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Observation of a Hole in the Laser-Induced Cesium Beam Spectrum

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ABSTRACT We experimentally observed a depression in the laser-induced cesium beam fluorescence spectrum. This paper describes the optical configuration we realized and proves that the hole can be used for laser frequency stabilization. The spectrum was recorded under different conditions. We theoretically analyzed the physical origin of this effect and found that the hole could have a sub-natural line width. This indicates that it could have great application prospects in atomic experiments and beam-type quantum precision instruments due to the simple construction.

INDEX TERMS Laser-induced spectrum, cesium beam, laser frequency stability.

I. INTRODUCTION

High-resolution spectroscopy plays a pivotal role in the development of atomic physics [1]. A variety of methods have been reported to open the possibility of utilizing the narrow spectroscopy in experiments [2]. Laser-induced fluorescence spectroscopy of atomic beams is a widely used type of Doppler-free spectroscopy (Fig.1). Due to the avoidance of collisions of the atoms and the careful control of the interaction angle between the laser and atomic beam, the atomic beam spectrum almost obtains the natural line width of the atomic transition line. For quantum instruments using atomic beams such as the cesium beam frequency standard, the atomic beam spectrum is the most convenient spectrum for locking the laser frequency [3]–[6].

We observed a depression in the atomic beam spectrum in the experiment, which was similar to the pump-probe technique in an atomic cell [7]–[9]. After theoretical analysis and calculation, we determined that its line width could be narrower than the natural atomic width. We proved that this depression could be used for laser stabilization in practice. Laser stabilization is one of the most common applications of high-resolution spectroscopy in physical research [10]–[16]. In general, frequency modulation (FM) is applied to a laser current, and a dispersive signal as feedback is obtained from spectrum demodulation (Fig.1). The narrower the line width

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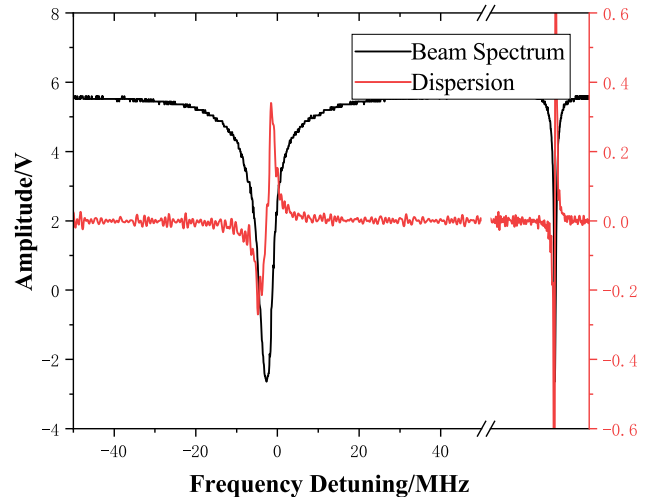


FIGURE 1. Laser-induced fluorescence beam spectrum and its first-order differential signal. The atomic beam spectrum is a Doppler-free spectrum. The dispersion is usually used to lock the laser frequency in an atomic beam experimental apparatus.

of the peak is, the larger the slope of the dispersion that can be obtained.

This paper presents a theoretical and experimental investigation of the hole in a fluorescence spectrum of an atomic beam. The organization of the paper is as follows: First, we show the configuration of our experiment, including the optical structure and atomic beam apparatus. We present some results of our measurement. Then, the physical origin

of the hole is discussed, and the line shape of the spectrum is presented. We conclude the features and prospects of the hole we have observed in the last section.

