Interference between atomic Rb $(5d_{5/2}–5p_{3/2})$ and $(5p_{3/2}–5s_{1/2})$ coherences: observation of an exceptional point by quantum beating at $\sim 2.1$ THz

W Goldshlag, R Su, S Park*, T O Reboli and J G Eden*

Laboratory for Optical Physics and Engineering, Department of Electrical and Computer Engineering, University of Illinois, Urbana, IL 61801, United States of America
E-mail: parksehyun0129@gmail.com and jgeden@illinois.edu

Received 30 May 2021, revised 27 July 2021
Accepted for publication 2 August 2021
Published 17 September 2021

Abstract
Coupled oscillators are prevalent in nature and fundamental to fields as disparate as astrophysics, photonics, the mechanical sciences, and geophysics. Theory has identified singularities in the response of coupled oscillators, known as exceptional points (EPs), that are associated with non-Hermitian operators and lie at the transition between weak and strong coupling of the oscillator. Although several EPs have been reported or predicted to exist in nanophotonic resonators and Feshbach resonances, for example, tuning the phase of two interfering atomic or molecular coherences near an EP has not been demonstrated previously.

We report the observation of an EP associated with a pair of interfering atomic coherences in Rb, oscillating at 386.3 and 384.2 THz, and confirm the theoretical prediction of an abrupt phase shift of $\sim \pi/4$ as the EP is traversed by independently varying two experimental parameters. Pairs (and trios) of coupled coherences in thermal Rb atoms are established among the $7s_{1/2}, 5d_{5/2}, 5p_{3/2}$, and $5s_{1/2}$ states in pump–probe experiments with <200 fs laser pulses, and observed directly in the temporal and spectral domains through the ensuing quantum beating in the $\sim 2–36$ THz interval. Interference between the $(5d_{5/2}–5p_{3/2})$ and $(5p_{3/2}–5s_{1/2})$ coherences is mediated by the $5p_{3/2}$ state and detected through quantum beating in the vicinity of the $(5d_{5/2}–5p_{3/2})–(5p_{3/2}–5s_{1/2})$ difference frequency of 2.11 THz which is monitored by a parametric four-wave mixing process. Phase of this composite atomic oscillator is first controlled by varying the mean Rb–Rb nearest neighbor distance $\langle R \rangle$ in a thermalized vapor. A discontinuous transition of $(0.8 \pm 0.2) \sim \pi/4$ radians in the phase of the coupled oscillator occurs when $\langle R \rangle$ is varied over the $\sim 80–90$ nm interval, a phase shift associated with the transformation of a broadband, dissipative oscillator (characterized by a Fano interference window) into a strongly-coupled system resonant at 2.1 THz.

Keywords: THz quantum beating, wavepacket interference, Fano resonance, exceptional point

* Authors to whom any correspondence should be addressed.

Original content from this work may be used under the terms of the Creative Commons Attribution 4.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
1. Introduction

Physical systems described by non-Hermitian operators are characterized by singularities in parameter space at which PT symmetry is broken. Known as exceptional points (EPs) [1–5], such singularities have proven challenging to observe experimentally [1, 6, 7] and virtually all of those documented to date have been identified in coupled-photon resonators such as photonic molecules [6, 8–13]. Classical and quantum models of coupled oscillators [1, 4, 6, 14, 15] demonstrate that: (1) EPs are situated at the transition between weak and strong coupling of the composite oscillator, and (2) their fundamental properties are captured in the spectral domain by a Fano lineshape which reflects the interference between competing resonances or exit channels [16–31].

Recently, coupled resonator models have also explored the common thread between Fano and Feshbach resonances associated with interfering decay channels [3], and have yielded generalized Fano spectral profile expressions [6, 31] which provide insight into the linkages between quantum optical phenomena such as electromagnetically-induced transparency and Autler–Townes splitting [4, 6, 32, 33]. Undoubtedly the most significant property of EPs is the predicted shift of $\sim \pi/4$ in the phase of a coupled system as an EP is approached by varying an appropriate system parameter [2]. Günther et al. have noted: ‘the phase-jump behavior can be used as an implicit indicator of a possible close location of an EP...’ [2]. In summary, despite the progress of the past decade in recognizing and analyzing EPs, few time-resolved studies of interfering atomic or molecular oscillators have appeared in the literature and the theoretically-predicted, abrupt shift in phase in the vicinity of an atomic EP has not been confirmed experimentally.

