

Noise in cavity ring-down spectroscopy caused by transverse mode coupling

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Abstract: In our continuous wave cavity ring-down spectroscopy (CW-CRDS) experiments, we have often observed that the decay time constant drops to a lower value at some cavity lengths or some intercavity pressures. The resulting instabilities lead to a reduction in the sensitivity of our CRDS system. We have deduced that the cause of this noise is the coupling between the TEM₀₀ mode that the laser excites, and the higher order transverse modes of the cavity. The coupling will cause anti-crossings as the modes tune with cavity length. A consequence is that the decay of light intensity leaving the cavity is no longer a single exponential decay, but the signal can be quantitatively fit to a two-mode beating model. With a 4 mm diameter intra-cavity aperture, the higher order modes are suppressed and the stability of the system improved greatly. One coupling mechanism is scattering from the mirror surfaces. This can explain some features of our data including the strength of this coupling and the relative tuning rate of the coupled modes. Remarkably, a scattering intensity between modes of $\sim 10^{-12}$ can produce observable changes in the cavity decay rate. However, the tuning rate between the TEM₀₀ mode and the higher order modes in a cavity pressure scan is larger than predicted and is still not explained. Images of higher order transverse modes excited at certain cavity conditions were recorded by an Indium Gallium Arsenide (InGaAs) area camera.

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OCIS codes: (280.3420) Laser sensors; (290.5880) Scattering, rough surfaces.

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1. Introduction

Cavity ring-down spectroscopy (CRDS) [1] has proven to be an extremely sensitive method of detecting small absorption levels of gaseous samples [2]. In this method, absorption of a sample is determined by an increase in the decay rate of an optical cavity that contains the sample of interest. Particularly advantageous is excitation with a continuous wave laser that is then rapidly turned off (compared to the decay time of the optical cavity), as this allows for much cleaner excitation of a single mode of the optical cavity. This method is known as CW-CRDS [3, 4, 5]. The sensitivity of CRDS to changes in sample absorption coefficient is equal to the speed of light in the sample times the noise in the determination of the cavity decay rate as determined by repetitive measurement of that decay as either the sample or excitation wavelength is changed. In the near-IR, where mirrors with $R \approx 99.999\%$ and distributed feedback (DFB) diode lasers are available, we can routinely achieve noise levels of about 1 part in 10^4 in the decay rate in a few seconds of signal averaging, which corresponds to a sensitivity of about 1 part in 10^9 absorption per pass of the cell.

We have often found, however, that there are times when the sensitivity of the instrument is quite a bit lower due to considerably increased instability of the decay rate at fixed wavelength and quite spectrally sharp variations in the decay rate with the length or inner pressure of the cavity. This paper presents our analysis of the cause of these instabilities and the method we have found that eliminates them. Briefly, we deduce that the noise arises from coupling between the TEM_{00} mode of the cavity we are exciting and higher order transverse modes which tune into resonance at specific cavity lengths. These modes are coupled by scattering on the mirrors which tends to excite each mode of the cavity. If the modes are not very nearly degenerate, the field amplitude transferred on each pass of the cavity destructively interferes and only a trivial amount of energy is transferred into the higher modes. However, in the case of exact degeneracy, the transfer is coherent on each pass and can add up constructively. Given the high signal to noise ratio of each individual decay ($\approx 10^3 : 1$), a power transfer $\sim 10^{-12}$ per pass will effect our determined decay rate.

In the rest of this paper we will first describe our experimental setup, then our observations, and finally present an analysis of our results.

2. Experimental setup

Our CW-CRDS setup is similar to that previously described [6] and also that adopted by other groups, so only the essentials will be described. Light is produced by a DFB semiconductor diode laser (NTT Electronics Corporation, NLK1U5EAAA) operating near 1652 nm. The linewidth is measured to be 10 MHz with Lorentzian line shape. The laser frequency is stable to 20 MHz on seconds due to laser temperature instability and it also drifts about 50 MHz per hour. The laser temperature tuning range (0 to 30 °C) corresponds to a change in wavelength of 3 nm. The laser comes mounted on thermoelectric cooler and fiber coupled with a 30 dB optical isolator. The light from the optical fiber is collimated, passed through an 47 dB (measured at 1652 nm) external optical isolator (IsoWave, I-15-UHP-4), and then an acousto-optic modulator (AOM) (IntraAction, ACM-802AA14). The laser frequency of the first order diffraction from

the AOM has a positive 80 MHz shift to the incident laser frequency in our system. This order passes through a two lens telescope and is mode matched into our CRDS cell.

