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CW Cavity Ring Down Spectroscopy

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Until now, applications of Cavity Ring Down Spectroscopy (CRDS) employed pulsed laser sources. Here, we demonstrate that a commercial single-frequency CW laser can also be conveniently employed, allowing to gain in spectral resolution, signal intensity and data acquisition rate. As a demonstration, we measured a section of the weak HCCH overtone transition near 570 nm and compare to existing photoacoustic data. Our high quality and reproducible Doppler-limited spectra display a (rms) noise-equivalent absorption of 10^{-9} /cm, or 5×10^{-8} per pass through the sample. Most interesting applications of CW-CRDS include high resolution spectroscopy at low pressure, sub-Doppler absorption spectroscopy in a supersonic jet, and trace-gas detection using compact diode laser sources.

1. Introduction

Cavity Ring Down Spectroscopy (CRDS) is a versatile high sensitivity absorption technique, exploited in an increasing number of investigations these last few years. Some of the published CRDS spectra correspond to absorption as small as 10^{-7} for one pass through the sample (a gas or a supersonic jet). With respect to the sensitivity, CRDS has a performance comparable to Photo-Acoustic Spectroscopy (PAS) [1] and Intra Cavity Laser Absorption Spectroscopy (ICLAS) [2]. One of the attractive features of CRDS is its conceptual and practical simplicity. This may well be considered as a multipass technique affording 10 to 1000 times longer pathlengths than traditional multiple reflection cells, therefore reliable absorption coefficients can be obtained directly [3–5]. In addition, even if the standard CRDS setup uses a pulsed laser source, the signal is not affected by the pulse to pulse intensity fluctuations, which are a serious source of noise when using a normal multipass cell. This allows CRDS to access the broad tunable range of pulsed lasers, which easily extends into the UV by the use of doubling crystals. Thus it is clear that this is a powerful and very versatile method. In fact, since the demonstrative detection of forbidden transitions of O_2 in the visible [6], CRDS was used to study metal clusters produced by laser ablation in supersonic jets [7], for the spectroscopy of weak transitions in a static gas cell [3, 8–10], to detect radical species (in the UV) in flames [11] or diamond deposition reactors [12], ions in a discharge [13], in association with Fourier transform spectral multiplexing [14] and, by exploiting its millisecond time resolution, to investigate reaction kinetics [15].

A detailed technical description of this method was given by Romanini and Lehmann [3], who discussed also the problem of the fundamental shot noise limitations of the sensitivity. Here, it is sufficient to outline what we will refer to as ‘standard CRDS’, where a pulsed laser is used to inject monochromatic radiation into a stable

optical resonator of length L (see e.g. Yariv [16] or Siegman [17] for the theory of optical cavities and their mode structure). This “ringdown cavity” may be vacuum tight to confine the sample of interest (a gas, a supersonic jet, etc). Injection and excitation of the ringdown cavity is effected by direct transmission of the laser pulse through one of the mirrors, along the cavity axis. Transmittivity of good dielectric mirrors [18] in the visible may be as small as $\mathcal{T} = 10^{-5}$ or even 10^{-6} . However, the small injected fraction of the laser pulse still may consist of more than 10^{10} photons, which remain trapped as the pulse propagates back and forth between the mirrors. At each pass, part of the trapped pulse is retransmitted outside the cavity and produces an easily detectable signal. The relevant feature of this ringdown signal is its first order decay envelope $\exp(-t/\tau)$, whose time constant τ may be as large as 100 μ s. Considering the surface and diffraction losses \mathcal{L} of the mirrors (usually, $\mathcal{L} < \mathcal{T}$) and the frequency dependent absorption coefficient of the sample α_s , the decay rate of the trapped photons is $1/\tau = (\mathcal{T} + \mathcal{L} + \alpha_s L)/(L/c)$, where c is the speed of light. This decay rate is the sum of the slowly frequency dependent term $c\alpha_c = c(\mathcal{T} + \mathcal{L})/L$ (in the visible, mirrors have more than 50 nm working range), plus the sample absorption spectrum $c\alpha_s(\nu)$. The first term may be considered as a broad-band smooth baseline which can be easily distinguished from the rapidly varying spectral features of the sample. If needed (e.g. in case of relatively broad-band spectral features), the baseline may also be independently measured after removing the sample. Finally, the sample absorption α_s (over the baseline α_c) is obtained directly in units of 1/cm simply from $\alpha_{\text{tot}} = \alpha_c + \alpha_s = 1/c\tau$, where $L_{\text{eq}} = c\tau$ is an equivalent absorption pathlength. For small signals, we can write $\delta\tau/\tau = \delta\alpha_{\text{tot}}/\alpha_{\text{tot}}$, so the noise equivalent absorption (detection limit) is immediately deduced from the noise on the measurement of τ , as $\delta\alpha_{\text{noise}} = \delta\tau_{\text{noise}}/\tau L_{\text{eq}}$.

