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## Theory for Quantum Interference Signal from an Inhomogeneously Broadened Two-Level System Excited by an Optically Phase-Controlled Laser-Pulse Pair

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**Abstract:** A useful expression for the quantum interference (QI) signal was derived for an inhomogeneously broadened two-level system when it was excited by an optically phase-controlled laser-pulse pair. It was shown that the QI signal oscillates as a function of a relative optical phase, with the reduced angular frequency given by the relation  $\omega_a = (\Gamma^2\omega_0 + \gamma_g^2\Omega)/(\gamma_g^2 + \Gamma^2)$ , where  $\gamma_g$  and  $\Gamma$  are standard deviations of the system's absorption and the laser spectra both having a Gaussian line shape, respectively, and  $\omega_0$  and  $\Omega$  are the center angular frequency of the system absorption and the carrier angular frequency of the laser, respectively.

### 1. Introduction

Control of quantum interference (QI) of molecular wavefunctions excited by a pair of femtosecond laser pulses that have a definite optical phase is one of the basic schemes for the control of versatile quantum systems including chemical reactions. The QI technique with the pulse pair, or the double pulse, has been applied to several atomic, molecular systems in the gas phase<sup>1–3</sup> and condensed phases.<sup>4–8</sup> A basic theory of the double-pulse QI experiment for a two-level molecular system in the gas phase has been given in the original paper by Scherer et al.<sup>1,2</sup> In their beautiful work, they derived the expression for the QI signal from a two-level system including a molecular vibration. However, the effect of inhomogeneous broadening, which is not very significant in the gas phase, has not been taken into account.

In this study, we derive a compact and useful expression for the QI signal for an inhomogeneously broadened two-level system in condensed phases, when the system was excited by an optically phase-controlled laser-pulse pair. In general, the homogeneous broadening gives a Lorentz profile:

$$S_L(\omega) = \frac{1}{2\pi} \frac{\gamma_l}{(\omega_0 - \omega)^2 + (\gamma_l/2)^2} \quad (1)$$

On the other hand, the inhomogeneous broadening gives a Gauss profile:

$$S_G(\omega) = \frac{1}{\sqrt{\pi} \gamma_g} e^{-(\omega - \omega_0)^2/\gamma_g^2} \quad (2)$$

When both the homogeneous and inhomogeneous broadening exist, the spectral profiles are given by a convolution of  $S_L(\omega)$  with  $S_G(\omega)$ , namely, a Voigt profile:

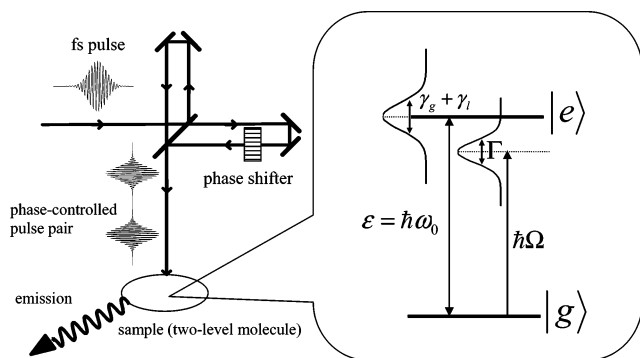
$$S_V(\omega) = \int_{-\infty}^{\infty} d\omega' S_L(\omega') S_G(\omega + \omega_0 - \omega') \quad (3)$$

As pointed out by Scherer et al., the QI signal is the free-induction decay and the Fourier transform of the optical spectral profile. According to the convolution theorem in the Fourier transform, the expression for the QI signal should, in principle, have the form:

$$\begin{aligned} \text{QI}(t_d) &= \text{FT}[S_V(\omega)] = \text{FT}[S_L(\omega)] \text{FT}[S_G(\omega)] \propto \\ &\cos[\omega_0 t_d] \exp\left[-\frac{\gamma_l t_d}{2}\right] \exp\left[-\frac{\gamma_g^2 t_d^2}{4}\right] \end{aligned} \quad (4)$$

where  $t_d$  is a time delay between the laser-pulse pair. However, in the above discussion, the laser pulse is assumed to be impulsive; that is, the effects of a finite time width or a spectral width of the actual laser pulse are not taken into account. The purpose of this paper is to derive the expression for the QI signal that includes the effects of nonimpulsive

