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Dispersion in a cw Optically Pumped FIR Laser

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Abstract. Direct frequency measurements of a $^{13}CO_2/^{15}NH_3$ OPFIRL output show that laser pulling effects give shifts of a few MHz, which agree well with a density matrix calculation.

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It has been known for some time that cavity frequency pulling can give large frequency shifts in the output of pulsed optically pumped FIR lasers (OPFIRL's) [1]. Because the line Q and the resonator Q are of the same order of magnitude, this effect can be quite pronounced in cw optically pumped lasers, and may be of importance for frequency synthesis or high-resolution laser spectroscopy applications [2–6]. We report here the direct measurement of the frequency of a cw OPFIRL as a function of FIR resonator tuning; significant pulling effects are observed which agree well with a density matrix calculation of two-photon dispersion.

We have previously made detailed measurements of the dependence of the output power of a cw $^{15}NH_3$ waveguide OPFIRL on $^{13}CO_2$ input power, $^{13}CO_2$ laser frequency and FIR resonator length [7]. The complex changes observed were attributed to laser light shifts in a Doppler broadened three-level system, although it was recognised that dispersion could be important. We have now monitored the OPFIRL frequency by beating with the output of a reference system.

The apparatus has been largely described elsewhere [7], with the addition of a second OPFIRL chain for the FIR beat detection measurements (Fig. 1). The cw CO_2 pump lasers (Edinburgh Instruments, PL3), PZT tunable over ±40 MHz, were monitored by heterodyning with respect to a reference laser, locked opto-galvanically to line centre. The FIR outputs were mixed in a helium cooled InSb Putley detector, and the beat frequency displayed on a spectrum analyser. The

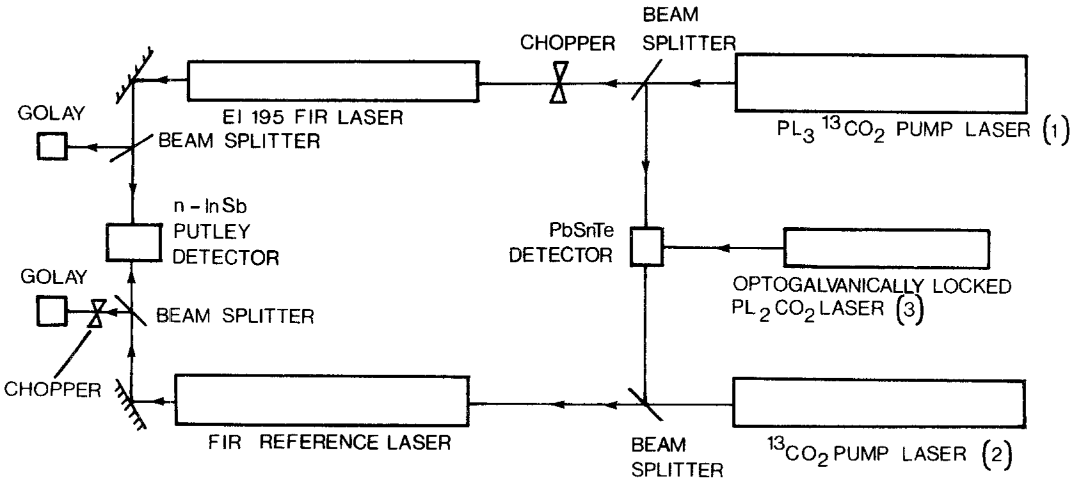

Fig. 1. FIR heterodyne system for direct frequency determination

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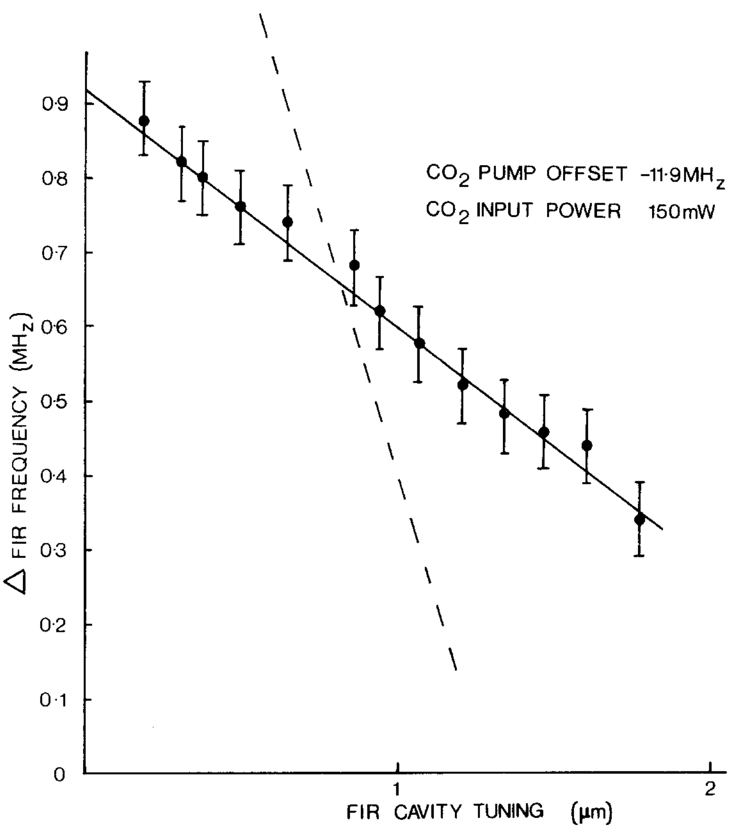

