Resonant processes in a frozen gas

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Abstract

We present a theory of resonant processes in a frozen gas of atoms interacting via dipole-dipole potentials that vary as r^{-3}, where r is the interatomic separation. We supply an exact result for a single atom in a given state interacting resonantly with a random gas of atoms in a different state. The time development of the transition process is calculated both on- and off-resonance, and the linewidth with respect to detuning is obtained as a function of time t. We introduce a random spin Hamiltonian to model a dense system of resonators and show how it reduces to the previous model in the limit of a sparse system. We derive approximate equations for the average effective spin, and we use them to model the behavior seen in the experiments of Anderson *et al.* [1] and Lowell *et al.* [3,4]. The approach to equilibrium is found to be proportional to $\exp(-\sqrt{\gamma_{eq} t}\ )$, where the constant γ_{eq} is explicitly related to the system's parameters.

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I. INTRODUCTION

Frozen gases are a new and in some ways ideal laboratory to test our understanding of quantum theory in a complex system. With present technology one can manipulate and detect electronic processes with an extraordinary selectivity and precision; furthermore, the translational temperature of the gas can be lowered to the point where it can be ignored when discussing electronic processes. Frozen Rydberg gases have the added advantages that their states are well understood and many processes occur on a microsecond time scale, easily allowing for time-resolved spectroscopy. Pioneering experiments on resonant processes in these gases have been carried out by Anderson *et al.* [1] and Mourachko *et al.* [2]. The present work was motivated by the desire to understand those experiments and the subsequent work of Lowell *et al.* [3,4] highlighting the dynamic aspects of these resonant, many particle systems. The measurements are of mixtures of ^{85}Rb atoms initially prepared in the $23s$ and $33s$ states (henceforth to be called the s and s' states, respectively). There are initially N atoms in the s state and N' atoms in the s' state. The transition $ss' \to pp'$ is monitored, where p refers to the $24p$ state and p' refers to the $34p$ state, at and near resonance, where $\epsilon_s' + \epsilon_s = \epsilon_p' + \epsilon_p$. Some of the experimental features we wish to understand are the rapid rise followed by slow approach to saturation of the signal and the width of the lineshape (in the detuning $\Delta = \epsilon_p' + \epsilon_p - \epsilon_s' - \epsilon_s$) as a function of time.

Each individual interaction leads to a (coherent) oscillatory behavior, but as we will see below, averaging the $ss' \to pp'$ interaction over the random positions of the atoms greatly smooths out the signal. The effective "incoherence" brought about by the $sp \to ps$ process (the "walking away" discussed by Mourachko *et al.* [2]) completes the smoothing out of the on-resonance signal but has less effect on the off-resonance signal. (The $s'p' \to p's'$ process is also present, but is smaller than $sp \to ps$ by a factor of 16 in this case and can therefore be neglected to the first approximation.)

It was surmised by Anderson *et al.* [1] that the linewidth must be of the order of the average interaction energy. This interaction splits the ss'-pp' degeneracy and the subsequent migration of the p state to other atoms broadens the energy of this "elementary excitation" into a band, as appropriate to an amorphous solid. Cluster calculations were performed to illustrate this band formation [3], but averaging over atom positions was done by order-of-magnitude arguments only. Mourachko *et al.* [2] pointed out, in connection with their experiments on a different system, that the "walking away" of the p excitation from its original location should be regarded as a diffusion process, and dynamical equations for the resonance transition in the presence of this diffusion were written down [5].

The present work builds on these earlier insights, but goes considerably further in the process of accurately averaging over atomic positions. It turns out that this averaging (effectively, a phase averaging) itself produces an $\exp(-\sqrt{\gamma_{eq} t})$ dependence at large times, characteristic of a diffusive process. A report of the newer experimental data by Lowell *et al.* in comparison with the results of the present theory is being submitted separately [4]. In this paper we give a full account of the general theory, and we discuss in detail in Section II a particular case that cleanly shows the effects of randomness and phase-averaging. In this simple case, the averaging process can be carried out exactly. For the practically important r^{-3} interaction potential, one can proceed in a mathematically elegant way and obtain closed formulas that can be evaluated by a computer package, or in some cases by hand. It appears to be a fortunate coincidence that the r^{-3} potential accurately describes the dipole-dipole interactions that are so prevalent in nature. Some aspects of this approach are easily extended to more general interactions; however, the r^{-3} dependence has special properties which, when coupled with the assumption of a random distribution of atoms, lead to relatively simple results.

Although this work was stimulated by experiments on Rydberg atoms, it is generally applicable to resonances induced by dipole-dipole interactions. These may be common in highly excited gases and in molecular systems. Frozen, resonant gases may even exist in interstellar clouds.

We develop the theory in three stages. First, in Section II, we discuss a single s' atom interacting resonantly with a surrounding gas of s atoms, without any possibility for the s excitation to "walk away" through an $sp \to ps$ process. When the result is averaged over the positions of the s atoms, it corresponds to a "sparse" system of s' atoms, i.e. a system where $N' \ll N$. We do not know of an existing experiment to which this treatment applies, so this section can be viewed as a theoretical prediction of the outcome of a possible experiment. Our purpose here is also one of exposition: this example allows us to introduce in the simplest context some of the mathematical techniques used throughout the paper. In Section III, we complicate things somewhat by allowing the s state to decay away exponentially (or more generally, but explicit formulas are given for the exponential decay). If the s state decays diffusively, our treatment here is related to that of Mourachko *et al.* [2], except for the introduction of mathematically exact phase averaging. Here, too, the treatment applies only to the case where $N' \ll N$. Finally, in Section IV we introduce an effective spin Hamiltonian which fully models the atomic system for all values of N and N', as well as for any strength of the resonant process ($ss' \to pp'$) and of the mixing processes ($sp \to ps$ and $s'p' \to p's'$). This system is relevant to the experiments of Anderson *et al.* [1] and Lowell *et al.* [3,4]. We show its equivalence to the previous results in the sparse limit ($N' \ll N$) and discuss briefly the approximate solution in the non-sparse case. The results in this section are summarized elsewhere [4], but the derivation is presented here for the first time, as are additional formulas. The limitations of the present theory and some of the many possible extensions are discussed briefly in Section V.

II. SPARSE $SS' \to PP'$ PROCESSES

We consider one atom, initially in the state s', in interaction with a gas of s atoms through an $ss' \to pp'$ process. Like Mourachko *et al.* [2], we describe the system by the equations:

$$i\ \dot{a}_0 = \Delta\ a_0 + \sum_k V_k\ c_k, \tag{1a}$$

$$i\ \dot{c}_k = V_k\ a_0. \tag{1b}$$

Here $a_0(t)$ is the amplitude of the state in which the atom at the origin is in state s' and all other atoms are in state s, while $c_k(t)$ (with k running from 1 to N) is the amplitude of the state in which the atom at the origin is in state p' and the atom at $\mathbf{r}_k$ is in state p, while all the others remain in state s. V_k is the interaction potential and $\Delta = \epsilon_p' + \epsilon_p - \epsilon_s' - \epsilon_s$ is the detuning from resonance. We will assume that V_k is of the dipole-dipole form

$$V_k = \frac{\mu\mu'}{r_k^3}. \tag{2}$$

The atoms are sufficiently cold that during the time scale of interest they move only a very small fraction of their separation, and therefore V_k can be taken to be independent of time. As the temperature increases one approaches the opposite limit, where binary collisions control the resonant process [8]. There must of course be a gradual transition, which is beyond the scope of this paper.

The set of differential equations (1) can easily be solved with the initial condition $a_0(t = 0) = 1$ and $c_k(t = 0) = 0$ for all k, yielding

$$c_k(t) = -\frac{2i\ V_k\ e^{-i\Delta t/2}\ \sin\left(\sqrt{\Delta^2 + 4\mathcal{V}^2}\, t/2\right)}{\sqrt{\Delta^2 + 4\mathcal{V}^2}}\ , \tag{3}$$

with

$$\mathcal{V}^2 = \sum_{k=1}^{N} V_k^2\,. \tag{4}$$

Eq. (1b) then gives $a_0(t)$. The experiment monitors an optical transition from the p' state (or, equivalently, from one of the p states), which is proportional to

$$\begin{aligned} S(t) &= 1 - |a_0(t)|^2 = \sum_k |c_k(t)|^2 \\ &= \frac{4\mathcal{V}^2 \sin^2\left(\sqrt{\Delta^2 + 4\mathcal{V}^2}\, t/2\right)}{\Delta^2 + 4\mathcal{V}^2}. \end{aligned} \tag{5}$$

We see that the signal $S(t)$ exhibits Rabi oscillations.