The cell consists of two 1 inch diameter super mirrors (Research Electro-Optics Inc., with reflectivity $R = 99.9987\%$ at $\lambda = 1652$ nm) that are held in mirror mounts with bellows between the fixed and variable plate. This allows for the mirrors to be adjusted while under vacuum and a seal to be made around the outside of the mirror. The input mirror is flat and the output one has a radius of curvature $R_c = 1$ m and the mirrors are separated by $L = 39.5$ cm. This gives a free spectral range $FSR = c/2L = 379.5$ MHz and a transverse mode spacing of 82.1 MHz for an empty cavity. The TEM₀₀ mode of the cavity has a focus on the flat mirror and a calculated beam waist of $\omega_0 = 0.507$ mm. On the output mirror, the mode has a beam waist of $\omega_1 = 0.652$ mm and, of course, a radius of curvature that matches that of the mirror. Both mirrors have the back side wedged by 0.55° to prevent feedback into the cavity which would lead to modulation of the mirror effective reflectivity. The input mirror mount has three piezoelectric transducers (PZT) that allow the cavity length to be scanned over $8 \mu\text{m}$.

Light leaving the ring-down cavity is focused onto a $300 \mu\text{m}$ diameter InGaAs detector using a 4 cm focal length off-axis parabolic mirror. The TEM₀₀ mode of the cavity is calculated to be focused to a spot with beam waist $28.4 \mu\text{m}$ on the detector. The detector is slightly tilted to prevent the reflection from its front surface from feeding back into the cavity, which could also be a source of instability in the decay rate.

In CW-CRDS, either laser (using current tuning) or cavity (using PZTs) is scanned into resonance. A trigger detector monitors the cavity transmission and when this exceeds a preset threshold, the radio frequency (RF) power to the AOM is turned off. Also, a data acquisition card is triggered, which captures the ring-down event for signal processing. The measured extinction ratio of our AOM is 49 dB, which was achieved by inserting a second RF switch between the RF oscillator and amplifier. Our detector has a transimpedance amplifier with a 3 dB bandwidth of about 1 MHz. The data acquisition digitizes the detector output with a 12 bit A/D and a digitization rate of 1 MHz. The detector noise is measured to be 2.5 mV (RMS), which based upon the transimpedance gain of the amplifier (5×10^6 V/A) and the optical sensitivity (0.9 A/W) corresponds to a detector noise of 0.6 nW .

By careful alignment of the cavity, we can reduce the transmission of higher order modes to 6% of that of the TEM₀₀ modes. See Fig. 1. This allows us to set the threshold such that we have essentially zero probability of triggering on a higher transverse mode. To maximize stability of the cavity decay rate, and thus CRDS sensitivity, one wants to insure that only decay of TEM₀₀ modes are observed.

The detector signal is digitized for 2 ms after each trigger. We determine the detector offset and noise by fitting the last 0.5 ms signal, by which time the cavity intensity has decayed to well below our detector noise. This baseline is subtracted from the earlier points and a weighted least squares fit [7] is done to the logarithm of the signal to determine the cavity decay rate. This is followed by one or two cycles of a nonlinear least squares fit [7] to the original data, which experience and modeling have shown us to be fully converged. The fitting model is