Fig. 2. FIR frequency of $^{15}NH_3/^{13}CO_2$ laser, measured relative to reference FIR laser, as a function of cavity length tuning for single, on-resonance, peak. The dashed line is the empty cavity tuning, and the solid line is the theoretical tuning, as described in the text

frequency of the reference OPFIRL was determined by the transferred Lamb-dip technique [8]. When the pump frequency is tuned through resonance the FIR output shows a narrow dip related to the Lamb dip of the pump transition, enabling us to make a precise measure of the centre frequency. For the particular case of $^{13}CO_2/^{15}NH_3$ a pump offset of -11.9 MHz from CO_2 line centre gives resonant pumping.

In Figs. 2 and 3 are shown the frequency of the FIR output-measured relative to the reference OPFIRL – as a function of FIR cavity-length tuning at fixed pump frequency. A single peak is obtained with on-resonance pumping (Fig. 2); two (Doppler split) peaks with off-resonance pumping (Fig. 3). The dashed lines show the tuning curves expected for an empty cavity, and the solid lines are those computed theoretically as described below.

Our analysis of the observed effects is based on the strong-signal 3-level model as applied to OPFIRL's [1, 2]. We have gone beyond these analyses in two main respects. First, we determine the FIR laser frequency and power (Rabi frequency) *self-consistently*; and second we have incorporated the bidirectional nature of the FIR waves to lowest orders by adding the co- and counterpropagating susceptibilities. We outline the present calculation only, since the basic equations are readily available [1, 2]. For simplicity, we assume small pump absorption, so that the pump intensity I_1 is constant along the tube; similarly we distribute the FIR losses so that the FIR intensity I_2 is constant. Since we are dealing with a Doppler-broadened pump transition, we velocity-average the calculated susceptibility. For any I_1, we can thus calculate the FIR susceptibility, either small-signal ($I_2=0$) or strong signal (I_2 finite). We typically find a double-peak gain structure, with splitting roughly equal to the expected Doppler-splitting $2(k_2/k_1)|\Delta_1|$, where k_2, k_1 are the FIR and pump wavevectors and Δ_1 the pump offset. One only of these peaks shows Autler-Townes splitting as I_1 is increased. This splitting is washed out, as expected, for large enough values of I_2.

In a laser, the intensity (power) and frequency are not set a priori, but adjust to satisfy the equilibrium conditions: (i) the gain must balance the cavity loss (5% per pass, measured in our system); (ii) the cavity length must be an integral number of half-wavelengths. These conditions on respectively the imaginary and real parts of the susceptibility fix the FIR intensity and frequency for fixed values of pump intensity and offset, linewidths and, of most interest here, cavity length L. When L is swept and other parameters held fixed, $\mathrm{Re}\{\chi\}$ must change to maintain the wavelength condition, and so, as is well known, the frequency is pulled

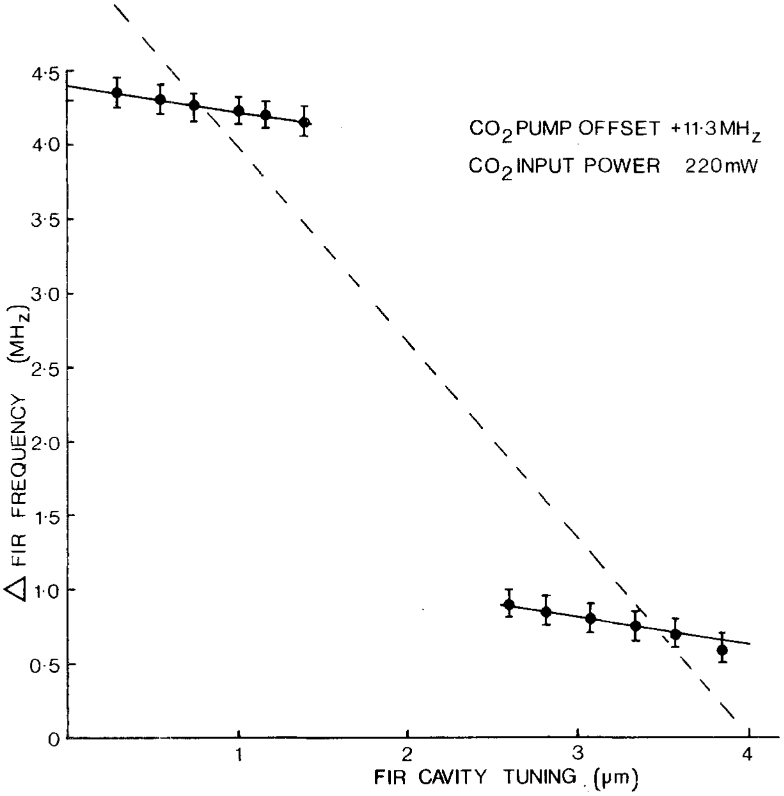

Fig. 3. As Fig. 2, for a fixed off-resonance pump setting, showing tuning curves for the Doppler split peaks. The dashed line is the empty cavity tuning, and the solid lines the theoretical curves