II. EXPERIMENT

The physical configuration is presented in Fig.2. A vacuum cesium beam tube containing a cesium oven and two interaction areas with spotlight bowls was built. There are three magnet shields in the tube to avoid the influence of an environmental magnetic field on the atomic beam. The vacuum degree of the tube is approximately 10^{-6} Pa. Two windows 22 cm apart on the tube allow lasers to enter. The temperature of the cesium oven is controlled at approximately 100°C with fluctuations less than 0.01°C. The collimator in the cesium oven is approximately 4 mm length and is formed by approximately 400 thin tubes 0.09 mm high and 0.18 mm wide. The divergence of the atomic beam is approximately 40 mrad. Therefore, the intensity of the beam from the collimator is approximately 4.13×10^{13} atoms every second in this case. The collision of cesium atoms can be ignored because of the large mean free path. The atoms in the cesium beam can be assumed to have a Maxwell velocity distribution, and the most probable velocity is approximately 200 m/s. The optical structure is shown below the tube in Fig.2. An 852 nm distributed feedback laser(DFB, EYP-DFB-0852) laser with an approximately 0.6 MHz line width generates the laser, and a PBS splits the laser into two. An AOM is used to shift the laser frequency, and the frequency difference between the two lasers is controlled to 251 MHz. This is the difference between the $|F = 4\rangle - |F' = 5\rangle$ line and the $|F = 4\rangle - |F' = 4\rangle$ transition line of the cesium atom. We can call the laser near the cesium oven the pump laser and the other laser the probe laser. The AOM ensures that the probe laser is scanned to the cesium $|F = 4\rangle - |F' = 5\rangle$ transition while the pump laser is at the $|F = 4\rangle - |F' = 4\rangle$ transition line. $|F = 4\rangle - |F' = 5\rangle$ is a cyclic transition(Fig.3). The intensities of the pump laser and probe laser are adjusted to 2 mW by the AOM and the phase retarder separately. The PD1 and the PD2 can give the laser power in real time. The laser diameter is approximately

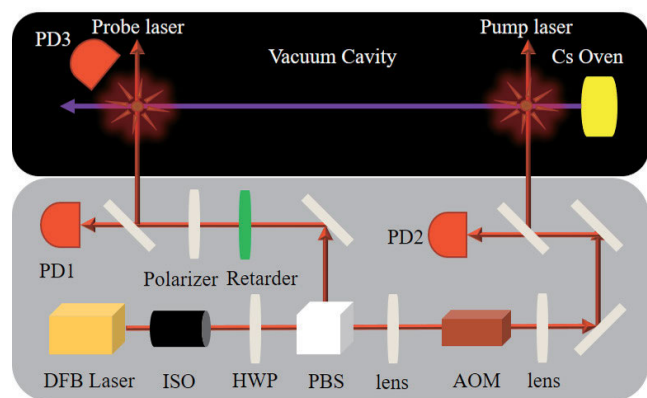


FIGURE 2. Structure of the optics. Black part: cesium beam tube. HWP: half-wave plate, ISO: isolator, PBS: polarizing beam splitter, PD: photodetector, AOM: acousto-optic modulator.

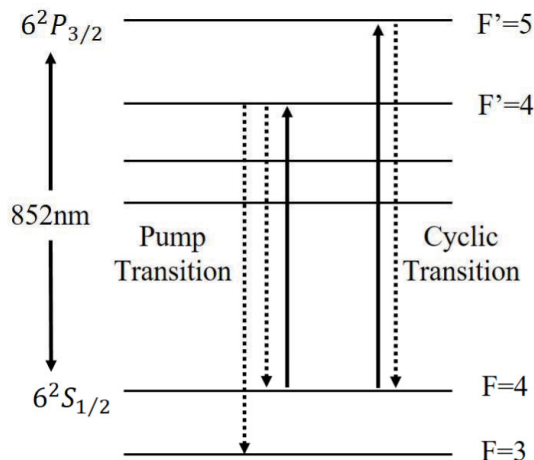


FIGURE 3. Energy levels of the cesium atom D2 line. The lifetime of the $^{133}\text{Cs} - \text{D2}$ line is 30.405(77) ns, and the $|F = 4\rangle - |F' = 4\rangle$ transition is a pumping transition. The atoms in the excited state $|F' = 4\rangle$ could drop to both $|F = 4\rangle$ and $|F = 3\rangle$, where $|F = 3\rangle$ is the "dark" state of the $|F = 4\rangle - |F' = 4\rangle$ laser. The $|F = 4\rangle - |F' = 5\rangle$ transition is a cyclic transition. The atoms in the excited state $|F' = 5\rangle$ can only drop to $|F = 4\rangle$ due to the transition rule $\Delta mF = \pm 1$. The transition will last until the atoms exit the interaction area.

6 mm, covering the atomic beam width to obtain higher pumping efficiency and detection efficiency. The two lasers entering the tube are adjusted carefully to ensure that they are perpendicular to the atomic beam. Then, interaction of the lasers and atoms occurs in two separated areas. The Doppler effects can almost be eliminated because the transverse velocity of atoms flying into the bowls is almost zero. The photodetector above the fluorescence collection bowl in the detection area collects the fluorescence signal induced by the probe laser. The frequency of the laser is scanned by the laser-induced current. It is clear that the atomic beam spectrum is obtained when probe laser is applied without the pump laser(Fig.1). This has significant benefits in terms of eliminating the Doppler background. With the pump laser entering the tube, the hole can be obtained. According to the signal amplified from the photodiode in the detection area, we can obtain its shape.