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**Figure 1.** Schematic drawing of the QI experiment with a phase-controlled laser-pulse pair.

laser pulses which possess a Gaussian pulse shape. The procedure for derivation has two steps: first, we derive the expression for the homogeneously broadened two-level system, and then we obtain the expression for the inhomogeneously broadened system by integrating the result of the homogeneously broadened system weighted by the inhomogeneous spectral distribution function.

## 2. Theory

**2.1. Homogeneously Broadened Two-Level System.** Let us consider a two-level electronic system interacting with a phase-controlled femtosecond-laser pulse pair (Figure 1). When the ground-state energy is assumed to be zero, that is, the system is referenced to the molecular frame, the electronic Hamiltonian for the two-level system with the homogeneous broadening is given by

$$\hat{H} = (\epsilon - i\gamma_1/2)|e\rangle\langle e| \quad (5)$$

where  $\gamma_1$  is a homogeneous relaxation constant that stands for a radiative or a nonradiative decay constant. An electronic transition dipole operator is expressed as

$$\hat{\mu} = \mu_{eg}(|e\rangle\langle g| + |g\rangle\langle e|) \quad (6)$$

The interaction Hamiltonian between the system and a photon field is given by

$$\hat{V} = -\hat{\mu} E(t) \quad (7)$$

where photoelectric field  $E(t)$  in the double-pulse QI experiments is given by the sum of  $E_1$  and  $E_2$ , each of which has a Gaussian profile:

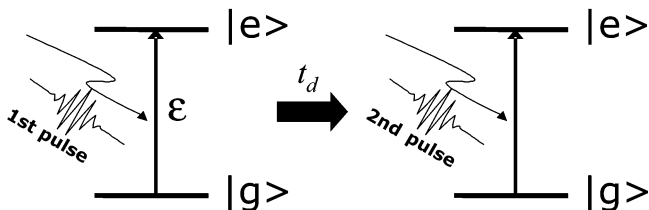
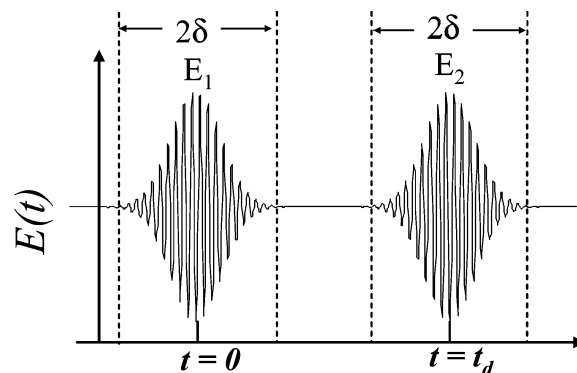
$$E(t) = E_1(t) + E_2(t) \quad (8)$$

where  $\tau$  is a standard deviation of each laser pulse in the

$$E_1(t) = E_0 \exp[-t^2/(2\tau^2)] \cos(\Omega t) \quad (9)$$

$$E_2(t) = E_0 \exp[-(t - t_d)^2/(2\tau^2)] \cos[\Omega(t - t_d)] \quad (10)$$

time domain and related to a standard deviation  $\Gamma$  of each laser pulse in the frequency domain by  $\tau = 1/\Gamma$ , and  $\Omega$  is a common carrier frequency of the laser pulses. The phase shift of the photon field is defined as delay time:<sup>9</sup> the delay time  $t_d$  between double pulses is finely controlled with attoseconds order in the optical phase-controlled experiments. This



**Figure 2.** Time domains: free evolution and interaction with laser pulses.

definition is natural in the optical phase-shift experiments as pointed out by Albrecht et al.<sup>10</sup>