2. Spectral resolution in CRDS

For the following, it is important to notice that impulsive ringdown cavity excitation is efficient only if the laser pulse spectrum overlaps one or more of the transmission modes of the cavity [19,20]. This is usually granted if one uses a commercial pulsed dye laser having a linewidth of about $0.1\text{ cm}^{-1}=3\text{ GHz}$, which is larger than the free spectral range (FSR) of a ringdown cavity few tens centimeters long. Achieving spectral resolution higher than the cavity FSR would open for CRDS new applications such as sub-Doppler supersonic slit-jet absorption spectroscopy. As it has been discussed [11], one way to increase the resolution could be to exploit the high spectral density of high order transverse modes of the cavity. However, it has been pointed out that side effects such as transverse mode beating, multiexponentiality of the ringdown or fluctuations of the decay rate would pose substantial experimental complications [5, 19, 20]. In order to avoid such problems, a Fourier-transform limited pulsed laser could be used to selectively excite one cavity mode [19], after accurate mode matching and frequency locking. The laser should be narrower than the cavity FSR, and its frequency should be tracked as the cavity length is tuned to maintain efficient single-mode cavity excitation. This would allow recording spectra with resolution limited by the mode width [20], which is as narrow as 60 kHz for a ringdown having $\tau = 10\mu\text{s}$.

Another possibility for very high resolution CRDS is to use a single frequency CW laser tuned to one of the cavity resonances. In this case a substantial buildup of the intracavity field occurs [21], and a strong ringdown signal may then be observed after the laser beam is quickly interrupted (faster than τ). In fact, the intensity of the beam transmitted through the cavity after buildup may be close to the incoming intensity, which permits to detect the ringdown signal with high signal to noise ratio. With respect to the resolution, this is in principle not determined by the laser linewidth but by the width of the excited cavity resonance, as in the single-mode pulsed case mentioned above. Increased performance should also be granted by the high repetition rate of the ringdown events one could generate, since this is limited by the ringdown time itself, and not by the repetition rate of a pulsed laser. This type of CRDS (CW-CRDS) looks especially attractive if tunable diode lasers are considered as a source, in particular external-cavity single-frequency models which are continuously tunable over more than 50 nm in the near IR and 10 nm in the visible. Compact and relatively inexpensive trace gas detector systems could be built based on diode-laser CW-CRDS [22].

Preliminary results, where ringdown signals were obtained using optical feedback locking of a diode laser to a ringdown cavity, where reported [23]. However, we are not aware of anybody being able to obtain an absorption spectrum by CW-CRDS. There are several reasons for that. The sub-MHz short term frequency jit-

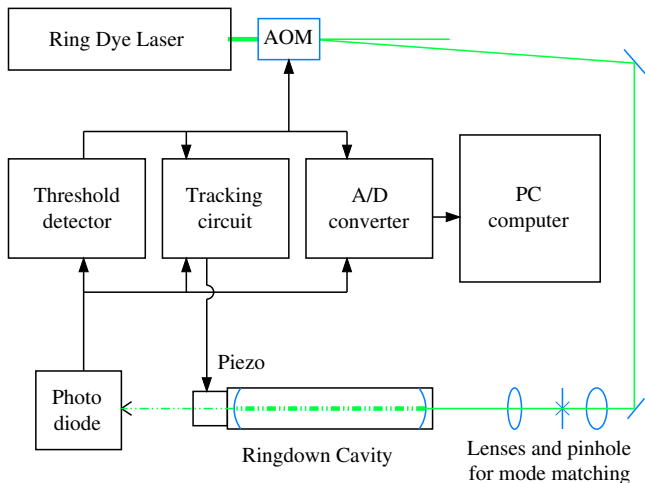
ter of most single frequency lasers is much larger than the linewidth of the cavity formed by two supermirrors. This makes frequency locking together the laser and the cavity extremely difficult. Even after succeeding in this task, for example by the especially performant technique of Drever *et al.* [24], one needs to switch off the laser beam each time the ringdown signal should be sampled. This would strongly disturb the locking electronics, introducing possibly long recovery times and other problems. Other methods of sensitive absorption detection by CW locking to optical cavities have been shown to work, which altogether avoid the problem of interrupting the laser beam and the locking loop [25, 26]. However, we devised a simpler approach, where locking is replaced by a less demanding and robust tracking scheme, and one can use directly the ringdown signal to measure the intracavity sample absorption.