We describe here the observation of an EP associated with a pair of interfering coherences produced simultaneously in the Rb atom. Furthermore, the phase of this coupled oscillator is found to change discontinuously by $(0.8 \pm 0.2) \sim \pi/4$ radians as the EP is traversed, thereby confirming one signature of the singularity. Both pairs and trios of coupled atomic oscillators are prepared by establishing quantum coherences among at least four states of atomic Rb through one- and two-photon excitation of the atom in the vapor phase with $< 200$ fs laser pulses. The temporal evolution of the coupled system is monitored by quantum beating in the $\sim 2$–36 THz frequency interval and is detected optically by parametric four-wave mixing (PFWM) [34, 35]. In pump–probe experiments, quantum beating is observed at multiple frequencies, including the $7s^2 5S_{1/2} \rightleftharpoons 5D_{3/2}$ (denoted $7s_{1/2} \rightarrow 5s_{3/2}$) atomic state difference frequency of 18.22(5) THz. Of particular interest, however, is interference between the Rb $(5d_{5/2} \rightarrow 5p_{3/2})$ and $(5p_{3/2} \rightarrow 5s_{1/2})$ coherences, coupled through the intermediate $5p_{3/2}$ state, that manifests itself as a generalized Fano lineshape [6, 31] associated with the $(5d_{5/2} \rightarrow 5p_{3/2})$–$(5p_{3/2} \rightarrow 5s_{1/2})$ resonance at 2.11 THz. Controlling the phase of this composite quantum system, denoted $(5d_{5/2} \rightarrow 5p_{3/2})$–$(5p_{3/2} \rightarrow 5s_{1/2})$, and its associated lineshape is first accomplished by varying the mean Rb–Rb interatomic separation $(\langle R \rangle)$ in a thermalized vapor (which is collision-free throughout the measured $(5d_{5/2} \rightarrow 5p_{3/2})$–$(5p_{3/2} \rightarrow 5s_{1/2})$ coherence time of $< 30$ ps), thus affirming the conclusion of Limonov et al [29] that the ability to manipulate the Fano profile ‘...provides a route toward understanding and controlling interference processes across several branches of physics’. The ability to tune the phase of the composite oscillator in this way is attributed to induced-dipole interactions with background thermal Rb atoms which define the interatomic potential at $(\langle R \rangle)$, $V(\langle R \rangle)$. Increasing $(\langle R \rangle)$ from $\sim 80$ nm to $90$ nm results in an abrupt shift of $\sim \pi/4$ radians in the Fano phase which is in accord with the calculations of reference [2] and is attributed to induced-dipole interactions $(C_3 R^{-3})$ with background atoms. Over the narrow region in $(\langle R \rangle)$ in which the phase transition occurs, the interference window in the Fano lineshape is suppressed, and the resonance peak at $\sim 2.1$ THz appears, the strength of $C_3 R^{-3}$ interactions increases by a factor of $\sim 2$–3. Tuning of the coupled-oscillator phase through an EP with the laser intensity and a three coupled-coherence system, $(7s_{1/2} \rightarrow 5d_{5/2})$–$(5d_{5/2} \rightarrow 5p_{3/2})$–$(5p_{3/2} \rightarrow 5s_{1/2})$, have also been observed. Regardless of the experimental parameter that is varied, interference structure dominating the Fourier spectrum of the non-resonant coupled oscillator collapses as the EP is traversed. Thus, the impact of the observed phase shift is to transform a broadband, dissipative oscillator into a strongly-coupled resonant system. Conducting these experiments in thermalized vapor–gas mixtures allows for the impact of atom–atom interactions on the coupled quantum coherences to also be observed. Indeed, comparison of Fano profiles near 2.1 THz in Rb vapor with those in Rb–Cs and Rb–Ar mixtures provides further support for the presence of the EP and demonstrates tuning of coupled atomic coherences through an EP by means of Rb state-specific interactions, at long range $(50–100$ nm), with background atoms.

2. Probing interfering atomic coherences by quantum beating and PFWM

Panel (a) of figure 1 is a partial energy level diagram for Rb, illustrating the optical processes responsible for generating and detecting quantum beating at 2.1 THz and other atomic state difference frequencies. Both PFWM and quantum beating are driven simultaneously by $< 200$ fs pulses from a Ti:Al$_2$O$_3$ laser which have sufficient bandwidth ($\sim 20$ nm FWHM) to encompass both the $7s_{1/2} \leftrightarrow 5s_{3/2}$ and
Figure 1. Optical processes responsible for producing and detecting coupled atomic coherences, and representative time- and frequency-domain data: (a) qualitative, partial energy level diagram for Rb, illustrating one- and two-photon excitation of the atom with <200 fs pulses (centered at ~768 nm) so as to produce multiple quantum oscillators. Quantum beating is monitored in pump–probe experiments through PFWM. The vertical red arrows represent coherences established between the $5d_{5/2}$, $5p_{3/2}$, and $5s_{1/2}$ states of Rb while one- and two-photon excitation transitions, as well as the PFWM idler waves, are indicated by black lines. For clarity, resonant excitation of the $5p_{3/2}$ state by a single-photon transition is not shown. By varying the pump–probe delay time ($\Delta t$), Ramsey fringes associated with the signal wave intensity are observed (shown in blue) which, in the Fourier domain, yield a generalized Fano spectral profile having an interference window lying at ~70.3 cm$^{-1}$ (~2.1 THz) which is ~0.1 cm$^{-1}$ above the resonance difference frequency for the $(5d_{5/2}→5p_{3/2})$: $(5p_{3/2}→5s_{1/2})$ oscillator. Also, dotted horizontal lines denote virtual states and the red shading represents twice the laser bandwidth; (b) and (c) Ramsey fringes, representative of those recorded throughout the experiments and observed by varying $\Delta t$ over the 4.5–6.5 ps and 598.8–600.2 ps intervals, respectively, while monitoring the signal wave intensity. In (b), oscillations having periods of ~35 fs and ~475 fs, corresponding to frequencies of 18.2 THz and 2.1 THz, respectively, are evident whereas 18.2 THz quantum beating dominates at long time-delays. These data were acquired for $[\text{Rb}] = 2.6 \times 10^{14}$ cm$^{-3}$.