To derive the expression for the QI signal, we divide the time region into the free-evolution regions and the interaction regions (Figure 2). Then, the time evolution of the system from the initial electronic state  $|\psi(t = -\infty)\rangle = |g\rangle$  is given by the equation

$$|\psi(t)\rangle = \hat{U}(t - t_d - \delta)\hat{W}_2\hat{U}(t_d - 2\delta)\hat{W}_1|g\rangle \quad (11)$$

where the time evolution operator in the absence of the photon field is defined by

$$\hat{U}(t, t') = \exp[-i\hat{H}(t - t')/\hbar] \quad (12a)$$

or by replacing as  $\Delta t = t - t'$

$$\hat{U}(\Delta t) = \exp[-i\hat{H}\Delta t/\hbar] \quad (12b)$$

Within the framework of the first-order perturbation theory,<sup>11</sup> the time evolution operator  $\hat{W}_j$  ( $j = 1, 2$ ) in the presence of the photon field is given by

$$\begin{aligned} \hat{W}_j &= \hat{U}(t_j + \delta, t_j - \delta) \\ &= \left\{ 1 - \frac{1}{i\hbar} \int_{t_j - \delta}^{t_j + \delta} dt' \hat{U}(t_j - t') \hat{\mu} E_j(t') \hat{U}(t' - t_j) \right\} \\ &= \hat{U}(2\delta) \left\{ 1 - \frac{1}{i\hbar} \int_{t_j - \delta}^{t_j + \delta} dt' \hat{U}(t_j - t') \hat{\mu} E_j(t') \hat{U}(t' - t_j) \right\} \\ &\equiv \hat{U}(2\delta) \left( 1 - \frac{1}{i\hbar} \hat{F}_j \right) \end{aligned} \quad (13)$$

The substitution of eq 13 into eq 11 yields

$$|\psi(t)\rangle = \hat{U}(\delta) \left[ \hat{U}(t) + \frac{i}{\hbar} \hat{U}(t) \hat{F}_1 + \frac{i}{\hbar} \hat{U}(t - t_d) \hat{F}_2 \hat{U}(t_d) \right] |g\rangle \quad (14)$$

where  $\hat{F}$  is defined as an electronic transition operator, and  $\hat{U}(\delta)$  a global phase factor, which will be neglected hereafter, because it does not affect final results in the state-density

matrix. The projection of eq 14 onto the excited state  $|e\rangle$  gives

$$\begin{aligned}\langle e|\psi(t)\rangle &= \left\langle e \left| \hat{U}(t) + \frac{i}{\hbar} \hat{U}(t) \hat{F}_1 + \frac{i}{\hbar} \hat{U}(t-t_d) \hat{F}_2 \hat{U}(t_d) \right| g \right\rangle \\ &= \frac{i}{\hbar} \langle e | [\hat{U}(t) \hat{F}_1 + \hat{U}(t-t_d) \hat{F}_2 \hat{U}(t_d)] | g \rangle \\ &= \frac{i}{\hbar} \{ \exp[-i\omega_0 - \gamma_1/2]t \langle e | \hat{F}_1 | g \rangle + \\ &\quad \exp[-i\omega_0 - \gamma_1/2](t-t_d) \langle e | \hat{F}_2 | g \rangle \} \quad (15)\end{aligned}$$

where  $\omega_0 = \epsilon/\hbar$ . The matrix element of an electronic transition operator  $\hat{F}_j$  is calculated as

$$\begin{aligned}\langle e | \hat{F}_j | g \rangle &= \langle e | \int_{t_j-\delta}^{t_j+\delta} dt' \hat{U}(t_j-t') \hat{\mu} \hat{U}(t'-t_j) | g \rangle E_j(t') \\ &= \mu_{eg} \langle e | \int_{t_j-\delta}^{t_j+\delta} dt' \hat{U}(t_j-t') (|g\rangle\langle e| + \\ &\quad |e\rangle\langle g|) \hat{U}(t'-t_j) | g \rangle E_j(t') \\ &= \mu_{eg} \int_{t_j-\delta}^{t_j+\delta} dt' \langle e | U(t_j-t') | e \rangle \langle g | U(t'-t_j) | g \rangle E_j(t') \\ &= \mu_{eg} \int_{t_j-\delta}^{t_j+\delta} dt' e^{-i(\omega_0 - i\gamma_2/2)(t_j-t')} E_j(t') \quad (16)\end{aligned}$$

