Nonlinear Fano resonance without a continuum

Y.-N. Lv,1,2, * A.-W. Liu,2,3, * Y. Tan,2,3, * C.-L. Hu,3 T.-P. Hua,3 X.-B. Zou,1,2 Y. R. Sun,2,3, † C.-L. Zou,1,2,4, ‡ G.-C. Guo,1,2 and S.-M. Hu2,3,5, §

1CAS Key Lab of Quantum Information, University of Science and Technology of China, Hefei, Anhui 230026, P. R. China
2CAS Center for Excellence in Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei 230026, China
3Department of Chemical Physics, University of Science and Technology of China, Hefei 230026, China
4State Key Laboratory of Quantum Optics and Quantum Optics Devices, and Institute of Opto-Electronics, Shanxi University, Taiyuan 030006, China
5Institute of Advanced Science Facilities, Shenzhen, 518107, China

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A new mechanism of Fano resonance induced by distant discrete levels in atoms or molecules is proposed and experimentally verified by Doppler-free Lamb-dip spectroscopy of vibration-rotational transitions of CO$_2$. The observed spectrum has an asymmetric profile and its amplitude increases quadratically with the probe laser power. Our results appeal to further investigations of Fano resonances and facilitate a broad range of topics based on narrow transitions.

Since first revealed by Ugo Fano in the spectroscopy of atomic helium near the ionization threshold [1, 2], Fano resonance has attracted tremendous attention in various systems [3–11]. Fano resonance is acknowledged as a universal phenomenon in resonant wave-scattering processes, for the transport of photons [3–6, 10, 11], electrons [7–9] and collective excitations (such as phonons) in a very wide range of systems [12]. The characteristic asymmetric Fano spectral profile is a consequence of the quantum interference between two transition paths, one towards a discrete excited state and the other towards a continuum [2, 8]. The Fano profile not only offers the information of the amplitude and the phase of distinct transition paths for unraveling the internal physical processes [13, 14], but also provides an alternative knob to control the scattering or the excitation processes in optical and electronic devices [8, 15, 16].

Although the Fano resonance has been studied extensively, it is commonly believed to be absent if the target transition is far from the continuum. In particular, for low-lying states of atoms and molecules, symmetric spectral profiles (mostly the Lorentz function) are assumed in practical precision measurements [17–22]. It was surprising that Fano-like asymmetric profile was observed in saturated absorption spectroscopy of an isolated rotation-vibration transition of CO$_2$ [19, 20, 23, 24], which could not be explained by the conventional Fano resonance. Up to now, the physics of this asymmetric profile has been not interpreted, thus the corresponding investigation is limited. Moreover, the puzzling spectra pose challenges for evaluating the spectral data and testing fundamental physics at high precision [20, 25], where various narrow transitions of atoms and molecules are involved.

Here, we propose and experimentally demonstrate a new mechanism of nonlinear Fano resonance (NFR) among a few discrete energy levels in atoms or molecules, while no continuum is necessarily involved. The effect is numerically investigated and experimentally verified with vibration-rotational transitions of the CO$_2$ molecules, without a continuum being included in the calculation but discrete levels with detuning up to $10^{20}$ times of the natural linewidth of the interested transition. This NFR mechanism can significantly change the shape of the spectrum and cause considerable deviations in precision measurements.
Model and principle.—The mechanism of NFR is illustrated by a conventional precision spectroscopy measurement set-up in Fig. 1(a), where a target narrow-linewidth transition ($|1\rangle \leftrightarrow |2\rangle$ with frequency $\omega_{12}$) is probed by a near-resonant laser ($\omega_p$). However, the probe would not only excite the target transition, but also inevitably couple with other excess transitions, particularly when a strong probe field is applied in measurements of the ultra-weak transition. For example, as shown in Fig. 1(b), considering a far-off resonance excited state $|3\rangle$, the probe field excites the molecule to both the $|2\rangle$ with a probability $P \propto \gamma_{12}/|\Delta_B|^2$ and the $|3\rangle$ through the transition $|2\rangle \leftrightarrow |3\rangle$ with a Rabi frequency $\Omega_{23} \propto \sqrt{\gamma_{23}}$. Here, $I$ is the laser intensity, $\Delta_B$ is the linewidth due to transit-time broadening, which is usually much larger than the natural linewidth, and $\gamma_{12(23)}$ denotes the spontaneous emission rate from $|2(3)\rangle$ to $|1(2)\rangle$ [26]. For the far-off resonance, $|\Delta_{23}| = |\omega_p - \omega_{23}| \gg \Omega_{23}, \gamma_{23}$, the excess state induces a modification of the frequency of $|2\rangle$ due to the AC Stark effect [27], which can be estimated as

$$\delta = \frac{\Omega_{23}^2}{\Delta_{23}} \approx \frac{\gamma_{23}^2}{\gamma_{12} \Delta_{23}} P. \quad (1)$$

