mathbf{k}_1\rangle$, as defined in Appendix B. The equivalence of these two approaches to scattering problems is widely known: these approaches are time-dependent harmonic perturbation theory on the one hand and time-independent perturbation theory when the quantum statistics of the driving fields are unimportant on the other.

[20] For the absorption calculation we are interested in the amplitude to be in state 4 with three distinct photons in the quantum field, $a_{4,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} = \langle\Psi|3,\{0\}\rangle$, where $|\Psi\rangle$ is the fourth-order perturbed wave function of the state $|4',\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4\rangle$ in the semiclassical dressed basis. The equivalent amplitude in the quantum dressed basis is $a_{4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} = \langle\Psi|3,N,N',\{0\}\rangle$, where $|\Psi\rangle$ is the stationary perturbed wave function of the state $|4,N-1,N'-1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4\rangle$, as defined in Appendix B.

[21] By analogy with formal scattering theory, where $\sigma = (1/F)\Sigma_{\{k\}}|\langle S\rangle|^2/t$, we can identify $a_{4,\{\mathbf{k}\}}$ in its time-independent form as proportional to the S-matrix element to go from state $|3,\{0\}\rangle$ to state $|4',\{\mathbf{k}\}\rangle$ after state 3 is incoherently pumped, $|\langle 4',\{\mathbf{k}\}|S|3,\{0\}\rangle|^2 = rt|a_{4,\{\mathbf{k}\}}|^2$.