When a rotating-wave approximation is used, the matrix element is further calculated as

$$\begin{aligned}\langle e | \hat{F}_j | g \rangle &= \frac{1}{2} E_0 \mu_{eg} \int_{-\infty}^{+\infty} dx e^{-i(\omega_0 - \Omega)x} e^{-\gamma_1 x/2 - x^2/(2\tau^2)} \\ &= \sqrt{\frac{\pi}{2}} E_0 \mu_{eg} \tau \exp\left[\frac{\{\gamma_1 + i2(\omega_0 - \Omega)\}^2 \tau^2}{8}\right] \equiv F \times \\ &\quad \exp\left\{\frac{\tau^2}{8} [\gamma_1^2 - 4(\omega_0 - \Omega)^2 + i4\gamma_1(\omega_0 - \Omega)]\right\} \quad (17)\end{aligned}$$

The substitution of eq 17 into eq 15 yields

$$\begin{aligned}\langle e | \psi(t, t_d) \rangle &= \frac{iF}{\hbar} \exp[-i\omega_0 - \gamma_1/2]t \{1 + \exp[(i\omega_0 + \\ &\quad \gamma_1/2)t_d]\} \exp\left[\frac{\{\gamma_1 + i2(\omega_0 - \Omega)\}^2 \tau^2}{8}\right] \quad (18)\end{aligned}$$

The absolute square of eq 18 gives the density matrix element  $\rho_{ee}(t, t_d, \omega_0, \Omega)$  for the excited state

$$\begin{aligned}\rho_{ee}(t, t_d, \omega_0, \Omega) &= \langle e | \psi(t, t_d) \rangle \langle \psi(t, t_d) | e \rangle \\ &= \frac{2F^2}{\hbar^2} e^{\gamma_1^2/(4\Gamma^2)} e^{-(\omega_0 - \Omega)^2/\Gamma^2} \{e^{-\gamma_1 t} + e^{-\gamma_1(t-t_d)} + \\ &\quad e^{-\gamma_1 t/2} e^{-\gamma_1(t-t_d)/2} \cos[\omega_0 t_d]\} \quad (19)\end{aligned}$$

The first and second term give population decays of the excited state created by the first and second pulses, respectively. The third term is the interference term that is the product of coherence decays and an oscillating term.

**2.2. Inhomogeneously Broadened System.** In the previous section, the inhomogeneous broadening was not taken into consideration. The effects of inhomogeneous decay can be taken into account by summing up  $\rho_{ee}$  that originates from inhomogeneously broadened spectral components.<sup>12</sup> When the inhomogeneous spectrum function is given by a Gaussian

in eq 2, the expectation value of the excited-state density function can be written as

$$\begin{aligned}\langle \rho_{ee}(t_d, \omega_0, \Omega) \rangle &= \int_{-\infty}^{+\infty} d\omega S_G(\omega, \omega_0) \rho_{ee}(t_d, \omega, \Omega) \\ &= \frac{F^2}{\hbar^2} e^{\gamma_1^2/(4\Gamma^2)} \int_{-\infty}^{+\infty} d\omega S_G(\omega, \omega_0) e^{-(\omega - \Omega)^2/\Gamma^2} \\ &\quad \{e^{-\gamma_1 t} + e^{-\gamma_1(t-t_d)} + \\ &\quad e^{-\gamma_1 t/2} e^{-\gamma_1(t-t_d)/2} \cos(\omega t_d)\} \quad (20)\end{aligned}$$