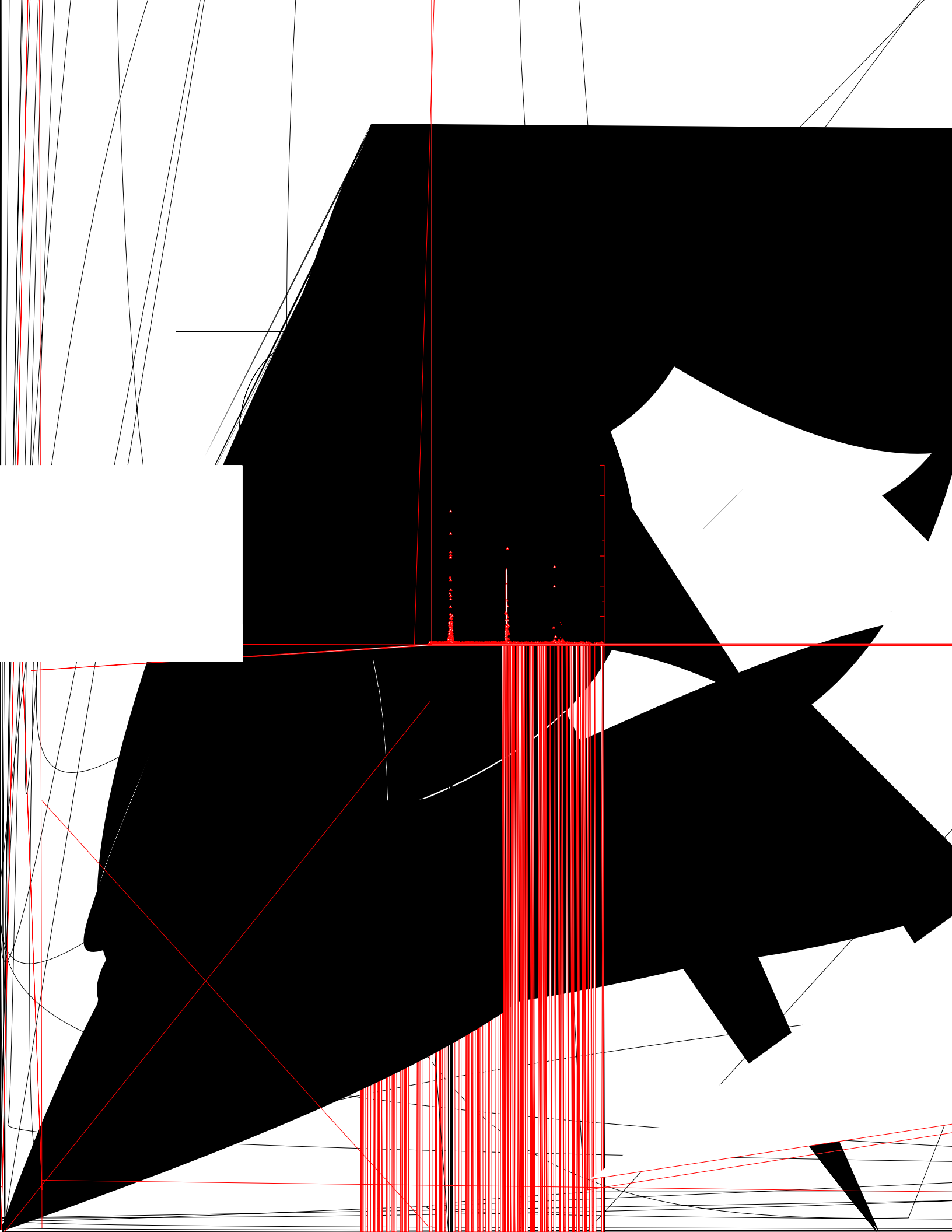
$$y(t) = A + B \exp(-kt) + \text{detector noise} \quad (1)$$

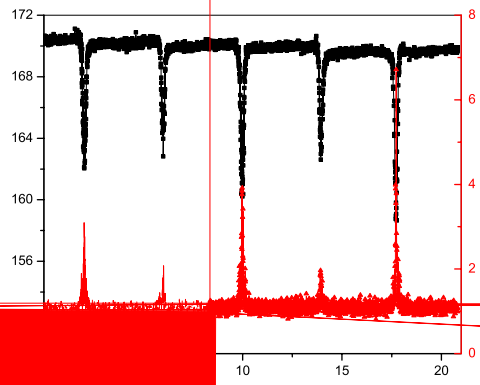
$y(t)$ is the detector voltage at time t , A is the detector DC offset, B is the amplitude at the beginning of the ring-down decay, and k is the cavity decay rate. We judge the quality of the fit by using the reduced χ^2 of the fit [7] given by:

$$\chi^2 = \frac{1}{(N-3)\sigma^2} \sum_{i=0}^{n-1} (y(i) - A - B \exp(-ki))^2 \quad (2)$$

where N is the number of data points in the observed decay (typically ~ 2000), A , B , and k are







In... on
d...
g...



