Nature of interference between Autler-Townes peaks in multi-level system

Dangka Shylla, Elijah Ogango Nyakango, Kirthanaa Indumathi and Kanhaiya Pandey

Department of Physics, Indian Institute of Technology Guwahati, Guwahati, Assam 781039, India

(Dated: July 1, 2019)

In this work we present a theoretical framework to identify the role and the nature of interference between Autler-Townes (AT) peaks (or dressed states) in generic multi-level system. The destructive interference between the AT peaks, gives rise to sharp transparency window known as electromagnetically induced transparency (EIT). In the three-level system, the two AT peaks interfere pair-wise with each other, almost similar to the two-slit interference. In the four-level system, the interference between the three AT peaks is also pair-wise analogous to three-slit interference but has a bit more complicated nature of interference. However, in many practical situations in atomic systems only the simple form of interference similar to three-level system dominates. In the three-level system, the nature of interference (i.e. constructive, destructive or zero/no interference) between the two AT peaks is purely determined by the natural decay rate of the states coupled by the control laser. However, in four-level system the nature of interference between the two extreme AT peaks can be tuned from constructive to destructive by tuning the power of the control laser.

PACS numbers:

I. INTRODUCTION

Modification of a weak probe laser absorption by a strong control laser in three-level system is due to two closely related phenomena, one is electro-magnetically induced transparency (EIT) and the other is Autler-Townes (AT) splitting. AT absorption peaks are due to the dressed states created by the control laser/s. In this paper we will use both terminologies i.e., AT peaks and dressed states which are basically the same. EIT in three-level system is AT splitting plus the interference [1] between them created by a control laser and has been addressed theoretically [2, 3] and experimentally using Akaike information criteria in real atomic system [4, 5] as well as in artificial atomic system [6]. The distinction between EIT and AT splitting has also been investigated based upon the quantum memory [8, 9].

In three-level system the strong control laser creates two AT peaks (dressed states) which are probed by a weak probe laser. The interference between these two AT peaks created by the control laser can be constructive, destructive or no interference depending upon the decay rate of the bare atomic states coupled by the control laser. Further in three-level system there are only two dressed states and hence there is a possibility of pair-wise interference only, almost similar to the two-slit interference. However, it is very interesting to investigate the nature of interference with more than two dressed states in a similar fashion to the three-slit interference [10, 12]. In order to probe the nature of interference between three dressed states or more, we need to consider four-level system and above respectively. The four-level system and above has been extensively studied for various applications [13, 24, 25], however the role and the nature of interference has not been addressed so far.

The absorption of the probe laser in the presence of the control lasers in multilevel systems are generally dealt by two approaches. One is laser induced coherence between the levels, which is also known as transfer of coherence (TOC) since simultaneous driving of different levels with lasers induces coherence between the levels which are not directly driven. The other approach is dressed states created by the control lasers and their excitation by the probe laser. In order to study the nature of interference between the AT peaks, first we provide the generic theoretical framework for the probe absorption using dressed state picture and verify this approach using TOC approach i.e. density matrix formalism in bare atomic state picture. Then we identify the interference and nature of it in the derived formula for the probe absorption using dressed state approach and by plotting the absorption and the AT peaks for a variety of systems.

II. MODEL

A. Dressed state

We consider the generic system as shown in Fig. 1 in which a weak probe laser is driving the transition $|1\rangle \rightarrow |2\rangle$ with Rabi frequency $\Omega_{12}$ and the detuning $\delta_{12}$. The strong control lasers are driving the transitions $|2\rangle \leftrightarrow |3\rangle$, $|3\rangle \leftrightarrow |4\rangle$,..., $|n-1\rangle \leftrightarrow |n\rangle$ with detunings $\delta_{23}, \delta_{34},..., \delta_{n-1,n}$ and the Rabi frequencies $\Omega_{23}, \Omega_{34},..., \Omega_{n-1,n}$. The Hamiltonian associated only with the control lasers in the rotating frame with rotating wave
The bare state |3⟩ is related to the probe laser and the incoherent decay rate becomes,

\[ H_c = -\hbar\delta_{23}|3⟩ \langle 3| - \hbar(\delta_{24} \pm \delta_{34})|4⟩ \langle 4| \]

\[ + \frac{\hbar\Omega_{23}}{2}|3⟩ \langle 4| + \frac{\hbar\Omega_{n-1n}}{2}|n-1⟩ \langle n-1| + h.c. \]

The sign ± of \( \delta_{i,j} \) is chosen according to the level structure. If the energy of the state \(|i⟩\) is higher than the state \(|i-1⟩\), then \( \delta_{i-1,i} \) will have + sign and if the energy of the state \(|i⟩\) is lower than the state \(|i-1⟩\), then \( \delta_{i-1,i} \) will have - sign. The eigenvalues \( (E_{d_i} = \hbar\Delta_{d_i}) \) of the \( H_c \) determines the position of the dressed states \( |d_i⟩ \) or AT absorption peaks. The eigenvectors will determine the dressed states which is a linear combination of the bare atomic states.

The generic form of the \( ith \) dressed state \( |d_i⟩ \) is given as

\[ |d_i⟩ = c_{i2}|2⟩ + c_{i3}|3⟩ + \ldots + c_{i,n-1}|n-1⟩ \]

and the incoherent decay rate of \( |d_i⟩ \) is

\[ \Gamma_{d_i} = |c_{i2}|^2\Gamma_2 + |c_{i3}|^2\Gamma_3 + \ldots + |c_{i,n-1}|^2\Gamma_{n-1} \]

The incoherent decay rate of the dressed states in addition to the decay rate of level \( 1 \) gives the linewidth, when they are probed by a laser. The coherent decay rate between two dressed states \( |d_i⟩ \) and \( |d_j⟩ \) is given as

\[ \kappa_{ij} = -\frac{1}{2} \left( c_{i2}^*\sqrt{\Gamma_2}|2⟩ + c_{i3}^*\sqrt{\Gamma_3}|3⟩ + \ldots + c_{i,n-1}^*\sqrt{\Gamma_{n-1}}|n-1⟩ \right) \]

\[ \left( c_{j2}\sqrt{\Gamma_2}|2⟩ + c_{j3}\sqrt{\Gamma_3}|3⟩ + \ldots + c_{j,n-1}\sqrt{\Gamma_{n-1}}|n-1⟩ \right) \]