In the above equation, the two-center Gaussian functions can be rewritten as a one-center Gaussian function:

$$\begin{aligned}S_G(\omega, \omega_0) e^{-(\omega - \Omega)^2/\Gamma^2} &= \frac{1}{\sqrt{\pi} \gamma_g} e^{-(\omega - \omega_0)^2/\gamma_g^2} e^{-(\omega - \Omega)^2/\Gamma^2} \\ &= \frac{1}{\sqrt{\pi} \gamma_g} \exp\left[-\frac{(\omega_0 - \Omega)^2}{\gamma_g^2 + \Gamma^2}\right] \times \\ &\quad \exp\left[-\frac{\gamma_g^2 + \Gamma^2}{\gamma_g^2 \Gamma^2} \left(\omega - \frac{\Gamma^2 \omega_0 + \gamma_g^2 \Omega}{\gamma_g^2 + \Gamma^2}\right)^2\right] \quad (21)\end{aligned}$$

By defining a reduced decay constant  $\gamma_a$  and a reduced frequency  $\omega_a$  equation 21 becomes a simple form:

$$\frac{1}{\gamma_a^2} \equiv \frac{\gamma_g^2 + \Gamma^2}{\gamma_g^2 \Gamma^2} \quad (22a)$$

$$\omega_a \equiv \frac{\Gamma^2 \omega_0 + \gamma_g^2 \Omega}{\gamma_g^2 + \Gamma^2} \quad (22b)$$

$$\begin{aligned}S_G(\omega, \omega_0) e^{-(\omega - \Omega)^2/\Gamma^2} &= \frac{1}{\sqrt{\pi} \gamma_g} \exp\left[-\frac{(\omega_0 - \Omega)^2}{\gamma_g^2 + \Gamma^2}\right] \times \\ &\quad \exp\left[-\frac{(\omega - \omega_a)^2}{\gamma_a^2}\right] \quad (23)\end{aligned}$$

By carrying out the Gauss integral and the Fourier-type integral of the Gaussian function, the final form of eq 20 becomes

$$\begin{aligned}\langle \rho_{ee}(t, t_d, \omega_0, \Omega) \rangle &= \frac{\gamma_a F^2}{\gamma_g \hbar^2} \exp\left[\frac{\gamma_1^2}{4\Gamma^2} - \frac{(\omega_0 - \Omega)^2}{\gamma_g^2 + \Gamma^2}\right] \{e^{-\gamma_1 t} + \\ &\quad e^{-\gamma_1(t-t_d)} + 2 \cos[\omega_a t_d] e^{-\gamma_1 t/2} e^{-\gamma_1(t-t_d)/2} e^{-(\gamma_a^2 t_d^2/4)}\} \quad (24)\end{aligned}$$

In the typical QI experiments, the QI signal is obtained as the total fluorescence integrated over time. Thus, the QI signal is calculated from eq 24 as the following:

$$\begin{aligned}\text{QI}(t_d) &= - \int_{t_d}^{\infty} dt \frac{d\langle \rho_{ee}(t, t_d, \omega_0, \Omega) \rangle}{dt} = \langle \rho_{ee}(t = t_d, t_d, \omega_0, \Omega) \rangle \\ &= \frac{\gamma_a F^2}{\gamma_g \hbar^2} \exp\left[\frac{\gamma_1^2}{4\Gamma^2} - \frac{(\omega_0 - \Omega)^2}{\gamma_g^2 + \Gamma^2}\right] \{1 + e^{-\gamma_1 t_d} + \\ &\quad 2 \cos[\omega_a t_d] e^{-(\gamma_1 t_d/2)} e^{-(\gamma_a^2 t_d^2/4)}\} \quad (25)\end{aligned}$$