We apply this result to a system of N' atoms, initially in state s', randomly dispersed among a much larger number, N, of s atoms. In this sparse limit, the signal is proportional to N' times the sample average of $S(t)$, and the sample average is equivalent to an ensemble average over the atomic positions $\mathbf{r}_k$ that are hidden in $\mathcal{V}^2$ (see Eq. (7) below). In general, averaging is more easily done on the Laplace transform of the signal, which in this case is

$$\begin{aligned} \tilde{S}(\alpha) &= \int_0^\infty e^{-\alpha t}\, S(t)\, dt \\ &= \frac{1}{\alpha}\frac{2\mathcal{V}^2}{\alpha^2 + \Delta^2 + 4\mathcal{V}^2} \\ &= \frac{1}{2\alpha}\left(1 - \frac{\alpha^2 + \Delta^2}{\alpha^2 + \Delta^2 + 4\mathcal{V}^2}\right) \end{aligned} \tag{6}$$

Later in this paper, more complex models are solved directly by Laplace (or Fourier) transforms; thus Eq. (6) is useful for comparison with these more general results and approximations.

A. Averaging over atom positions

To compute ensemble averages, we use the following result, valid in the limit $N \gg 1$, for a random distribution of the variables $\mathbf{r}_k$:

$$\begin{aligned}
\left(e^{-\beta \mathcal{V}^2}\right)_{av} &= \frac{1}{\Omega^N} \int d^3r_1 \ldots d^3r_N \exp\left[-\beta \sum_{k=1}^{N} V^2(r_k)\right] \\
&= \left\{\frac{1}{\Omega} \int d^3r \exp\left[-\beta V^2(r)\right]\right\}^N \\
&= \left\{1 - \frac{1}{\Omega} \int d^3r \left[1 - e^{-\beta V^2(r)}\right]\right\}^N \\
&\longrightarrow \exp\left\{-\frac{N}{\Omega} \int d^3r \left[1 - e^{-\beta V^2(r)}\right]\right\},
\end{aligned} \tag{7}$$

where Ω is the volume of the gas in the trap. In particular, for the dipolar interaction $V(r) = \mu\mu'/r^3$, we have

$$\int d^3r \left[1 - e^{-\beta(\mu\mu')^2/r^6}\right] = \frac{4\pi^{3/2}}{3} \mu\mu' \sqrt{\beta}, \tag{8}$$

leading to

$$\left(e^{-\beta \mathcal{V}^2}\right)_{av} = e^{-v\sqrt{\beta}}, \tag{9}$$

where

$$v = \frac{4\pi^{3/2}}{3} \frac{N}{\Omega} \mu\,\mu'. \tag{10}$$

For the typical densities in the experiments of Anderson *et al.* [1] and Lowell *et al.* [3,4], v is on the order of MHz. (In Appendix A, we repeat the arguments above with an interaction that includes the angular dependence due to the relative positions of the dipole moments and assumes that all of the dipole moments point in the same direction.) Since v has the units of an energy, it must be of order $\mu\mu' N/\Omega$ on dimensional grounds. It is still remarkable, however, that Eq. (10) gives simply and exactly the quantity v that will enter in all the averaged quantities in this section.

Using Eq. (9), we can evaluate the average of any function $F(\mathcal{V}^2)$ provided that it can be represented as

$$F(\mathcal{V}^2) = \int_0^\infty d\beta\ \tilde{F}(\beta)\ e^{-\beta \mathcal{V}^2}. \tag{11}$$

Further, we can use the following identity

$$e^{-v\sqrt{\beta}} = \frac{1}{\sqrt{\pi}} \int_0^\infty dy\ \exp\left\{-\frac{y^2}{4} - \frac{v^2\beta}{y^2}\right\}, \tag{12}$$

so that

$$
\begin{aligned}
F(\mathcal{V}^2)_{av} &= \int_0^\infty d\beta\, \tilde{F}(\beta)\, e^{-v\sqrt{\beta}} \\
&= \frac{1}{\sqrt{\pi}} \int_0^\infty dy\, e^{-y^2/4} \int_0^\infty d\beta \tilde{F}(\beta) e^{-v^2\beta/y^2} \\
&= \frac{1}{\sqrt{\pi}} \int_0^\infty dy\, e^{-y^2/4} F\left(\frac{v^2}{y^2}\right) .
\end{aligned} \tag{13}
$$

This last set of relations implies that we can average a function $F(\mathcal{V}^2)$ over the positions of the interacting dipoles (atoms in our case) by replacing $\mathcal{V}^2$ with v^2/y^2 and integrating over the kernel $\exp\left(-y^2/4\right)/\sqrt{\pi}$. This trick is not always useful, as it can lead to highly oscillatory integrals. It is, however, convenient in determining the lineshape in the saturation ($t \to \infty$) limit (as shown below).

B. The averaged signal and lineshape

Starting from Eq. (6) in the form

$$
\tilde{S}(\alpha) = \frac{1}{2\alpha} - \frac{\alpha^2 + \Delta^2}{2\alpha} \int_0^\infty d\beta \; e^{-\beta(\alpha^2+\Delta^2+4\mathcal{V}^2)}, \tag{14}
$$

and using Eq. (9), we obtain

$$
\begin{aligned}
\tilde{S}(\alpha)_{av} &= \frac{1}{2\alpha} - \frac{A^2}{2\alpha} \int_0^\infty d\beta \; e^{-\beta A^2 - 2v\sqrt{\beta}} \\
&= \frac{\sqrt{\pi}}{2} \frac{v}{\alpha A} \; \exp\left(\frac{v^2}{A^2}\right) \operatorname{erfc}\left(\frac{v}{A}\right),
\end{aligned} \tag{15}
$$

where $A^2 = \alpha^2 + \Delta^2$. Expanding in v/A and performing the inverse Laplace transform leads to

$$
S(t)_{av} = \frac{\sqrt{\pi}}{2} vt \sum_{n=0}^{\infty} \frac{(-vt)^n}{\Gamma\left(n+2\right)\Gamma\left(\frac{n+2}{2}\right)} \; {}_1F_2\left[\frac{n+1}{2}; \frac{n+2}{2}, \frac{n+3}{2}; -\frac{\Delta^2 t^2}{4}\right], \tag{16}
$$

where ${}_nF_m(a_1, \ldots, a_n; b_1, \ldots, b_m; z)$ is a generalized hypergeometric function. It is known that ${}_1F_2$ is related to the integral of a power multiplied by a Bessel function. Expanding ${}_1F_2$ yields the double series

$$
S(t)_{av} = \frac{\sqrt{\pi}}{2} vt \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \frac{(-1)^{n+m} v^n \Delta^{2m} t^{n+2m}}{4^m n! m! \Gamma\left(\frac{n+2m+2}{2}\right)(n+2m+1)}, \tag{17}
$$

which can also be obtained directly by expanding Eq. (15) in $1/\alpha$ and then taking the inverse Laplace transform. The double series can be rearranged to obtain the following expansion in the detuning:

$$
S(t)_{av} = \sqrt{\frac{vt}{2}} \int_0^\infty \frac{dp}{\sqrt{p}} e^{-p^2} \sum_{m=0}^{\infty} \frac{1}{m!} \left(-\frac{\Delta^2 pt}{2v}\right)^m J_{2m+1}\left(\sqrt{8vpt}\right). \tag{18}
$$

The first term in this expansion, the on-resonance signal, can be written as

$$S(t)_{av}\Big|_{\Delta=0} = \left[\frac{\sqrt{\pi}}{2} vt \; {}_0F_2\left(1, \frac{3}{2}; \frac{v^2t^2}{4}\right) + \frac{1}{2} - \frac{1}{2} \; {}_0F_2\left(\frac{1}{2}, \frac{1}{2}; \frac{v^2t^2}{4}\right)\right], \tag{19}$$

which can also be obtained by setting Δ equal to zero in Eq. (16) and summing the resulting series. Plots of $S(t)_{av}$ for several values of Δ are shown in Fig. 1. Notice that the initial slope of the signal is independent of Δ. This feature translates into resonance widths that vary as $1/t$ for small t - the so-called transform broadening discussed by Thomson *et al.* [6,7]. With v on the order of MHz, this initial rise occurs in a fraction of a μs. Another point of interest is that the averaging is much more effective at smoothing out the oscillations for $\Delta = 0$ than it is for $\Delta \neq 0$.

We can construct a measure of the width from the first two terms in the Δ-expansion, Eq. (18). If

$$S(t)_{av} = S_0\left(t\right) + \Delta^2 S_1\left(t\right) + \mathcal{O}\left(\Delta^4\right), \tag{20}$$

and we define

$$w = \sqrt{-\frac{S_0\left(t\right)}{S_1\left(t\right)}}, \tag{21}$$

then $2w$ would be the FWHM if the lineshape were Lorentzian. The quantity w/v is plotted in Fig. 2. For small vt, we have explicitly $w = \sqrt{12}/t$. While the $1/t$ behavior follows from the transform broadening argument, or even simply from dimensional analysis, the coefficient $\sqrt{12}$ is a prediction of the detailed theory.