from that of peak gain towards the empty cavity resonance frequency and vice versa. For the ranges of parameters considered, the pulling is nearly linear, and in good accord with the observations (Figs. 2 and 3). For the case where the gain is double-peaked, the output frequency first lingers close to one peak as *L* is swept, then abruptly tunes to the vicinity of the second peak. For the case where the FIR laser is above threshold throughout the inter-peak region, the present model shows hysteresis in this switch, i.e. it occurs at different cavity lengths depending on the direction of which *L* is varied.

At first sight, one would suppose that peak FIR output power be obtained where the laser frequency coincides with the empty cavity frequency, i.e. at the intersections of the tuning lines in Figs. 2 and 3. This is not so, as can be seen in Fig. 4. Here we show, as a function of pump offset, the change of *L*, i.e. of empty cavity frequency, required to tune from one power peak to the other. If peak power coincided with zero pulling, these circles would lie on a straight line, but in fact there are deviations from the straight line, with a dispersion-like spectrum versus pump offset. In contrast, the directly measured frequency splitting of the peaks (crosses in Fig. 4) show a much smaller deviation from linearity. The phenomenon is thus predominately a pulling effect. The explanation lies in the fact that the FIR field is in resonance with the excited velocity group of NH$_3$ molecules for only half a round trip: on the return half, the Doppler shift switches these molecules out of resonance, so that the FIR field experiences little gain; but because the real part of the susceptibility decays more slowly off-resonance than the imaginary part, there is a phase shift on this "dead" pass, which is responsible for the excess splitting in Fig. 4.

This effect arises naturally, with the correct sign and spectral signature, from the present model in which the effects of bidirectionality of FIR (and pump) are included. The phase shift is of opposite sign for the two power peaks because of the population inversion, meaning that the cavity must be tuned *further* than expected between the peaks. The size of the anomalous splitting is governed by the FIR homogeneous linewidth and the pump offset in a manner consistent with Fig. 4. When the effect is measured in terms of true FIR frequency, it would be expected to be *less* than $2(k_2/k_1)\Delta_1$, because the gain lines partially overlap, leading to merging at small enough pump offsets. This is not as observed experimentally (Fig. 4). We report elsewhere a treatment in the weak probe limit, which includes coherent effects [9]. We have identified a new coherent effect, a "two-photon light shift", which pushes the peaks apart and opposes their collapse into each other near line centre.

In conclusion we have made direct frequency measurements of an OPFIRL output and compared them with that expected from an empty cavity. Pronounced pull-

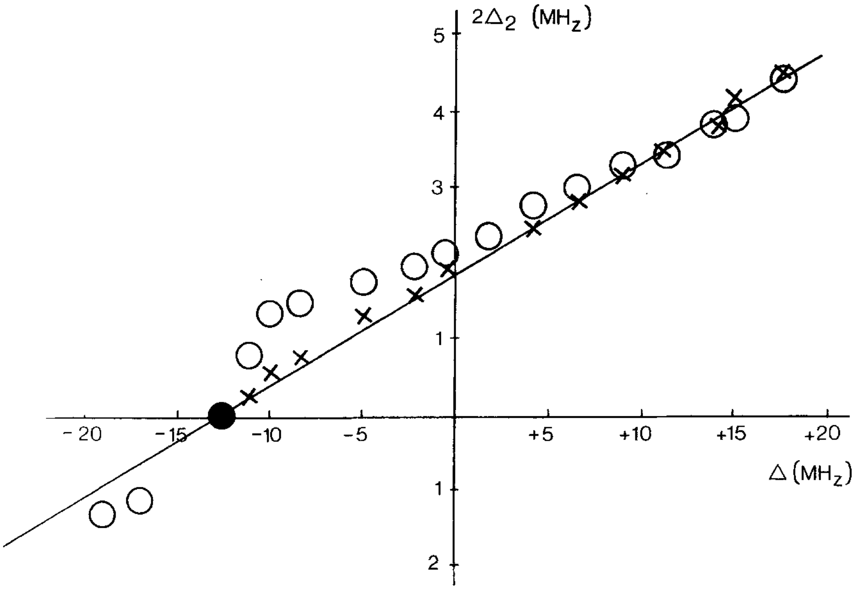

Fig. 4. Gain peak splitting of $^{15}NH_3/^{13}CO_2$ laser OPFIRL at 152.9 µm as a function of fixed pump offset. The crosses are from the direct frequency measurements; the circles are deduced from cavity scans, and the solid circle from the transferred Lamb dip measurement. The line is $\Delta_2 = (k_2/k_1)\Delta_1$

ing effects are observed which agree well with our density matrix calculation. Dispersion of the FIR laser gain medium is shown to markedly effect the Doppler splitting of the output spectrum, and is important for the fine tuning characteristics of such systems.

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