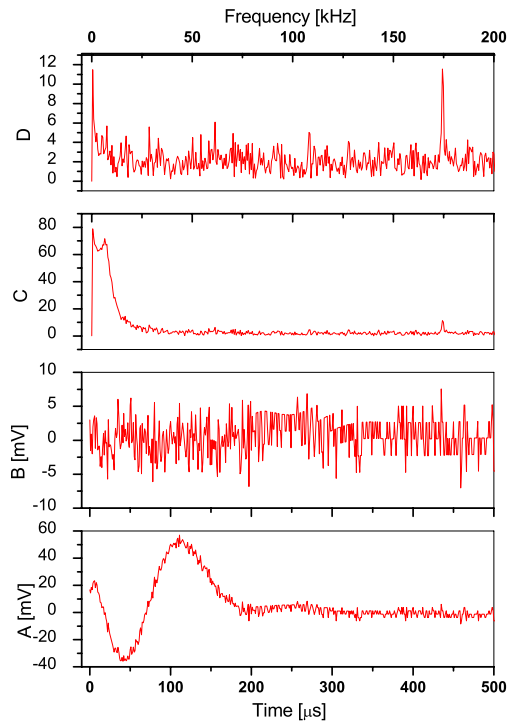


Fig. 6. A is the residuals of the fit to a single exponential decay and C is the noise spectrum of it; B is the residuals of the fit to two-mode beating model and D is the noise spectrum of it. A and B have the same horizontal axis. C and D have the same horizontal axis. The peak ~ 175 kHz in C and D is from the computer system. The time zero point has been shifted because the first 10 points of each decay are always skipped in the fitting.

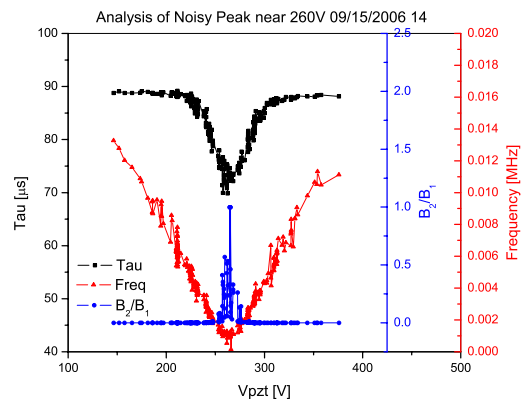


Fig. 7. Analysis of one of the noisy peaks in Fig. 2. The cavity was filled with 4.43 torr Nitrogen gas. The slope of $\Delta\nu$ changing with V_{pzt} is ≈ 120 Hz/V, or 7.215 kHz/ μm .