The lineshape at saturation ($t \to \infty$) can be obtained using the relation in Eq. (13), which yields

$$S(t)_{av} = \frac{1}{\sqrt{\pi}} \int_0^\infty dy \; e^{-y^2} \frac{\left[1 - \cos\left(\sqrt{\Delta^2 + \frac{v^2}{y^2}}\; t\right)\right]}{1 + \frac{\Delta^2}{v^2} y^2}. \tag{22}$$

For large times the cosine term above averages to zero, giving

$$S(t \to \infty)_{av} = \frac{\sqrt{\pi}}{2} \frac{v}{\Delta} \exp\left(\frac{v^2}{\Delta^2}\right) \operatorname{erfc}\left(\frac{v}{\Delta}\right), \tag{23}$$

which can also be extracted from the small α behavior of Eq. (15). This lineshape is plotted in Fig. 3. The FWHM is approximately $4.6v$. Also plotted is a Lorentzian lineshape with the same height and FWHM. Notice that this lineshape is sharper than the Lorentzian for small Δ and falls off more slowly for large Δ.

III. SPARSE $SS' \to PP'$ IN A BATH OF $SP \to PS$

We again consider one atom, initially in state s', in interaction with a gas of s atoms via the $ss' \to pp'$ process, but we now allow for $sp \to ps$ processes by modifying Eqs. (1) to

$$i\ \dot{a}_0 = \Delta\ a_0 + \sum_k V_k\ c_k, \tag{24a}$$

$$i\ \dot{c}_k = V_k\ a_0 + \sum_l U_{kl} c_l\,. \tag{24b}$$

where $U_{kl} = \mu^2/r_{kl}^3$ and the other symbols have the same meaning as in Eqs. (1). Although Eqs. (24) are exact, they are difficult to solve exactly. We therefore introduce the equation

$$i\ \dot{c}_k = V_k\ a_0 - i\gamma c_k\,, \tag{25}$$

in place of Eq. (24b), where γ is an effective inverse lifetime. In a crude way, the $-i\gamma c_k$ term in Eq. (25) describes the "walking away" of the p states from the neighborhood of the resonating atom at the origin. One can see from Eq. (24b) that the model of Section II is recovered in the limit $\mu \ll \mu'$.

When Eq. (1b) of the last section is replaced with (25), the solution of the new system can be obtained by replacing Δ with $\Delta + i\gamma$ in Eq. (3) and in the corresponding equation for $a_0(t)$. Then

$$\begin{aligned} L(t) &\equiv \sum_{k=1}^{N} \left|c_k\,(t)\right|^2 \\ &= \frac{2\mathcal{V}^2\ e^{-\gamma t}\ \left[\cosh\left(yt\right) - \cos\left(xt\right)\right]}{x^2 + y^2}, \end{aligned} \tag{26}$$

where x and y are real numbers that satisfy

$$(x + iy)^2 = (\Delta + i\gamma)^2 + 4\mathcal{V}^2. \tag{27}$$

At resonance, we have

$$(x, y) = \begin{cases} \left(\sqrt{4\mathcal{V}^2 - \gamma^2}, 0\right) & \text{if } \gamma < 2\mathcal{V}, \\ \left(0, \sqrt{\gamma^2 - 4\mathcal{V}^2}\right) & \text{if } \gamma > 2\mathcal{V}. \end{cases} \tag{28}$$

Note that $L\,(t)$ decays more slowly in the latter case. Hence the large-t limit of $L\,(t)_{av}$ will be dominated by small $\mathcal{V}$. This remains true off-resonance as well. The small-$\mathcal{V}$ expansion for y is

$$y = \gamma - \frac{2\mathcal{V}^2\gamma}{\Delta^2 + \gamma^2} \tag{29}$$

Thus, we can conclude that for the leading behavior at large times,

$$\lim_{t\to\infty} L\,(t) \propto \exp\left[-\frac{2\mathcal{V}^2\gamma}{\Delta^2 + \gamma^2} t\right]. \tag{30}$$

Using Eq. (9), we obtain an asymptotic behavior of the average

$$\lim_{t\to\infty} L\left(t\right)_{av} \propto \exp\left(-\sqrt{\gamma_{eq}t}\right), \tag{31}$$

where

$$\gamma_{eq} = \frac{2v^2\gamma}{\Delta^2 + \gamma^2}. \tag{32}$$

Let us now examine the full time dependence. We begin with the Laplace transform of $L(t)$ which is

$$\tilde{L}(\alpha) = \frac{2s\mathcal{V}^2}{s^4 + \left(x^2 - y^2\right)s^2 - x^2y^2}, \tag{33}$$

where $s = \alpha + \gamma$. Substituting x and y from Eq. (27) gives

$$\tilde{L}(\alpha) = \frac{2s\mathcal{V}^2}{s^4 + \left(\Delta^2 - \gamma^2 + 4\mathcal{V}^2\right)s^2 - \Delta^2\gamma^2}. \tag{34}$$

The signal is still proportional to $N'S(t)$, with $S(t) = 1 - |a_0(t)|^2$. It is no longer true that $1 - |a_0|^2 = \sum_k |c_k|^2$, because $\sum_k |c_k|^2$ is not the total probability of finding a p state, but only the probability of finding a p state that has not "walked away". However, the relation

$$\frac{d}{dt}\left(|a_0|^2 + \sum_k |c_k|^2\right) = -2\gamma\sum_k |c_k|^2, \tag{35}$$

with the usual initial conditions, enables us to obtain

$$S(t) = L(t) + 2\gamma\int_0^t L(t')\,dt', \tag{36}$$

and

$$\tilde{S}(\alpha) = \left(1 + \frac{2\gamma}{\alpha}\right)\tilde{L}(\alpha). \tag{37}$$

For $\gamma > 0$, $S(t)$ rises from 0 to 1 (or more accurately to $(1+ N'/N)^{-1}$, as seen in the next section), while for $\gamma = 0$ it saturates at $2\mathcal{V}^2/\left(\Delta^2 + 4\mathcal{V}^2\right)$. What happens for small γ is a quick rise (on the average time scale of v^{-1}) to $2\mathcal{V}^2/\left(\Delta^2 + 4\mathcal{V}^2\right)$, followed by a slow rise to 1.

Averaging these equations is done as in Section II. For instance, averaging $L\left(\alpha\right)$ yields

$$\widetilde{L}\left(\alpha\right)_{av} = \frac{\sqrt{\pi}}{2}\frac{v}{sB}\,\exp\left(\frac{v^2}{B^2}\right)\mathrm{erfc}\left(\frac{v}{B}\right), \tag{38}$$

where

$$B^2 = \left(s^2 + \Delta^2\right)\left(s^2 - \gamma^2\right)/s^2, \tag{39}$$

which is the analog of Eq. (15). The inverse Laplace transform of this expression is rather complicated, but it simplifies immensely at resonance, becoming

$$L(t)_{av} = \frac{\sqrt{\pi}}{2} v \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \left(\frac{2v^2}{\gamma}\right)^{n/2} e^{-\gamma t} \int_0^t d\tau \, \tau^{n/2} I_{n/2}(\gamma\tau) \,. \tag{40}$$

This result can also be expressed in terms of generalized hypergeometrics, and in that form appears similar to Eq. (16). One can use the asymptotic behavior of the Bessel function in Eq. (40) to confirm the large-t dependence argued earlier in Eq. (31). An expansion in inverse powers of α, leading to an expression similar to Eq. (17), is easily obtained by computer. Fig. 4 shows the average signal $S(t)_{av}$ for $\gamma = v$, and Δ/v having the values 0, 1, and 2. One can see that the approach to one becomes slower as the detuning is increased, as predicted by Eqs. (31) and (32). This behavior is the same as that found by Mourachko *et al.* [2] under the assumption that the time development of the c_k is governed by a diffusion equation. We see that in the present approach it arises simply from phase averaging. The result for the on-resonance signal is shown in Fig. 5. As one would expect, for small values of γ the small-t features of Fig. 5 resemble those in Fig. 1 of Section II, where the $sp \to ps$ process is neglected entirely.

IV. INTERACTING RESONANT PROCESS

We now consider N atoms at positions $\mathbf{r}_k$ which are initially in state s and N' atoms at positions $\mathbf{r}_{k'}$ which are initially in state s'. We represent the s and p states at $\mathbf{r}_k$ with the down and up states of an effective spin σ_k, and similarly represent the s' and p' states at $\mathbf{r}_{k'}$ with a spin $\sigma_{k'}$. The Hamiltonian is then

$$\begin{aligned} &\sum_k \left[\varepsilon_s + (\varepsilon_p - \varepsilon_s)\sigma_k^+\sigma_k^-\right] + \sum_{k'} \left[\varepsilon_s' + (\varepsilon_p' - \varepsilon_s')\sigma_{k'}^+\sigma_{k'}^-\right] \\ &+ \sum_{kk'} V_{kk'} \left[\sigma_k^+\sigma_{k'}^+ + \sigma_k^-\sigma_{k'}^-\right] + \sum_{k,l\neq k} U_{kl}\sigma_k^+\sigma_l^- , \end{aligned} \tag{41}$$

where

$$V_{kk'} = \frac{\mu\mu'}{\left|\mathbf{r}_k - \mathbf{r}_{k'}\right|^3} \quad \text{and} \quad U_{kl} = \frac{\mu^2}{\left|\mathbf{r}_k - \mathbf{r}_l\right|^3}. \tag{42}$$

For the experiments under consideration $\mu \approx 4\mu'$, and the *sp*-*ps* coupling μ^2, which leads to "spin diffusion", is larger than the ss'-pp' coupling $\mu\mu'$, which is responsible for the resonant energy transfer.