It's anticipated that for long-lived energy level $|2\rangle$ with $\gamma_{12} \ll \Delta_B$, the excess effect should not be ignored if $\delta$ is comparable with $\Delta_B$. For example, considering a typical requirement of $P \sim O (10^{-4})$ to enable an observable saturating absorption of a narrow transition, if $\Delta_B/2\pi \sim 10^9$ Hz for room-temperature gases, an excess transition has $\gamma_{23}/2\pi \sim 10^8$ Hz and $\Delta_{23}/2\pi \sim 10^{15}$ Hz, then the excess effect beyond the conventional two-level approximation should be considered when $\gamma_{12}/2\pi \lesssim 10^{-4}$ Hz.

As shown in Fig. 1(c), when molecules travel through the standing-wave field, periodic excitation and periodic energy level modulation on $|2\rangle$ appear simultaneously. Thereafter, the system is described by the Floquet Hamiltonian

$$H/\hbar = \sum_j \left\{ \left[ \Delta_{12} - \delta \cos^2 (k_j z_j) \right] \sigma_{22}^j + \Omega_{12} \cos (k_j z_j) \left( \sigma_{12}^j + \sigma_{21}^j \right) \right\}. \quad (2)$$

Here, superscript or subscript $j$ indicates the $j$-th molecule, $\Delta_{12} = \omega_{12} - \omega_p$, $k_j$ is the wave vector of the probe field, $z_j = V_j t$ is the instant location of the molecule with longitudinal velocity $V_j$, $\sigma_{ab} = \langle a | b \rangle$, and $\Omega_{12}$ is the Rabi frequency. Intuitively, the Floquet interaction in the laboratory frame [Fig. 1(c)] can be converted to the interaction in the molecular frame [Fig. 1(d)] and the corresponding Hamiltonian becomes $H/\hbar = \sum_j \left\{ \left[ \Delta_{12} - \frac{\delta}{2} \right] \sigma_{22} + \Omega_{12} \cos (k_j z_j) \sum_m \left[ J_m (r_j) e^{i2m k_j V_j t} \sigma_{12}^j + h.c \right] \right\}$, where $J_m (\cdot)$ is the Bessel function of order $m$, $r_j = \delta \lambda/8\pi V_j$ [26]. Therefore, the moving molecules in the standing-wave probe field can be treated as static molecules probed by a comb, which is significantly different from the conventional two-level treatment of the system in Fig. 1(a).

Asymmetric spectral profile.—Although the above discussions hold for all types of absorption spectroscopy, we focus on atoms or molecules in a gas cell. For $\zeta_j \ll 1$, only the low order sidebands $(J_0, \pm 1, \pm 2)$ of the comb are considered. As schematically shown in Fig. 2(a), an asymmetric absorption spectrum emerges due to the $\pi$ phase difference between sidebands $(J_{\pm 1} (\zeta_j) \approx \pm \zeta_j/2)$. Analytically, a Doppler-free fine spectral profile on the Doppler-broadened background, could be approximated solved as [26]

$$\Delta P \propto I^2 F^2 (\Delta_{12}) + \sqrt{2 \hbar \omega_p^3} \frac{\delta I \Delta_{23}^2}{\gamma_{12}} \frac{\delta F (\Delta_{12})}{\delta \Delta_{12}}. \quad (3)$$