It is curious to note that back in 1984, one of the first ringdown applications was based on a CW source, without locking. Anderson, Frisch and Masser [27] exploited the occasional coincidences of a He-Ne laser with the cavity modes. When these coincidences would produce by buildup sufficient intensity transmitted by the cavity, a threshold circuit was interrupting the laser and allowing to observe the free ringdown signal. These authors could not tune their laser to obtain absorption spectra, nor they tried to control the occurrence of buildup-ringdown events. In fact, this pioneering work is usually not considered as a first demonstration of CRDS since the authors neither mentioned the potential of the technique for spectroscopy nor were interested in other than characterizing the reflectivity of mirror coatings. In contrast to that, our intent was to use CW-CRDS for spectroscopic applications. Therefore, in order to obtain a high repetition rate in sampling the ringdown signals while tuning the laser we had to introduce a regularity into the laser to cavity mode coincidences.

3. Experimental

Our experimental setup (figure 1) was very similar to that of Anderson *et al.* [27], except that we used an acousto-optic modulator (AOM) to interrupt the laser beam and our cavity was linear with two supermirrors of reflectivity 99.990% around 570 nm, where we tested the technique using weak acetylene overtone transitions. The mirrors had a radius of curvature of 100 cm, were placed at a distance of 50 cm, and one of them was mounted on a piezoelectric transducer. We used a Coherent CR-699 single frequency ring dye laser, stabilized within about 1 MHz to an external etalon and capable of continuous 30 GHz scans. We also used a threshold circuit to interrupt the beam when sufficient buildup would be detected. Interruption of the laser beam is essential to allow observing a clean ringdown decay [28].

The ringdown signals were registered by a silicon photodiode and digitized by a 1 MHz 12 bits PC interface card (model CIO-DAS 16/M1 by ComputerBoards Inc.).

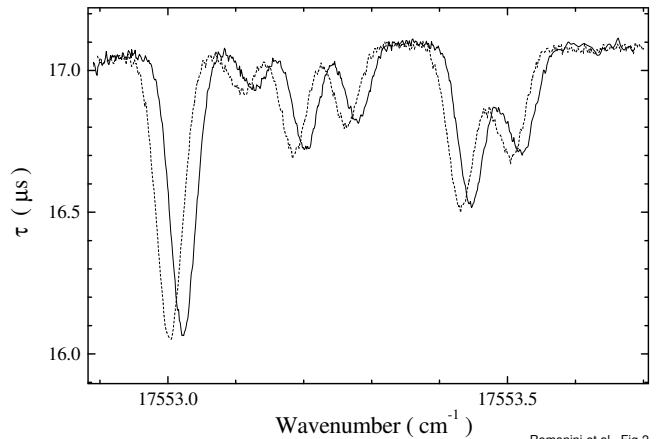


Romanini et al., fig. 1

Fig. 1. Simplified scheme of the CW-CRDS setup.

The AOM was used in deflection to have maximum attenuation contrast, and served also the purpose of isolating the laser from the strong optical feedback coming from the input mirror of the cavity. The beam reflected from this mirror back to the laser is shifted in frequency by twice the acoustic modulation of the AOM (80 MHz), therefore falling in between two of the laser cavity modes. Such an isolation was sufficient, as we did not observe any perturbation of the dye laser by the optical feedback.

