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Subnatural Linewidth Spectroscopy by Double Optical Resonance with Two-Photon Pumping

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(Received 18 December 1979)

A variant of the double-optical-resonance method is proposed in which the pumping step is a two-photon transition, either stimulated Raman scattering or two-photon absorption. It is shown that the probe linewidth can be minimized by proper choice of pump frequencies and assignment of pump beam propagation directions relative to the probe beam. The minimum linewidth is smaller than the value obtained by the usual one-photon pumping technique, and the method does not require any relations among level spacings.

PACS numbers: 33.70.Jg, 32.80.Kf, 33.80.Kn, 42.65.Cq

Linewidths narrower than the natural width have been observed by the use of Ramsey fringes,[1-7] time-delayed fluorescence,[8-10] and double optical resonance.[11-17] The last method was recently used by Hackel and Ezekiel[18,19] to obtain subnatural linewidths by two-step resonant scattering in I_2 vapor, with the level scheme shown in Fig. 1(a). In their experiment the transition $a \to b$ is driven by a pump field E of frequency Ω, and the transition $b \to c$ is probed by a collinearly propagating field E' of frequency Ω' which satisfies $|\mu_{bc}E'| \ll |\mu_{ba}E|$. In this Letter we present a variation on this idea in which the three levels are chosen so that the pumping transition $a \to b$ can be a two-photon process, either stimulated Raman scattering, Fig. 2(a), or two-photon absorption, Fig. 2(b). The principal advantage of this scheme is that the two pump frequencies are only constrained by one resonance condition; thus one frequency is freely adjustable. This freedom can be used to reduce the theoretically predicted linewidth below the minimum value possible for conventional double resonance with single-photon processes. In the interests of definiteness we will only describe the analysis for the level scheme of Fig. 1(a), but we should point out that two-photon pumping schemes, with the advantage just mentioned, cal also be devised for line-narrowing experiments suing the level schemes of Figs. 1(b) and 1(c).

The calculation of the probe linewidth for a two-photon pumping scheme is easily done by exploiting the formal similarity between the theory for Fig. 1(a) and the theory for Figs. 2(a) or 2(b). The object in each case is to find the steady-state solution for the three-level density matrix, this in turn determines the nonlinear susceptibility which is measured in experiments. This was

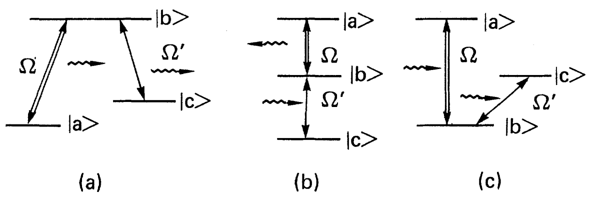

FIG. 1. Three-level arrays for double optical resonance. Double arrows represent pump waves, single arrows represent probe waves, and wavy arrows represent the relative propagation directions required for minimum linewidth.

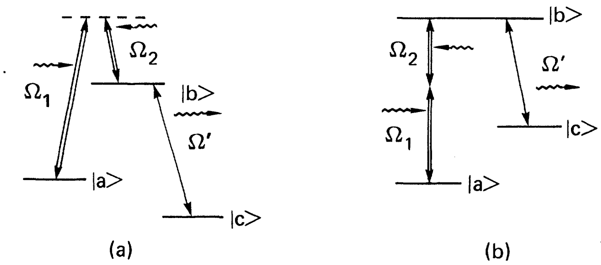

FIG. 2. Double optical resonance with two-photon pumping: (a) stimulated Raman scattering; (b) Two-photon absorption. Wavy arrows represent the relative propagation directions required for minimum linewidths.

done for Fig. 1(a) by solving the rotation-wave approximation to the Bloch equations.[20] The rotating-wave approximation is the simplest application of the general method of averages,[21, 22] which has also been used to deal with two-photon processes.[23] For Fig. 2 the method of averages yields Bloch equations which have exactly the same form as the equations for Fig. 1(a). In all three cases the Bloch equations have the form

$$i\dot{\rho}_{\alpha\beta}+\Delta_{\alpha\beta}\rho_{\alpha\beta} = i\gamma_{\alpha\beta}N_\alpha^{(0)}\delta_{\alpha\beta}+[V,\rho]_{\alpha\beta}\quad(\alpha,\beta=a,b,c),\quad(1)$$

where the γ's are phenomenological damping constants, and the $N^{(0)}$'s are equilibrium populations.

