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Resonance fringes in the two-photon absorption with a Doppler shift

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The two-photon absorption initiated by two coherent beams is considered. It is shown that the resonance fringe variation, which appeared in the Doppler-free two-photon absorption of pulsed laser beams, can also be observed in absorption with a Doppler shift.

Current research activities in the two-photon absorption of multiple coherent beams have been centered mainly on the Doppler-free processes. The chief characteristic of this process is that after absorbing two opposite-momentum photons, the atom's momentum remains unchanged. In an experiment proposed by Li,[1] the Doppler-free two-photon absorption is initiated by two crossed laser beams. It was shown that the atomic transition rate depends on the relative phase and intersecting angle of these coherent beams. Through a study of these dependences, one can obtain a direct mechanism for revealing the phase of the two-photon Doppler-free transition amplitude.

Experimental efforts on the Doppler-free two-photon absorption of coherent beams have been centered on high-resolution spectroscopy. These experiments can be classified either as the separated-beam or crossed-beam experiments.[2] In the separated-beam experiment proposed and performed by Chebotayev *et al.*,[3] atoms interact with parallel coherent laser beams. The atomic absorption depends on the external phase induced by the spatial separation; this dependence provides a mechanism for overcoming transit-time broadening and obtaining optical Ramsey fringes. The observation of these fringes leads to the improvement of spectral resolution. In the crossed-beam experiments of Teets *et al.*,[4] and Salour *et al.*,[5] a pulsed laser is used and beams are overlaid. In this case the atomic absorption depends on the external phase induced by the path length difference of laser pulses. This dependence provides a mechanism for overcoming laser pulse broadening and obtaining resonance fringe variation.

The above-mentioned transit-time broadening and pulse broadening are associated with the transverse and longitudinal uncertainties of the laser beam momentum, respectively. The underlying mechanics in the separated-beam and crossed-beam experiments for overcoming these uncertainties are different. The difference has already been discussed in a recent publication.[2] In this paper I shall consider a crossed-beam experiment of two-photon absorption with a Doppler shift. It will be shown that the resonance fringe variation observed in the Doppler-free two-photon absorption of pulsed laser beams can likewise be observed in the two-photon absorption with Doppler shift.

Let us consider a gas of three-level atoms resonantly interacting with two laser beams from independent sources. Initially, atoms are excited by the laser beam 1 from the ground state 1 to the intermediate state 0. Then, they are further excited by the laser beam 2 to the upper state 2, as shown in Fig. 1. The experimental arrangement for the process under consideration is depicted in Fig. 2. The independent laser beams are propagated in opposite directions. Each beam is split to form a detour and recombined again before entering the atomic cell. This is accomplished through the use of half-silvered mirrors SM and reflecting mirrors M. Optical isolators OW are used to reduce mutual disturbance between laser sources. A detector is mounted above the atomic cell to detect the fluorescence from the upper state 2. Laser beams have wave vectors $\vec{k}_1$ and $\vec{k}_2$ with

$$|\vec{k}_2| = k_1 = \omega_1/c \tag{1}$$

and

$$|\vec{k}_2| = k_2 = \omega_2/c\ , \tag{2}$$

where ω_1 and ω_2 are their frequencies and c is the velocity of light. After absorbing a photon from laser beam 1, an atom in the atomic cell changes its momentum from $\vec{p}_1$ to $\vec{p}_0$,

$$\vec{p}_0 = \vec{p}_1 + \hbar\vec{k}_1 \tag{3}$$

and its internal energy from ϵ_1 to ϵ_0,

$$\hbar\omega_{10} = \epsilon_0 - \epsilon_1\ . \tag{4}$$

The transition amplitude has the form

$$g_1(\omega_{10})(1 + a_1 e^{ik_1L_1})\langle\vec{p}_0, \epsilon_0|t|\vec{p}_1, \epsilon_1; \vec{k}_1\rangle\ , \tag{5}$$

where $g_1(\omega_{10})$ describes the atomic line profile. The real quantities a_1 and L_1 are the relative strength and the path-length difference of the laser beam 1 in detour 1, $\langle\vec{p}_0, \epsilon_0|t|\vec{p}_1, \epsilon_1; \vec{k}\rangle$ is the

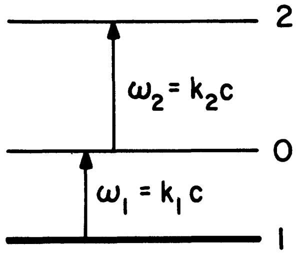

FIG. 1. A three-level atomic system.

