Coherent Transient Multiphoton Scattering in a Resonant Two-Level System

Naohiro Tan-no and Kazuhide Ohkawara
Department of Electronics, Yamagata University, Yonezawa 992, Japan

and

Humio Inaba
Research Institute of Electrical Communication, Tohoku University, Sendai 980, Japan
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We report theoretically and experimentally a new class of multiphoton scattering in the coherent transient regime associated with a resonant two-level system that is excited by two noncollinear laser beams. We find that its excitation, described by the nonperturbative solutions for the optical Bloch equation, gives rise to a detectable photon scattering of up to 27th-order wave-vector coupling in resonant sodium vapor.

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There have been many interesting studies on various coherent transient phenomena arising from the interaction between coherent optical fields and a two-level system in the limit of homogeneous relaxation time. Such modern phenomena[1-4] are generally understood by their optical Bloch equation[5] in combination with the Maxwell equation. On the other hand, classical higher-order processes, which are described by the perturbation method in terms of a power series of optical fields,[6] have already offered various techniques for spectroscopy. Furthermore, multiphoton scattering in solids[7] and in a gas[8] can be interpreted by the nonlinearity based on perturbation analysis.

In this report[9] we present an entirely novel class of multiphoton coupling in coherent transient interaction with a resonant two-level system that is coherently excited by two noncollinear beams of the same frequency field. This coherent transient multiphoton scattering is analyzed by solving the optical Bloch equation. Experimentally we find that a resonant excitation of atoms in a gaseous sample leads to the observation of up to 27th-order scattering with a momentum coupling of $(14\vec{k}_2 - 13\vec{k}_1)\hbar$ and also of anomalous momentum scattering such as $\frac{1}{2}(\vec{k}_1 + \vec{k}_2)\hbar$, etc.

We consider the coherent interaction of atoms with two optical fields coming from a laser oscillator. The two fields with noncollinear wave vectors cross each other at the atomic system with a small angle. Here, we take into account the two linearly polarized fields $\vec{E}_1$ and $\vec{E}_2$ which are parallel to each. Their total is

$$\vec{E} = \vec{E}_1 + \vec{E}_2 = \vec{e}\mathcal{E}_1 \cos\Phi_1 + \vec{e}\mathcal{E}_2 \cos\Phi_2, \tag{1}$$

with $\Phi_i = \omega t - k_i r$ $(i = 1, 2)$, where $\mathcal{E}_i$ and $\vec{e}$ are the amplitude of the field and the basic vector, respectively. Using the rotating-wave approximation and an unitary transformation, we obtain an effective Hamiltonian which does not depend on the time explicitly. Then, the equation of motion for the density matrix is given and is transformed into the optical Bloch equation as follows

$$\partial\vec{\rho}/\partial t = \vec{\Omega} \times \vec{\rho}, \tag{2}$$

with the Bloch vectors $\vec{\rho}(u, v, w)$ and $\vec{\Omega}(-\lambda_2, -\lambda_1, \Delta\omega)$; $\lambda_1 = (\chi_1 - \chi_2)\sin\delta$, $\lambda_2 = (\chi_1 + \chi_2)\cos\delta$, $\chi_i = p_{12}\mathcal{E}_i/\hbar$, p_{12} is the matrix element of dipole moment, $\delta = \frac{1}{2}(\vec{k}_2 - \vec{k}_1)\vec{r}$, $\Delta\omega = \Omega_{12} - \omega$, and Ω_{12} is the transition frequency.

Now, considering the constant field amplitudes $\mathcal{E}_{10}$ and $\mathcal{E}_{20}$, we can easily obtain the solutions for the optical Bloch equation. Furthermore, in order to simplify the problem, we consider only the case of equal amplitudes $\mathcal{E}_{10} = \mathcal{E}_{20} = \mathcal{E}_0$. Thus, a macroscopic polarization is obtained by means of an inverse unitary transformation. We can define a Rabi frequency Ω from the solution as

$$\Omega = \{\Delta\omega^2 + 4[(p_{12}/h)\mathcal{E}_0]^2 \cos^2\delta\}^{1/2}. \tag{3}$$