The one-photon processes of Fig. 1(a) are described by the matrix elements $V_{ba}=-\mu_{ba}E=V_{ab}^*$, $V_{bc}=-\mu_{bc}E'=V_{cb}^*$, where $\mu_{\alpha\beta}$ is a dipole matrix element, and the complex detunings

$$\Delta_{ba}=\Omega-kv-\omega_{ba}+i\gamma_{ab}=-\Delta_{ab}^*,$$
$$\Delta_{bc}=\Omega'-k'v-\omega_{bc}+i\gamma_{bc}=-\Delta_{cb}^*,$$
$$\Delta_{ac}=(\Omega'-k'v)-(\Omega-kv)-\omega_{ac}+i\gamma_{ac} =-\Delta_{ca}^*,\Delta_{\alpha\alpha}=i\gamma_{\alpha\alpha},$$

where $\omega_{\alpha\beta}\equiv\omega_\alpha-\omega_\beta$, and ω_α is an unperturbed energy. The wave vectors point in the z direction with z components k,k' and v is the z component of the atomic velocity. The stationary solution of Eq. (1) for each velocity must be averaged over the velocity distribution to get the susceptibility. In the limit of large Doppler broadening, low pressure, and weak pump field, i.e., $|\mu_{ba}E|\ll\gamma_{ab}$, the velocity integrals can be done analytically.[20] The total susceptibility is $\chi=\chi_1+\chi_3|E|^2$, and the experimentally measured third-order susceptibility $\chi_3=\chi_{3B}+\chi_{3N}$. The half width at half maximum due to χ_{3B} is of the order of the natural linewidth for any choice of relative propagation directions of the probe and pump beams. The term χ_{3N} vanishes for $k/k'<0$, i.e., counterpropagating pump and probe beams, but in the copropagating case $k/k'>0$, it yields a strong narrow line with width

$$\Gamma_N=\tfrac{1}{2}\gamma_c+\tfrac{1}{2}\beta\gamma_a+\tfrac{1}{2}|1-\beta|\gamma_b,\qquad(2)$$

where $\beta\equiv|k'/k|$, γ_α is the decay rate for level α, and we have used the approximation $\gamma_{\alpha\beta}=\frac{1}{2}(\gamma_\alpha+\gamma_\beta)$. For $\gamma_a=\gamma_c=0$, this becomes

$$\Gamma_N\cong\tfrac{1}{2}|1-\Omega'/\Omega|\gamma_b\cong\tfrac{1}{2}|\omega_{ac}/\omega_{bc}|\gamma_b,$$

and so for $\Omega'=\Omega$ a large reduction in linewidth is possible. These line narrowing effects have been attributed to frequency correlations imposed by conservation of energy and momentum in the compound $a\to c$ transition.[14]

The application of the method of averages to Fig. 2(a) yields Bloch equations described by $V_{ba}=Q_{ba}E_1E_2^*$,

$$\Delta_{ba}=\Omega_1-\Omega_2-(k_1-k_2)v-\omega_{ba}+i\gamma_{ba},$$
$$\Delta_{ac}=(\Omega'-k'v)-[\Omega_1-\Omega_2-(k_1-k_2)v]+i\gamma_{ac},$$

where the Raman matrix element is

$$Q_{ba}={\sum_\lambda}'\left[\frac{1}{\omega_{a\lambda}-\Omega_2}+\frac{1}{\Omega_1-\omega_{\lambda a}}\right]\mu_{b\lambda}\mu_{\lambda a},$$

and the primed sum runs over all states of the atom except for those giving resonant denominators. The pump beams are collinear with the probe beam and the amplitude, frequency and wave vector of the jth pump beam are, respectively, denoted by E_j, Ω_j, and k_j ($j=1,2$). The remaining matrix element V_{bc} and detuning Δ_{bc} are unchanged. Since the equations for Fig. 2(a) have the same form as those for Fig. 1(a), the susceptibility for Fig. 2(a) can be obtained from the results for Fig. 1(a) by the transformations $\mu_{ba}\to Q_{ba}$, $E\to E_1E_2^*$, $\Omega\to\Omega_1-\Omega_2$, and $k\to k_1-k_2$. This leads to $\chi=\chi_1+\chi_5|E_1E_2^*|^2$, and $\chi_5=\chi_{5B}+\chi_{5N}$.