$5d_{3/2} ← 5s_{1/2}$ two-photon transitions at 760.1 nm and 778.1 nm, respectively. Photoexciting both transitions with each pulse establishes coherent superpositions of states, and quantum beating at several atomic difference frequencies ensues. Of these, 18.2 THz ($7s_{1/2}→5d_{5/2}$) is most prominent but single-photon excitation of Rb also produces a coherent superposition of the $5p_{3/2}$ fine structure level and ground. Consequently, the $(5d_{5/2}→5p_{3/2})$ and $(5p_{3/2}→5s_{1/2})$ coherences are also produced which correspond to frequencies of 386.3 THz and 384.2 THz, respectively. As shown in figure 1(a), quantum beating is monitored by driving the PFWM process: $5s_{1/2} → (7s_{1/2}, 5d_{5/2}) → 6p_{3/2} → 5s_{1/2}$ and interference between the coherent idler waves (generated near the $7s_{1/2}, 5d_{5/2}→6p_{3/2}$ transitions) modulates the PFWM signal wave (6$p_{3/2} → 5s_{1/2}$) intensity at ~420.2 nm (shown in blue in figure 1(a)). Accordingly, the temporal evolution of the quantum beating associated with each observed coherence is monitored by scanning the time delay between the pump and probe pulses ($\Delta t$) while recording the relative PFWM signal wave intensity, as illustrated in figure 1(a). In the Fourier domain, the temporal dynamics of the coherences driving the PFWM process are reflected by spectra near ~2.1, 10.7, and 18.2 THz. It should be mentioned that these quantum oscillators may also be detected by six-wave parametric mixing [36]. Quantum beating at the $7s_{1/2}→5d_{5/2}$ difference frequency of Rb was first reported in reference [34] and sidebands were observed by Shen et al [35], but not resolved. As discussed in the text to follow, most of the experimentally-observed coherences appear in the Fourier domain as spectrally-narrow (i.e. high-Q) peaks. An exception is the $(5d_{5/2}→5p_{3/2})$: $(5p_{3/2}→5s_{1/2})$ coupled oscillator. In the vicinity of the $(5d_{5/2}→5p_{3/2})$: $(5p_{3/2}→5s_{1/2})$ difference frequency of 2.1 THz, a Fano lineshape having the classic interference window is observed (cf figure 1(a)). The physics of this pair of coupled coherences is of interest because the $5p_{3/2}$ state determines the coupling strength between the 386.3 and 384.2 THz oscillators, but it is the $5d_{5/2}$ state that serves as the interface between the coherences and the PFWM process. The broad, generalized Fano profile at lower right in figure 1(a), observed in the vicinity of 2.1 THz, indicates that the $(5d_{5/2}→5p_{3/2})$: $(5p_{3/2}→5s_{1/2})$ oscillator is weakly-coupled for this value of Rb number density ($[\text{Rb}] = 2.6 \times 10^{14}$ cm$^{-3}$). Finally, as noted above, varying the pump–probe delay time yields Ramsey fringes associated with the PFWM signal wave (figure 1(a), blue curve). Representative examples of the signals recorded in different $\Delta t$ intervals are given in panels (b) and (c) of figure 1.

3. Experimental arrangement and results

3.1. Experimental parameters and data acquisition

In these experiments, optical coherences are established among the $5s_{1/2}$, $5p_{3/2}$, $5d_{5/2}$, and $7s_{1/2}$ states of Rb through simultaneous one- and two-photon excitation of the atom with 50–200 fs pulses provided by a coherent Ti:sapphire (Ti:Al$_2$O$_3$) laser system. A schematic diagram of the experimental arrangement is given in figure 2. In order to maximize the amplitude of $7s_{1/2}→5d_{5/2}$ quantum beating (for the purpose of having the $7s_{1/2}→5d_{5/2}$ difference frequency serve as an experimental reference), the peak of the laser spectrum was intentionally set with custom internal optics to ~768 nm which lies slightly to the blue side of the mid-point between the $7s_{1/2} ← 5s_{1/2}$ and $5d_{5/2} ← 5s_{1/2}$ two-photon transitions (owing to the relative cross-sections for $\Delta l = 0$ and 2 two-photon transitions), and negative chirp was imposed upon the laser pulses (monitored by the FROG unit
of figure 2). The experiments reported here employed natural abundance or isotopically-enriched Rb vapor thermalized at temperatures of 423–488 K which correspond to Rb number densities of [Rb] = 1.0–9.2 × 10^{14} cm^{-3}. Rubidium vapor or Rb–Cs or Rb–Ar mixtures were contained in sapphire or fused silica cells with a length of 2.5 cm. Conducting these experiments in thermalized vapor/gas mixtures allows for the impact of atom–atom interactions on the coupled oscillators to be observed directly, and quantum beating to be detected through PFWM.