Using the orthonormality condition \( \langle i|j⟩ = \delta_{ij} \) the coherent decay rates becomes,

\[ \kappa_{ij} = -\frac{1}{2} \left( c_{i2}^*c_{j2}\Gamma_2 + c_{i3}^*c_{j3}\Gamma_3 + \ldots + c_{i,n-1}^*c_{j,n-1}\Gamma_{n-1} \right) \]

The coherent decay rise to interference between the dressed states. The dressed states \( |d_i⟩ \) couples with \( |1⟩ \) through a probe laser with coupling strength written in terms of Rabi frequency \( \Omega_{p1} = -\langle 1|\hat{D}\hat{E}_0^q|d_i⟩/\hbar \)

where, \( \hat{E}_0^q \) is the electric field amplitude associated with probe laser and \( \hat{D} \) is the electric dipole moment operator. With the rotating wave approximation \( \langle 1|\hat{D}\hat{E}_0^q|d_i⟩/\hbar = \langle 1|\hat{D}\hat{E}_0^q c_{23}|2⟩/\hbar \) and hence \( \Omega_{p1} = c_{23}\Omega_{12} \). The amplitude of the excitation path for AT peaks corresponding to the dressed states \( |d_i⟩ \) will be proportional to the \( \Omega_{p1} \).

The amplitude for the probe absorption corresponding to this peak will be proportional to \( |\Omega_{p1}|^2 \).

The equation of motion for the dressed states \( |d_i⟩ \) and the bare state \( |1⟩ \) will be given as

\[ \frac{d}{dt} \begin{bmatrix} C_1 \\ C_{d_1} \\ \vdots \\ C_{d_{n-1}} \\ C_{d_{n+1}} \end{bmatrix} = \begin{bmatrix} 0 & \Omega_{p1} & \Omega_{p2} & \ldots & \Omega_{p,n-1} \\ \Omega_{p1}^* & -i\gamma_{d_1} & i\kappa_{12} & \ldots & i\kappa_{1,n-1} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ i\kappa_{n-1,1} & i\kappa_{n-1,2} & \ldots & -i\gamma_{d_{n-1}} & 0 \end{bmatrix} \begin{bmatrix} C_1 \\ C_{d_1} \\ \vdots \\ C_{d_{n-1}} \end{bmatrix} \]

where \( \gamma_{d_i} = \Gamma_1 + \Gamma_2 + i\delta_{d_i} \) with \( \delta_{d_i} = \delta_{12} + \Delta_{d_i} \). We consider the steady state for the dynamics of all the dressed states \( |d_i⟩ \) i.e. \( \frac{dC_{d_i}}{dt} = 0 \). The absorption of the probe laser is given as \( \frac{dC_{1}}{dt} = \frac{dC_{d_1}}{dt} = C_1 + \frac{dC_{d_1}}{dt} \). For the weak probe we consider that \( C_1 \approx 1 \) and hence, \( \frac{dC_{1}}{dt} = \frac{dC_{d_1}}{dt} + \frac{dC_{d_1}}{dt} \). The normalized absorption of the probe laser is given as \( -2(\Gamma_1 + \Gamma_2)/((\Omega_{12})^2)(\frac{dC_{d_1}}{dt} + \frac{dC_{d_1}}{dt}) \). The normalization is done such that in the absence of the all the control lasers, the probe absorption is 1 at the resonance \( (\delta_{12} = 0) \).

**B. Bare state TOC approach**

The other approach which is commonly used to deal with multi-level system is the transfer of coherence (TOC) between the bare states using the density matrix approach and is very well described in our previous work [22]. The absorption of the probe laser is written in terms of the density matrix element between level \( |1⟩ \) and \( |2⟩ \) i.e. \( \rho_{12} \). The solution for \( \rho_{12} \) can be written in the continued fractional form given below:
The absorption of the probe laser is determined by the eigenvalues of the Hamiltonian associated with the control laser and is given below:

\[
H_c = \hbar \begin{bmatrix}
0 & \Omega_{p1} & \\
\Omega_{p2} & 0 & \\
-\delta_{23} & 0 & 0
\end{bmatrix}
\]  \tag{9}

From Eq. 9, the equation of motion for the coefficients of \(|1\rangle\), \(|d_1\rangle\) and \(|d_2\rangle\) are given below:

\[
\begin{align*}
\frac{i}{\hbar} \frac{dC_1}{dt} &= \frac{\Omega_{p1}}{2} C_{d1} + \frac{\Omega_{p2}}{2} C_{d2} \\
\frac{i}{\hbar} \frac{dC_{d1}}{dt} &= -\gamma_{d1} C_{d1} + \frac{\Omega_{p1}^*}{2} C_{1} + i\kappa_{12} C_{d1} \\
\frac{i}{\hbar} \frac{dC_{d2}}{dt} &= -\gamma_{d2} C_{d2} + \frac{\Omega_{p2}^*}{2} C_{1} + i\kappa_{12}^* C_{d1}
\end{align*}
\]  \tag{10}

We consider the steady state case for the time evolution of the dressed states i.e., \(\frac{dC_{d1}}{dt} = \frac{dC_{d2}}{dt} = 0\) which leads to the following equation

\[
\frac{dC_1}{dt} = -\frac{1/4}{1 - \frac{\Omega_{p1}^2}{\gamma_{d1}} \frac{\Omega_{p2}^2}{\gamma_{d2}}} \begin{bmatrix}
|\Omega_{p1}|^2 & |\Omega_{p2}|^2 & \kappa_{12} \Omega_{p1}^* \Omega_{p2} + \text{c.c.} \\
\gamma_{d1} & \gamma_{d2} & \gamma_{d1} \gamma_{d2}
\end{bmatrix}
\]  \tag{11}