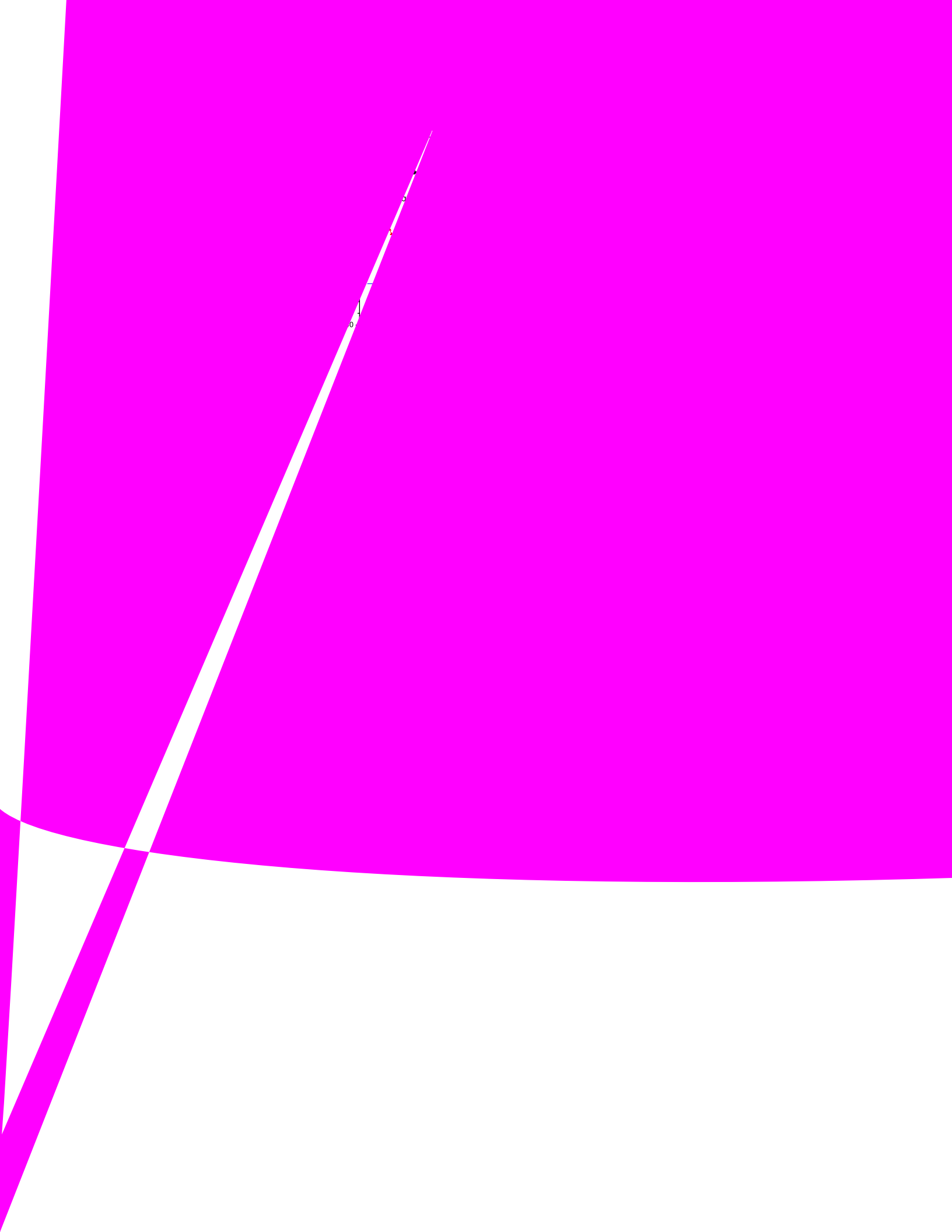


Fig. 1

—A—

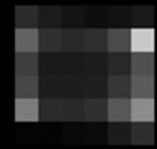
Chi Squ

—B—

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A



B

with a mode diameter of 100 μm and a diameter of 100 μm.

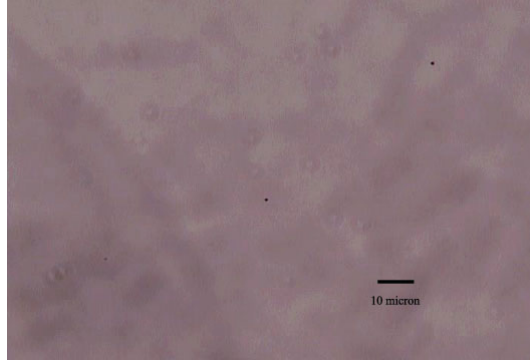


Fig. 11. Image of damaged spots on one of the old supermirror surfaces. The picture represents an area of $146(H) \times 100(V) \mu\text{m}$ near the center of the HR surface of the mirror. The three black dots are damaged spots. The size of the spots is $\sim 1 \mu\text{m}$. The nonuniform background is because the objective lens is dirty. This type of damaged spots were not observed for new mirrors.

frequency ν_{qnm} [8], given by

$$\nu_{qnm} = \frac{c}{2n_0L} \left(q + (m+n+1) \frac{\cos^{-1}(\sqrt{1-L/R_c})}{\pi} + \delta\varphi \right) \quad (4)$$

where $\delta\varphi$ is a constant proportional to the phase shift of the light upon reflection off the mirror. This gives a longitudinal mode spacing $FSR = 379.5 \text{ MHz}$ and a transverse mode spacing of $\Delta\nu_T = 82.1 \text{ MHz}$. Thus for two modes to be degenerate, we require that $\Delta q\Delta\nu_L + \Delta(m+n)\Delta\nu_T = 0$. The lowest order near resonances calculated by this expression have $\Delta(n+m) = 37, 74$ with mode detunings of 1.553, 3.105 MHz respectively. The radial size of a TEM_{nm} mode is given approximately by [8] $\sqrt{n+m+1} \omega$ where ω is the beam radius of the Gaussian shaped TEM_{00} mode (where the field drops to $1/e$ of its peak value). For our cavity, the limiting aperture has a radius of 7.6 mm near the curved mirror where the beam radius is 0.652 mm. Thus, our cavity can support modes with $n+m < 135$. One thing to note is that a change in n_0 holding L constant shifts all modes proportional to their frequency and thus does not lead to any mode crossings.