Again χ_{5B} contributes a broad line for any choice of propagation vectors. The condition guaranteeing $\chi_{5N} \neq 0$ is given by $k/k' > 0 \rightarrow (k_1 - k_2)/k' > 0$. Choosing $k' > 0$ by convention reduces this to $k_1 - k_2 > 0$ which, combined with the resonance condition $\Omega_1 - \Omega_2 = \omega_{ba}$, requires $k_1 > 0$; that is, the higher-frequency pump beam must copropagate with the probe in order to produce a narrow line. The width of this line is given by Eq. (2) with $\beta = |k'/(k_1 - k_2)|$. If the second pump also copropagates, i.e., $k_2 > 0$, then $\beta = \Omega'/(\Omega_1 - \Omega_2) \cong \omega_{bc}/\omega_{ba}$ and the linewidth is

$$\Gamma_N \cong \tfrac{1}{2}\gamma_c + \tfrac{1}{2}(\omega_{bc}/\omega_{ba})\gamma_a + \tfrac{1}{2}|\omega_{ac}/\omega_{ba}|\gamma_b .$$

Thus with all three beams copropagating the linewidth is the same as that seen in the one-photon pumping scheme, Fig. 1(a). If the second pump counterpropagates, $k_2 < 0$, then $\beta = \Omega'/(\Omega_1 + \Omega_2)$ and the linewidth now depends on $\Omega_1 + \Omega_2$ which is not constrained by the Raman resonance condition. For $\gamma_a < \gamma_b$ the minimum of Γ_N occurs at $\beta = 1$, i.e., for $\Omega_1 + \Omega_2 = \Omega'$. This condition can be restated as $k_1 = k' + k_2$, which in turn implies that no net momentum is transferred to the atom. Combining this with the resonance conditions yields $\Omega_1 = \frac{1}{2}(\omega_{ba} + \omega_{bc})$ and $\Omega_2 = \frac{1}{2}\omega_{ac}$. Since Ω_2 is positive by convention, $\omega_{ac} = \omega_a - \omega_c > 0$ is a necessary condition for attaining the minimum linewidth. This corresponds to the configuration shown in Fig. 2(a). The conclusion is that the level scheme of Fig. 2(a) when pumped by stimulated Raman scattering, with the higher-frequency pump beam copropagating and the lower-frequency pump beam counterpropagating with respect to the probe beam, can produce a probe linewidth as small as $\frac{1}{2}(\gamma_a + \gamma_c)$. This result does not depend on the relative level spacings. This is another advantage over the one-photon pumping scheme, which is only useful for $|\omega_{ac}/\omega_{ba}| \ll 1$.

The corresponding results for Fig. 2(b) can be derived by the argument given above together with the replacements $\Omega_2 \rightarrow -\Omega_2$, $k_2 \rightarrow -k_2$, and $E_2^* \rightarrow E_2$. The required choice of propagation vectors is the same in both cases, but the condition for attaining the minimum linewidth is changed to $\omega_{ac} < 0$, corresponding to the level scheme in Fig. 2(b).

The narrow linewidths shown above are produced by frequency correlations which are possible only for a narrow band in the atomic velocity distribution. In this connection it may be useful to point out that the Doppler-free, three-photon method proposed by Cagnac, Grynberg, and Biraben[24] can produce the narrow width γ_{ac} without any restriction on atomic velocities. This is done by (i) detuning the pump lasers so that there is no resonant intermediate level $|b\rangle$, (ii) imposing a three-photon resonance condition for the transition $a \rightarrow c$, and (iii) choosing a noncollinear geometry for the three propagation vectors which leads to an exact cancellation of the overall Doppler shift. Since all levels other than $|a\rangle$ and $|c\rangle$ are virtual and Doppler broadening is eliminated, the width of the three-photon transition is γ_{ac}. The ability to use all the atoms rather than a small fraction can be an advantage for the three-photon Doppler-free method. On the other hand, the double-resonance method uses a collinear beam geometry and produces an enhanced signal strength by virtue of the resonant intermediate level $|b\rangle$. In many cases the resonant enhancement can more than compensate for the small fraction of useful atoms.

The utility of sharp spectral lines for Doppler-free spectroscopy, study of collisional effects, and frequency standards is well known. In this Letter we have proposed to produce such lines by a modified optical double-resonance technique using a two-photon pumping process. This method has two important advantages: (i) The probe linewidth can be minimized by adjusting the pump frequencies; (ii) no accidental coincidences between level spacings, e.g., $\omega_{ba} = \omega_{bc}$ for Fig. 1(a), are required. The importance of these features is that they make line narrowing by double resonance applicable to many more atomic and molecular targets. This should facilitate the establishment of frequency standards in a large range of the electromagnetic spectrum and also allow more extensive high-precision measurements of collisional effects.

These advantages are obtained at the cost of a more elaborate pumping scheme requiring two tunable lasers which may have to operate in widely separated spectral regions. We should also point out that the scheme does not escape the general limitations of line narrowing by double resonance. In particular, the use of the compound transition $a \rightarrow b \rightarrow c$ means that the narrow lines cannot be used in absolute determinations of level splittings with uncertainties less than the natural widths. With these reservations in mind we believe that the two-photon pumping scheme will provide a flexible and general method for the generation of subnatural linewidths.

This work was performed under the auspices of the U. S. Department of Energy by the Lawrence Livermore Laboratory under Contract No. W-

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