A Michelson interferometer having a temporal resolution of 3.3 fs (0.5 μm spatial resolution) has been developed that allows for ∆τ to be scanned up to 1100 ps while reducing noise in the Fourier domain by at least two orders of magnitude, relative to previous interferometers [33–35]. For each of the ~200000 steps in a typical ∆τ = 0–670 ps scan, for example, 20 measurements of the relative PFWM signal wave intensity (recorded with a Si photodiode) were averaged. Data were acquired in >50 scans of ∆τ, each of which required 17 h despite the laser pulse repetition frequency being set at 1 kHz. The data acquisition rate is constrained, not by the laser but rather by the piezoelectric driver of the interferometer’s scanning translation stage and, specifically, the time necessary for the driver to settle reliably at a new position. Because the sampling frequency is 300 THz, the Nyquist frequency is 150 THz which provides more than sufficient bandwidth to observe the atomic difference frequencies of interest here. Extensive tests confirm that, given the interferometer spatial resolution and setting reproducibility, the experimental resolution in the Fourier (spectral) domain is <0.05 cm^{-1} and the dynamic range was measured to be >50 dB. By averaging the peak positions measured from 21 scans, the 7_s1/2–5d_{5/2} difference frequency was determined to be 18.22(5) THz which corresponds to 607.93(7) cm^{-1}. It must be emphasized that the precision of this measurement (which improves slightly on the value reported by Sansonetti [37]; 607.93 ± 0.02 cm^{-1}) is possible only because of the stability of the interferometer and the exceptional coherence time associated with 7_s1/2–5d_{5/2} quantum beating which is found to be >1 ns. Throughout the experiments, the pump and probe pulse energies were fixed at discrete values in the 150–350 μJ range and the phase history of both pulses was monitored continuously by frequency-resolved optical gating (FROG). No significant changes in the cross-sections and transverse intensity distributions of the pump and probe pulse laser beams were observed as the pulse energy was varied. For the maximum single-pulse energy available with the present Ti:sapphire laser system, the peak intensity at the optical cell is estimated to be ~1.9 GW cm^{-2} for 200 fs pulses. Most of the experiments to be described here were conducted with single pulse energies of 300 μJ.

3.2. Temporal and Fourier domain data of quantum beating at ∼2.1, 10.7, and 18.2 THz

Typically, scans of the dependence of the relative signal wave (420 nm) intensity on ∆τ were recorded over the 0–670 ps interval, and representative data are shown in figures 1(b) and (c) for the 4.5 < ∆τ < 6.5 ps and 598.8–600.2 ps intervals, respectively. The results presented in both panels are representative of those acquired over the course of >50, 17 h scans. Several frequency components are evident in the oscillographic signal of figure 1(b), the most prominent of which has a period of ~475 fs (corresponding to ~2.1 THz) and is clearly modulating the amplitude of the PFWM signal. The higher frequency structure in panel (b) has a periodicity of ~55 fs which is associated with the 7_s1/2–5d_{5/2} energy difference. Ramsey fringes for this quantum oscillator have been found to persist for >1 ns which corresponds to more than 1.8 × 10^4 oscillation periods, thereby enabling the precise measurement of the 7_s1/2–5d_{5/2} energy difference (607.93(7) cm^{-1}). An example of the data routinely recorded beyond ∆τ = 500 ps is presented in figure 1(c). Note that although 2.1 THz oscillations dominate the waveform early (figure 1(b)), this frequency component of the (5d_{5/2}–5p_{3/2})–(5p_{3/2}–5s_{1/2}) coupled quantum oscillator dephases quickly and is no longer detectable for ∆τ ~ 600 ps. In fact, as will be evident in figure 3, the 2.1 THz frequency component has vanished by ∆τ = 100 ps. The Fourier (spectral) domain representation of temporal data such as that of figure 1 is given in figure 3(a) which identifies the observed quantum beating frequencies. The most prominent of these are the (7_s1/2–5d_{5/2}), (8_s1/2–6d_{5/2}), and (5d_{5/2}–5p_{3/2})–(5p_{3/2}–5s_{1/2}) difference frequencies at 18.2, 10.7, and 2.1 THz, respectively. As mentioned previously, the latter is unique in that one state (5p_{3/2}) is common to two interacting oscillators. Both the second and third harmonics of 18.2 THz are also observed, and coupling between the 7_s1/2–5d_{5/2} and 8_s1/2–6d_{5/2} coherences is responsible for the presence of several frequency components described by (7_s1/2–5d_{5/2}) ± nβ, where n is an integer and β = (7_s1/2–5d_{5/2})–(8_s1/2–6d_{5/2}) = 250.2 ± 0.1 cm^{-1} (7.5 THz). The locations of 4 such overtones are indicated in figure 3(a).