reduced transition amplitude. The excited atom in the intermediate state 0 is further excited by the laser beam to the upper state 2. The atom changes its momentum from $\vec{p}_0$ to $\vec{p}_2$,

$$\vec{p}_2 = \vec{p}_0 + \hbar\vec{k}_2 , \tag{6}$$

and its internal energy from ϵ_0 to ϵ_2,

$$\hbar\omega_{20} = \epsilon_2 - \epsilon_0 . \tag{7}$$

The upper state 2 is reached from the lower state 1 through the absorption of two photons with momenta $\hbar\vec{k}_1$ and $\hbar\vec{k}_2$. The transition amplitude has the form

$$g_2(\omega_{20})g_1(\omega_{10})(1 + a_2 e^{ik_2L_2})(1 + a_1 e^{ik_1L_1}) \times \langle \vec{p}_2, \epsilon_2 | t | \vec{p}_0, \epsilon_0; \vec{k}_2 \rangle \langle \vec{p}_0, \epsilon_0 | t | \vec{p}_1, \epsilon_1; \vec{k}_1 \rangle , \tag{8}$$

where $g_2(\omega_{20})$ describes the atomic line profile of the upper state 2. Similarly the real quantities a_2 and L_2 are the relative strength and the path-length difference of the laser beam 2 in detour 2. $\langle \vec{p}_2, \epsilon_2 | t | \vec{p}_0, \epsilon_0; \vec{k}_2 \rangle$ is the reduced transition amplitude for the transition from the intermediate state 0 to the upper state 2.

The amplitude in Eq. (8) is for the atomic transition under two continuous-wave laser beams. The transition amplitude under two pulsed laser beams can be written as

$$\int dk_2' dk_1' G_2(k_2') G_1(k_1') g_2(\omega_{20}) g_1(\omega_{10}) \times (1 + a_2 e^{ik_2'L_2})(1 + a_1 e^{ik_1'L_1}) \times \langle \vec{p}_2, \epsilon_2 | t | \vec{p}_0, \epsilon_0; \vec{k}_2' \rangle \langle \vec{p}_0, \epsilon_0 | t | \vec{p}_1, \epsilon_1; \vec{k}_1' \rangle , \tag{9}$$

where $G_2(k_2')$ and $G_1(k_1')$ describe the profiles of laser pulses. To simplify the calculation, functions $G_2(k_2')$, $G_1(k_1')$, $g_2(\omega_{20})$, and $g_1(\omega_{10})$ are assumed to have Gaussian shapes:

$$G_2(k_2') = \exp[-A_2(k_2' - k_2)^2] , \tag{10}$$

$$G_1(k_1') = \exp[-A_1(k_1' - k_1)^2] , \tag{11}$$

$$g_2(\omega_{20}) = \exp[-B_2(\omega_2' + \vec{p}_0^2/2\hbar m - (\vec{p}_0 + \hbar\vec{k}_2')^2/2\hbar m - \omega_{20})^2] , \tag{12}$$

and

$$g_1(\omega_{10}) = \exp[-B_1(\omega_1' + \vec{p}_1^2/2\hbar m - (\vec{p}_1 + \hbar\vec{k}_1')^2/2\hbar m - \omega_{10})^2] , \tag{13}$$

where

$$\omega_1' = k_1'c , \tag{14}$$

$$\omega_2' = k_2'c , \tag{15}$$

and m is the mass of the atom. Constants A_i and B_i for $i = 1, 2$ are related to the durations of laser pulses and to the lifetimes of atomic states. After performing integrations in Eq. (9) and eliminating irrelevant factors, one has

$$\begin{aligned} &\langle \vec{p}_2, \epsilon_2 | t | \vec{p}_0, \epsilon_0; \vec{k}_2 \rangle \langle \vec{p}_0, \epsilon_0 | t | \vec{p}_1, \epsilon_1; \vec{k}_1 \rangle \\ &\quad \times \prod_{j=1}^{2} \exp\left[-\frac{A_j B_j c^2}{A_j + B_j c^2}\left(\frac{\omega_{j0}}{c} - k_j\right)\left(\frac{\omega_{j0}}{c} - k_j - (-1)^j 2\frac{v}{c}\frac{B_j c^2 \omega_{j0}/c + A_j k_j}{A \ + B_j c^2}\right)\right] \\ &\qquad \times \left(1 + a_j \exp\left[-\frac{L_j^2}{4(A_j + B_j c^2)}\left(1 - (-1)^j 2\frac{v}{c}\frac{B_j c^2}{A_j + B_j c^2}\right)\right]\right. \\ &\qquad\quad \left.\times \exp\left\{i\frac{L_j}{A_j + B_j c^2}\left(B_j c^2 \frac{\omega_{j0}}{c} + A_j k_j\right) - i(-1)^j \frac{v}{c}\frac{B_j c^2 L_j}{(A_j + B_j c^2)^2}\left[B_j c^2 \frac{\omega_{j0}}{c} - A_j\left(\frac{\omega_{j0}}{c} - 2k_j\right)\right]\right\}\right), \end{aligned} \tag{16}$$