If we consider a change in cavity length, then all the modes will change frequency, but not at the same rates.

$$\frac{d\nu_{qnm}}{dL} = -\frac{\nu_{qnm}}{L} + \frac{c}{2n_0L} \frac{n+m+1}{2\pi\sqrt{L(R_c-L)}} \quad (5)$$

For our cavity, the frequency difference of two modes will shift at a rate of $123\Delta(n+m) \text{ Hz}/\mu\text{m}$. Comparing with the data presented in Fig. 7, where a slope of $7.215 \text{ kHz}/\mu\text{m}$ is observed, we conclude that for this resonance, the mode crossing the TEM_{nm} mode must have transverse mode numbers $n+m \sim 59$. If the cavity length is 39.604 cm, based on Eq. (4), the index of the lowest order mode approaching the TEM_{00} mode is 60, with a mode detuning of 31 kHz. If we consider the pressure scans, the modes move relative to one another only due to strain induced change in the cavity length. As discussed earlier, we can estimate the magnitude of this effect from the pressure change required for one FSR change in the cavity compared to the calculated index shift. For both gases, the cavity length increases about $0.1 \mu\text{m}$ per torr, while with Xenon, 81% of the pressure scan is done by refractive index changing and with Nitrogen, the same number is only 59%. Using this factor, we can convert the slope of the mode crossing

observed in Fig. 8 to a shift per unit cavity length change, which is $307 \text{ kHz}/\mu\text{m}$. This predicts that for this resonance the transverse mode numbers of the crossing mode are $n + m \sim 2500$, which is unphysically high.

Comparing with the experimental results in Fig. 10, we can see that the calculation above explains the PZT scan very well but the pressure scan badly. Although the diffraction losses

fit would vary evenly both above and below the decay time of the TEM₀₀ mode as the initial relative phase of the mode excitations changed from 0 to π . This lack of symmetry in the variation in decay times was for a long time a point of confusion.

Figure 7 and 8 also show the ratio of the two mode amplitudes as a function of tuning. We note that the relative amplitudes are strongly dependent upon tuning, peaking when the modes are as nearly degenerate as possible. This result strongly suggests the analogy to the situation of quantum beat between two quantum levels that tune through resonance *when there is a coupling between the states* [11], and that the excitation of the the higher order modes of the cavity is not due to errors in the mode matching of the input beam, but rather due to some coupling of these modes inside the cavity.

In the standard theory of the resonance modes of an optical cavity, each mode is independent and there is no coupling between modes – it is a linear system like the normal modes of a set of harmonic oscillators. If we consider scattering on each mirror of the cavity, this will create a speckle pattern that will tend to couple the field amplitude of different modes. On each mirror reflection, one will have a matrix that relates the amplitude in different transverse modes before and after reflection, with small off-diagonal scattering elements. These will have a Gaussian random distribution if one assumes that we have a large number of independent isotropic scattering centers, which is speckle scattering [12]. The importance of scattering leading to coupling and mode repulsion in a highly degenerate cavity was discussed by Klaassen et al. [13], who demonstrated that this leads to an apparent width of a transmission peak that was significantly wider than that predicted from the observed cavity decay time.

The assumption that there is a scattering coupling of the modes explains some of the curious features of our observations. Once we excite the TEM₀₀ mode of the cavity, we will have transfer of electric field amplitude from the TEM₀₀ to other modes of the cavity. However, the field transferred on each round trip will change in relative phase by $\Delta\nu t_r$, where $\Delta\nu$ is the difference in frequency between the modes and $t_r = 2L/c$ is the cavity round trip time. For typical mode spacings, this will lead to destructive interference after a few round trips and we will get very weak excitation of these other modes for small scattering amplitudes. However, if one of the modes, TEM_{*nm*}, is very nearly resonant with the excited TEM₀₀ mode, then the field amplitude in it will initially build up linearly with time (intensity of that mode increase quadratically in time) and in fact the power in the TEM₀₀ will decay both due to intrinsic cavity losses and due to this coherent transfer. Thus, we expect that the observed decay will generally no longer be a single exponential function and will decay faster than when the TEM_{*nm*} mode is off resonance, if the energy in TEM_{*nm*} is not totally collected by the detector. This clearly agrees with our observations. In addition, when these two transverse modes are nearly on resonance, they will mix with each other severely through the coupling, generating two new eigenmodes which are mixtures of the TEM₀₀ and the TEM_{*nm*} modes. These two new eigenmodes can be excited together, and decay simultaneously because the spacing between them (\sim kHz) is much smaller than the laser linewidth (10 MHz). Thus, beating can happen between them as an analogy of a quantum beat, if the orthogonality between TEM₀₀ and TEM_{*nm*} is broken, or the detector has different sensitivity for these modes. Eq. (3) describes one extreme case of this beating, which is, all the energy of the TEM_{*nm*} mode is not collected by the detector while no loss for the TEM₀₀ mode. B_1 and B_2 are not the initial intensities of these two transverse modes, but those of the two diabatic states. In our system, only modes with the index $m + n$ less than 27 still can be efficiently collected by the detector. The 60th order transverse mode is focused to a size much larger than that of the detector, leading to a calculated energy loss of 54%. Our data shows even with this number Eq. (3) still describes the noisy data very well because the intensity of TEM_{*nm*} in the cavity is much smaller than that of the TEM₀₀ mode (see next paragraph). $\Delta\nu$ is like that of an avoided crossing with a minimum value given by the