We mode matched the laser to the TEM_{00} modes of the cavity, and piezoelectrically modulated the cavity length to make the frequency of one of these modes to oscillate around the laser line. The modulation amplitude was chosen to be less than the mode spacing, so that ringdown events could be observed twice per modulation period, each time the mode passed through the laser line. This provided a good and steady repetition rate for signal averaging, as long as the ringdown events were centered with respect to the modulation range. When the laser frequency was fixed, these regular ringdowns could be observed during the time of a few seconds, until the thermally unstabilized cavity would drift out of the laser frequency. We could compensate this drift by a feedback circuit, which measures the distance of the ringdown events from the center of the modulation range and by integration provides a correction bias voltage to the piezoelectric transducer. This circuit makes the cavity to follow reliably the laser for scanning speeds up to 10 GHz per minute. Given the limited piezoelectric excursion, the circuit also resets the bias voltage by applying a jump which corresponds to about one full cavity FSR, so that the tracking continues without appreciable interruptions. This system allowed to observe stable and regular ringdown signals during the whole 30 GHz excursion of one laser scan, but it could work for arbitrar-



Romanini et al., Fig 2

Fig. 2. Ringdown time as a function of laser frequency. The absorption lines belong to a weak overtone transition of HCCH [1], which was placed in the ringdown cavity at the pressure of 5 Torr. Each 30 GHz scan took 6 minutes. Two successive scans of the same spectral region show the good reproducibility. Their relative shift is for easier visual comparison.

ily large scans. We stress the fact that these ringdown decays are due exclusively to excitation of the TEM_{00} longitudinal modes.

With the data acquisition card installed into a 486DX33 PC computer, we could record and analyze separately each of the ringdown signals, at a rate of about 200 Hz. After baseline subtraction and conversion to logarithm, a linear fit would give τ for each decay. Shot to shot reproducibility of τ was about 0.5%, but some events showed larger deviations. It turned out that events having larger deviations had also a larger standard deviation of the fit. In this cases it was possible to observe some oscillations in the decay, which are likely produced by high order transverse modes lying within a laser bandwidth from the excited TEM_{00} mode [19]. Even if mode matching was effective in reducing the excitation of higher order modes to 1% of the fundamental mode, this residual excitation is sufficient to explain the observed ringdown profile deviations from exponentiality. Fast fluctuations in the laser frequency may be responsible for variations in the degree of simultaneous excitation of these modes from event to event. To significantly reduce this effect, we used the standard deviation of the fit to discriminate against the bad events during the data acquisition.

Another (occasional) problem we should mention, was the presence of baseline oscillations, having period close to the FSR of the cavity. At this time, we are not sure of the origin of these oscillations. However, they seem to depend on the alignment of laser beam with respect to the center of the cavity mirrors. By aligning the beam so that it would pass closer to the edge of the mirror

coatings, we observed that this effect disappeared.

To demonstrate the performance of this CW-CRDS system, we recorded part of the overtone transition of HCCH near 570 nm, which was previously studied by PAS [1]. The pressure in our sample was 5 Torr, for which the collisional broadening of acetylene is negligible compared to the Doppler width. In figure 2 we show the ringdown time recorded as described above for 30 GHz scans. Each data point was averaged over 50 ringdown events, after elimination of bad events, and the tuning rate was 20 GHz/minute. The excellent repeatability of CRDS measurements is demonstrated by superposing two scans taken in the same conditions (shifted in frequency for clarity). In order to record a larger portion of the HCCH spectrum we took five consecutive (partially overlapping) scans together with iodine fluorescence spectra for absolute calibration. The voltage ramp used to tune the laser was also recorded during the scan. Since this voltage controls linearly the length of the temperature stabilized external cavity to which the laser is locked, it is a reliable reference for linearization of the frequency scale. The joined spectrum, after conversion to the total absorption coefficient $\alpha_{\text{tot}} = 1/c\tau$, is shown in figure 3. We would like to point out that no baseline adjustments for different pieces of this spectrum were done. The repeatability and baseline stability of this technique is a very valuable feature for quantitative absorption measurements, both in the determination of transition intensities and in the realization of accurate trace element detection systems.

To illustrate the linearity of the recorded signal amplitude, in figure 4 we show a Gaussian fit of one of the isolated lines in the recorded HCCH spectrum. One can well appreciate that the lineshape did not suffer any distortion. The FWHM is found to be 1.275 GHz, which is less than 1% away from the calculated HCCH Doppler width at 25 °C. We also did a fit of the water line (due to sample impurity) at 17551.46 cm^{-1} , and obtained a Doppler FWHM of 1.54 GHz. The ratio of this width to that of the HCCH line is only 1% larger than the expected value, which depends only on the molecular masses and not on the temperature.