The strategy is to write down the evolution equations for the spin variables, take the expectation values over the initial state $|i\rangle$ (which consists of all spins down), discard the expectation values of quantities that fluctuate incoherently, solve the resulting equations, and finally average over the atomic coordinates $\mathbf{r}_k$ and $\mathbf{r}_{k'}$. The experimental signal is proportional to the number of p' states, $\sum_{k'} n_{k'}$, where

$$n_{k'} = \frac{1}{2}\left(1 + \langle\sigma_{k'}^z\rangle\right). \tag{43}$$

It should be noted that $|i\rangle$ is not the ground state, nor does the system relax to the ground state, because it is essentially decoupled from other degrees of freedom for the duration of the experiment.

We start with basic equations of motion such as

$$i\frac{d}{dt}\sigma_{k'}^{+} = \Delta\sigma_{k'}^{+} + \sigma_{k'}^{z}\sum_{k} V_{k'k}\sigma_{k}^{-}, \tag{44}$$

$$i\frac{d}{dt}\sigma_{k}^{+} = \sigma_{k}^{z}\sum_{k'} V_{kk'}\sigma_{k'}^{-} + \sigma_{k}^{z}\sum_{l\neq k} U_{kl}\sigma_{l}^{+}, \tag{45}$$

$$i\frac{d}{dt}\sigma_{k'}^{z} = 2\sum_{k} V_{k'k}\left(\sigma_{k}^{+}\sigma_{k'}^{+} - \sigma_{k}^{-}\sigma_{k'}^{-}\right), \tag{46}$$

where Δ is the detuning from resonance [9], and obtain

$$\begin{aligned}\frac{1}{2}\frac{d^2}{dt^2}\sigma_{k'}^{z} = &-\sum_{kl'} V_{k'k}V_{kl'}\sigma_{k}^{z}\left(\sigma_{k'}^{-}\sigma_{l'}^{+} + \sigma_{k'}^{+}\sigma_{l'}^{-}\right)\\ &-\sum_{kl} V_{k'k}V_{lk'}\sigma_{k'}^{z}\left(\sigma_{k}^{-}\sigma_{l}^{+} + \sigma_{k}^{+}\sigma_{l}^{-}\right)\\ &-\sum_{k,l\neq k} V_{k'k}U_{kl}\sigma_{k}^{z}\left(\sigma_{k'}^{+}\sigma_{l}^{+} + \sigma_{k'}^{-}\sigma_{l}^{-}\right)\\ &-\Delta\sum_{k} V_{k'k}\left(\sigma_{k'}^{+}\sigma_{k}^{+} + \sigma_{k'}^{-}\sigma_{k}^{-}\right).\end{aligned} \tag{47}$$

We reduce the expectation value of this equation as follows. In the first line, we assume that $\left\langle\sigma_{k}^{z}\left(\sigma_{k'}^{-}\sigma_{l'}^{+} + \sigma_{k'}^{+}\sigma_{l'}^{-}\right)\right\rangle$ is negligible unless $k' = l'$, in which case it reduces to $\langle\sigma_{k}^{z}\rangle$. Similarly, $\left\langle\sigma_{k'}^{z}\left(\sigma_{k}^{-}\sigma_{l}^{+} + \sigma_{k}^{+}\sigma_{l}^{-}\right)\right\rangle$ is replaced by $\langle\sigma_{k'}^{z}\rangle\,\delta_{kl}$. The third line contains only incoherent terms, which we treat by assuming that they lead to a Lorentzian broadening by letting

$$\sigma_{k}^{z}\sum_{l\neq k} U_{kl}\sigma_{l}^{+} \to i\gamma\sigma_{k}^{+} \tag{48}$$

when computing these terms. With this crude approximation and Eq. (46), we obtain

$$\sum_{kl} V_{k'k}U_{kl}\left\langle\sigma_{k}^{z}\left(\sigma_{k'}^{+}\sigma_{l}^{+} + \sigma_{k'}^{-}\sigma_{l}^{-}\right)\right\rangle = \frac{\gamma}{2}\frac{d}{dt}\langle\sigma_{k'}^{z}\rangle\ . \tag{49}$$

Collecting results, we obtain

$$\begin{aligned}\frac{1}{2}\frac{d^2}{dt^2}\langle\sigma_{k'}^{z}\rangle\ = &-\sum_{k} V_{k'k}V_{kk'}\left(\langle\sigma_{k}^{z}\rangle + \langle\sigma_{k'}^{z}\rangle\right)\\ &-\frac{\gamma}{2}\frac{d}{dt}\langle\sigma_{k'}^{z}\rangle - \Delta g_{k'}\,,\end{aligned} \tag{50}$$

where

$$g_{k'}(t) = \sum_{k} V_{k'k}\left\langle\sigma_{k'}^{+}\sigma_{k}^{+} + \sigma_{k'}^{-}\sigma_{k}^{-}\right\rangle. \tag{51}$$

Using arguments similar to those used to derive Eq. (50), we obtain the approximate equation

$$\frac{dg_{k'}}{dt} = \Delta \frac{dn_{k'}}{dt} - \gamma \left(g_{k'} - \bar{g} \right), \tag{52}$$

where $\bar{g} = \frac{1}{N'} \sum_{k'} g_{k'}$ and the $\gamma \bar{g}$ term has been put in by hand to preserve conservation. These equations imply

$$\frac{d\bar{g}}{dt} = \Delta \frac{dn'}{dt}, \tag{53}$$

where

$$n' = \frac{1}{N'} \sum_{k'} n_{k'}, \tag{54}$$

with the initial condition $g_{k'}(0) = 0$.

At this point, it is more convenient to work with the p' occupation $n_{k'}$, defined in Eq. (43), and with the corresponding p occupation, n_k. For comparison with experiment we will need the sample average, which corresponds to $S(t)$ of Sections II and III. Taking Laplace transforms of Eqs. (50) and (52), we obtain

$$\begin{aligned} &\left(\alpha^2 + 2 \sum_k V_{kk'}^2 + \gamma \alpha + \frac{\alpha \Delta^2}{\alpha + \gamma} \right) \tilde{n}_{k'} \\ &= \frac{2}{\alpha} \sum_k V_{kk'}^2 - 2 \sum_k V_{kk'}^2 \tilde{n}_k - \Delta^2 \frac{\gamma}{\alpha + \gamma} \tilde{n}', \end{aligned} \tag{55}$$

where the tildes denote Laplace transforms and $\tilde{n}'(\alpha)$ is the sample average of $\tilde{n}_{k'}(\alpha)$.

We can obtain a similar equation for $\tilde{n}_k$, and we are then left with coupled equations which we are not able to solve. However, if $N \approx N'$ we can simplify the problem by assuming that the strong *sp*-*ps* coupling acts to randomize the σ_k spins, while the resonant ss'-pp' processes are entirely coherent. Accordingly, in Eq. (55) we replace $\tilde{n}_k$ with its sample average $\tilde{n}$ and obtain an equation for $\tilde{n}_{k'}$ alone by using the exact relation $Nn = N'n'$, which follows from the fact that p and p' states are created in pairs by the $V_{kk'}$ term in Eq. (41). We solve for $\tilde{n}_{k'}$ and average over a random distribution of atoms, replacing the sample average $(1/N') \sum_{k'} n_{k'}$ with an ensemble average, and obtain

$$\tilde{n}' = \frac{(1/\alpha) \left(1 - C^2 \tilde{F}_{av} \right)}{1 + \frac{N'}{N} - \left(\frac{N'}{N} C^2 - \frac{\gamma \Delta^2}{\alpha + \gamma} \right) \tilde{F}_{av}}, \tag{56}$$

where

$$C^2 = \alpha^2 + \alpha \gamma + \frac{\alpha \Delta^2}{\alpha + \gamma}, \tag{57}$$

and $\tilde{F}_{av}$ is the ensemble average of

$$\tilde{F} = \frac{1}{C^2 + 2\sum_k V_k^2} \,. \tag{58}$$

For $N = N'$, Eq. (56) simplifies to

$$\tilde{n}' = \frac{(1/\alpha)\left(1 - C^2 \tilde{F}_{av}\right)}{2 - \left(C^2 - \frac{\gamma\Delta^2}{\alpha+\gamma}\right)\tilde{F}_{av}}, \tag{59}$$

$\tilde{F}_{av}$ is computed in the same way as the averages of $\tilde{S}$ in Section II and $\tilde{L}$ in Section III. The analog of Eq. (15) is

$$C^2 \tilde{F}(\alpha)_{av} = 1 - \sqrt{\frac{\pi}{2}} \frac{v}{C} \exp\left(\frac{v^2}{2C^2}\right) \operatorname{erfc}\left(\frac{v}{\sqrt{2}C}\right), \tag{60}$$

with v given by Eq. (10) and C by Eq. (57). One way to compute $n'(t)$ is to expand $\tilde{n}'(\alpha)$, Eq. (56), in inverse powers of α. This gives the analog of Eq. (17), but we have not found a simple expression for the coefficients.