The eigenvalues of the two dressed states \(|d_1\rangle\) and \(|d_2\rangle\) corresponding to Hamiltonian in Eq. 9 and various other

\[
\begin{align*}
C. \text{ Three-level system}
\end{align*}
\]
parameters are given below:

\[ E_{d1} = \frac{\hbar}{2} [-\delta_{23} - \sqrt{\delta_{23}^2 + \Omega_{23}^2}]; |d_1\rangle = -\cos \theta |2\rangle + \sin \theta |3\rangle \]

\[ \Gamma_{d1} = \cos^2 \theta \Gamma_2 + \sin^2 \theta \Gamma_3; \]

\[ E_{d2} = \frac{\hbar}{2} [-\delta_{23} + \sqrt{\delta_{23}^2 + \Omega_{23}^2}]; |d_2\rangle = \sin \theta |2\rangle + \cos \theta |3\rangle \]

\[ \Gamma_{d2} = \sin^2 \theta \Gamma_2 + \cos^2 \theta \Gamma_3; \]

\[ \kappa_{12} = \frac{1}{4} (\Gamma_2 - \Gamma_3) \sin 2\theta; \Omega_{p_1} = -\cos \theta; \Omega_{p_2} = \sin \theta; \]

where, \( \tan 2\theta = \frac{\Omega_{23}}{\delta_{23}} \) (12)

The nature of interference between the two dressed states \( |d_1\rangle \) and \( |d_2\rangle \) is govern by the parameter \( \kappa_{12} \). It is possible that the interference parameter, \( \kappa_{12} \) can change its sign for given \( \Gamma_2 \) and \( \Gamma_3 \) by tuning the control laser detuning, \( \delta_{23} \) from positive to negative and vice versa. However, it is very important to note that the amplitude of excitation, \( \Omega_{p_2} \) of the dressed state \( |d_2\rangle \) also changes its sign if \( \delta_{23} \) is changed from positive to negative and vice versa. So, overall the interference term \( \kappa_{12}\Omega_{p_1}\Omega_{p_2} \) will not change sign and hence it is not possible to change the sign of interference by changing the detuning of the control laser.

After putting the value of \( \Omega_{p_1}, \Omega_{p_2} \) and \( \kappa_{12} \) in Eq. (11)

\[ \frac{dC_1}{dt} = \left[ \frac{|\Omega_{12}|^2/4}{1 - \frac{2\delta_{12}^2}{\gamma_1\gamma_2}} \right] \left[ \cos^2 \theta \gamma_{d1} + \sin^2 \theta \gamma_{d2} - \frac{\Gamma_2 - \Gamma_3}{4} \frac{\sin 2\theta \gamma_{d1}\gamma_{d2}}{\gamma_{d1}\gamma_{d2}} \right] \]

\[ \rho_{12} = \frac{i \Omega_{12}}{2 \gamma_{12}} \left[ \frac{1}{1 + \frac{2\delta_{12}^2}{\gamma_1\gamma_2}} \right] \]

We compare both approaches and find complete match for various parameters.

From Eq. (11) it is clear that for \( \Gamma_2 > \Gamma_3 \) there is destructive interference, for \( \Gamma_2 = \Gamma_3 \) there is no/zero interference, and for \( \Gamma_2 < \Gamma_3 \) there is constructive interference between the AT peaks. The value of \( \Gamma_3 = 0 \) which is the case for \( \Lambda \)-system, there is destructive interference between two AT peaks as the dashed green curve is higher than the solid red curve in Fig. 4a. At resonance the interference is maximum and is completely destructing the absorption due to the two AT peaks. From Fig. 4b with \( \Gamma_3 = 0.5 \Gamma_2 \) \( (\Gamma_2 - \Gamma_3 > 0) \) which is valid for the ladder system, it implies that the interference is destructive but not completely as we can see finite absorption by solid red curve at resonance. For \( \Gamma_2 = \Gamma_3 \) there is no interference between the two AT peaks. The complete match between dashed green and red solid curve in Fig. 4b also implies this. Also we consider \( \Gamma_1 = \Gamma_3 = \Gamma \) and \( \Gamma_2 = 0 \), which is valid for V-system, the interference between two AT peaks is constructive. We can see in Fig. 4d the dashed green curve is lower than the solid red curve near resonance. It is clear that the nature of interference between the two AT peaks can not be tuned from constructive to destructive interference for given atomic levels and can not be tuned by the laser parameters.

D. Four-level system

The various four-level system has been studied for different applications in two configurations namely in the chain configurations such as N [13, 15], Ladder-Lambda [16, 17], Ladder [18], and in the branching configuration such as Y [19], Tripod [20, 22], Inverted Tripod [23] as shown in Fig. 5. The different names has been given for the four-level system depending upon the energy levels of |1>, |2>, |3> and |4> and other possible four-level systems can also be constructed by changing the energy levels. The dressed state picture of all possible four-level systems will be same as shown in Fig. 5.
The response of the weak probe laser in steady state is regularly analysed by the element of density matrix between $|1\rangle$ and $|2\rangle$, $\rho_{12}$ which involves the TOC between $|1\rangle$ and $|3\rangle$ and $|1\rangle$ and $|4\rangle$ by the following equation

$$\rho_{12} = \frac{i \Omega_{12}}{2 \gamma_{12}}$$

For branch system at branching level $|2\rangle$ the above equation will have following form

$$\rho_{12} = \frac{i \Omega_{12}}{2 \gamma_{12}}$$

The above density matrix solution is a good cross check of our calculation for the four-level system in the dressed state picture and identification of the nature of interference between the various AT peaks.

The Hamiltonian associated with the control lasers for the four-level chain system is written as

$$H_c = \hbar \begin{bmatrix} 0 & \Omega_{p1} & 0 & 0 \\ \Omega_{p1} & 0 & -\delta_{23} & 0 \\ 0 & -\delta_{23} & 0 & \Omega_{p3} \\ 0 & 0 & \Omega_{p3} & 0 \end{bmatrix}$$

The Hamiltonian associated with the control lasers for the four-level system branching at level $|2\rangle$ such as Y, Tripod and Inverted Tripod as shown in Fig. 4 is written as

$$H_c = \hbar \begin{bmatrix} 0 & \Omega_{p1} & \Omega_{p2} \\ \Omega_{p1} & 0 & -\delta_{23} \\ \Omega_{p2} & -\delta_{23} & 0 \end{bmatrix}$$

The rate equation for the bare state $|1\rangle$ and of the dressed states $|d_1\rangle$, $|d_2\rangle$, $|d_3\rangle$ for both types of system mentioned above are given below

$$\frac{dC_1}{dt} = -\gamma_1 C_1 + \Omega_{p1}^2 C_1 + \Omega_{p3}^2 C_3 + \Omega_{p3} \Omega_{p2} C_2$$

$$\frac{dC_1}{dt} = -\gamma_1 C_1 + \Omega_{p1}^2 C_1 + \Omega_{p3}^2 C_3 + \Omega_{p3} \Omega_{p2} C_2$$

