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Hole Burning in the Resonance Fluorescence of Impurity Centers

by

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HOLE BURNING IN THE RESONANCE FLUORESCENCE OF IMPURITY CENTERS

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Abstract

The system of an impurity electron coupled to localized phonons is considered under the strong excitation of an external laser field. The resonance fluorescence spectrum is calculated for a two-level impurity atom. A hole and an antihole are found on the sidebands of the spectrum. Characteristic features of these novel structures are examined, and their physical origin is discussed.

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Resonance fluorescence (RF) has been a topic of wide interest. Since it often involves the interaction of intense laser fields with atomic electrons, it serves as an important example of quantum electrodynamics. The first complete treatment of the problem was given by Mollow¹ in 1969. Three peaks in the spectrum of the scattered light were predicted for an atom driven near resonance by a monochromatic classical electric field. The theory was confirmed experimentally a few years later.²

When the incident field is sufficiently weak, the scattering of photons from an atom is perfectly elastic and can be accurately described by classical electrodynamics. Increasing the field intensity leads to an appearance of an inelastic component consisting of a Lorentzian peak centered on the incident field frequency with a width of the natural line width γ . When the incident field is so strong that the Rabi frequency $R > \gamma$, the spectrum splits symmetrically into three peaks with two sidebands of width $3\gamma/2$ and a central peak of natural width at the frequency of the incident field. The displacement R of the sidebands from the center is a result of the dynamical ac Stark effect. There have also been many attempts to investigate the RF spectra with a noncontinuous wave,³ and a variety of peaked structures have been observed.

It has been known for a long time that spontaneous emission can be significantly influenced by the environment.⁴ When the emitting atom is situated near a solid surface⁵ or in a cavity,^{4,6} the emission line shape and position are qualitatively different from those from an isolated atom. More recently, similar behavior of spontaneous radiation following electron-hole recombination has been found in GaAs/AlGaAs double heterostructures.⁷ In the case of RF, the resulting spectrum is expected to depend upon the environment as well. In fact, an RF spectrum with asymmetric sidebands has been

predicted⁸ and experimentally verified⁹ for gas atoms colliding with foreign atoms. A subnatural or supernatural linewidth of the central peak has been predicted for a two-level atom in a squeezed vacuum.¹⁰ For an atom inside an optical cavity, the sideband width is found to be given by the atom-cavity coupling coefficient.¹¹ In addition, there has also been a theoretical investigation related to the surface-dressed RF spectrum.¹²

We study the RF spectrum, in this Letter, for an impurity center¹³ formed by an atom embedded in a semiconductor. The electron bound to the impurity center couples with both the external laser field and localized phonon modes. Studies of light interaction with impurity states in ruby has led to the first laser.

Localized phonon modes considered in this system represent oscillations of the impurity atom which are assumed to be harmonic.¹⁴ Their frequencies are normally far from the resonance of the plasma excitation, and hence the coupling between the atomic motion and electronic excitations is expected to play an important role in the light scattering process. The electron-phonon coupling represents one example of the interaction between boson and fermion fields and shows up in many different contexts in solid-state physics.¹⁵

Let us assume that the impurity center has only two nondegenerate states $|+\rangle$ and $|-\rangle$ with an energy separation ω_e . When the external laser field $E(t) = E_L e^{-i\omega_L t} + \text{c.c.}$ with $\omega_L \approx \omega_e$ splits each of the two states, and when the splitting is on resonance with one of the phonon modes, it is then found that phonons can burn a hole on one of the sidebands and an antihole on the other in the RF spectrum.

Let $a^\dagger$ (a) be the fermion raising (lowering) operator for the two-level atom and $b_i^\dagger$ (b_i) the boson creation (annihilation) operator for the i -th

phonon mode. Since ω_i is much lower than ω_e , the electron-phonon coupling will not induce an electronic transition, and hence the Hamiltonian of our interacting system can be written as

$$H = \omega_e a^\dagger a + \sum_j \omega_j b_j^\dagger b_j - [\mu a^\dagger E(t) + \mu^* a E^*(t)] - \sum_j \lambda_j a^\dagger a (b_j^\dagger + b_j) \quad (1)$$

where μ represents the matrix element of the electronic transition dipole and λ_j is the electron-phonon coupling constant. The number of phonon modes is usually limited in realistic systems,¹⁶ and it is sufficient to consider a single phonon mode. This simplification does not change the main physics involved in the problem. In fact, as we shall see later, only one phonon mode whose frequency coincides with the ac Stark splitting is very important. Therefore, we drop the subscript j for all phonon quantities from now on.