where high-order Doppler-shift contributions are neglected, and only the first-order contribution is kept. v is the projection of the atomic velocity in the direction of the wave number $\vec{k}_1$. In reaching Eq. (16), it is assumed that the reduced transition amplitudes are slowly varying functions of wave vectors $\vec{k}_1$ and $\vec{k}_2$.

From Eq. (16), the experimentally observed transition rate can be obtained. It is accomplished through squaring the transition amplitude and then taking an average with respect to the atomic velocity distribution $f(v)$. The resonance fringe variation in the atomic absorption of coherent beams is a variational dependence of the transition rate on the path-length differences. The dependence comes from the last term in Eq. (16). However, this term contains a velocity-dependent Doppler phase. For arbitrary path-length differences L_1

and L_2, the random atomic motion through the Doppler phase will suppress the fringe variation in the atomic transition rate. To overcome such random motion, one has to set

$$\frac{B_1c^2L_1}{(A_1+B_1c^2)^2}\left[B_1c^2\frac{\omega_{10}}{c}-A_1\left(\frac{\omega_{10}}{c}-2k_1\right)\right] = \frac{B_2c^2L_2}{(A_2+B_2c^2)^2}\left[B_2c^2\frac{\omega_{20}}{c}-A_2\left(\frac{\omega_{20}}{c}-2k_2\right)\right]. \qquad (17)$$

From Eqs. (16) and (17), the resonance fringe variation can be expressed as

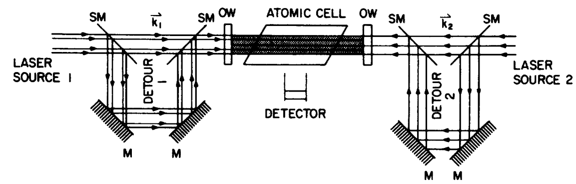

FIG. 2. The experimental setup of the proposed experiment. Half-silvered mirrors SM are used to split and combine laser beams. M denotes reflecting mirrors and OW optical isolators. Laser beams from independent sources are counterpropagating through the atomic cell. The detector is mounted above the atomic cell.

$$\cos\left[\sum_{j=1}^{2}\frac{L_j}{A_j+B_jc^2}\left(B_jc^2\frac{\omega_{j0}}{c}+A_jk_j\right)\right] \times\int dv\, f(v)\exp\left[-\sum_{j=1}^{2}\frac{L_j^2}{4(A_j+B_jc^2)}\left(1-(-1)^j2\frac{v}{c}\frac{B_jc^2}{A_j+B_jc^2}\right)\right] \times\left\{\sum_{j=1}^{2}\exp\left[-\frac{A_jB_jc^2}{A_j+B_jc^2}\left(\frac{\omega_{j0}}{c}-k_j\right)\left(\frac{\omega_{j0}}{c}-k_j-(-1)^j2\frac{v}{c}\frac{B_jc^2\omega_{j0}/c-A_jk_j}{A_j+B_jc^2}\right)\right]\right\}^2. \qquad (18)$$

When the laser pulse durations are much shorter than the lifetimes of the atomic states

$$A_j \ll B_jc^2, \quad \text{for } j=1,2. \qquad (19)$$

Equations (17) and (18) can be simplified to

$$L_1\omega_{10}=L_2\omega_{20}, \qquad (20)$$

and

$$\cos\left(\frac{L_1\omega_{10}}{c}+\frac{L_2\omega_{20}}{c}\right)\int dv\, f(v)\exp\left[-\frac{L_1^2}{4B_1c^2}\left(1+2\frac{v}{c}\right)-\frac{L_2^2}{4B_2c^2}\left(1-2\frac{v}{c}\right)\right] \times\left\{\prod_{j=1}^{2}\exp\left[-A_j\left(\frac{\omega_{j0}}{c}-k_j\right)\left(\frac{\omega_{j0}}{c}-k_j-(-1)^j2\frac{v}{c}\frac{\omega_{j0}}{c}\right)\right]\right\}^2. \qquad (21)$$

The resonance fringe variation in Eq. (18) is very similar to that in the Doppler-free two-photon absorption of the crossed-beam experiment,[2] namely, the variations with respect to the laser-beam frequencies are not centered in Bohr frequencies, and the fringe contrasts become very poor at large path-length differences.[6] At the short-pulse limit, the fringe variation in Eq. (20) depends only on the path-length differences, not the laser-beam frequencies. It is due to this, in the crossed-beam experiment, that the direct phase is introduced into the coherent transition amplitude through the displacement of a part of the quantum system.