In the steady state, the dynamics of the dressed states i.e. $\frac{dC_1}{dt} = \frac{dC_2}{dt} = \frac{dC_3}{dt} = 0$, we get following equation

$$\frac{dC_1}{dt} = -\frac{1}{N} \left( \frac{\Omega_{p1}^2}{\gamma_1} + \frac{\Omega_{p2}^2}{\gamma_2} + \frac{\Omega_{p3}^2}{\gamma_3} \right)$$

where,

$$N = 1 - \frac{\Omega_{p1}^2}{\gamma_1 \gamma_2 \gamma_3} \frac{\Omega_{p2}^2}{\gamma_2 \gamma_3} \frac{\Omega_{p3}^2}{\gamma_1 \gamma_3}$$

The above equation represents the absorption of the probe laser. In this equation the terms denoted as “Three AT peaks” below curly underline represents the absorption of the probe laser due to individual three dressed states or AT peaks, $|d_1\rangle$, $|d_2\rangle$ and $|d_3\rangle$. The amplitude of the individual AT peaks will be proportional to $|\Omega_{p1}|^2$, $|\Omega_{p2}|^2$ and $|\Omega_{p3}|^2$. Further, the AT peaks amplitude are modified as $-\frac{|\Omega_{p1}|^2}{\gamma_2 \gamma_3}$, $-\frac{|\Omega_{p2}|^2}{\gamma_1 \gamma_3}$ and $-\frac{|\Omega_{p3}|^2}{\gamma_1 \gamma_2}$ respectively and
is denoted as “Correction for AT peaks”. The interference between the AT peaks denoted as “Interference1” in the expression, is very much similar to the three-slit interference. The magnitude and sign of the interference is proportional to \( \kappa_{12}, \kappa_{23} \) and \( \kappa_{13} \) respectively. However, we also observe the interference between the AT peaks which are a bit more complicated and denoted as “Interference2”.

1. Chain configurations with all the control lasers at resonance

The analytical expression for the eigenvalues and eigenvectors of Hamiltonian given by Eq. 17 are complicated for general detunings. However, it becomes very simple when all the control lasers are at resonance and it is also easy to interpret the nature of interference between the AT absorption peaks. In this particular case the eigenvalues of the dressed states and the associated parameters are listed below:

\[
\begin{align*}
E_{d_1} &= -\sqrt{\frac{\Omega_{23}^2 + \Omega_{34}^2}{2}}; \\
|d_1\rangle &= \frac{1}{\sqrt{2}} \left( \frac{\Omega_{23}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} |2\rangle - \frac{\Omega_{34}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} |4\rangle \right); \\
\Gamma_{d_1} &= \frac{1}{2} \frac{|\Omega_{23}|^2}{\Omega_{23}^2 + \Omega_{34}^2} \Gamma_2 + \frac{1}{2} \frac{|\Omega_{34}|^2}{\Omega_{23}^2 + \Omega_{34}^2} \Gamma_4; \\
E_{d_2} &= 0; |d_2\rangle = -\frac{\Omega_{34}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} |2\rangle + \frac{\Omega_{23}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} |4\rangle; \\
\Gamma_{d_2} &= \frac{1}{2} \frac{|\Omega_{23}|^2}{\Omega_{23}^2 + \Omega_{34}^2} \Gamma_2 + \frac{1}{2} \frac{|\Omega_{34}|^2}{\Omega_{23}^2 + \Omega_{34}^2} \Gamma_4; \\
E_{d_3} &= \frac{\Omega_{23}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}}; \\
|d_3\rangle &= \frac{1}{\sqrt{2}} \left( \frac{\Omega_{23}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} |2\rangle + \frac{\Omega_{34}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} |4\rangle \right); \\
\Gamma_{d_3} &= \frac{1}{2} \frac{|\Omega_{23}|^2}{\Omega_{23}^2 + \Omega_{34}^2} \Gamma_2 + \frac{1}{2} \frac{|\Omega_{34}|^2}{\Omega_{23}^2 + \Omega_{34}^2} \Gamma_4; \\
\kappa_{12} &= \frac{\Gamma_2 - \Gamma_4}{2\sqrt{2}} \Omega_{23} \Omega_{34}; \\
\kappa_{13} &= -\frac{\Gamma_2 + \Gamma_4}{4} \Omega_{23}^2 + \frac{\Gamma_2 + \Gamma_4}{4} \Omega_{34}^2; \\
\kappa_{23} &= \frac{\Gamma_2 - \Gamma_4}{2\sqrt{2}} \Omega_{23} \Omega_{34}; \\
\Omega_{p_1} &= \Omega_{p_3} = \frac{1}{\sqrt{2}} \frac{\Omega_{23}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} \Omega_{12}; \Omega_{p_2} = -\frac{\Omega_{34}}{\sqrt{\Omega_{23}^2 + \Omega_{34}^2}} \Omega_{12}.
\end{align*}
\]

Now we consider the various combinations of \( \Gamma_2, \Gamma_3 \) and \( \Gamma_4 \) to see the nature of interference. If \( \Gamma_2 = \Gamma_3 = \Gamma_4 \) then \( \kappa_{12} = \kappa_{23} = \kappa_{13} = 0 \) from Eq. 22 and there is no interference between any of the AT absorption peaks. The complete overlap between absorption shown by the solid red curve (AT peaks including the interference) and the dashed green curve (only AT peaks) in Fig. 6a is an indication of no interference.

For \( \Gamma_2 < \Gamma_4 \equiv \Gamma_3 \), the terms \( \kappa_{12} \) and \( \kappa_{23} \) are negative while \( \kappa_{13} \) is positive for \( \Omega_{23} = \Omega_{34} \) from Eq. 22. In addition, \( \Omega_{p_1} \) and \( \Omega_{p_3} \) are positive and \( \Omega_{p_2} \) is negative. This implies that corresponding to “Interference1” and “Interference2” all the terms are positive and hence there is constructive interference between three AT peaks. The contribution from “Interference2” as compared to “Interference1” is much smaller as the individual terms are small. Further, the modification of the individual AT peaks due to “Correction for AT peaks” terms are also small. In Fig. 6b, we can see that the dashed cyan curve corresponding to the terms \( \text{AT1+AT2+AT3+Interference1} \) is almost overlapping with the solid red curve which includes all the terms. Near the resonance i.e., \( \delta_{12} = 0 \), the dashed green curve is lower than the solid red curve which is due to constructive interference between \( |d_1\rangle \) and \( |d_2\rangle, |d_2\rangle \) and \( |d_3\rangle \) and \( |d_1\rangle \) and \( |d_3\rangle \).