Similar to the Markoff approximation discussed in Ref. 1, we assume that the density matrix ρ of the system can be expressed as a direct product of density matrices for the electron and phonon subsystems, that is, $\rho = \rho_e \otimes \rho_b$. This implies that we can take the mean values of the operators involved and obtain their equations of motion from the Hamiltonian (1) through the normal procedure.¹⁷ Defining the mean values $\eta = \langle a^\dagger a \rangle$, $\alpha = \langle a \rangle$, and $\beta = \langle b \rangle$, we find the steady-state solutions in the rotating frame as

$$\eta_0^3 + \Delta \frac{\omega^2 + \gamma^2}{\omega \lambda^2} \eta_0^2 + \frac{1}{4} \left(\frac{\gamma_e^2}{4 + \Delta^2 + 2\Omega^2} \right) \left(\frac{\omega^2 + \gamma^2}{\lambda^2 \omega} \right)^2 \eta_0 - \frac{\Omega^2}{4} \left(\frac{\omega^2 + \gamma^2}{\lambda^2 \omega} \right)^2 = 0 \quad (2)$$

$$\alpha_0 = ikE_L(2\eta_0 - 1) / \left[-\frac{\gamma_e}{2} + i \left(\Delta + \frac{2\lambda^2 \omega \eta_0}{\omega^2 + \gamma^2} \right) \right] \quad (3a)$$

$$\beta_0 = i\lambda\eta_0/(i\omega + \gamma) \quad , \quad (3b)$$

where we have introduced phenomenologically the damping rates γ_e and γ to account for the energy dissipation, and have defined the Rabi frequency $\Omega = \mu E$ and detuning $\Delta = \omega_L - \omega_e$. The subscript 0 denotes the steady-state value of the corresponding quantity. It should be pointed out that only the positive real root of Eq. (2) is the physically meaningful expectation value of the upper level occupation probability. Since the coupling constant λ is generally much smaller than any other energy involved in the problem, and we only consider a strong excitation $\Omega \gg \lambda$ here, to solve for the spectrum we may use a linearization procedure to simplify the calculation.^{18,19} Similar to Refs. 13 and 19, we obtain the equations of motion for the deviation of the mean values from their corresponding steady-state values as

$$\dot{\delta\eta}(t) = -\gamma_e \delta\eta(t) - i\Omega^* \delta\alpha(t) + i\Omega \delta\alpha^*(t) \quad (4a)$$

$$\begin{aligned} \dot{\delta\alpha}(t) = & -2i\Omega^* \delta\eta(t) - \left[\frac{\gamma_e}{2} - i\Delta - i\lambda(\beta_0 + \beta_0^*) \right] \delta\alpha(t) \\ & + i\lambda\alpha_0 [\delta\beta(t) + \delta\beta^*(t)] \end{aligned} \quad (4b)$$

$$\dot{\delta\beta}(t) = i\lambda\delta\eta(t) - i(\omega - i\gamma)\delta\beta(t) \quad , \quad (4c)$$

where second- and higher-order terms are neglected.

The power spectrum of the scattered field $S(\nu)$ is the Fourier transform of the correlation function $\langle a^\dagger(t)a \rangle_0$, and it can be decomposed into coherent and incoherent parts as

$$S(\nu) = S_{\text{coh}}(\nu) + S_{\text{inc}}(\nu) \quad , \quad (5)$$

where

$$S_{\text{coh}}(\nu) = \langle a \rangle_0^2 \delta(\nu - \omega_L) \quad , \quad (6a)$$

$$S_{\text{inc}}(\nu) = \frac{1}{\pi} \int_{-\infty}^{\infty} dt \langle \delta a^\dagger(t) \delta a \rangle_0 \exp[-i(\nu - \omega_L)t] \quad . \quad (6b)$$