The temporal history of the Fourier amplitude for several frequency components of figure 3(a) have been calculated with the short-time Fourier transform (STFT) by adopting a Hamming or rectangular window having a temporal width of ∆τ = 3.9 ps which fixes the spectral resolution at 0.265 THz. Results are represented in panel (b) of figure 3 for the Rb 7_s1/2–5d_{5/2}, (5d_{5/2}–5p_{3/2})–(5p_{3/2}–7_s1/2), and 8_s1/2–6d_{5/2} difference frequencies when the number density is maintained at 9.2 × 10^{14} cm^{-3}. For this value of [Rb], the 7_s1/2–5d_{5/2} quantum beating amplitude peaks at ∆τ = 80–90 ps and a revival (not shown) occurs between ∆τ = ~400 and 600 ps. Undulations superimposed onto this curve are real and are the result of interference between the (7_s1/2–5d_{5/2}) and (7_s1/2–5d_{3/2}) coherences, thus yielding oscillations at the 5d_{5/2}–5d_{3/2} fine structure-splitting frequency of 2.96 cm^{-1}. In the [Rb] interval of (2.6–5.0) × 10^{14} cm^{-3}, 7_s1/2–5d_{5/2} quantum beating continues to be strong at the ∆τ range limit of the interferometer (1100 ps). Although the appearance of 8_s1/2–6d_{5/2} quantum beating was unexpected, >50 time-delay scans yield results essentially identical to the lower of the two black curves of figure 3(b). To illustrate the reproducibility of the data, the red trace of figure 3(b) was recorded for [Rb] = 2.6 × 10^{14} cm^{-3}, or a factor of ~3.5 lower than that for the black profile. The
to be largely due to the mismatch of coupled oscillator is estimated to be 30 ps. Both are presumed rapidly and the upper limit on the coherence time for this shows the $10^{14}$ cm of reproducibility. 

most of these data, [Rb] was fixed at 9.2 THz, 2.1 THz, and 10.7 THz, respectively. For black traces represent different [Rb] values (2.6 $\times$ 10$^{14}$ cm$^{-3}$, respectively), indicating that the 5d3/2–5p3/2 coherence drives quantum beating at 18.2 THz. We, therefore, also observe three coupled-atomic coherence ((7s1/2–5d3/2): [5d5/2–5p3/2]: [5p3/2–5s1/2]) in which the interaction between the 18.2 and 2.1 THz oscillators is now mediated by the 5d3/2 state.

3.3. Generalized Fano lineshapes for the (5d5/2–5p3/2)–(5p3/2–5s1/2) oscillator

Since the dynamic range of experimental spectra (such as that of figure 3(a)) is >50 dB, spectral features lying near the noise floor and not detectable previously are now accessible. As one example, consider the spectral region lying near the base of the 18.2 THz resonance shown as that of figure 3(a)) is $\sim$0.5–55 THz resonance shown in the left-hand portion of figure 4. Note the symmetry of the spectrum with respect to the 7s1/2–5d5/2 frequency, a characteristic of most of the observed spectra which we attribute to amplitude modulation of the 18.2 THz peak by other quantum beating frequencies. Several of the weaker features in panel (a) may be associated with a shallow electronic excited state of the Rb dimer. These, as well as the strong pairs of lines displaced by $\sim$1.0 and $\sim$2.4 cm$^{-1}$ from the atomic resonance, will be discussed in detail elsewhere but, for present purposes, it must be emphasized that the quantum beating spectral features observed throughout the $\sim$0.5–55 THz spectral region in figure 4 are consistently reproducible, and virtually all are spectrally-narrow. The sharp spectral features of the left portion of figure 4 contrast with the broad Fano lineshapes observed for the (5d5/2–5p3/2)–(5p3/2–5s1/2) resonance, one of which is shown by the right-hand side of figure 4.

Recorded over the $\sim$1.8–2.4 THz interval for [Rb] = 1.0 $\times$ 10$^{14}$ cm$^{-3}$ ($T = 423$ K), this Fano profile exhibits
a spectral breadth (FWHM) of 5 cm\(^{-1}\) (150 GHz), or \(\sim 2\) orders of magnitude larger than that associated with the \(7\)\(s_{1/2}\)–\(5\)\(d_{5/2}\) resonance. Two weak interference windows superimposed onto the lineshapes are also observed at \(\sim 69\) and \(\sim 70.4\) cm\(^{-1}\), the latter of which lies \(\sim 0.1\) cm\(^{-1}\) above the \(5\)\(d_{5/2}\)–\(5\)\(p_{1/2}\)–\(5\)\(s_{1/2}\) resonance difference frequency. Both windows intensify at larger values of Rb number densities. Spectra similar to figure 4 (right) have been acquired over a range in [Rb], and examples of those obtained at \(T = 423, 433,\) and \(453\) K (corresponding to [Rb] = (1.0, 1.6, and 4.0) \(\times\) \(10^{14}\) cm\(^{-3}\)) are overlaid in panel (a) of figure 5. For the sake of clarity, each is displayed separately in panels (b)–(d) of figure 5 and the mean Rb–Rb interatomic separation \(\langle R \rangle\), calculated from the nearest-neighbor distributions associated with each vapor cell temperature [38, 39], is indicated. Superimposed onto the experimental data of figures 5(b)–(d) are generalized Fano lineshapes (shown in red), calculated on the basis of the wave operator theory of Durand et al [31] and fitted to experimental spectra. Specifically, equation (29) of [31] describes the interference of two resonances and their interaction with a single continuum, and allows each of the observed spectra of figure 5 to be described by a single, modified Fano expression. For the \(T = 423\) and \(433\) K profiles, for example, the resonance–resonance and resonance–continuum decay (damping) frequencies to be: \((\gamma_1, \gamma_2) = (7, 7, 3.5, 3.5)\) and \((0.4, 8)\) cm\(^{-1}\), respectively. Values of \(\langle R \rangle\) were determined from the nearest-neighbor distributions [38, 39] corresponding to each Rb number density, and the laser pump and probe pulse energies were fixed at 300 \(\mu\)J throughout.