For \( \Gamma_2 > \Gamma_4 = \Gamma_3 \), the terms \( \kappa_{12} \) and \( \kappa_{23} \) are positive and \( \kappa_{13} \) is negative for \( \Omega_{23} = \Omega_{34} \) from Eq. 22. Similarly, \( \Omega_{p_1} \) and \( \Omega_{p_3} \) are positive and \( \Omega_{p_2} \) is negative. This implies that all the terms corresponding to “Interference1” are negative and hence there is destructive interference between the three AT peaks. Corresponding to “Interference2” all three terms are positive. Again, the contribution from “Interference2” as compared to “Interference1” is much smaller as individual terms are small. Further, the modification of the individual AT peaks due to “Correction for AT peaks” terms are also small. In Fig. 6c, we can see the dashed cyan curve corresponding to the terms \( \text{AT1+AT2+AT3+Interference1} \) is almost overlapping with solid red curve which includes all the terms. Near the resonance i.e., \( \delta_{12} = 0 \), the dashed green curve is lower than the solid red curve which is due to destructive interference between \( |d_1\rangle \) and \( |d_2\rangle, |d_2\rangle \) and \( |d_3\rangle \) and \( |d_1\rangle \) and \( |d_3\rangle \).
green curve is higher than the solid red curve which is due to destructive interference between \(|d_1\) and \(|d_3\), \(|d_2\) and \(|d_3\) and \(|d_1\) and \(|d_3\).

For the parameters relevant to the N- system in Fig. 6 i.e. \(\Gamma_3 = 0, \Gamma_4 = \Gamma_2\), both \(\kappa_{12}\) and \(\kappa_{23}\) are zero but \(\kappa_{13}\) is negative. This implies that the first two terms in “Interference1” are zero and the third term is negative and hence there is destructive interference between \(|d_1\) and \(|d_3\). All the terms in “Interference2” are zero but there is contribution from the surviving term of “Correction for the AT peaks” which starts to play a role as we can see from the deviation of the dashed cyan curve from the solid red curve.

For the parameters relevant to the ladder-lambda system i.e., \(\Gamma_3 = \Gamma_2, \Gamma_4 = 0\) in Fig. 6 \(\kappa_{12}, \kappa_{23}\) and \(\kappa_{13}\) are all positive. Hence, the first two terms in “Interference1” are negative and the third term is positive and also corresponding to “Interference2” the first two terms are negative and the third term is positive. In the said figure near the resonance i.e., \(\delta_{12} = 0\), the dashed green curve is higher than the solid red curve which is due to domination of destructive interference between the three AT peaks. In this case both the “Interference2” and “Correction for the AT peaks” starts playing a significant role as we can see from the mismatch between the dashed cyan curve and solid red curve.

Now we consider the possibility of tuning the interference from constructive to destructive by tuning the control laser parameter such as the Rabi frequencies. As we noticed that the nature of interference in the three-level system can not be tuned from negative to positive or zero by changing the laser parameters. However, if we consider a simple case in four-level-system where \(\Gamma_3 < \Gamma_2\) and \(\Gamma_4 = 0\) then it is possible to tune the interference from negative to positive between \(|d_1\) and \(|d_3\). In Fig.
and $\Omega_5$ is negative, zero and positive for low, $1/\sqrt{2}\Gamma_3$ and high value of $\Omega_3$ respectively. It is also noted that with this increase of $\Omega_3$, there is no change in sign of $\Omega_m$, $\Omega_m^2$ and $\Omega_m$ and hence it is possible to tune the interference between $|d_1\rangle$ and $|d_3\rangle$ from constructive, to zero and to destructive.

2. Branching configurations with all control lasers at resonance

We consider the case when all the control lasers are at the resonance. In this particular case the energy of the dressed states and the related parameters are listed below:

$$E_{d_1} = -\frac{1}{2} \Omega_{23}^2 + \Omega_{12}^2;$$

$$|d_1\rangle = \frac{1}{\sqrt{2}} \Omega_{23} \Gamma_3 + \frac{1}{\sqrt{2}} \Omega_{12} \Gamma_4;$$

$$\Gamma_{d_1} = \frac{1}{2} \Omega_{23} \Gamma_3 + \frac{1}{2} \Omega_{12} \Gamma_4;$$

$$E_{d_2} = 0; \ |d_2\rangle = -\frac{1}{2} \Omega_{23} \Gamma_3 + \frac{1}{2} \Omega_{12} \Gamma_4;$$

$$\Gamma_{d_2} = \frac{1}{2} \Omega_{23} \Gamma_3 + \frac{1}{2} \Omega_{12} \Gamma_4;$$

$$E_{d_3} = \frac{1}{2} \Omega_{23} \Gamma_3 + \frac{1}{2} \Omega_{12} \Gamma_4;$$

$$|d_3\rangle = \frac{1}{\sqrt{2}} \Omega_{23} \Gamma_3 + \frac{1}{\sqrt{2}} \Omega_{12} \Gamma_4;$$

$$\Gamma_{d_3} = \frac{1}{2} \Omega_{23} \Gamma_3 + \frac{1}{2} \Omega_{12} \Gamma_4;$$

$$\kappa_{12} = \frac{\Gamma_3}{2 \Omega_{23}^2 + \Omega_{12}^2};$$

$$\kappa_{13} = \frac{\Gamma_3}{2 \Omega_{23}^2 + \Omega_{12}^2};$$

$$\kappa_{23} = \frac{\Gamma_3}{2 \Omega_{23}^2 + \Omega_{12}^2};$$

The interesting point is that the central AT peak, shown by dashed blue curve (corresponding to eigenvalue 0) in Fig. 9 has zero amplitude as $\Omega_m = 0$ and hence only two dressed states $|d_1\rangle$ and $|d_3\rangle$ can be excited by the probe laser. Thus, it behaves similar to the three-level system. For Tripod system, all the terms corresponding to the “Interference2” and “Correction for AT peaks” are zero and there is destructive interference between $|d_1\rangle$ and $|d_3\rangle$ as the “Interference1” term $\kappa_{13}\Omega_{23}\Omega_{12}$ is negative. It is also seen in Fig. 9 as the solid red curve is lower than the dashed green curve.