Here $F (\Delta_{12}) = \exp \left[ -(\Delta_{12} - \frac{\delta}{2})^2 / \Delta_B^2 \right]$ and $c$ is the speed of light. The first term corresponds to the symmetric saturated absorption spectrum (SAS), and the second term indicates an additional anti-symmetric spectral profile due to the NFR. Since $\delta \propto I$, both terms are quadratically proportional to $I$, in sharp contrast with the conventional Fano resonance. Obviously, by increasing $\delta/\gamma_{12}$, the second term becomes dominant and the spectrum changes from a symmetric to an
asymmetric profile. The mechanism was also verified numerically with Eq. (2). In Fig. 2(b), the typical Doppler-broadened spectrum for molecule ensemble coupling with a standing wave is simulated by taking \(\gamma_{12}/2\pi = 2 \times 10^{-5}\text{Hz}, \delta/2\pi = 0.02\text{MHz},\) and the most probable transit-time broadening \(\Delta_B/2\pi = 0.4\text{MHz}.\) The Doppler-free profile of Fig. 2(b) is magnified and shown in Fig. 2(c), which reveals the asymmetric spectral feature. If we change the modulation amplitude \(\delta/2\pi\) from 0.02 MHz to 0.12 MHz, the asymmetric feature becomes even more significant [Fig. 2(d)].

**Benchmark of asymmetry.**—The degree of the asymmetry could be quantified by the Fano factor \(q\), which is derived by fitting the spectrum using the Fano function [2]:

\[
P = A \frac{q^2 - 1}{1 + (\omega - \omega_q)^2} + 2A \frac{q(\omega - \omega_q)}{1 + (\omega - \omega_q)^2} + B , \tag{4}
\]

where \(A, B, \omega_q\), and \(\Gamma\) are the fitting parameters. In particular, \(\omega_q\) corresponds to a fitting frequency shift with respect to the unperturbed transition frequency, and \(\Gamma\) represents the linewidth. In Figs. 2(c) and 2(d), the Doppler-free spectra are perfectly fitted by the Fano function, with \(q = 0.19\) and 0.67, respectively. The physics unraveled by the asymptotic solution [Eq. (3)] is also numerically examined with an effective model which only includes low-order sidebands \(j_0 \pm 1\). Although the dynamics of the molecule ensemble are complicated, the results of the effective model agree well with the full model thus further validating the underlying mechanism of NFR Comparing Eq. (3) with Eq. (4), the Fano factor can be evaluated as [26]

\[
\tilde{q} \approx \frac{\sqrt{2} \omega_{12}^3}{32 \pi^2 c^2} \frac{\delta}{\gamma_{12}} = \frac{3 \sqrt{2} \omega_{12}^3 \gamma_{23} \Delta_B}{4 \pi \omega_{12}^3 \gamma_{12} \Delta_B} \tag{5}
\]

for \(q \ll 1\), which indicates that the degree of asymmetry increases with the dimensionless parameter \(\delta/\gamma_{12}\) and \(\Delta_B\).

The mechanism of NFR and the predictions of \(\tilde{q}\) are further tested by systematically studying the Fano factor via the full model, and the numerical results are summarised in Fig. 3. Apparently, \(q\) monotonously increases with \(\delta\) but decreases with \(\gamma_{12}\), as shown in Fig. 3(a) and (b). Moreover, the fitted frequency shift \(\omega_q\) in Fig. 3(a) implies a systematic deviation in the transition frequency obtained from the fitting when probing the weak transition. Because \(\delta\) varies with the transverse speed \(V_t\) of the molecule, the dependence of \(q\) on \(V_t\) is investigated and the results are shown in Fig. 3(c). These numerical results of \(q\) suggest a linear dependence on \(\delta, \Delta_B,\) and \(1/\gamma_{12}\) when \(q \ll 1\), and agree well with the prediction of \(\tilde{q}\). Such linear dependence of \(q\) on \(\delta/\gamma_{12}\) is also directly confirmed in Fig. 3(e), which shows that the contours of \(q\) almost overlap with those of \(\delta/\gamma_{12}\). Furthermore, typical absorption profiles for the noted points \((p_1, p_2)\) in Fig. 3(c) are shown in Fig. 3(d), and the spectrum drastically changes from weakly asymmetric to almost anti-symmetric when \(q\) increases from 0.24 to 0.87. It is also found that \(q\) saturates to 1 for increasing \(\delta \Delta_B/\gamma_{12}\). The character of \(q \leq 1\) manifests a fundamental difference between the asymmetric profile of our NFR with the conventional Fano resonance [2], because the asymmetry arises from the modulation-induced anti-symmetric components in Eq. (3). Therefore, by including the saturation behaviour of the fitted \(q\) in practice, we can provide a rule of thumb for observing the Fano profiles of weak transitions as \(q \approx \tilde{q}/(1 + \tilde{q})\). According to this formula, we derived the \(q\) for the parameters in Figs. 2(c) and 2(d), the noted points \(p_1\) and \(p_2\) in Fig. 3(d) as 0.16, 0.54, 0.15 and 0.76 respectively. Obviously, the above formula provides a reasonable estimation of the Fano factor, and it is studied in more detail in [26].