It is evident from figure 5 that the Fano profiles describe well the observed spectral lineshapes but do not capture the interference structure apparent on either side of the resonance at 70.3 cm\(^{-1}\) in figure 5(d), however, requires \(\gamma_1\) to fall to 0.4 cm\(^{-1}\) but \(\gamma_2\) increases to 8 cm\(^{-1}\). The Fano phase \(\delta\) is also extracted from the fitting of equation (29) of [31] to experiment. For the lower [Rb] values of figures 5(b) and (c), \(\delta\) is found to be 3.1 radians for both spectra whereas the corresponding value for figure 5(d) is 2.7 radians. The fittings of equation (29) of [31] to the experimental spectra in figure 5, and their associated constants, are not unique but are representative of the parameter sets that best describe the data.

Figure 4. Comparison of representative Fourier spectra associated with the \(7\)\(s_{1/2}\)–\(5\)\(d_{5/2}\) and \((5\)\(d_{5/2}\)–\(5\)\(p_{1/2}\))–\(5\)\(s_{1/2}\)) difference frequencies: (left) magnified view of the spectrum observed in the 603.5–612.5 cm\(^{-1}\) (\(\sim\) 18.10–18.35 THz) region, at the base of the \(7\)\(s_{1/2}\)–\(5\)\(d_{5/2}\) resonance; (right) Fano lineshape recorded between 45 and 95 cm\(^{-1}\) (\(\sim\) 1.4–2.8 THz) and associated with the \((5\)\(d_{5/2}\)–\(5\)\(p_{1/2}\))–\(5\)\(s_{1/2}\)) coupled oscillator. The spectra at left and right were acquired for [Rb] maintained at 1.6 and 95 cm\(^{-1}\) \((T = 433\) K) and \(1.0 \times 10^{14}\) cm\(^{-3}\) \((T = 423\) K), respectively.

Figure 5. Comparison of experimental and calculated Fano spectral lineshapes: (a) series of three spectral (Fourier domain) profiles, recorded between 62 and 78 cm\(^{-1}\), for vapor cell temperatures of \(T = 423, 433,\) and \(453\) K which correspond to [Rb] = (1.0, 1.6, and 4.0) \(\times\) \(10^{14}\) cm\(^{-3}\), respectively; (b)–(d) comparison of generalized Fano profiles (red curves, based on equation (29) of Durand et al [31]) with experimental data for each of the spectra of (a). Simulations show the resonance–resonance and resonance–continuum decay (damping) frequencies to be: \((\gamma_1, \gamma_2) = (7, 7, 3.5, 3.5)\) and \((0.4, 8)\) cm\(^{-1}\), respectively. Values of \(\langle R \rangle\) were determined from the nearest-neighbor distributions [38, 39] corresponding to each Rb number density, and the laser pump and probe pulse energies were fixed at 300 \(\mu\)J throughout.
collisions in the thermalized vapor. Consequently, the evolution of the Fano lineshape displayed in figure 5 cannot be attributed to collisions.

3.4. Transition of the Fano phase and spectral profile near an EP

The spectra of figure 5 reveal a dramatic and rapid transformation in the Fano profile over a narrow range in $\langle R \rangle$ (factor of 2–3 interval in $[Rb]$). This lineshape conversion process results (with increasing $[Rb]$) in a single, narrow bandwidth of 2–3 interval in $[Rb]$. This lineshape conversion process and extracted from generalized Fano profile fittings to the data. For the Rb–Ar experiments, $[Ar]$ was constant at 3.2×10$^{14}$ cm$^{-3}$, whereas the Rb–Cs spectra acquired at the same temperature (443 K) cannot be attributed to collisions. Clearly, neither Ar nor Cs perturbs the Fano spectrum of the (5$d_{3/2}$–5$p_{3/2}$)–(5$p_{3/2}$–5$s_{1/2}$) coupled oscillator to the degree observed for a Rb atom-only background, but the high-frequency portion of the relative that to figure 6(a). Simulations of the lineshapes of the Rb, Rb–Ar, and Rb–Cs experimental profiles with the modified Fano lineshape expressions of reference [31] yield the red curves of figures 6(a) and (b). For the $T = 443$ K spectrum of figure 6(a), the damping frequencies were determined to be $(\gamma_1, \gamma_2) = (0.2, 5.5)$. In contrast, the Rb–Ar and Rb–Cs experimental spectra show broadening of the interference window while sharpening the 70.3 cm$^{-1}$ peak. Before leaving the data of figures 6(a) and (b), it should be emphasized that the Rb/Cs and Rb/Ar spectra of panel (b) differ significantly from each other and the Rb/Rb spectrum of 6(a). Although both show a suppressed response in the 70–74 cm$^{-1}$ interval (relative to figures 6(a), 5(b) and (c), and 4), the effect is most pronounced for Cs and we presume that spectra similar to those of figure 6(b) offer a new diagnostic of the long-range interactions between excited Rb atoms and Cs, Ar, or other perturber species.