Fig. 6 shows the signal as calculated in Eq. (59) for $\gamma = v$, and $\Delta/v = 0$, 1, and 2. We see that the on-resonance signal is devoid of any oscillations. Off-resonance, the oscillations are not completely washed out by the phase averaging, and some nonmonotonicity is evident. Consequently, the dip in the width seen in Fig. 2 is also present in this calculation, provided γ is not too large.

Fig. 6 differs from the corresponding Fig. 4 of Section III in one important respect: the on-resonance signal saturates to 1/2 rather than to 1. For general values of N and N', Eq. (56) predicts that as $t \to \infty$ (i.e., as $\alpha \to 0$) $N'n'$ approaches $NN'/(N+N')$, as expected from simple kinetics for the two-body reaction $ss' \to pp'$.

As in the previous sections, the energy scale is set by v, which according to Eq. (10) has the value $5.72\,\mu\mu' N/\Omega$. As discussed by Anderson *et al.* [1], this quantity is of the correct order of magnitude to account for the data.

A. Reduction to the sparse limit

We now show explicitly how the general spin-variable formalism relates to the coefficients a_0 and c_k of Sections II and III when there is only a single atom that can be in the s', p' pair of states (i.e., when only a single spin variable of the type $\sigma_{k'}$ is present.) The key to the correspondence is that in this case the initial state $|i\rangle$ (which consists of all spins down) evolves to

$$|t\rangle = \left(a_0(t) + \sum_q c_q(t)\sigma_{k'}^{+}\sigma_q^{+}\right)|i\rangle \tag{61}$$

at time t, where the spin variables now denote time-independent (Schrödinger) operators. With the understanding that $\mathbf{r}_{k'} = 0$, we have:

$$\langle \sigma_{k'}^z \rangle \equiv \langle t|\,\sigma_{k'}^z\,|t\rangle = 1 - 2\,|a_0(t)|^2 \,, \tag{62}$$

$$\langle \sigma_k^z \rangle = 2\,|c_k(t)|^2 - 1, \tag{63}$$

$$\left\langle \sigma_{k'}^{+} \sigma_k^{+} \right\rangle = a_0(t) c_k^{\dagger}(t). \tag{64}$$

It is then clear that the expectation value of Eq. (46) is equivalent to the following, which is itself a consequence of Eqs. (1):

$$\frac{d}{dt}\,|a_0(t)|^2 = \sum_k V_k \left(a_0 c_k^{\dagger} - a_0^{\dagger} c_k \right), \tag{65}$$

where V_k is short for $V_{k'k}$. Similarly, the equations for $\langle \sigma_k^z \rangle$ and $\left\langle \sigma_{k'}^{+} \sigma_k^{+} \right\rangle$ are equivalent to the equations for $|c_k(t)|^2$ and $a_0(t)c_k(t)^{\dagger}$ that follow from Eqs. (24b).

When the Lorentzian approximation is made in Eq. (47) or its analog for σ_k^z, it is possible to obtain a closed set of equations for $\langle \sigma_k^z \rangle$ and $\langle \sigma_{k'}^z \rangle$ that give the results reported in Sections II and III for a sparse system. As indicated by the Laplace transforms in Eqs. (34) and (37), the equation for $L(t) = \frac{1}{2}\left(1 + \sum_k \langle \sigma_k^z \rangle\right)$ must be of the fourth order, and that for $S(t) = \frac{1}{2}\left(1 - \sum_k \langle \sigma_k^z \rangle\right)$ of fifth order.

If Eq. (56) were exact, it would correctly give the signal in the limit $N' \ll N$, and $n'(t)$ would be equal to $S(t)_{av}$ of Section III if the values for γ used in the two models were assumed to be equal. In addition, $n'(t)$ would reduce to the exact $S(t)_{av}$ of Section II if γ were taken to be zero. Since the result for $n'(t)$ is based on approximations that are certainly not valid for $N' \ll N$, it is already comforting that, in that limit, it shows a general resemblance to the correct behavior. We are currently working on better approximations to apply to the spin-variable formalism.

V. DISCUSSION

The system we are studying can exhibit a wide range of behaviors, depending on the values of the parameters μ/μ' and N/N'. At the same time, there are features that persist (at least qualitatively) throughout the range of parameters.

In Section II, we determine the exact behavior of a frozen system consisting of a single s' state interacting resonantly with a sea of s states. That is to say, we treat exactly the limit $\mu \ll \mu'$ and $N' \ll N$. A single ss' pair produces, of course, a signal with a sinusoidal behavior, and the same is true, less obviously, for a single s' interacting with a sea of s that are not mutually interacting. The phase interference brought in by averaging over configurations causes the signal from a sparse system of s' to take on the form shown in Fig. 1. Because the averaging does not completely wash out the oscillations at small times, especially for nonzero values of the detuning, the resonance linewidth has the nonmonotonic behavior seen in Fig. 2. The initial rise of the averaged signal is linear in time, even though the unaveraged signal is quadratic, and is given by $\sqrt{\pi} v t/2$, with v defined in Eq. (10). The linewidth at large times is approximately $4.6\,v$ [10]. The fact that v is sizably larger than the simple estimate $\mu\mu'\,(N/\Omega)$ reflects quantitatively the fact that “close pairs” of atoms are more heavily weighted in the average.

In Section III we try to allow for any ratio μ/μ', but we still keep $N' \ll N$: physically, we consider a sparse distribution of (initially) s' states in a gas where *sp*-*ps* flips take place at a non-negligible rate. The first question we should answer is: after the $ss' - pp'$ transition, does the p state "walk away" to infinity or remain localized? In this paper, we have assumed that localization does not take place, or, if it does, is important only in the very sparse limit where the localization length is smaller than $(\Omega/N)^{1/3}$. Leaving much room for future work, we have simply introduced a lifetime γ^{-1} for the p state to "stick around". The spectral function for the propagating *sp*-*ps* flip is then simply a Lorentzian. We have obtained tentative estimates of γ when it is entirely due to the *sp*-*ps* flips, and we plan to report these results in the near future. There can also be other contributions to γ, arising for instance from translational motion of the atoms. (The gas can be treated as frozen to a good approximation when the typical collision time τ exceeds v^{-1}, and certainly τ^{-1} contributes to γ.) In this paper, we in effect add a damping term to the equations of Section II, making the problem soluble again. The effect of γ is to further smooth out and wash away the oscillatory behavior of the unaveraged signal. For large γ all traces of the oscillations are wiped out and the signal saturates according to $\exp(-\sqrt{\gamma_{eq} t})$, where γ_{eq} is given by Eq. (32). The saturated linewidth is infinite, because the s' state eventually decays for all values of Δ. However, the signal at finite t varies significantly with Δ, indicating that the linewidth has a minimum at intermediate times. For small t the signal is always independent of Δ, consistent with a linewidth proportional to t^{-1}, as seen already in Fig. 2.

In Section IV we introduce an effective spin Hamiltonian to describe the resonating frozen gas for all values of the parameters μ/μ' and N/N', and we discuss in particular the case $N \approx N'$, which is relevant to the experiments carried out so far. The only simplification we make in this spin Hamiltonian is to neglect the potential responsible for the $s'p'$-$p's'$ interaction. This coupling can easily be included, although the resulting equations of motion contain more terms and would be more difficult to handle; it is in fact small for the experiments of Anderson *et al.* [1,3]. Other resonant systems can be described by similar spin Hamiltonians. The experiment of Mourachko *et al.* [2], for example, consists of a $pp - ss'$ resonance in the presence of *sp*-*ps* and $s'p - ps'$ flips. A spin-one Hamiltonian therefore describes this system, with the -1, 0, and 1 spin states corresponding to the s, p, and s' atomic states, respectively. In this case, there is only one set of spins and all spins are initially in the 0 state. The spin model we have discussed in this paper consists of two sets of spins, which complicates matters but at the same time allows one to consider the limit in which one set is sparse.

There is practically no end to the variety of phenomena that this type of spin Hamiltonian can describe, combining features from many branches of physics. Atomic physics, random systems, spin systems and magnetic resonance, and many-body theory are all represented. The equilibrium behavior poses interesting problems of statistical mechanics, but we are interested in the quantum dynamics, starting from a prepared state. We have obtained and solved approximate dynamical equations for the case $N \simeq N'$, using the Lorentzian approximation as in Section III to model the *sp*-*ps* interaction. Much more work can be done and is now in progress, and some of the results may change when better approximations are introduced. In particular, in a better theory the linewidth is likely to depend on the *sp*-*ps* interaction as well as on the ss'-pp'. Nevertheless, the qualitative behavior of the signal is common to all approximations we have tested so far and matches what is seen in the experiments that initially motivated this work. The important features that we already discussed are all there: the sharp initial rise, the slow saturation, the dependence on Δ that is less pronounced at small times and corresponds to a time-dependent linewidth.