In the above derivation, the pure dephasing was not taken into account, and a transverse relaxation time constant  $T_2$  and a longitudinal relaxation constant  $T_1$  are related by

$$\frac{1}{T_2} = \frac{\gamma_l}{2} = \frac{1}{2T_1} \quad (26)$$

However, in general, there also exists a pure dephasing  $\gamma^*$  that is brought about from elastic solute–solvent collisions.<sup>13</sup> Thus, the transverse relaxation time constant should be rewritten as

$$\frac{1}{T_2} = \frac{\gamma_l}{2} + \gamma^* = \frac{1}{2T_1} + \frac{1}{T_2^*} \quad (27)$$

The final expression for the QI signal is given by

$$\text{QI}(t_d) = \frac{\gamma_a F^2}{\gamma_g \hbar^2} \exp \left[ \frac{1}{(2T_1 \Gamma)^2} - \frac{(\omega_0 - \Omega)^2}{\gamma_g^2 + \Gamma^2} \right] \{ 1 + e^{-(t_d/T_1)} + 2 \cos[\omega_a t_d] e^{-(t_d/T_2)} e^{-(\gamma_a^2 t_d^2/4)} \} \quad (28)$$

### 3. Discussion

By comparing the third term in eq 28 with eq 4, we notice that  $\omega_0$  and  $\gamma_g$  in the impulsive excitation are replaced by  $\omega_a$  and  $\gamma_a$ , respectively, in the nonimpulsive excitation. These reduced constants, of course, approach  $\omega_0$  and  $\gamma_g$  in the limiting case of impulsive laser pulses; that is, when  $\Gamma \gg \gamma_g$ , the following relations can be deduced:

$$\omega_a \cong \omega_0, \gamma_a \cong \gamma_g \quad (29)$$

In the reverse limiting case of  $\gamma_g \gg \Gamma$ , that is, in the case of a quasi continuum wave (CW) laser, we notice that

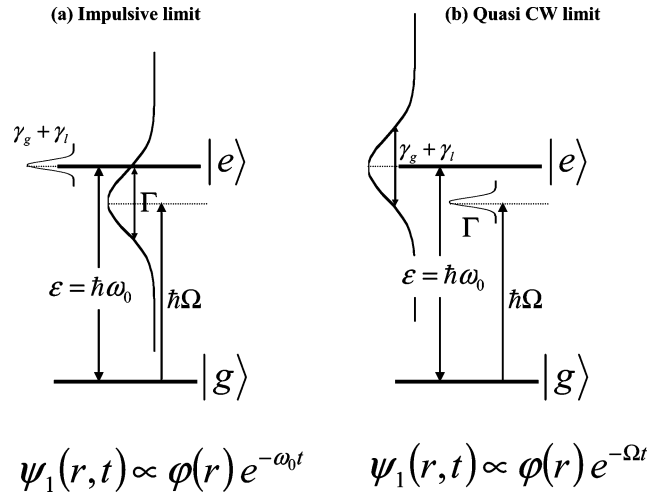
$$\omega_a \cong \Omega, \gamma_a \cong \Gamma \quad (30)$$

Under this condition, if we further assume that  $1/T_2 \gg \Gamma$ , the QI signal can be approximately written as

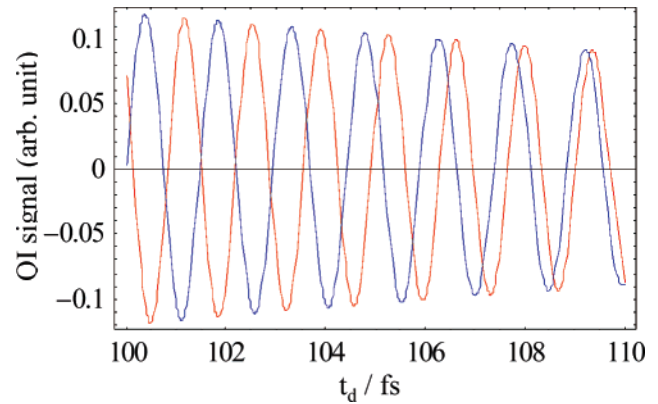
$$\text{QI}(t_d) = \frac{\Gamma F^2}{\gamma_g \hbar^2} \exp \left[ \frac{1}{(2T_1 \Gamma)^2} - \frac{(\omega_0 - \Omega)^2}{\gamma_g^2 + \Gamma^2} \right] \{ 1 + e^{-t_d/T_1} + 2 \cos[\Omega t_d] e^{-(t_d/T_2)} \} \quad (31)$$