It is noted that Eqs. (6) have been written in the rotating frame. Since the coherent part is of no interest to us, we consider only S_{inc} which originates from inelastic scattering processes. According to quantum regression theory,^{20,21} the steady-state value $\langle \delta a^\dagger(t) \delta a \rangle_0$ can be obtained by making the following replacements in Eqs. (4) and relevant conjugate,

$$\delta \eta(t) \rightarrow \Delta \eta(t) = \langle [a^\dagger(t) a(t) - \langle a^\dagger a \rangle_0] [a - \langle a \rangle_0] \rangle_0 \quad (7a)$$

$$\delta \alpha^*(t) \rightarrow \Delta \alpha_2(t) = \langle [a^\dagger(t) - \langle a^\dagger \rangle_0] [a - \langle a \rangle_0] \rangle_0 \quad (7b)$$

$$\delta \alpha(t) \rightarrow \Delta \alpha_1(t) = \langle [a(t) - \langle a \rangle_0] [a - \langle a \rangle_0] \rangle_0 \quad (7c)$$

$$\delta \beta^*(t) \rightarrow \Delta \beta_2(t) = \langle [b^\dagger(t) - \langle b^\dagger \rangle_0] [a - \langle a \rangle_0] \rangle_0 \quad (7d)$$

$$\delta \beta(t) \rightarrow \Delta \beta_1(t) = \langle [b(t) - \langle b \rangle_0] [a - \langle a \rangle_0] \rangle_0 \quad , \quad (7e)$$

with the initial values $\Delta \eta(0) = -\eta_0 \alpha_0$, $\Delta \alpha_2(0) = \eta_0 - |\alpha_0|^2$, $\Delta \alpha_1(0) = -\alpha_0^2$, $\Delta \beta_2(0) = 0$ and $\Delta \beta_1(0) = 0$.

Although we only need the steady-state value of the correlation function $\Delta\alpha_2(t)$, we still have to solve the coupled Eqs. (4) with initial conditions specified above. This is done by means of the Laplace transform. The calculation is standard but tedious, and the result is

$$\Delta\alpha_2(p) = G(p)/D(p) \quad , \quad (8)$$

where the numerator and denominator are given respectively by

$$\begin{aligned} G(p) = & -\Delta\alpha_2(0)(p+\gamma_e)(p+w^*)(p+z_0)(p+z_0^*) \\ & -2\Omega^2(p+z_0)(p+z_0^*)[\Delta\alpha_1(0)+\Delta\alpha_2(0)] \\ & +2\Omega\lambda^2\omega[\alpha_0^*\Delta\alpha_1(0)+\alpha_0\Delta\alpha_2\omega] \\ & -2i\Delta\eta(0)(p+w^*)[\Omega(p+z_0)(p+z_0^*)-\lambda^2\omega\alpha_0^*] \quad , \end{aligned} \quad (9a)$$

$$\begin{aligned} D(p) = & -(p+\gamma_e)(p+w)(p+w^*)(p+z_0)(p+z_0^*) \\ & -4\Omega^2(p+z_0)(p+z_0^*)(p+\gamma_e/2) \\ & +2\Omega\lambda^2\omega[\alpha_0(p+w)+\alpha_0^*(p+w^*)] \quad . \end{aligned} \quad (9b)$$

with $z_0 = \gamma + i\omega$, $w = \frac{\gamma_e}{2} + i(\Delta + \lambda Q_0)$ and $Q_0 = \beta_0 + \beta_0^*$.

The denominator D is a fifth-order polynomial in p , and its zeros cannot be solved analytically in general. In our numerical study, however, we

find that the last term of (9b) is four orders of magnitude smaller than the first two terms and hence can safely be neglected when the conditions are such that $\Delta \ll \gamma_e$, $\gamma \gtrsim 0.025\gamma_e$ and $\lambda \ll \omega, \omega_e, \omega_L, \Omega$, which are satisfied in the present case. Thus we can factorize (9b) and rewrite

$$D(p) = -(p+z_0)(p+z_0^*)[(p+\gamma_e)(p+w)(p+w^*) + 4\Omega^2(p+\frac{\gamma_e}{2})] \quad (10)$$

Comparing the last factor of (10) with the power spectrum for a system that does not involve phonons,¹ we see that the only difference is the extra term $i\lambda Q_0$ in w and w^* . Physically, this term represents the energy correction due to the electron-phonon coupling. Since a third-order polynomial can always be factorized, the power spectrum (8) can be decomposed into five terms as

$$\begin{aligned} \Delta\alpha_2(p) &= \frac{A_1}{p-p_+} + \frac{A_2}{p-p_-} + \frac{A_3}{p-p_3} + \frac{A_4}{p+z_0} + \frac{A_5}{p+z_0^*} \\ &= h_1(p) + h_2(p) \quad , \end{aligned} \quad (11)$$

where h_1 and h_2 represent the first three and last two terms of the spectrum, respectively, and the numerators A_i are defined by

$$A_i = \lim_{p \rightarrow p_i} (p-p_i)G(p)/D(p) \quad , \quad i = 1, 2, 3, 4, 5 \quad (12)$$