From simulations of spectra represented by figures 5 and 6, the Fano phase $\delta$ was extracted, and the results are given by the open circles of figure 6(c) for Rb vapor when $\langle R \rangle$ is between 55 and 120 nm. For the Rb–Ar and Rb–Cs data of figure 6(b), $\delta$ was found to be almost identical for both perturbers and is represented by a single open triangle (Δ). One concludes from figures 6(a) and (c) that both the Fano spectral profile and the phase of the composite (5$d_{3/2}$–5$p_{3/2}$)–(5$p_{3/2}$–5$s_{1/2}$) oscillator undergo a sharp transition in the range of $\langle R \rangle = 80–90$ nm. Not only does the Fano phase change discontinuously near $\langle R \rangle = 87$ nm, but the magnitude of the phase shift is $(0.8 ± 0.2) \sim \pi/4$ radians.
Results similar to those of figures 5 and 6 are obtained if the laser intensity is varied. The left-hand portion of figure 7 shows Fourier spectra recorded for the same Rb number density and, hence, $\langle R \rangle$ value of figure 6 but the laser pulse energy ($E_p$) is now increased incrementally from 100 $\mu$J to 300 $\mu$J. One notices immediately that the spectrum is a continuum at the lowest optical field intensity (violet trace, figure 7), and the resonant peak at $\sim$70.3 cm$^{-1}$ is first noticeable when $E_p$ reaches 150 $\mu$J (blue). The latter spectral profile is a Fano lineshape exhibiting a null between $\sim$67 and 68.8 cm$^{-1}$. As $E_p$ is increased further, the interference structure discussed earlier strengthens, particularly on the high frequency side of the resonance. Above $E_p \sim$ 200 $\mu$J, however, the undulation-modulated continuum quickly diminishes in intensity as energy is transferred into the narrow feature at 2.11 THz. Indeed, the right-half of figure 7 illustrates the initial nonlinear growth of the 70.3 cm$^{-1}$ peak amplitude when $E_p$ is increased above 200 $\mu$J. The ordinate at right represents the fraction of the Fourier power spectrum $\langle |F(\omega)|^2 \rangle$, where $F(\omega)$ is the Fourier transform of the $f(\Delta t)$ experimental waveform attributable to the 2.1 THz resonance line (denoted ‘RES’ in the integral of figure 7 and having a spectral width of $\sim$0.4 cm$^{-1}$). When normalized to the power spectrum integrated over the $\sim$65–88 cm$^{-1}$ interval (denoted ‘TOT’), the value of the integral rises rapidly between $E_p = 200 \mu$J and $\sim$250 $\mu$J but saturates thereafter. We attribute this behavior to the onset of strong coupling of the composite ($5d_{5/2}$–$5p_{3/2}$) coupled oscillator in the vicinity of the EP. At still higher pulse energies, the background undulations and continuum strength remain unchanged.

5. Conclusions

In summary, an EP (singularity) associated with the response of a coupled pair of atomic coherences has been observed in thermalized Rb vapor. Quantum beating in Rb in the vicinity of the ($5d_{5/2}$–$5p_{3/2}$)–($5p_{3/2}$–$5s_{1/2}$) difference frequency resonance at $\sim$2.1 THz exhibits a generalized Fano profile that is transformed by independently varying two experimental parameters, $\langle R \rangle$ of the background vapor or the laser intensity. In the $\langle R \rangle$ interval of $\sim$80–90 nm, the measured phase of the coupled ($5d_{5/2}$–$5p_{3/2}$): ($5p_{3/2}$–$5s_{1/2}$) quantum system abruptly shifts by $\sim$0.8 radians. The
discontinuous change in the phase and the experimentally-observed Fourier domain spectra confirm the theoretical formalisms of references [1–3, 6, 31]. Altering the degree of coupling between the two constituent oscillators \([5(3\ell_2 - 5p\ell_2)\) and \((5p\ell_2 - 5s\ell_2)\)] through long-range \((\sim 55–120 \text{ nm})\) interactions with background atoms is enabled by the induced-dipole term contributing to the interatomic potential, \(V(R)\). The impact of the phase shift is profound, transforming a broadband, dissipative oscillator into a strongly-coupled system locked to the \(\Delta R\) frequency difference of \(70.3 \text{ cm}^{-1}\) (2.1 THz). Also, the undulatory \((\text{interference})\) structure superimposed onto the Fano line-shapes collapses as the phase transition is traversed. Similar results were observed when varying the laser pump/probe pulse energy, and a trio coupled-atomic oscillator system, \((\text{5}^{\text{ss}}\text{s–5}^{\text{ss}}\text{p})\), is also reported. These data suggest that specifically-selected, coupled quantum coherences can now be examined in a variety of atoms and molecules, and a new approach to quantifying atom–atom or atom–optical field interactions is available by observing the phase of an atomic oscillator as the relative strength of \(\Delta R\) and \((\text{5}^{\text{ss}}\text{s–5}^{\text{ss}}\text{p})\) is changed.

**Author contributions**

WG, RS, and JGE conceived the experiments, and WG, RS, and TR acquired the data. SP, WG, TR, and JGE analyzed the results, and SP and JGE wrote the manuscript.

**Competing interests**

The authors declare that they have no competing interests.

**Acknowledgments**

The support of this work by the US Air Force Office of Scientific Research under Grant Nos. FA9550-1-14-0002 and FA9550-18-1-0380 (H Schlossberg, W Roach, G Pomrenke) is gratefully acknowledged.