It is worthwhile to note that the present Rabi frequency depends on a degree of wave-vector coupling $\vec{k}_2 - \vec{k}_1$ and spatial position $\vec{r}$. Accordingly, a distinctive feature of this coherent interaction is that an atom located at a position experiences an inherent phase of two-beam interference which differs from that for adjacently located atoms. For anticollinear beams $(\vec{k}_1 = -\vec{k}_2)$, its excitation scheme seems to resemble that giving rise to a transient standing wave which contributes to the standing-wave echo formation as reported by Le Gouët and Berman[10] and Mossberg *et al.*[11]

Here, we consider the exact resonance case. Then, the macroscopic polarization can be written as

$$P(\vec{r},t) = -N_0 p_{12}\,\mathrm{Re}\{i\sin[\theta\cos\tfrac{1}{2}(\vec{k}_2-\vec{k}_1)\vec{r}]\exp[-i\,\tfrac{1}{2}(\vec{k}_1+\vec{k}_2)\vec{r}+i\omega t]\}$$
$$= N_0 p_{12}\,\mathrm{Re}\Big[i\sum_{n=0}^{\infty}(-1)^{n+1}J_{2n+1}(\theta)\Big(\exp\{-i[(n+1)\vec{k}_1-n\vec{k}_2]\vec{r}\}+\exp\{-i[(n+1)\vec{k}_2-n\vec{k}_1]\vec{r}\}\Big)\exp(i\omega t)\Big]. \quad (4)$$

Here, we define an area $\theta = 2p_{12}\tau\mathcal{E}_0/\hbar$, with τ the pulse width; J_{2n+1} is the Bessel function of order $2n+1$. When one expands, for example, only the lower-order terms, a novel aspect is revealed: The first term corresponds to the sum of the phase factors of the two incident plane-wave fields, but its amplitude is modulated as the Bessel function of order 1. The second term originates from the degenerate three-wave parametric coupling. Further higher-order terms appear to be related to the fifth- and seventh-order photon coupling, respectively, and so on. It is to be noted that each amplitude is more complicated because the Bessel function is expanded in terms of a power series of the area θ, and therefore an order of the wave-vector coupling as recognized from the phase factor does not coincide with that of the product of electric field amplitudes in this expansion. Consequently, the expansion of the macroscopic polarization entirely differs from that for the conventional analysis of the optical nonlinearity based on the perturbation approach. Since we do not know how to specify an order of such nonlinear coupling, we simply choose the order of the wave-vector coupling as the order.

When slightly detuning the applied frequency from the exact resonance and assuming the magnitude to be $\Delta\omega\tau \leq \theta$, the macroscopic polarization is given approximately by the following expansion:

$$P(r,t) \propto J_0(\theta)\exp[-i\,\tfrac{1}{2}(\vec{k}_1+\vec{k}_2)\vec{r}]$$
$$+\tfrac{1}{2}\sum_{n=1}^{\infty}(-1)^n J_{2n}(\theta)\Big(\exp\{-i[(n+\tfrac{1}{2})\vec{k}_1-(n-\tfrac{1}{2})\vec{k}_2]\vec{r}\}+\exp\{-i[(n+\tfrac{1}{2})\vec{k}_2-(n-\tfrac{1}{2})\vec{k}_1]\vec{r}\Big). \quad (5)$$

It should be noted that the present equation reveals the appearance of anomalous scattering associated with the phase factors accompanied with half-integer wave-vector coupling. Such a new type of half-integer wave-vector coupling has never been observed to our knowledge, but it is easily understood by considering a formation of phase grating due to the two-beam interference and the conventional parametric photon coupling. For the present case, each signal wave should be observed in half-angle direction between the mth- and the $(m+1)$th-order signal waves.