For $\Omega \gg \gamma_e$, the three zeros are

$$p_{\pm} = -\frac{\gamma_e}{2} \frac{6\Omega^2 + (\Delta + \lambda Q_0)^2}{4\Omega^2 + (\Delta + \lambda Q_0)^2} \pm i\sqrt{4\Omega^2 + (\Delta + \lambda Q_0)^2} \quad , \quad (13a)$$

$$P_3 = -\frac{\gamma_e}{2} \frac{4\Omega^2 + 2(\Delta + \lambda Q_0)^2}{4\Omega^2 + (\Delta + \lambda Q_0)^2} \quad (13b)$$

Since $\Delta\alpha_2(p)$ is the Laplace transform of $\langle \delta a(t) \delta a \rangle_0$, and since $\langle \delta a(-t) \delta a \rangle_0^* = \langle \delta a(t) \delta a \rangle_0$, it is not difficult to show that¹

$$S(\nu) = \frac{2}{\pi} \text{Re} \{ \Delta\alpha_2[i(\nu - \omega_L)] \} \quad (14)$$

where we have replaced p by $i(\nu - \omega_L)$.

Since we are only interested in the relative values of the scattered field spectrum, we define for convenience the spectrum

$$g(\nu) = \pi S(\nu) = g_1(\nu) + g_2(\nu) \quad (15)$$

$$g_j(\nu) = 2 \text{Re} \{ h_j[i(\nu - \omega_L)] \} \quad , \quad j = 1, 2 \quad (16)$$

From the expressions (11)-(14), one can easily analyze the physical origin of the spectral shape. $g_1(\nu)$ represents the dominant part that originates from phonon-assisted optical dynamical splitting, and $g_2(\nu)$ represents the other part that is solely phonon-mediated. The former has three absorption peaks at $\nu = \omega_L$ and $\nu = \omega_L \pm [4\Omega^2 + (\Delta + \lambda Q_0)^2]^{\frac{1}{2}}$. This indicates that the dynamical splitting induced by the driving laser field for each of the two electron levels is $R = [4\Omega^2 + (\Delta + \lambda Q_0)^2]^{\frac{1}{2}}$, which depends not only on the intensity of the laser but also on the electron-phonon coupling strength. Evidently, stronger coupling leads to wider dynamical splitting, a fact that has never been reported in ordinary resonance fluorescence processes.^{1,19} The latter, $g_2(\nu)$,

yields a hole, an antihole or a dispersive structure at $\nu = \omega_L \pm \omega$. We believe that these structures of the spectrum are due to the absorption on creation of one phonon in the interaction. In what follows, we present some of our results computed numerically with the parameters $\gamma = 0.05$, $\lambda = 1$, $\omega = 10$ and $\Delta = 0$, similar to those in Refs. 13 and 14. The unit adopted in our calculation is such that $\gamma_e/2 = 1$.

The spectrum of scattered light for $\Omega = 5.12$ is shown in Fig. 1. The value of Ω is so chosen that the phonon-mediated hole and antihole appear at the peaks of the sidebands. The full spectrum calculated from (8) and (14) is plotted in 1(a), while $g_1(\nu)$ and $g_2(\nu)$ are plotted separately in 1(b). The figure demonstrates that the third term of Eq. (9) is indeed negligible for the case with specified parameters. The appearance of phonon-induced dips results in the asymmetric sidebands. The situation is basically the same as Ω changes. Our numerical study shows that the sidebands of $g_1(\nu)$ are farther away from the central peak as Ω increases, while the dip and cap of $g_2(\nu)$ remain at the same position.

We now turn our attention to the behavior of $g_2(\nu)$. Figure 2 shows the scattered light spectra $g_1(\nu)$ and $g_2(\nu)$ separately for different intensities of the driving field. When Ω is such that the Stark splitting is off resonance with the phonon frequency of $\omega \neq R$, the spectra are depicted in 2(a) and 2(c). The corresponding spectrum for the resonance case is shown in 2(b). It is seen that the phonon contribution in 2(b) is at least an order of magnitude stronger than those in 2(a) and 2(c). What has to be emphasized here is that the function $g_2(\nu)$ in Fig. 2 has been amplified by a factor of ten to make the structures in 2(a) and 2(c) clearly observable. In addition, we also find that the phonon-induced structures diminish as the side bands of $g_1(\nu)$ move farther away from the phonon resonance.