For $\Gamma_1 = \Gamma, \Gamma_2 = 0, \Gamma_3 = \Gamma, \Gamma_4 = \Gamma$ in Fig. 9, which is valid for the inverted tripod system, all the terms corresponding to the “Interference2” and “Correction for AT peaks” are zeros and there is constructive interference between $|d_1\rangle$ and $|d_3\rangle$ as the “Interference1” term $\kappa_{13}\Omega_{23}\Omega_{12}$ is positive. For Ladder-lambda system...
\[ \Gamma_1 = 0, \Gamma_2 = \Gamma, \Gamma_3 = 0 \text{ and } \Gamma_4 = 0.5\Gamma \] the “Interference1” term \( \kappa_{13} \Omega \rho_{p_1} \Omega \rho_{p_3} \) between \( |d_1\rangle \) and \( |d_3\rangle \) is negative and hence there is destructive interference. The non-zero term of “Interference2” \( \kappa_{12} \kappa_{23} \Omega \rho_{p_1} \Omega \rho_{p_3} \) is negative and hence contributing as more destructive interference. Further, the “Correction for the AT peaks” is non-zero for the two dressed states and hence we see further reduction in the absorption as shown by the solid red curve which is lower than the dashed cyan curve (AT peaks+Interference1) in Fig. 9c.

For \( \Gamma_2 = \Gamma_3 = \Gamma_4 = \Gamma \) which is valid for the Y-system, “Interference1”, “Interference2” and “Correction for AT peaks” are zero as \( \kappa_{12} = \kappa_{13} = \kappa_{23} = 0 \) and hence there is no interference between any of the AT peaks. In Fig. 9b there is complete overlap between solid red curve and green dash curve.

E. Four-level loopy system

The four-level loopy system is as shown in Fig. 10. It can be of various types depending upon the energy level of the states \( |1\rangle, |2\rangle, |3\rangle, |4\rangle \) as we discussed in the previous section. The study of the loopy system is very important as it further authenticates our approach for the dressed states as in this case the interference terms, \( \kappa \)'s and \( \Omega \)'s can be complex. The various loopy system has been discussed previously [24, 25].

The density matrix element for the probe absorption in the four-level loopy system as shown in Fig. 10 is given by the following equation,

\[ \rho_{12} = \frac{1}{1 + \frac{1}{4} \left| \Omega_{23} \right|^2 + \frac{1}{4} \left| \Omega_{24} \right|^2 + \frac{1}{8} \left| \Omega_{23} \Omega_{24} \right|^2 + c.c.} \]

The Hamiltonian associated with the control lasers for this system is given below

\[ H_c = h \begin{pmatrix} 0 & \frac{\Omega_{23}}{2} & \frac{\Omega_{24}}{2} \\ \frac{\Omega_{23}}{2} & -\delta_{23} & 0 \\ \frac{\Omega_{24}}{2} & 0 & -\delta_{34} \end{pmatrix} \]

The general control laser Rabi frequencies will be \( \Omega_{23} = |\Omega_{23}| e^{i\phi_{23}}, \Omega_{34} = |\Omega_{34}| e^{i\phi_{34}} \) and \( \Omega_{24} = |\Omega_{24}| e^{i\phi} \) which can be considered (without any loss of generality) as \( \Omega_{23} = |\Omega_{23}| \) and \( \Omega_{34} = |\Omega_{34}| \) and \( \Omega_{24} = |\Omega_{24}| \) i.e. considering \( \Omega_{23} \) and \( \Omega_{34} \) as real and \( \Omega_{24} \) as complex where \( \phi = \phi_{24} - \phi_{23} - \phi_{34} \). The eigenvalue and eigenvector corresponding to general parameters in the above Hamiltonian in Eq. 25 is complicated however it is relatively simpler for the case when all the control lasers are on the resonance and \( \Omega_{23} = \Omega_{34} = \Omega \) and \( \Omega_{24} = \Omega_1 e^{i\phi} \) where \( \Omega \) and \( \Omega_1 \) are real quantities. The eigenvalues, eigenvectors and various other parameters for \( \phi = 0 \) are listed below:

\[
\begin{align*}
E_{d_1} &= -\frac{\Omega_1}{2}; |d_1\rangle = -\sqrt{\frac{1}{2}} |2\rangle + \sqrt{\frac{1}{2}} |4\rangle; \\
E_{d_2} &= \frac{\Omega_1 - \Omega'}{2}; |d_2\rangle = \sqrt{\frac{1}{2}} |2\rangle - \sqrt{\frac{1}{2}} |4\rangle; \\
E_{d_3} &= \frac{\Omega_1 + \Omega'}{2}; |d_3\rangle = \sqrt{\frac{1}{2}} |2\rangle + \sqrt{\frac{1}{2}} |4\rangle; \\
E_{d_4} &= \frac{\Omega_1 + \Omega''}{2}; |d_4\rangle = \sqrt{\frac{1}{2}} |2\rangle + \sqrt{\frac{1}{2}} |4\rangle;
\end{align*}
\]

Now we discuss by plotting the probe absorption and the AT peaks for specific systems. The relevant parameters are mentioned in the captions of the plots. First, we consider the case with \( \Gamma_1 = \Gamma_3 = \Gamma_4 = 0 \), \( \Gamma_2 = \Gamma \) which is valid for loopy-tripod-system and we take \( \Omega_{23} = \Omega_{34} = \Omega_{24} = 0.5\Gamma \) for all the cases. Hence, all the individual terms of “Interference1” are negative and
of “Interference2” are positive. The terms corresponding to “Interference1” are higher than that of “Interference2” and so effectively there is destructive interference between the AT peaks, shown in Fig. 11a as the probe absorption goes to zero at the crossing points of the AT peaks. Further, the probe absorption is also lower than the sum of all the AT peaks in the overlapping region. In the case for $\Gamma_2 = \Gamma_4 = \Gamma$, $\Gamma_1 = \Gamma_3 = 0$ valid for the loopy N-system, corresponding to the terms in “Interference1” there is no interference between the AT peaks $|d_1\rangle$ and $|d_2\rangle$ while there is destructive interference between $|d_3\rangle$ and $|d_4\rangle$. For this system all the terms corresponding to “Interference2” are zero. The probe absorption and AT peaks for this particular system are shown in Fig. 11b. For $\Gamma_1 = \Gamma_3 = \Gamma_4 = \Gamma$, $\Gamma_2 = 0$ which is valid for loopy inverted-tripod system as shown in Fig. 11c, all the individual terms of the “Interference1” and “Interference2” are positive and hence there is constructive interference between the AT peaks.