The noise level in this HCCH spectrum, measured as the standard deviation of a baseline section from a straight line (rms noise level), corresponds to an absorption coefficient of $10^{-9}/\text{cm}$. This is almost the same as the detection limit previously obtained by the first author using a pulsed CRDS system with 2 or 3 times longer ringdown time [8]. While the ringdown signal level is here much larger than in that previous work, the number of averages per point are about the same. Since the pulsed CRDS data were recorded using a boxcar, we are tempted to attribute the failure to obtain much better performance to extra noise in our digitization system. In fact, we tested our data acquisition system by feeding to it synthetic ringdown signals generated by the discharge of a capacitor. The pulse to pulse fluctuations we observed were 0.3%, practically the same as in the real

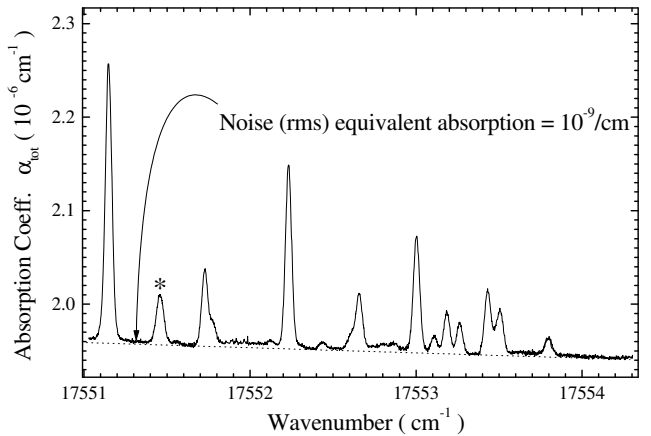


Fig. 3. Section of the HCCH overtone transition [1] near 570 nm at 5 Torr and room temperature. As pressure broadening was negligible, lines are Doppler broadened. The line marked by an asterisk is due to water impurity in the sample. No baseline correction was applied to the five 30 GHz laser scans which were joined to give this spectrum.

ringdown signal measurements.

While measuring this spectrum, we averaged only 50 ringdown events per data point, at the repetition rate of 200 Hz. We tested that averaging up to 1000 events gives a signal to noise ratio improvement that goes like the square root of the number of samples. Since the maximum repetition rate can be of the order of a few kHz, limited by the ringdown time, we can expect a factor of 3 improvement in the signal to noise ratio by sampling 500 events per point at the same scanning rate. Further improvements are to be expected both by optimizing the electronics and by using better mirrors. It is difficult to estimate the performance of different digitizing or alternative analog data processing schemes. However, better mirror coatings are certainly available in the range from $1.5 \mu\text{m}$ to about 400 nm. Decay times of $100 \mu\text{s}$ would give at once another factor 5 improvement of the detection limit. Here, we are scaling the sensitivity linearly with the ringdown time since we are limited by the digitizing electronics and not by detector or shot noise.

4. Discussion

While no ICLAS spectra were available for direct comparison, we could compare our data to the same spectral portion recorded by PAS which was kindly provided to us by Halonen and co-workers [1]. Even if their signal to noise ratio appears to be 2 times better, one should take into account that their pressure was 50 mbar (37 Torr) and that in conditions of negligible collisional broadening line intensities are proportional to pressure. We should also take into account that Halonen *et al.* used slightly longer averaging per data point. While it is diffi-

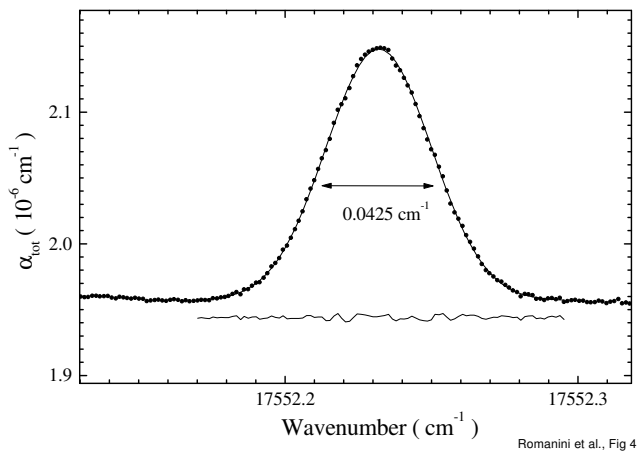


Fig. 4. Gaussian fit of one of the isolated lines in the recorded CW-CRDS HCCH spectrum. The FWHM is found to be 1.275 GHz, in perfect agreement with the HCCH Doppler width at 25 °C.