To study the physical origin of the hole and antihole, we look at the level splitting of the two-level atom. As shown schematically in Fig. 3, each level is split by the strong laser field into two with a distance R between them. The four energy levels of the electron-field system then provide three possible lines. The three spontaneous emission lines are represented by $g_1(\nu)$. When this splitting matches the phonon frequency ω , the phonons can excite the electron field system from $|2\rangle$ to $|1\rangle$, and hence results in reduced probability for transitions a and c but enhanced probability for transitions b and d. This explains why there is a dip on the left side band, a cap on the right side band, and the central peak remains unchanged. This research was supported in part by the Office of Naval Research and the National Science Foundation under Grant CHE-9016789.

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Figure Captions

1. Scattered light spectrum for $\Omega = 5.12$: (a) the whole spectrum $g(\nu)$ calculated from Eq. (8); (b) components from the dynamical level splitting $g_1(\nu)$ and phonon coupling $g_2(\nu)$, calculated from Eq. (11).
2. Components of the scattered light spectrum $g_1(\nu)$ and $g_2(\nu)$ calculated from Eq. (11) for different excitation field intensity: (a) $\Omega = 7.0$; (b) $\Omega = 5.12$; (c) $\Omega = 3.0$.
3. Schematic explanation for phonon-assisted spontaneous emission.

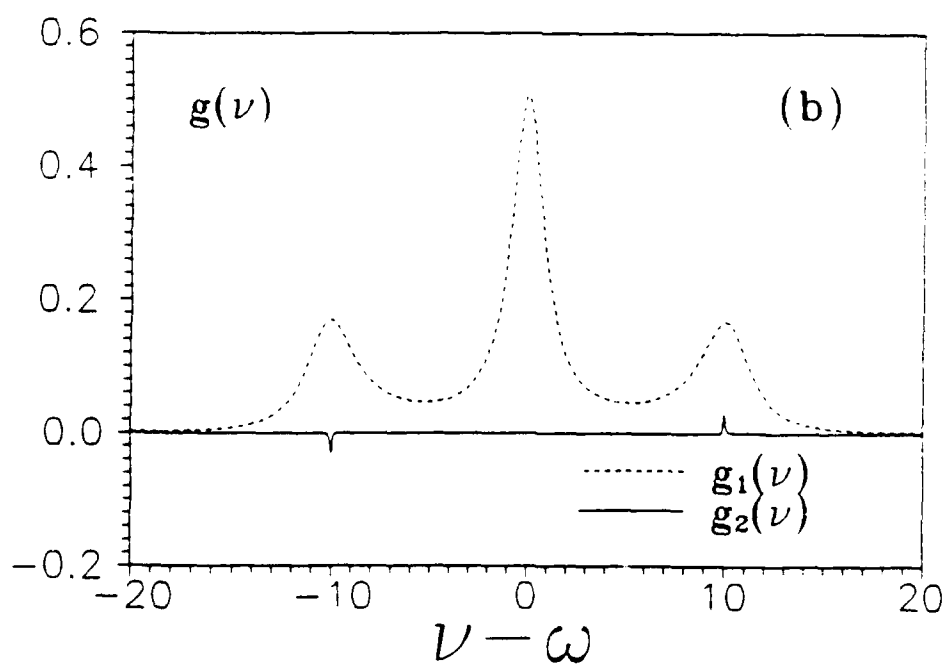
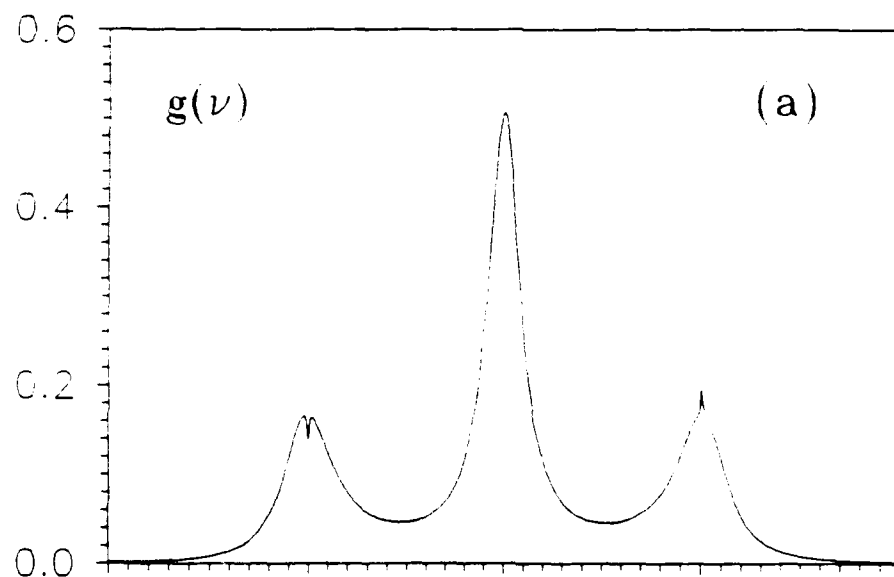