For $\phi = \pi/2$ the different parameters for the dressed states are given below:

\[ E_{d_1} = \frac{\Omega_1^\prime}{2}; |d_1\rangle = \sqrt{\Omega_1^\prime + \Omega_2^\prime} \left( |\Omega_1^\prime + \Omega_1^\prime| \right) - 2 \Omega_1^\prime |\Omega_1^\prime + \Omega_1^\prime| + |\Omega_1^\prime + \Omega_1^\prime| \]

\[ E_{d_2} = 0; |d_2\rangle = - \frac{\Omega_1^\prime}{2} |\Omega_1^\prime + \Omega_1^\prime| + \frac{\Omega_1^\prime}{2} |\Omega_1^\prime + \Omega_1^\prime| \]

\[ E_{d_3} = \frac{\Omega_1^\prime}{2}; |d_3\rangle = \left( \frac{\Omega_1^\prime + \Omega_3^\prime}{\sqrt{2}} \right) |\Omega_1^\prime + \Omega_3^\prime| \]

\[ E_{d_4} = \frac{\Omega_1^\prime}{2}; |d_4\rangle = \left( \frac{\Omega_1^\prime + \Omega_3^\prime}{\sqrt{2}} \right) |\Omega_1^\prime + \Omega_3^\prime| \]

Now we consider specific systems with specific laser parameters. First, we consider the case for $\Gamma_1 = \Gamma_3 = \Gamma_4 = 0$, $\Gamma_2 = \Gamma$ which is valid for the Loopy-tripod system with $\Omega_{23} = \Omega_{34} = \Omega_{24} = 0.5\Gamma$ which is for all the cases. For this case, all the individual terms of “Interference1” and “Interference2” are negative and hence there is prominent destructive interference between AT peaks shown in Fig. 12a as the probe absorption goes to zero at the crossing points of the AT peaks. Further, the probe absorption is also lower than the sum of all the AT peaks in the overlapping region. This case is also similar for $\Gamma_2 = \Gamma_4 = \Gamma$, $\Gamma_1 = \Gamma_3 = 0$ which is valid for loopy N-system as shown in Fig. 12b, however the magnitude of the interference terms are less as compared to the loopy-tripod system. For $\Gamma_1 = \Gamma_3 = \Gamma_4 = \Gamma$, $\Gamma_2 = 0$ which is valid for loopy inverted-tripod system as shown in Fig. 12c all the individual terms of the “Interference1” and “Interference2” are positive and hence there is constructive interference between the AT peaks.

### III. CONCLUSION

In conclusion, we present the theoretical frame work to identify the nature and the role of interference between the Autler-Townes peaks (dressed states) in multi-level system. In three-level system the two AT peaks interferes pair-wise and the nature of interference is very simple which can be constructive, destructive or no interference depending upon the decay rate of the states coupled by
Figure 12: AT peaks and effect of interference for probe absorption vs its detuning ($\delta_{12}/\Gamma$) with $\delta_{34} = \delta_{24} = 0$, $\Omega_{23} = \Omega_{34} = \Omega_{24}$ i.e. $\Omega_{p1} = \frac{1}{\sqrt{2}} (\sqrt{\Omega_{23}} + \Omega_{12}$, $\Omega_{p2} = \frac{1}{\sqrt{2}} (\sqrt{\Omega_{34}} - \sqrt{\Omega_{24}}$, $\Omega_{p3} = \frac{1}{\sqrt{2}} (\sqrt{\Omega_{34}} - \Omega_{24}$ and $\phi = \pi/2$. a) $\kappa_{12} = \kappa_{23} = \sqrt{\frac{\Gamma}{\Gamma - \delta_{12}}}$, $\kappa_{13} = -\left(\frac{\sqrt{\Omega_{12}}}{\sqrt{\Omega_{23}} - \delta_{23}}\right)^{2} \frac{\Gamma}{\delta_{12}}$, $\Gamma_{\text{d}_1} = \Gamma_{\text{d}_2} = \Gamma_{\text{d}_3} = \frac{\Gamma}{2}$ b) $\kappa_{12} = \kappa_{23} = \frac{\Gamma}{\sqrt{\Omega_{23} - \delta_{23}}}$, $\kappa_{13} = -\left(\frac{\sqrt{\Omega_{12}}}{\sqrt{\Omega_{23}} - \delta_{23}}\right)^{2} - 1 \frac{\Gamma}{\delta_{12}}$, $\Gamma_{\text{d}_1} = \Gamma_{\text{d}_2} = \Gamma_{\text{d}_3} = \frac{\Gamma}{2}$ c) $\kappa_{12} = \kappa_{23} = \frac{\Gamma}{\sqrt{\Omega_{23} - \delta_{23}}}$, $\kappa_{13} = \left(\frac{\sqrt{\Omega_{12}}}{\sqrt{\Omega_{23}} - \delta_{23}}\right)^{2} - 1 \frac{\Gamma}{\delta_{12}}$, $\Gamma_{\text{d}_1} = \Gamma_{\text{d}_2} = \Gamma_{\text{d}_3} = \frac{\Gamma}{2}$.

IV. ACKNOWLEDGMENT

E.O.N. would like to acknowledge Indian Council for Cultural Relations (ICCR) for the PhD scholarship. K.P. would like to acknowledge the funding from SERB of grant No. ECR/2017/000781. We would like to thank David Wilkowski for his intriguing comments.