To demonstrate the existence of the coherent resonant multiphoton scattering, as analyzed above, we have performed an experiment on the 589.6 nm D_1 line transition of atomic sodium vapor. Linearly polarized exciting pulses were obtained from the output of a N_2-laser–pumped dye-laser system by virture of a Glan-prism polarizer. A single longitudinal-mode dye-laser pulse with a 0.03 cm^{-1} bandwidth and a 6 nsec pulse width was suitably divided into two beams by a beam splitter. The two beams crossed each other with a small angle φ of approximately 3 mrad, without a delay time, in a 2-cm-long sodium vapor cell of the heat-pipe type maintained at 500 K. The two beams were collimated with about 1 mm diameter. Each beam had 10 $\mathrm{kW/cm^2}$ peak power and was suitably reduced with use of filter attenuators. The transmitted laser beams and the scattered signals were detected with a photomultiplier detector through a 100-μm-width slit placed in front of the detector. The detector system was smoothly movable in both directions of scattering angle. A signal integrator and a recorder were employed to process and display scattered signal intensities as a function of scattered positions.

Figure 1 presents a typical photograph of the far-field pattern of the scattered light for an exact resonance. In this pattern, two bright spots near the center show the two transmitted beams and a sequence of weak spots on both sides reveals the third- and fifth-order scatterings. Since signals of the higher-order scattering were weak, the film sensitivity was not enough for their display. However, the detector system scanned along the scattering angle direction easily explored signals of the higher-order scattering as shown in Fig. 2.

A number attached to each signal peak in Fig. 2 indicates the order of the wave-vector coupling at every integer multiple of the angle φ. For instance, "27" means a wave-vector coupling scheme described by $14\vec{k}_1 - 13\vec{k}_2$, which was the

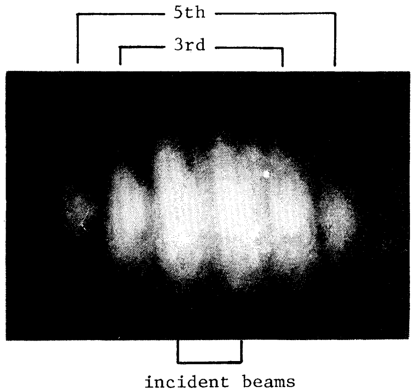

FIG. 1. A typical photograph of the far-field pattern of scattered and transmitted laser light for exact resonance.

highest-order coupling process observed in our experiment.

Remarkable reduction of the observed scattered light intensity by about a factor of 2 in the higher-order coupling can be explained by a phase mismatching due to the long cell length.

Figure 3 shows a typical photograph of the observed scattering pattern for near resonance at a detuned frequency of $\Delta\omega = 0.03\ \mathrm{cm}^{-1}$. We can see easily from this pattern a central, vertically long bright spot that gives evidence of the anomalous half-integer wave-vector coupling. A signal intensity trace of this scattering is shown in Fig. 4. In this figure, smaller signals appeared at every multiple of the half-integer angle between larger peaks, corresponding to light scattered by the half-integer wave-vector coupling scheme. We note that it was difficult to detect the multiple-phase scattering higher than the eleventh order, since it was caused by the detuning of the laser

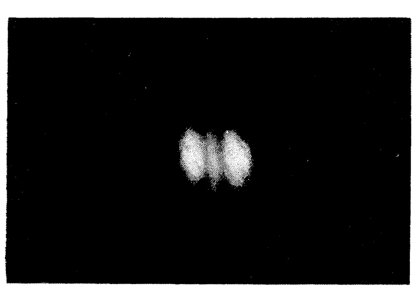

FIG. 3. A typical photograph of the far-field pattern of scattered and transmitted light for the near-resonance at the detuning frequency $\Delta\omega = 0.03\ \mathrm{cm}^{-1}$.

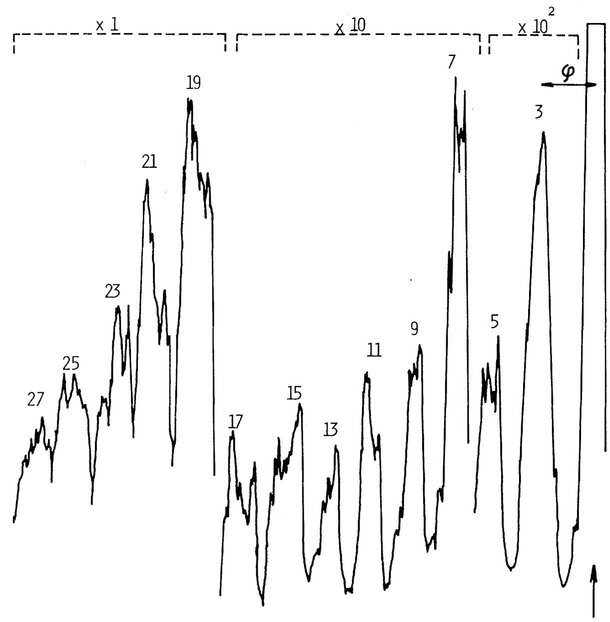

FIG. 2. A signal intensity trace of the observed scattered light as a function of the scattered position. An arrow indicates one of the transmitted laser beams.