In the case of a weak intensity of the pump laser, there is no strong pumping effect, and the cesium atoms show a fluorescence emission spectrum. When the intensity of the pump laser strengthens, the hole appears as soon as some of the atoms in the beam are pumped to the $|F = 3\rangle$ energy level. By scanning the frequency of the DFB laser, the hole(Fig.4) is obtained. Due to the pumping effect of the pump laser, the cesium atoms are prepared to the $|F = 3\rangle$ state. Stimulated absorption and spontaneous radiation in the detection area will reduce as the probe laser is tuned in the $|F = 4\rangle - |F' = 5\rangle$ transition. When the pump laser is weak and cannot cause full pumping efficiency, the hole is small but has a quite narrow line width. When nearly all the atoms are pumped, the peak of the hole will be as high as the signal background. If the pump laser becomes even stronger, then the peak will have a flat top. We measured the line width of the hole in the cesium beam spectrum. The results

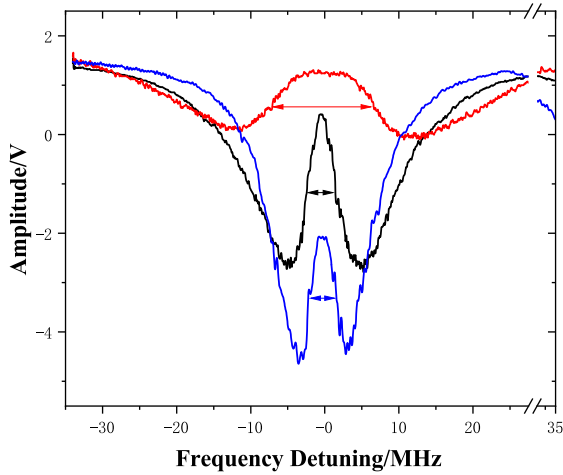


FIGURE 4. Holes with different intensities of pump laser will have different sizes. Red line(6 mW): flat-top peak hole with a 13.13 MHz line width. Black line(2 mW): hole at a moderate pumping efficiency with a 3.33 MHz line width. Blue line(1.5 mW): hole at a low pumping efficiency with a 3.18 MHz line width.

showed that the line width of the hole could be approximately 3 MHz, smaller than the $^{133}\text{Cs} - D2$ natural line width of approximately 5 MHz. The phenomena can be observed when the pumping efficiency is nearly 80%. The radio frequency in the AOM can be used to adjust the peak position of the depression in the atomic beam spectrum. If the frequency of the AOM is lower than the difference of the levels used (here, approximately 251 MHz for $|F' = 4\rangle$ and $|F' = 5\rangle$), then the hole appears with a frequency shift (Fig.5), and vice versa. Considering the angular offset caused by the AOM, when a 6 MHz frequency variation occurred, the separation angle between the first-order and zero-order beams changed by less than 1.5 mrad. This caused a frequency shift of the hole less than 0.4 MHz [17], which is much less than the frequency variation.

In most precision laser application scenarios, frequency stabilization is indispensable. Generally, the most convenient and effective method is to use the atomic line to stabilize the laser frequency. The line width of commonly used spectral lines is limited by the natural line width of the atom, which is an important factor limiting the instability of the laser frequency. Therefore, one of the major applications of the hole is laser stabilization. After careful adjustment, we locked the frequency of the DFB laser to the peak of the depression (Fig.6). To the best of our knowledge, this is the simplest structure that a laser can be stabilized using a sub-natural peak. Then, we found that holes can be observed in the cesium beam spectrum under other combinations of the pump laser and the probe laser using this structure, but the hole mentioned above was the largest. We conclude that the features of the hole are as follows:

- 1) The pump laser provides a pumping transition for the atoms.
- 2) The probe laser can provide a pumping transition or a cyclic transition for the atoms, but a cyclic transition can afford a larger signal.