**Experiment.**—The proposed mechanism could explain the asymmetric profile observed in the cavity-enhanced absorption spectroscopy of HD [19, 20, 26]. However, the
The presence of the splitting due to the hyperfine structure in the HD molecule [28], which is comparable to the observed line width, complicates the interpretation of the asymmetric line profile. To circumvent the ambiguity, we further tested the NFR in two independent experiments by measuring the Doppler-free absorption spectra of a series of transitions of CO$_2$. The CO$_2$ molecule is chosen because its energy structure is well-understood and the hyperfine splitting of the vibration-rotational levels in CO$_2$ is also negligible, which avoids the influence of other near-resonance energy levels. The first setup employs the cavity ring-down spectroscopy (CRDS) method, which measures the direct absorption coefficients for different weak transitions of $^{13}$C$^{16}$O$_2$ [29], as shown in Fig. 4(a), and an example spectrum is shown in the inset. Infrared transitions with $\gamma_{12}$ values spanning in the range of $10^{-5} - 10^{-2}$ Hz, weaker than typical CO$_2$ electronic transitions by over one billion times, are investigated under the same experimental conditions. All these selected weak transitions are confirmed to be well isolated, therefore possible distortion of the profile due to nearby transitions is excluded. The extracted $q$ values are shown in Fig. 4(c). Quantitative agreement between the experimental and theoretical results is achieved by considering the AC stark shift $\delta/2\pi = 0.07 \pm 0.02$ MHz, as noted in the pink area of the figure.

The second setup implements the wavelength-modulated cavity enhanced absorption spectroscopy (WM-CEAS) method [24], which is shown in Fig. 4(b). Transitions of the main isotopologue $^{12}$C$^{16}$O$_2$ are studied. Note that neither $^{12}$C nor $^{16}$O nucleus has nuclear spin, and the $^{12}$C$^{16}$O$_2$ molecule has no hyperfine structure. The inset of Fig. 4(b) shows the WM-CEAS spectrum of the $^{12}$C$^{16}$O$_2$ transition at 6413.9526 cm$^{-1}$ with an Einstein $A$-coefficient of $1 \times 10^{-5}$ s$^{-1}$, corresponding to $\gamma_{12}/2\pi = 1.6 \mu$Hz. To the best of our knowledge, this is the molecular absorption line with the smallest transition moment ever detected by Doppler-free spectrum [30]. Different intra-cavity optical powers up to 1570 W were used in this experiment, allowing us to reveal the dependence of $q$ on $I$ shown in Fig. 4(d), which is in good agreement with our numerical simulation for $\delta/2\pi = 0.07 \pm 0.02$ MHz. In addition, the result implies the relation that $\delta$ is proportional to $I$ and $\tilde{\gamma}$ is insensitive to $I$, that further verifying the predictions of $\tilde{\gamma}$. It is worth noting that the same parameter $\delta$ was used in the simulations of both $^{13}$C$^{16}$O$_2$ and $^{12}$C$^{16}$O$_2$ spectra, since far off-resonance electronic states are same for both isotopologues. Therefore, the agreement between the theory and experimental results unambiguously validates our model and confirms the mechanism of NFR without a continuum when measuring weak transitions.

**Conclusion.**—We theoretically predicted and experimentally demonstrated a new type of Fano resonance without involving a continuum. Such a mechanism can significantly change the shape of the spectrum, appealing for more careful evaluation and investigation of the two-level approximation when performing spectral measurements on ultra-narrow resonances. Meanwhile, our work provides an approach to evaluate such a systematic shift for acquiring a more accurate line center. Moreover, the observed Fano factor could serve as an indicator of perturbation from unknown strong transitions that could not be effectively probed. Ultimately, the results provide new insights into Fano resonance and affect a variety of important applications using these narrow transitions, including optical clocks [17, 18], long-lived memory of quantum information [31, 32], testing of quantum theory [19, 20], determination of fundamental constants [21] and searching for new physics beyond the Standard Model [22].

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