This result may be the time-domain expression for the hole-burning experiments. These two extreme situations are schematically drawn in Figure 3. Figure 3 infers that the overlap of the laser-pulse spectrum with the absorption spectrum plays a role in the effective spectral width for the system excited by the nonimpulsive laser pulse.

Figure 4 shows the interference term of QI signals calculated for intermediate cases. The red sinusoidal curve of the QI signal was calculated for  $\gamma_g = 100 \text{ cm}^{-1}$  and  $\Gamma = 200 \text{ cm}^{-1}$ , while the blue one was calculated for  $\gamma_g = 200 \text{ cm}^{-1}$  and  $\Gamma = 100 \text{ cm}^{-1}$ . All the other parameters were common for the two calculations. The frequency of the QI signal is altered by the ratio of  $\gamma_g$  to  $\Gamma$  for the cases of nonzero detuning (e.g.,  $\omega_0 - \Omega \neq 0$ ). This dependence of



**Figure 3.** Limiting cases: impulsive laser (left) and quasi CW laser (right).



**Figure 4.** QI signals simulated for the intermediate cases. The parameters specific for each curve were  $\gamma_g = 100 \text{ cm}^{-1}$  and  $\Gamma = 200 \text{ cm}^{-1}$  (for the red curve),  $\gamma_g = 200 \text{ cm}^{-1}$  and  $\Gamma = 100 \text{ cm}^{-1}$  (for the blue curve). The common parameters for the two curves were  $\omega_0 = 25\,000 \text{ cm}^{-1}$ ,  $\Omega = 22\,000 \text{ cm}^{-1}$ ,  $\gamma_l = 100 \text{ cm}^{-1}$ , and  $\gamma^* = 25 \text{ cm}^{-1}$ .

the QI frequency  $\omega_a$  on  $\gamma_g$  can be usefully applied to the determination of an inhomogeneous decay component; that is, in the QI experiments, the precise determination of each frequency parameter,  $\omega_a$ ,  $\omega_0$ , and  $\Omega$ , will aid to deduce the inhomogeneous decay constant by the relation

$$\gamma_g = \left| \frac{\omega_0 - \omega_a}{\omega_a - \Omega} \right|^{1/2} \Gamma \quad (32)$$

which can be derived from eq 22b.

### 4. Summary

We have shown that the decays of the QI signal obtained by the nonimpulsive excitation can be written as the product of exponential decay and Gaussian decay, and the Gaussian decay constant is given by

$$\frac{1}{\gamma_a^2} \equiv \frac{\gamma_g^2 + \Gamma^2}{\gamma_g^2 \Gamma^2}$$

We have also shown that the frequency of the QI signal is given by

$$\omega_a \equiv \frac{\Gamma^2 \omega_0 + \gamma_g^2 \Omega}{\gamma_g^2 + \Gamma^2}$$

In general, it is often difficult to fit the optical absorption spectrum with Voigt functions in the frequency domain, because the Voigt function includes the convolution integral, and one often finds several parameter sets of the least-squared fits. This situation makes it difficult to separate homogeneous components from inhomogeneous components in the frequency-domain spectrum. In contrast, the fitting procedure is rather easier in the QI experiment, once the expression that includes the effect of laser-pulse width is given. This is because the homogeneous and inhomogeneous components are the simple product in the QI experiment. By analyzing the frequency-domain spectrum and the time-domain QI profile simultaneously (e.g., global fit), the reliable determination of homogeneous and inhomogeneous components of relaxations becomes possible.

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