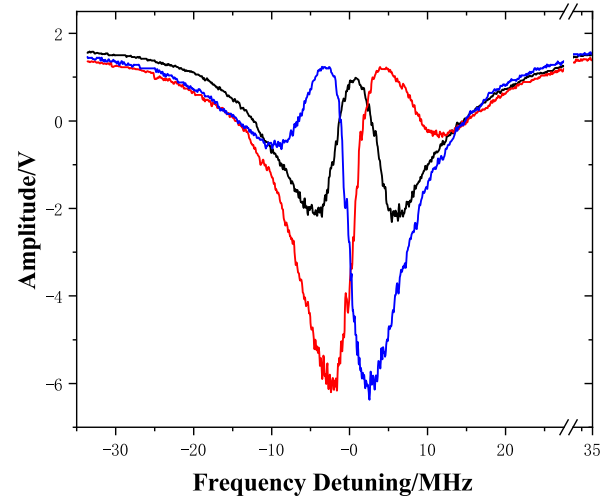


FIGURE 5. Holes with different frequency detunings of the pump laser will have different positions. Red line: RF of the AOM is 254 MHz. Black line: RF of the AOM is 251 MHz. Blue line: RF of the AOM is 248 MHz.

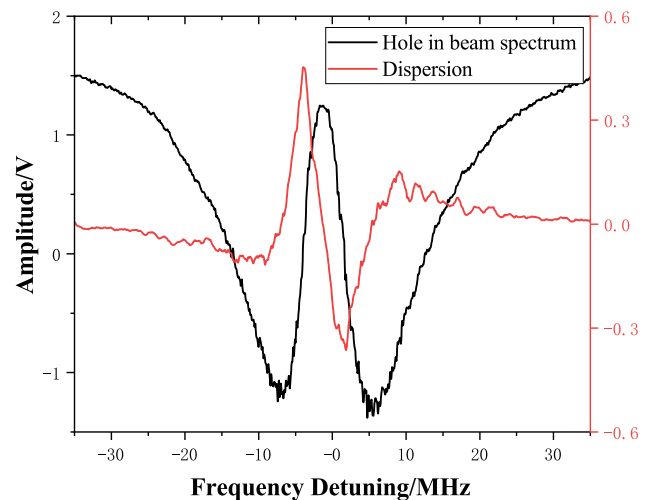


FIGURE 6. Hole in the cesium beam spectrum used for laser stabilization and its error signal. The frequency of the DFB laser could be stabilized by the feedback of the induced current.

- 3) The pump laser should pump the ground state to a state without an interaction induced by the probe laser.

This phenomenon can be similarly realized in rubidium atomic beams or other atomic beams.

III. THEORETICAL ANALYSIS

If a laser with a frequency close to the atomic transition line interacts with the atomic beam, then stimulated absorption and spontaneous radiation will probably occur. The ground state population of the atoms may be changed in this process [18]. In the experiment we described above, the interaction with the atomic beam will cause a strong pumping effect when the pump laser has a high intensity. When the atoms in beam enter part of the pumping area, first, the number of atoms in the corresponding ground energy level will change. The atoms will be totally or partly pumped to a “dark state” of the probe laser (cesium atoms in $|F = 4\rangle$ will be pumped to $|F = 3\rangle$).

When the atoms enter the detection area, the interaction will not occur. If the laser frequency is not tuned to the transition line of atoms exactly right, then the laser-induced fluorescence is not affected. When the detuning frequency becomes zero, the fluorescence should be the largest originally, but at this time, the atoms “vanish”. Therefore, the photons emitted by the atoms may decrease sharply if the pumping effect is strong enough. The fluorescence in the detection area will show a depression at this moment and the spectrum will be nearly zero.

The fluorescence emitted from the detection area can be calculated with a simplified atom-level model, and the line shape of the hole can be presented using the parameters we measured. Considering the interaction under actual conditions, the theoretical signal S is

$$S(\omega) = A\hbar\omega_0\beta N\Gamma_{probe}(\omega) \times \int_{\tau_{min}}^{\tau_{max}} \tau_{probe} \exp(-\eta\Gamma_{pump}(\omega) \int_{\tau_{min}}^{\tau_{max}} \tau_{pump}) \quad (1)$$

where

$$\begin{cases} \Gamma_{probe}(\omega) = \frac{\Gamma_{probe}(\omega_0) (\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2} \\ \Gamma_{pump}(\omega) = \frac{\Gamma_{pump}(\omega_0) (\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2} \end{cases} \quad (2)$$

$$\begin{cases} \tau_{probe} = d_{probe}/v \\ \tau_{pump} = d_{pump}/v \end{cases} \quad (3)$$

where A is the magnification of the diode and circuit, \hbar is the Planck constant, ω_0 is the resonant frequency, Δ is the frequency detuning of the laser and γ is the spontaneous emission coefficient. β depends on the laser frequency we use. N is the total number of atoms available, and Γ is the transition probability in the interaction conditions. d_{probe} and d_{pump} are the diameters of the lasers. We assume that the line width of the lasers is several times smaller than the atomic transition line. Then, we can deduce the line width $\Delta\nu$ of the spectrum:

$$\Delta\nu \approx \gamma \sqrt{\eta\Gamma_{pump}(\omega_0) \int_{\tau_{min}}^{\tau_{max}} \tau_{pump} - 1} \quad (4)$$