**Data availability statement**

The data that support the findings of this study are available upon reasonable request from the authors.

**ORCID iDs**

S Park [https://orcid.org/0000-0003-2734-8034](https://orcid.org/0000-0003-2734-8034)

**References**

[1] Heiss W D 2012 *J. Phys. A: Math. Theor.* **45** 444016

[2] Günther U, Rotter I and Samsonov B F 2007 *J. Phys. A: Math. Theor.* **40** 8815

[3] Heiss W D and Wunner G 2014 *Eur. Phys. J. D* **68** 284

[4] Peng B, Özdemir Ş K, Chen W, Nori F and Yang L 2014 *Nat. Commun.* **5** 5082

[5] Zhen B, Hsu C W, Igarashi Y, Lu L, Kaminer I, Pick A, Chua S-L, Joannopoulos J D and Soljačić M 2015 *Nature* **525** 354

[6] Casucci N et al 2018 *Nat. Commun.* **9** 396

[7] Atabek O, Lefebvre R, Lepers M, Jaouadi A, Dulieu O and Kokouline V 2011 *Phys. Rev. Lett.* **106** 173002

[8] Papadopoulos I, Wagner P, Wunner G and Main J 2007 *Phys. Rev. A* **76** 053604

[9] Longhi S 2010 *Phys. Rev. Lett.* **105** 013903

[10] Lee S-B, Yang J, Moon S, Lee S-Y, Shim J-B, Kim S W, Lee J-H and An K 2009 *Phys. Rev. Lett.* **103** 134101

[11] Kim K-H, Hwang M-S, Kim H-R, Choi J-H, No Y-S and Park H-G 2016 *Nat. Commun.* **7** 13893

[12] Brandstetter M, Liertzer M, Deutsch C, Klang P, Schöberl J, Türeci H E, Strasser G, Unterrainer K and Rotter S 2014 *Nat. Commun.* **5** 4034

[13] Rütter C E, Makris K G, El-Ganainy R, Christodoulides D N, Segev M and Kip D 2010 *Nat. Phys.* **6** 192

[14] Joe Y S, Satanin A M and Kim C S 2006 *Phys. Sez.* **74** 259

[15] Assawaworrarit S, Yu X and Fan S 2017 *Nature* **546** 387

[16] Fano U 1961 *Phys. Rev.* **124** 1866

[17] Connerade J-P and Lane A M 1988 *Rep. Prog. Phys.* **51** 1439

[18] Kim S B, Kane D J and Eden J G 1992 *Phys. Rev. Lett.* **68** 1311

[19] Fan P, Yu Z, Fan S and Brongersma M L 2014 *Nat. Mater.* **13** 471

[20] Luk’yanchuk B, Zholudev N I, Maier S A, Halas N J, Nordlander P, Giessen H and Chong C T 2010 *Nat. Mater.* **9** 707

[21] Galli M, Portalupi S L, Belotti M, Andreani L C, O’Faolain L and Krauss T F 2009 *Appl. Phys. Lett.* **94** 071101

[22] Ott C et al 2014 *Nature* **516** 374

[23] Zhang S, Genov D A, Wang Y, Liu M and Zhang X 2008 *Phys. Rev. Lett.* **101** 047401

[24] Miroshnichenko A E, Flach S and Kivshar Y S 2010 *Rev. Mod. Phys.* **82** 2257

[25] Verslegers L, Yu Z, Ruan Z, Catryssse P B and Fan S 2012 *Phys. Rev. Lett.* **108** 083902

[26] Khanikaev A B, Wu C and Shvets G 2013 *Nanophotonics* **2** 247

[27] Stern L, Grajower M and Levy U 2014 *Nat. Commun.* **5** 4865

[28] Yu Y, Xue W, Semenova E, Yvind K and Mork J 2017 *Nat. Photon.* **11** 81

[29] Limonov M F, Rybin M V, Poddubny A N and Kivshar Y S 2006 *Phys. Rev. Lett.* **100** 083902

[30] Sun P, Zhang R, Chen W, Braun P V and Eden J G 2019 *Appl. Phys. Rev.* **6** 041406

[31] Durand P, Pálová I and Gadéa F X 2001 *J. Phys. B: At. Mol. Opt. Phys.* **34** 1953

[32] Harris S E, Field J E and Imamoglu A 1990 *Phys. Rev. Lett.* **64** 1107

[33] Aspuru-Guzik A, Head-Gordon M and Schatz G C 2005 *Phys. Rev. Lett.* **95** 206401

[34] Tran H C, John P C, Gao J and Eden J G 1998 *Opt. Lett.* **23** 70

[35] Shen F, Gao J, Senin A A, Zhu C J, Allen J R, Lu Z H, Xiao Y and Eden J G 2007 *Phys. Rev. Lett.* **99** 143201

[36] Zhu C-J, Xiao Y, Senin A A, Gao J, Eden J G, Varzhapsetyan T S and Sarkisyan D H 2007 *Phys. Rev. A* **75** 053405

[37] Sanz and M and Dalgarno A 1995 *Phys. Rev. A* **52** 311

[38] Torquato S, Lu B and Rubinstein J 1990 *Phys. Rev. A* **41** 2059

[39] Marinescu M and Dalgarno A 1995 *Phys. Rev. A* **52** 311