[1] A. Imamoglu, Phys. Rev. A 40, 2835 (1989), URL https://link.aps.org/doi/10.1103/PhysRevA.40.2835
[2] Y.-q. Li and M. Xiao, Phys. Rev. A 51, 4959 (1995), URL https://link.aps.org/doi/10.1103/PhysRevA.51.4959
[3] G. S. Agarwal, Phys. Rev. A 55, 2467 (1997), URL https://link.aps.org/doi/10.1103/PhysRevA.55.2467
[4] S. Khan, V. Bharti, and V. Natarajan, Physics Letters A 380, 4100 (2016), ISSN 0375-9601, URL http://www.sciencedirect.com/science/article/pii/S0375960116313433
[5] P. M. Anisimov, J. P. Dowling, and B. C. Sanders, Phys. Rev. Lett. 107, 163604 (2011), URL https://link.aps.org/doi/10.1103/PhysRevLett.107.163604
[6] L. Giner, L. Veissier, B. Sparkes, A. S. Shremet, A. Nicolas, O. S. Mishina, M. Sherman, S. Burks, I. Shomroni, D. V. Kupriyanov, et al., Phys. Rev. A 87, 013823 (2013), URL https://link.aps.org/doi/10.1103/PhysRevA.87.013823
[7] B. Peng, a. K. Ozdemir, W. Chen, F. Nori, and L. Yang, 5 (2014), URL https://www.nature.com/articles/ncomms6082#supplementary-informations
[8] E. Saglamyurek, T. Hrushevskiy, A. Rastogi, K. Heshami, and L. J. LeBlanc, Nature Photonics 12, 774 (2018), URL https://doi.org/10.1038/s41566-018-0279-0
[9] A. Rastogi, E. Saglamyurek, T. Hrushevskiy, S. Hubele, and L. J. LeBlanc, arXiv preprint arXiv:1902.02815
[10] U. Sinha, C. Couteau, T. Jennewein, R. Laflamme, and M. Asad Siddiqui and T. Qureshi, Progress of Theoretical Physics 132, 774 (2018), URL https://arxiv.org/abs/1902.02815
[11] M. Asad Siddiqui and T. Qureshi, Progress of Theoretical and Experimental Physics 2015 (2015), ISSN 2050-3911, http://oup.prod.sis.lan/ptep/article-pdf/2015/8/083A02/7698190/ptp112.pdf, URL https://doi.org/10.1093/ptep/ptv112

the strong control lasers. In four-level system the nature of interference is more complicated but again all the three AT peaks interferes pair-wise. There are two terms for the interference, one that is similar to the three-level system and another that is a little bit more complicated. We have also done similar calculation for five level system which involves further complicated nature of the interference. For any system, if the decay rate of the levels coupled by the control lasers are equal then there is no interference between any of the AT peaks.
12

[12] A. Sinha, A. H. Vijay, and U. Sinha, Scientific Reports 5, 10304 (2015), URL https://doi.org/10.1038/srep10304

[13] M. G. Bason, A. K. Mohapatra, K. J. Weatherill, and C. S. Adams, Journal of Physics B: Atomic, Molecular and Optical Physics 42, 075503 (5pp) (2009), URL http://stacks.iop.org/0953-4075/42/075503

[14] Y. Chen, X. G. Wei, and B. S. Ham, Journal of Physics B: Atomic, Molecular and Optical Physics 42, 065506 (2009), URL http://stacks.iop.org/0953-4075/42/i=6/a=065506

[15] J. Sheng, X. Yang, U. Khadka, and M. Xiao, Opt. Express 19, 17059 (2011), URL http://www.opticsexpress.org/abstract.cfm?URI=oe-19-18-17059

[16] T. Hong, C. Cramer, W. Nagourney, and E. N. Fortson, Phys. Rev. Lett. 94, 050801 (2005).

[17] J. A. Sedlacek, A. Schwettmann, H. Kubler, R. Low, T. Pfau, and J. P. Shaffer, Nature physics 8, 819 (2012), URL http://link.aps.org/doi/10.1038/nphys2247

[18] B. Zhang, J.-H. Wu, X.-Z. Yan, L. Wang, X.-J. Zhang, and J.-Y. Gao, Opt. Express 19, 12000 (2011), URL http://www.opticsexpress.org/abstract.cfm?URI=oe-19-13-12000

[19] A. Ghosh, K. Islam, S. Mondal, D. Battacharya, N. Pal, and A. Bandyopadhyay, Journal of Physics B: Atomic, Molecular and Optical Physics 51, 145001 (2018), URL https://doi.org/10.1088%2F1361-6455%2Faac6f5

[20] S. Kumar, T. Lauprêtre, F. Bretenaker, F. Goldfarb, and R. Ghosh, Phys. Rev. A 88, 023852 (2013), URL http://link.aps.org/doi/10.1103/PhysRevA.88.023852

[21] F. Leroux, K. Pandey, R. Rehbi, F. Chevy, C. Miniatura, B. Gremaud, and D. Wilkowski, Nature Communications 9, 3580 (2018), URL https://doi.org/10.1038/s41467-018-05865-3

[22] Y.-X. Hu, C. Miniatura, D. Wilkowski, and B. Grémaud, Phys. Rev. A 90, 023601 (2014), URL http://link.aps.org/doi/10.1103/PhysRevA.90.023601

[23] K. Pandey, C. C. Kwong, M. S. Pramod, and D. Wilkowski, Phys. Rev. A 93, 053428 (2016), URL http://link.aps.org/doi/10.1103/PhysRevA.93.053428

[24] L. Li, H. Guo, F. Xiao, X. Peng, and X. Chen, J. Opt. Soc. Am. B 22, 1309 (2005), URL http://josab.osa.org/abstract.cfm?URI=josab-22-6-1309

[25] D. Shylla, E. N. Ogaro, and K. Pandey, Scientific Reports 8, 8692 (2018), URL https://doi.org/10.1038/s41598-018-27011-1

[26] D. Shylla and K. Pandey, arXiv:1802.09935 (2018), URL https://arxiv.org/abs/1802.09935

[27] L. Hao, Y. Jiao, Y. Xue, X. Han, S. Bai, J. Zhao, and G. Raithel, New Journal of Physics 20, 073024 (2018), URL https://doi.org/10.1088%2F1367-2630%2Faaf4b0

[28] M. T. Simons, M. D. Kautz, C. L. Holloway, D. A. Anderson, G. Raithel, D. Stack, M. C. St. John, and W. Su, Journal of Applied Physics 123, 203105 (2018), https://doi.org/10.1063/1.5020173, URL https://doi.org/10.1063/1.5020173