ACKNOWLEDGMENTS

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APPENDIX A: ANGULAR AVERAGING

In Section II we averaged the quantity $e^{-\beta\mathcal{V}^2}$, where $\mathcal{V}^2 = \sum_{k=1}^{N} V_k^2$ and $V_k = \mu\mu'/r_k^3$. In this appendix we include the angular dependence of the dipole-dipole interaction assuming that all of the dipoles point in the same direction, i.e. we replace V_k with

$$V(r,\theta) = -\frac{\mu_1\mu_2}{r^3}\left(3\cos^2\theta - 1\right). \tag{1}$$

The analog of Eq. (8) now has a nontrivial angular part; it becomes

$$\begin{aligned} &2\pi\int_0^\infty r^2 dr \int_{-\pi/2}^{\pi/2} \sin\theta d\theta \left[1 - e^{-\beta V^2(r,\theta)}\right] \\ &= \frac{16\pi^{3/2}}{9\sqrt{3}}\ \mu\mu'\ \sqrt{\beta}. \end{aligned} \tag{2}$$

The additional angular dependence changes v by a factor $4/3\sqrt{3}$.

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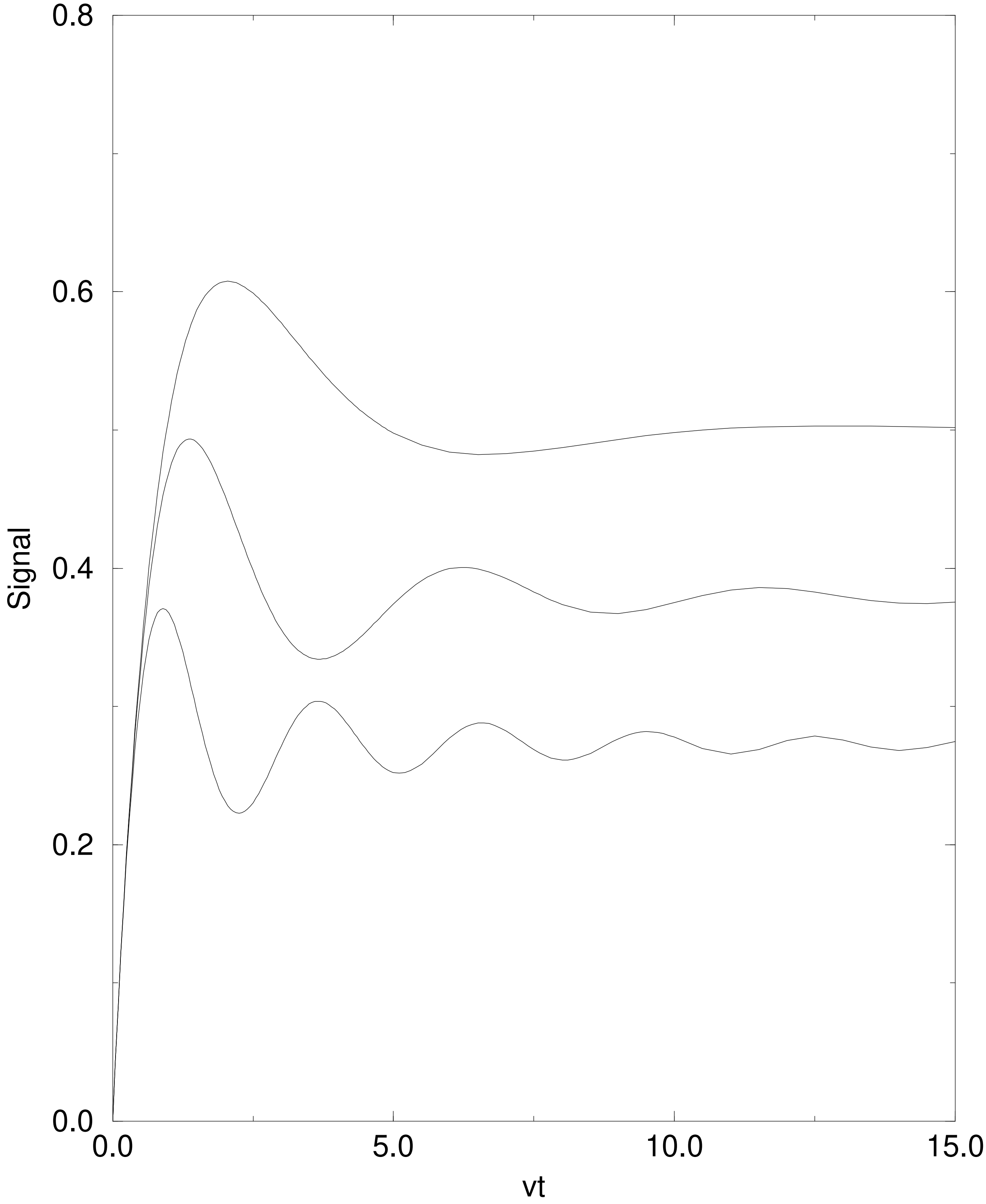
[10] All widths discussed in this paper are in energy units. There is an additional factor of $1/2\pi$ when computing the width in frequency units.

FIGURES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0.6
0.4
0.2
0.0
Signal
0.0
5.0
10.0
15.0
vt

FIG. 1. The averaged signal, $S(t)_{av}$, for $\Delta/v = 0$, 1, and 2 (the upper, middle, and lower curves, respectively) calculated from Eq. (17) for the sparse limit. The ss'- pp' process is averaged; the sp-ps process neglected. The Rabi oscillations are not completely washed out by the averaging process and are more pronounced off-resonance.