frequency.

To summarize, we have shown that various wave-vector coupling schemes can induce transient higher-order multiphoton scattering in a gaseous sample by the resonant excitation of two

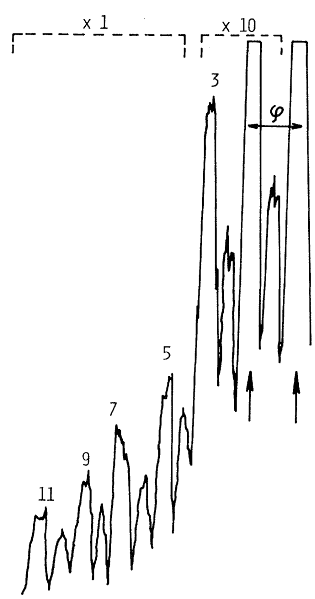

FIG. 4. A signal intensity trace of the observed scattering of the half-integer wave-vector coupling, corresponding to Fig. 3, for near resonance.

noncollinear beams. The theoretical analysis agrees fairly well with the observed results. Thus, this clearly demonstrates the validity of the analysis which does not involve any perturbation method as is conventionally used in classical nonlinear optics. The present analysis and experimental technique should be applicable to other phenomena such as free-induction decay, coherent Raman scattering, two-photon resonance and self-induced transparency, etc.

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[1]R. G. Brewer and R. L. Shoemaker, Phys. Rev. Lett. 27, 631 (1971), and Phys. Rev. A 6, 2001 (1972).

[2]N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Lett. 13, 567 (1964); I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. 141, 391 (1966).

[3]D. Grischkowsky and J. A. Armstrong, Phys. Rev. A 6, 1566 (1972).

[4]S. L. McCall and E. L. Hahn, Phys. Rev. Lett. 18, 908 (1967), and Phys. Rev. 183, 457 (1969).

[5]J. H. Eberly and L. Allen, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1974).

[6]N. Bloembergen, *Nonlinear Optics* (Benjamin, New York, 1965).

[7]W. Yu and R. R. Alfano, Phys. Rev. A 11, 188 (1975).

[8]C. V. Heer and N. C. Griffen, Opt. Lett. 4, 239 (1979).

[9]It is our note that our first observation on the present transient resonant multiphoton scattering was reported in Proceedings of the Annual Meeting of the Physical Society of Japan, Ohsaka, 31 March 1979 (unpublished).

[10]J.-L. LeGouët and P. R. Berman, Phys. Rev. A 20, 1105 (1979).

[11]T. W. Mossberg, R. Kachru, E. Whittaker, and S. R. Hartmann, Phys. Rev. Lett. 43, 851 (1979).

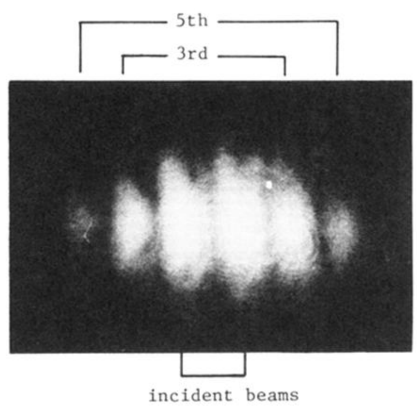

FIG. 1. A typical photograph of the far-field pattern of scattered and transmitted laser light for exact resonance.

FIG. 3. A typical photograph of the far-field pattern of scattered and transmitted light for the near-resonance at the detuning frequency $\Delta\omega = 0.03\ \mathrm{cm}^{-1}$.