The experiment proposed here might be extended to the multiple-pulse experiment for the purpose of increasing fringe contrast and narrowing fringe width. For a multiple pulse experiment, one replaces the two detours in Fig. 1 by two overlapping loops and places the atomic cell in the overlapping region. Multiple pulses are generated by going around the loops. The condition for observing the resonance fringe variation in the multiple-pulse experiment is similar to that of Eq. (20) with L_1 and L_2 denoting the circumference of each loop.

Atomic absorption processes initiated by coherent laser beams are relatively new. The main character of these processes is that the atomic absorption amplitude depends on an external phase, which can be varied through the change of frequency, intersection angle, and path-length difference of coherent beams.[1] By varying the external phase, one can obtain a direct determination of the transition amplitude and super high-resolution spectroscopy. However, due to the random motion of atoms, without a proper experimental arrangement, coherent incident beams do not lead to coherent atomic processes. The reason is as follows. During absorption the atom changes its momentum which induces a Doppler phase in the coherent term of the transition amplitude. After averaging with respect to the atomic velocity distribution, the coherent term vanishes. In order to restore the coherent term, a mechanism is needed to suppress the velocity-dependent Doppler phase. Several mechanisms are known at the present. In one of them, the atom does not change its momentum in the absorption process; then the velocity-dependent Doppler phase is automatically zero. An example of this is the appearance of the resonance fringe variation in the Doppler-free two-photon absorption process.[1-5] In a second mechanism, the Doppler phase is suppressed through the absorption and emission of equal- and opposite-momentum photons. This mechanism leads to the Ramsey fringes in the saturation spectroscopy[7] and to that described in the experiment originally proposed by Baklanov *et al.*,[8] on the atomic absorption of a two-level system in two separated fields. The mechanism in this paper is yet another one. The Doppler phase is suppressed through a proper coordination among the path-length differences.

In conclusion, the author wishes to express his appreciation to Professor J. A. Jacobs and Professor D. Kaplan for many interesting discussions.

Note added. In the articles published by M. M. Salour, he repeatedly stated that to obtain resonance interference fringes in the profile of a two-photon absorption, two important requirements must be fulfilled. First, the probability amplitude for absorbing two counterpropagating photons is proportional to $e^{i(\omega t-kz)}e^{i(\omega t+kz)}=e^{2i\omega t}$ and does not depend on the spatial position of the atom. Second, the experiment must be done not with two independent pulses. In the works by Chebotayev *et al.*, they insisted on using the standing fields to obtain the resonance fringe. In the present note, none of the above requirements were provoked. Hence, their views only have a limited validity.

The resonance fringe is a coherent and interference effect. In the most physical processes, to observe the direct interference effects, some rigidities have to be maintained. Thus, those previous workers exclusively limited themselves to the Doppler free processes, for which the nonrigid Doppler effect is eliminated. However, there are other physical processes which were initiated by Brown and Twiss. The rigidity requirement in observing the direct interference effect can be relaxed. The present note provides a new mechanism for overcoming the nonrigid effect in the two-photon absorption process with a Doppler shift. Hence, by using a coherent-beam method, one not only can overcome the pulse broadening, but also the Doppler broadening.

The physical process proposed in the present note might be simply viewed as follows: The pulses from laser source 1 excite the initial state $|1\rangle$ to intermediate state $|0\rangle$. Due to the proper spacing between laser pulses a variation is induced in the population of state $|0\rangle$. However, this variation depends on the atomic velocity and cannot be observed directly. Later pulses from laser source 2 further excite the intermediate state $|0\rangle$ to the final state $|2\rangle$. The population variation of state $|2\rangle$ depends on the spacing between later pulses as well as the spacing between initial pulses. With a proper coordination among these spacings, which is given by Eq. (20), the atomic velocity dependence appearing in the population variation of state $|2\rangle$ can be suppressed and the resonance fringe variation becomes experimentally observable.

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