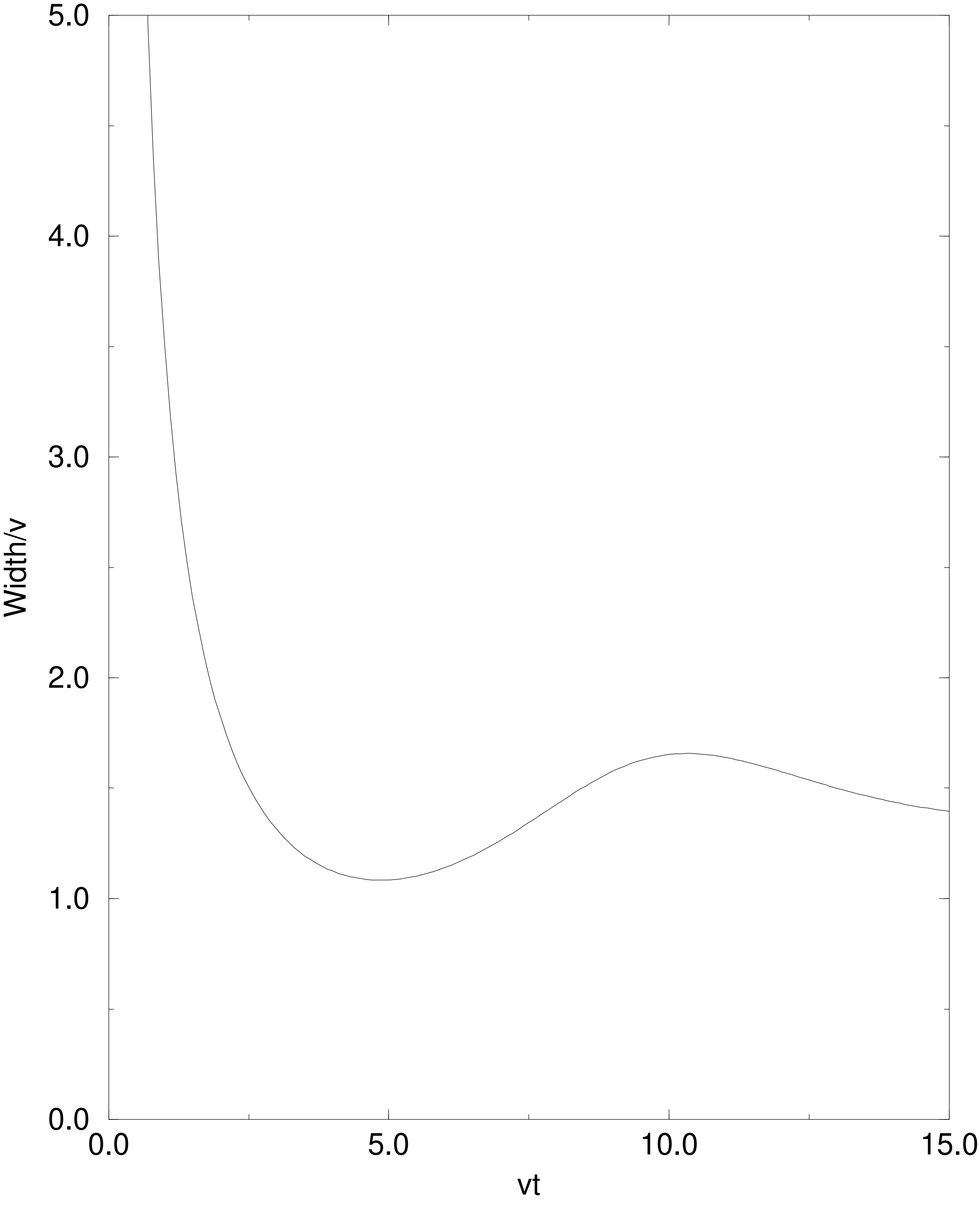
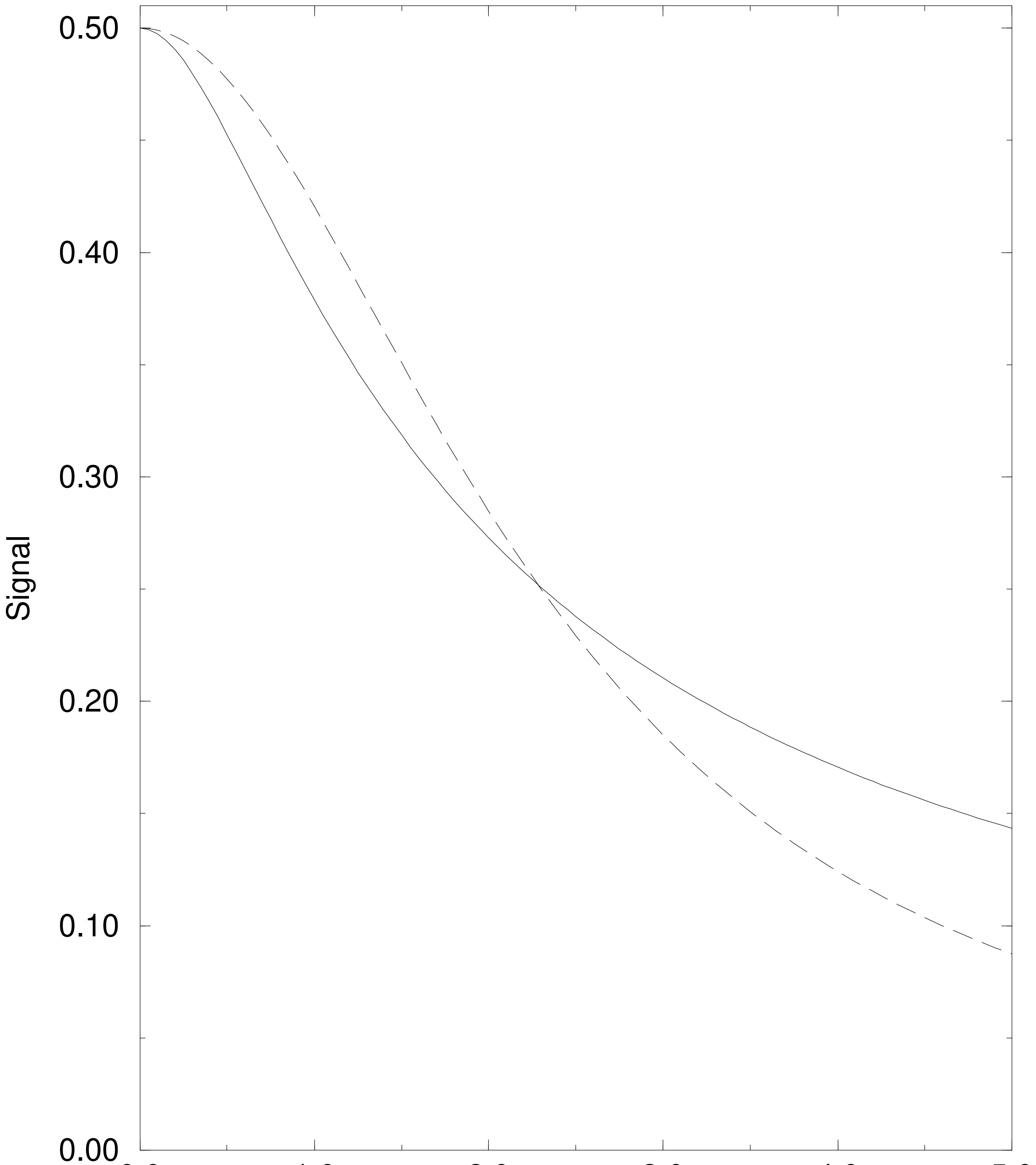

5.0
4.0
3.0
2.0
1.0
0.0
Width/v
0.0
5.0
10.0
15.0
vt

FIG. 2. The "width" versus time, as given by Eq. (21), shows a dip which is associated with the stronger and higher frequency oscillations seen at larger values of Δ as shown in Fig. 1.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0.20
0.30
0.40
0.50
0.0
1.0
2.0
3.0
4.0
5.0
Δ/v

FIG. 3. The saturation ($t \to \infty$) lineshape, Eq. (23). It is not a Lorentzian but a Lorentzian averaged as in Eq. (22). The dashed line is a Lorentzian with the same height and FWHM, drawn for comparison.

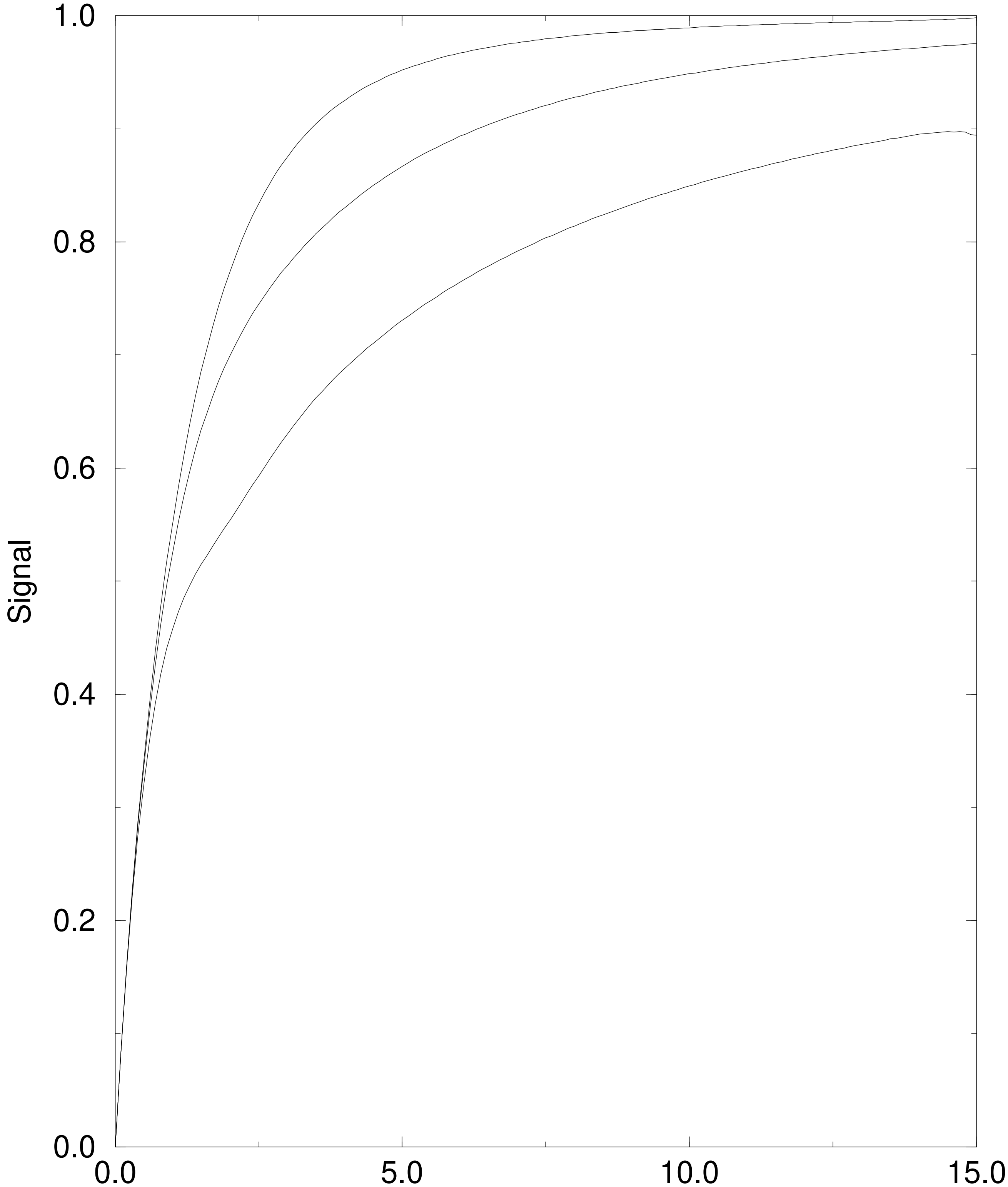
1.0
0.8
0.6
0.4
0.2
0.0
Signal
0.0
5.0
10.0
15.0
vt

FIG. 4. The averaged signal, $S(t)_{av}$, for $\gamma = v$, and $\Delta/v = 0$, 1, and 2 (the upper, middle, and lower curves, respectively) calculated from $\tilde{S}(\alpha)_{av} = \left(1 + \frac{2\gamma}{\alpha}\right) \tilde{L}(\alpha)_{av}$ and Eq. (38). The approach to saturation goes like $\exp\left(-\sqrt{\gamma_{eq} t}\right)$, with γ_{eq} given by Eq. (32).