scattering coupling. Based upon the quantum analogy, we can predict $\Delta v_{min} = 2|S_1 + S_2|/t_r$, where S_1, S_2 are the scattering amplitudes for each mirror. Based upon our observation that $\Delta v_{min} \sim 1$ KHz, we can estimate $S_1, S_2 \sim 10^{-6}$, which translates a scattering intensity of only $\sim 10^{-12}$ per pass. The point that two new eigenmodes beat with each other is also verified by experimental data. At two wings of each dropout peak, τ_1 (inverse of k_1) is substantially larger than τ_2 (inverse of k_2), and they can be regarded as the lifetime of the TEM₀₀ and TEM_{nm} modes respectively because the interference between them is not as strong as at the peak. However, when approaching the peak, τ_1 decreases and τ_2 increases to new values which are between τ_1 and τ_2 near dropout peak wings. These two new time constants are very close with each other and can be the same. They are decay time constants of the two mixing eigenmodes. These two mixing modes decay faster (τ decreases) than the TEM₀₀ mode, and slower (τ increases) than the higher order mode. The lifetime τ_{nm} of the higher order mode is smaller than that of the TEM₀₀ mode (diffraction losses of both are totally negligible). Possible explanations are that higher order modes can have larger scattering losses because of larger mode size, or that they sample different parts of the mirrors, which may not be spatially uniform.

In terms of the two state model we used to fit the decays above, the peak value of the excitation of the TEM_{nm} mode in the two adiabatic modes, which are mixtures of the TEM₀₀ and TEM_{nm} modes, even on resonance, will no longer be equal to $(1/\sqrt{2})$, but $\Delta v_{min} \tau_{nm}$. The maximum decrease in the TEM₀₀ intensity due to coupling to this TEM_{nm} mode is proportional to $(\Delta v_{min} \tau_{nm})^2$. Without the intracavity aperture, τ_{nm} is $\sim 50 \mu\text{s}$ for our system. Therefore the peak intensity of the TEM_{nm} mode is only $\sim 10^{-3}$ of that of the TEM₀₀ mode even on resonance (using the $\Delta v_{min} \sim 1$ kHz). With the intracavity aperture, the TEM_{nm} mode will have much higher diffraction loss. If we assume the diffraction loss reduces τ_{nm} down to $\sim 100t_r$, this maximum intensity loss will be $\sim 10^{-10}$. This is well below the signal to noise in the decay and thus will not effect the observed mode decay rate.

We can estimate the size we expect for the scattering coupling. Let the total scattering power loss of each mirror be L_s . Assuming that this mode is isotropic, the fraction of scattering intensity into a given mode should be on the order of $L_s \Delta\Omega / 2\pi$, where $\Delta\Omega \sim \pi(\lambda / \pi\omega_0)^2 = 3 \cdot 10^{-6}$ is the solid angle subtended by the TEM₀₀ mode in far field. Thus for a total scattering loss of a few parts per million per reflection ($= 1 - R - T$, T is the transmission of the mirror), we can estimate that the scattering power coupled into other modes will be on the order of $\sim 10^{-12}$, of the same order as required to explain the strength of our observed anticrossings between the lowest order and higher order modes. Given that many of the observed defects on the surface of the mirrors are visible under a modest power microscope, the assumption of isotropic scattering is perhaps crude. This estimate supports our model of coupling through surface scattering. Also, as pointed out before, new mirrors give smaller magnitude of depth and reduced χ^2 at dropout peaks in both cavity pressure scans and PZT scans (not shown here). This means that Δv_{min} of new mirrors is smaller than that of old mirrors, just as what we observed in the experiments.