η is the possibility of pumping, and τ_{pump} and τ_{probe} are the times of atoms flying through the pump and probe lasers. From equation(4), we find out that the line width of the spectrum is not limited by the natural line width of the atoms. We can also conclude that the conditions of the hole appear:

$$\eta\Gamma_{pump}(\omega_0) \int_{\tau_{min}}^{\tau_{max}} \tau_{pump} > 1 \quad (5)$$

This method can be a sub-natural line width spectroscopy under moderate conditions. The line width of the hole can be quite narrow when the hole just appears(Appendix). If the probe laser has a pumping effect($\beta \approx 2$), then the probing interaction can be easily saturated. This is the reason why the hole observed under the combination of pumping transitions

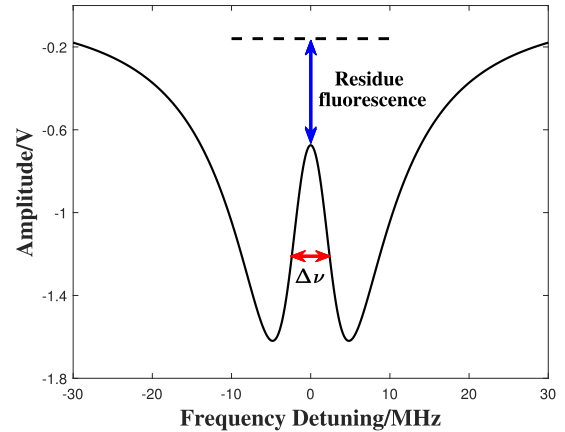


FIGURE 7. Theoretical line shape and line width of the spectrum. The background signal is an atomic beam spectrum signal, and the hole appears in the center of the spectrum.

is small. Cyclic transitions are normally caused by transition rule $\Delta mF = \pm 1$. The atoms are stimulated to an excited state and can only quickly return to the original ground state through spontaneous emission. There will be many photons emitted in all directions. A high probing efficiency will make the signal quite large, and laser stabilization will be realized. Compared to the pumping transition, laser-induced fluorescence can be promoted by $\beta = 240$.

In the actual situation, τ_{probe} and τ_{pump} will be different for different atoms because the atomic beam has a velocity distribution. This can be equivalent to the correction of the velocity distribution [19]:

$$I(v) = \frac{2}{v_p^{n+1}} v^n \exp\left(-\frac{v^2}{v_p^2}\right) \quad (6)$$

where

$$v_p = \sqrt{\frac{2kT}{m}} \quad (7)$$

I is the normalized intensity of the atomic beam. v is the velocity of the atom, and v_p is the most probable velocity in the atomic beam. k is the Boltzmann constant, and T is the temperature of the atomic beam oven. m is the mass of the atoms and depends on the atoms used. n is a constant and describes the different pumping and detection combination. $n = 3$ is the simplest case that uses a pumping transition for pumping and probing. For the case using a cyclic transition, $n = 2$ is applied. Because avoidance of the saturated effect of the probe laser is achieved, atoms with low speed could provide more fluorescence photons. In the end, we obtain Fig.7 in theory.

IV. CONCLUSION AND DISCUSSION

In this paper, a line depression caused by the laser pumping effect is experimentally found in a cesium beam. We studied the features of the spectrum at different intensities and frequencies of the laser. Then, we analyzed its cause and gave theoretical expressions of the line shape and width. The line width of this spectrum is not limited by the natural line

width of the cesium atom. Based on the obtained spectrum, we stabilized the frequency of a DFB laser. This is proof that the depression can have great applications in the laser field. Saturated absorption spectroscopy(SAS) [20], [21], free of Doppler broadening, is one of the most famous techniques in the high-resolution spectroscopy field [10]–[16], [22]. The principle of the hole in the atomic beam spectrum is different from the dip in SAS(see APPENDIX). At this time, there are clear advantages:

- 1) Higher potential in terms of the line width.
- 2) High utilization rate of atoms compared to an atomic cell.
- 3) Higher potential in terms of the signal-to-noise ratio.