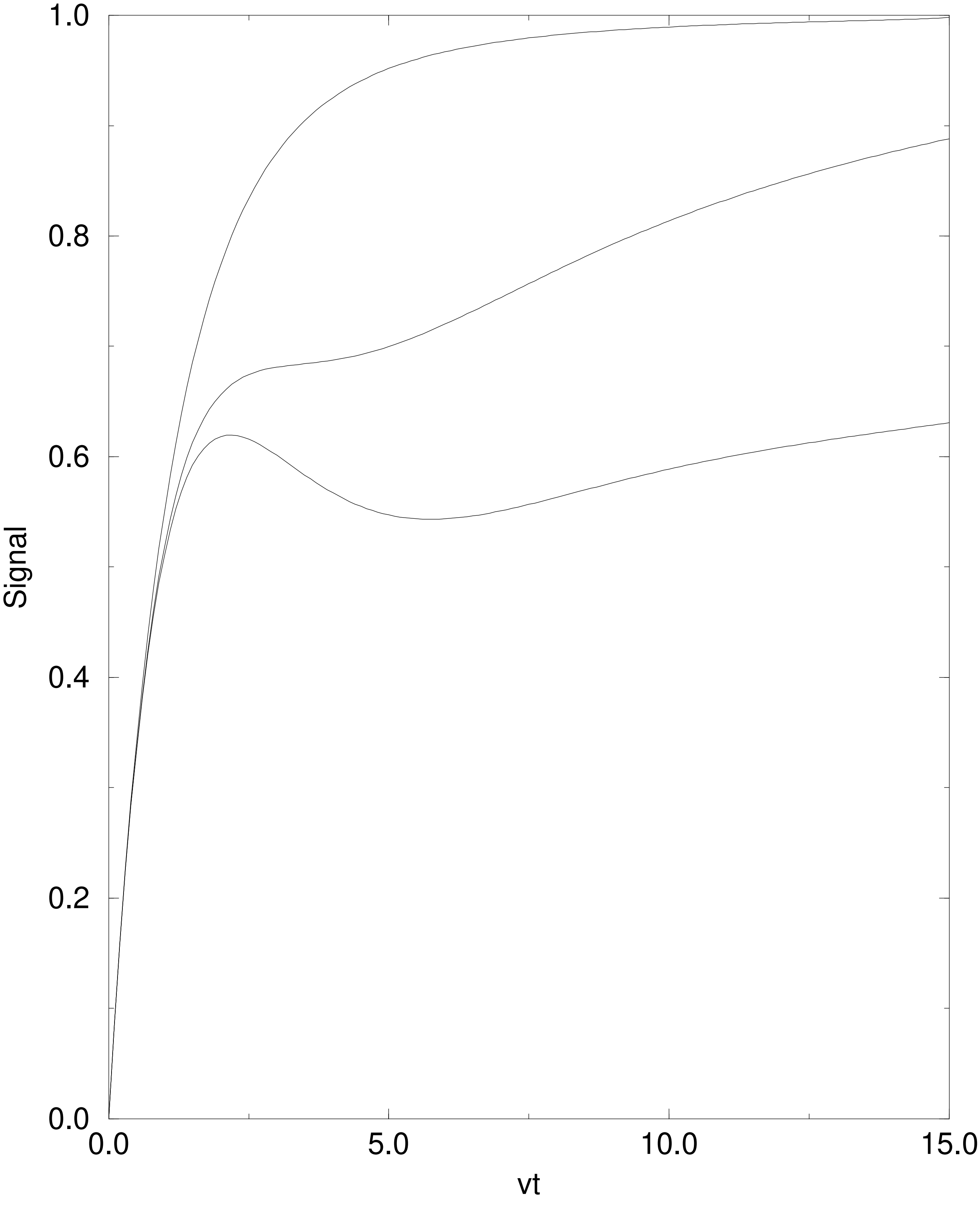
1.0
0.8
0.6
0.4
0.2
0.0
Signal
0.0
5.0
10.0
15.0
vt

FIG. 5. $S(t)_{av}$, as in Fig. 4, calculated at resonance ($\Delta = 0$), for inverse lifetimes $\gamma/v = 1$, 0.1, and 0.02 (the upper, middle, and lower curves, respectively). For the largest value of γ the oscillation is completely gone.

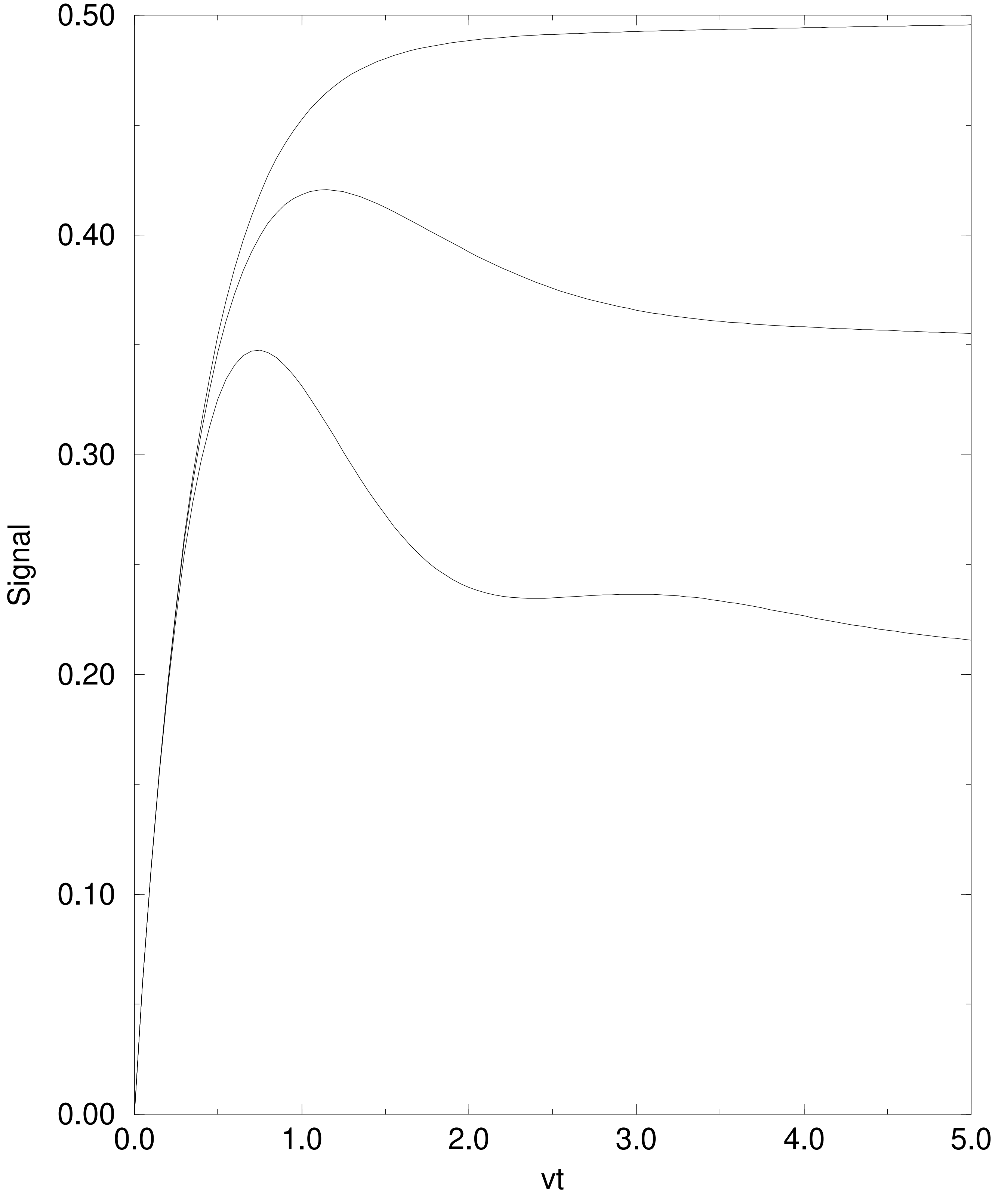
0.50
0.40
0.30
0.20
0.10
0.00
Signal
0.0
1.0
2.0
3.0
4.0
5.0
vt

FIG. 6. The averaged signal, $n'(t)$, for $N = N'$, $\gamma = v$, and $\Delta/v = 0$, 1, and 2 (the upper, middle, and lower curves, respectively), as calculated from Eq. (59). While the oscillations are "damped out" for $\Delta = 0$, they remain for $\Delta/v = 1$ and 2.