Fig. 2

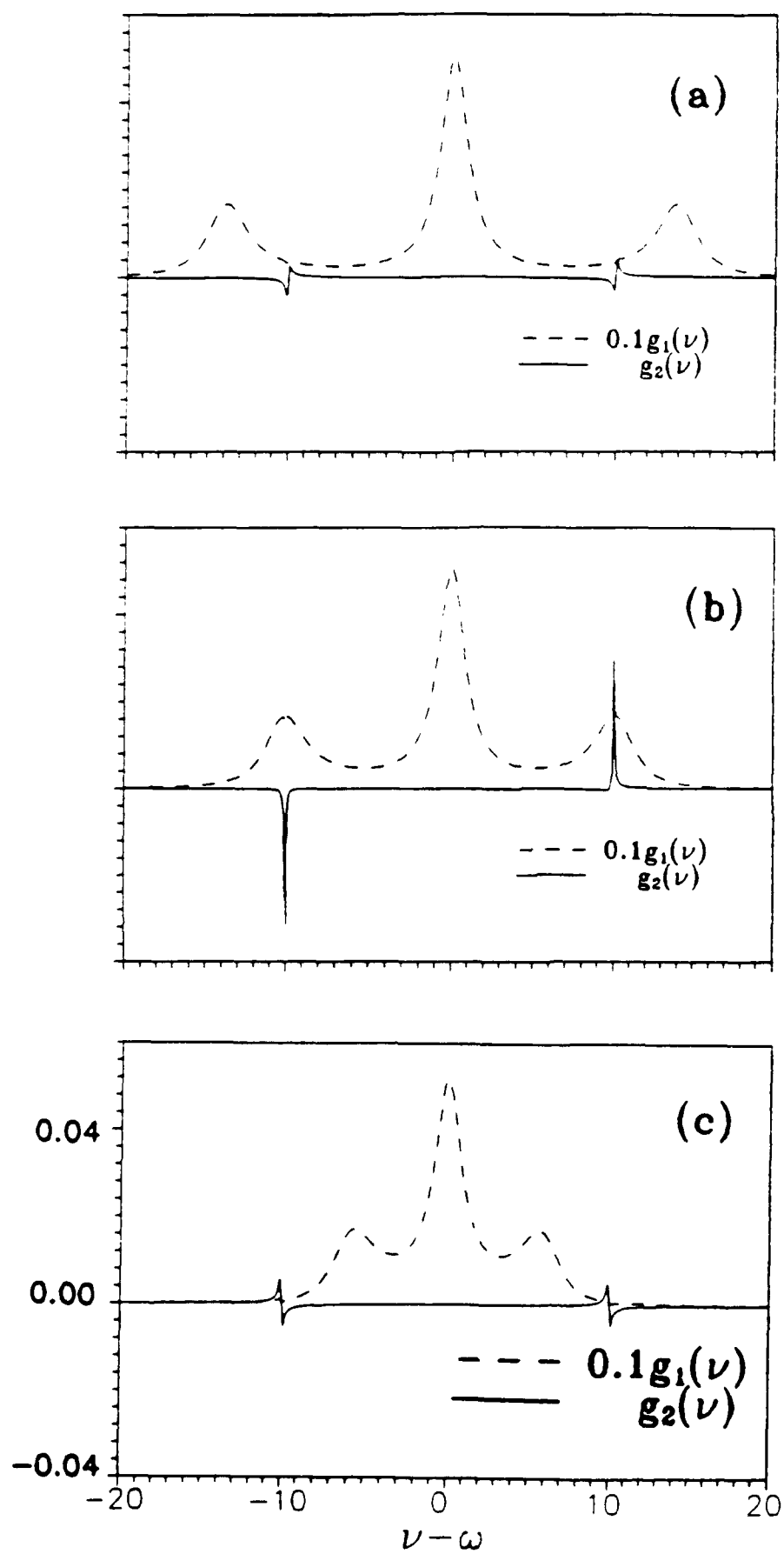


Fig. 3