Another possible coupling mechanism between TEM₀₀ modes and higher order modes is the reflection from the anti-reflective (AR) coating surface of each wedged mirror. This reflection will generate feedback to the TEM₀₀ mode. For a mirror with HR coating reflectivity of 99.999% and that of 1% for AR coating, the feedback again is on the order of 10^{-12} . However, the 0.55° wedged angle and the refractive index of the substrate will produce an angle displacement of 29 mrad to this weak feedback compared with the TEM₀₀ mode in the cavity. With this angle the transverse displacement of this feedback beam near the curved mirror is about 11 mm, which is larger than the limiting aperture size (7.6 mm) of the cavity.

In our experiments we also recorded decay signals of TEM₀₁ and TEM₁₀ modes, which can be triggered when their intensities are increased to reach the threshold by misaligning the cavity away from the TEM₀₀ mode. We found they have different decay times from TEM₀₀ modes,

likely because they sample different places on the mirrors which are not perfectly homogenous (calculated diffraction losses are completely negligible). We also found those decay signals are no longer single exponential decays but can also be fitted to the modified Eq. (3) quantitatively ($2\sqrt{B_1 B_2}$ replaced by B_3). Here B_1 and B_2 are initial amplitudes of TEM₁₀ and TEM₀₁ modes. B_3 is twice of the field overlap integration of these two modes. We found B_3 does not equal to zero. This suggests these two modes are no longer orthogonal, possibly because the transmission of the mirrors or the detector quantum efficiency are not spatially uniform. $\Delta\nu$ equals to 261.3 kHz, which is much larger than that of the cavity length scan and pressure scan situations and can not be explained by the mirror surface scattering coupling. We believe it is due to the lifting of the degeneracy of TEM_{*nm*} modes with $n + m = \text{constant}$ due to weak astigmatism in the cavity induced by stress on the mirrors, or from the broken cylindrical symmetry of the curvature when the mirrors were made. From Eq. (4) we can estimate the frequency difference between TEM₀₁ and TEM₁₀ modes if the radius of curvature of the mirror in x direction is no longer the same as that in y direction.

$$\frac{d\nu_{qnm}}{dR_c} = -\frac{c}{2n_0L} \frac{n+m+1}{2\pi R_c} \sqrt{\frac{L}{R_c-L}} \quad (6)$$

For our cavity and $n + m = 1$, $\Delta\nu$ of 261.3 kHz gives $\Delta R_c \sim 2.6$ mm according to Eq. (6), which corresponds to a $\frac{\Delta R_c}{R_c}$ change of 0.26%.

5. Conclusion

In conclusion, we have found the time constant of ringdown decays drops to a much lower number at some cavity length or some inner pressure of the cavity, and we deduced the cause of this noise is the coupling between the TEM₀₀ mode and the higher order transverse modes of the cavity. Images of the higher order modes excited at certain cavity conditions were captured by an InGaAs area camera. The noise signals are no longer single exponential decays but can be fitted to the two-mode beating model (Eq. (3)) quantitatively with the reduced χ^2 very close to one. The noise in both cavity length scans and cavity pressure scans can be removed by inserting a 4 mm diameter aperture in the cavity. One possible coupling explanation is scattering from the mirror surfaces because of the coating imperfections of them. This coupling will cause an anti-crossing as transverse modes tune with cavity length. For the cavity length scans, this mechanism explains the experimental data successfully. The strength of this scattering coupling calculated from the experimental data matches with the estimation of it by using the measured scattering loss of the mirrors, which supports our model of surface scattering coupling. We also found decay signals of TEM₀₁ and TEM₁₀ modes are no longer single exponential decays but can also be fitted to the modified Eq. (3) quantitatively. We believe this splitting is due to the lifting of the degeneracy of TEM_{*nm*} modes with $n + m = \text{constant}$ due to weak astigmatism in the cavity induced by stress on the mirrors, or from the broken cylindrical symmetry of the curvature when the mirrors were made.

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