cult to generalize from this single comparison, we point out that Halonen’s intracavity resonant acoustic cell is one of the most performant PAS schemes. Therefore, we believe that CW-CRDS is as performant or better than PAS in terms of sensitivity.

With respect to spectral resolution, we stated above that this is in principle limited by the width of the excited cavity mode. It is clear however that in our setup the optical frequency averaged over several ringdown events was determined by the laser, since all we know is that these events occur somewhere inside the laser bandwidth. An effect that should also be considered is that the frequency of the trapped light adiabatically follows the mode frequency, which in our measurements shifts in time. Given the slow cavity modulation we used, we estimated that during the time of the ringdown, the total mode frequency shift was still much smaller than the laser linewidth. In any case, one should recognize that MHz resolution is indeed more than sufficient for most applications, in particular slit jet spectroscopy where the residual Doppler width for transitions in the visible is typically 100 MHz. Indeed, we believe that our setup is well adapted for jet spectroscopy as it is relatively insensitive to vibrations induced by the vacuum system.

Also, we would like to point out that it is quite simple to exploit the passive cavity stability to reach the limit of the cavity mode width. It is sufficient to passively stabilize the cavity from fast temperature drifts and vibrations and then dither the laser frequency instead of the cavity modes. The same electronics would then allow to track the laser as the cavity is smoothly tuned. There are some applications where sub-MHz resolution would be needed. Among those few, we think that one could use the high intracavity power to drive nonlinear phenomena, such as Doppler-free saturated absorption or

two-photon electronic transitions. UV rovibronic states could be studied with visible tunable CW sources with a resolution sufficient to study even large molecules. Two-photon transition measurements were already obtained in the past using fluorescence detection [29]. However, important information would be deduced from the direct CW-CRDS absorption measurement of two-photon transition strengths.

The high intracavity power depends on the efficiency of the buildup. In the limit when the relative modulation of cavity and laser is slow, buildup is limited by fast frequency fluctuations of the laser. This is equivalent to consider the average laser linewidth compared with the mode width. In our case, with a laser power before the ringdown cavity of 100 mW, we could observe buildups such that the transmitted cavity intensity was on the order of 0.1 mW, which corresponded to about 1 W intracavity. Using the transverse section of the TEM₀₀ mode, this gives an intensity of about 300 W/cm². Use of other single frequency laser sources and mirror coatings could provide substantially different buildup performance, so that much larger intensities could also be achieved.

5. Conclusions

We have demonstrated a new version of cavity ring down spectroscopy employing a tunable CW single frequency laser source. A tracking system has been developed which locks automatically and then enables the ringdown cavity to follow the laser frequency for arbitrarily large scans. The experimental setup is rather simple and any commercial single frequency laser source can be used without modification, with its linewidth determining the spectral resolution. In particular, the combination of CW-CRDS with diode lasers is very promising for application to trace detection. By measuring Doppler-limited weak absorption spectra in acetylene, we demonstrated a detection limit (rms noise) of 10⁻⁹/cm, or 5 × 10⁻⁸/pass, which is as good as in the standard pulsed CRDS. As we are limited at the moment by noise in the digitizing electronics, the detection limit improves linearly with the mirror quality. We are confident that faster data acquisition rate, optimized electronics, and better mirror coatings will allow at least a factor 10 further performance improvement. Our CW-CRDS setup is quite insensitive to external perturbations and vibrations, which makes it promising for supersonic jet spectroscopy. Finally, together with the high power fluxes (> 100 Watts/cm²) to which the sample may be subjected, CW-CRDS should be considered for the investigation of nonlinear phenomena such as Doppler-free saturated absorption or Doppler-free two-photon absorption.

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