In addition to frequency stabilization, high-resolution spectroscopy will have many practical applications in scientific measurement, biomedical imaging and materials science [23]. The technology constructing depressions mentioned above can be extended to most alkalis that have pumping and cycling transition lines, such as rubidium. The principle of laser stabilization using the hole can be widely suitable for optical pumping magnetometers, optical pumping atomic frequency standards, ultracold physics and almost all atomic physics experiments.

APPENDIX

Calculation of the line width of the atomic beam spectrum or saturated absorption spectrum. S_b is the line shape of the atomic beam spectrum,

$$S_b(\omega) = A\hbar\omega_0\beta N\Gamma_{probe}(\omega) \int_{\tau_{min}}^{\tau_{max}} \tau_{probe} \quad (8)$$

where the transition probability is

$$\Gamma_{probe}(\omega) = \frac{\Gamma_{probe}(\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2} \quad (9)$$

We find that

$$S_b = S_{bmax}/2 \quad (10)$$

where S_{bmax} is the maximum of the signal, when

$$\Delta^2 = \left(\frac{\gamma}{2}\right)^2 \quad (11)$$

Thus,

$$\Delta\nu = \gamma \quad (12)$$

Calculation of the line width of the hole in the cesium atomic beam spectrum: To avoid confusion, we use S_h , which is equal to S in Section 3.

$$S_h(\omega) = A\hbar\omega_0\beta N\Gamma_{probe}(\omega) \int_{\tau_{min}}^{\tau_{max}} \tau_{probe} \exp\left(-\eta\Gamma_{pump}(\omega) \int_{\tau_{min}}^{\tau_{max}} \tau_{pump}\right) \quad (13)$$

where the atomic transition rates of the probe and pump laser beams are

$$\begin{cases} \Gamma_{probe}(\omega) = \frac{\Gamma_{probe}(\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2} \\ \Gamma_{pump}(\omega) = \frac{\Gamma_{pump}(\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2} \end{cases} \quad (14)$$

and

$$\begin{cases} \tau_{probe} = d_{probe}/v \\ \tau_{pump} = d_{pump}/v \end{cases} \quad (15)$$

To obtain the line width of the hole, we find the first derivative of the previous formula and set it to zero,

$$\frac{\partial}{\partial\omega} S_h = 0 \quad (16)$$

Then,

$$\left[\frac{\Gamma_{probe}(\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2} \exp\left(-\eta\frac{\Gamma_{pump}t_{pump}(\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2}\right) \right]' = 0 \quad (17)$$

where

$$t_{pump} = \int_{\tau_{min}}^{\tau_{max}} \tau_{pump} \quad (18)$$

Equation(18) is equal to

$$\left[\frac{\Gamma_{probe}(\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2} \frac{2\eta\Gamma_{pump}t_{pump}(\frac{\gamma}{2})^2\Delta}{\left(\Delta^2 + (\frac{\gamma}{2})^2\right)^2} - \frac{2\Gamma_{probe}(\frac{\gamma}{2})^2\Delta}{\left(\Delta^2 + (\frac{\gamma}{2})^2\right)^2} \exp\left(-\eta\frac{\Gamma_{pump}t_{pump}(\frac{\gamma}{2})^2}{\Delta^2 + (\frac{\gamma}{2})^2}\right) \right] = 0 \quad (19)$$

Thus, the line width of the hole is

$$2\eta\Gamma_{probe}\Gamma_{pump}t_{pump}\Delta\left(\frac{\gamma}{2}\right)^4 - 2\Gamma_{probe}\Delta\left(\frac{\gamma}{2}\right)^2\left(\Delta^2 + \left(\frac{\gamma}{2}\right)^2\right) = 0 \quad (20)$$

Simplifying,

$$\Delta^2 = \left(\frac{\gamma}{2}\right)^2(\eta\Gamma_{pump}t_{pump} - 1) \quad (21)$$

Thus,

$$\Delta\nu \approx \gamma\sqrt{\eta\Gamma_{pump}(\omega_0) \int_{\tau_{min}}^{\tau_{max}} \tau_{pump} - 1} \quad (22)$$

(22) shows that the line width $\Delta\nu$ is independent of the Lorentz profile, so it could be smaller than the natural line width γ , when $0 < \sqrt{\dots} < 1$.

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