

## Module 4 : Third order nonlinear optical processes

### Lecture 27 : Third-order nonlinearity measurement techniques: ARINS

#### Objectives

##### This lecture describes

- Theory of Antiresonant Ring Interferometric Nonlinear Spectroscopy (ARINS) technique for the measurement of third order nonlinear susceptibility  $\chi^{(3)}$ .
- Its experimental details and
- Its merits and demerits.

#### Third-order nonlinearity measurement techniques: ARINS

As mentioned in Lecture#26, Z-scan technique can not differentiate between the electronic and other mechanism of nonlinearity. Also, it does not provide high enough sensitivity for the measurement of weakly nonlinear materials at moderate and safe intensity levels. Here we describe the Antiresonant Ring Interferometric Nonlinear Spectroscopy (ARINS) Technique.

Apart from being a highly sensitive technique, it has the unique capability of discriminating between electronic and other mechanisms of nonlinearity.

#### Antiresonant Ring Interferometric Nonlinear Spectroscopy (ARINS) Technique:

The ARINS technique utilizes the dressing of two unequal-intensity counter-propagating pulsed beams with differential nonlinear phases, which occurs upon traversing the sample. This difference in phase manifests itself in the intensity dependent transmission. Photodetection of the transmission of the ARINS yields spatially and temporally integrated response. Consequently, it is the pulse energy and not the instantaneous power that is detected. Figure 27.1 shows the schematic diagram of ARINS setup.

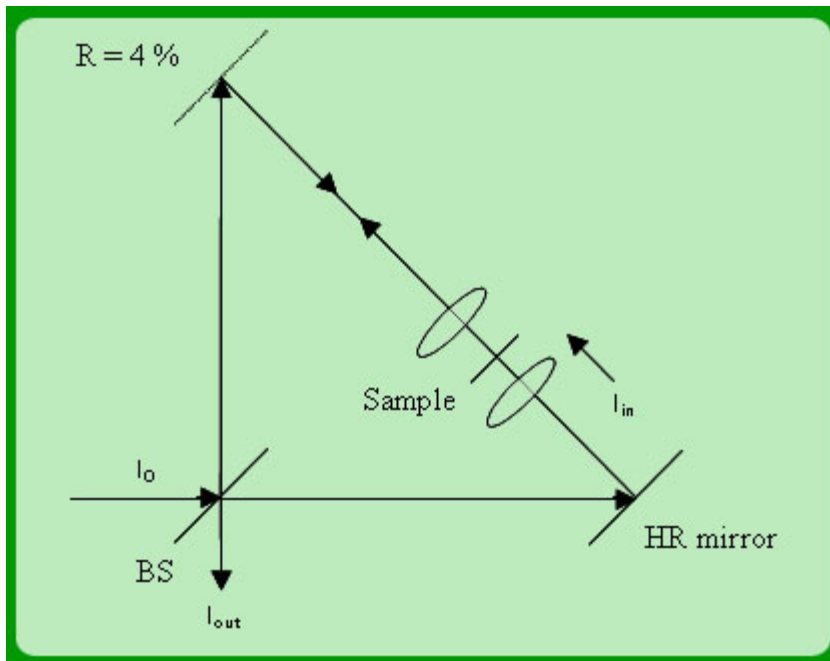


Figure 27.1: Schematic diagram of ARINS setup.

A 50-50 beam splitter divides the incoming pulsed beam into two counter-propagating pulses having a  $\pi$  phase difference. The pulses propagating in the clockwise (CW) direction are reflected by an uncoated flat with  $6^\circ$  wedged rear surface while that propagating in the counterclockwise (CCW) direction are reflected by a high reflectivity mirror. Thus this technique is based on the dressing of two unequal intensity counter-propagating pulses. These two pulses again recombine at the beam splitter to yield

ARINS transmission,  $|E_{out}|^2 \propto |E_{cw} + E_{ccw}|^2$ , where  $E_{cw}$  and  $E_{ccw}$  are the optical fields travelling in clockwise and counterclockwise directions respectively. Inside the ring there is a unit magnification telescope comprising of a pair of identical lenses in 2f configuration. One of the lenses focuses the pulses into the sample placed at its focus while the other one re-collimates them. The two counter propagating fields traversing the ring acquire linear as well as intensity dependent nonlinear phase shifts (if the sample exhibits nonlinear response). Since both the fields traverse the same optical path through the ring and encounter the same interactions with the optical elements, linear interactions would affect their amplitude and phase in identical manner. For an exactly 50% beam splitter, in the absence of any nonlinear interactions, the two returning fields will be having the same amplitude and phase difference between them and consequently they will interfere destructively at the beam splitter to yield zero transmission. In this 'balanced' condition all the input power is reflected along the incident direction. Measurement against this dark background provides the basis for the improved sensitivity essential for measuring relatively weak signals. Any small deviation from the ideal splitting ratio ( $\delta$ ) results in a leakage from the ARINS and is responsible for the background that limits the sensitivity of the measurement.

If the sample under investigation exhibits nonlinear response, then the two unequal intensity counter propagating pulses will undergo different phase changes after passing through the sample. Their superposition on the beam splitter will result in the intensity dependent transmission of the ARINS, which is related to the nonlinear response of the sample. However, differential dressing of the two counter propagating pulses with nonlinear phases is possible only when the two do not interact simultaneously in the sample. Else, the same nonlinear phase will be impressed on both by the cross-action phenomenon. Therefore, we prevent the temporal overlap of the two pulses in the sample by spatially offsetting it with respect to the center of the ARINS in such a way that the intense beam (i.e. CCW beam in our geometry) should reach the sample first in order to initiate nonlinear processes. The time difference between the arrivals of the two pulses ( $\Delta\tau_{off}$ ) determines the nature of the nonlinear optical process that can be studied depending on its response time. Nonlinear processes with decay time longer than  $\Delta\tau_{off}$  do not contribute to the intensity dependent transmission of the ARINS as both the pulses will be affected simultaneously in that case. The delay window thus acts as an ultrafast gate. Hence this technique has the unique ability to filter the nonresonant electronic contributions from integrating (or slow, e.g. thermal) nonlinearities or those arising from long lived (resonant) states and makes it ideal for time-resolved studies and ultrafast gating. If the arrival times of the two pulses in the sample were reversed, one would measure the unfiltered response.

### Theoretical Formalism:

Let us consider a collimated, spatially and temporally Gaussian pulse with electric field amplitude  $E_0$ , incident on the beam splitter, where it splits into two counter-propagating beams with electric fields  $E_{cw}$  and  $E_{ccw}$ . The general form for the electric field of a Gaussian beam is given by

$$E(Z, r, t) = E_0 \frac{w_0}{w(Z)} \exp\left[\frac{-r^2}{w^2(Z)}\right] \exp[i\phi(Z)] F(t) \quad (27.1)$$

where  $Z$  is the distance of propagation,  $r$  is the transverse coordinate,  $w_0$  is the beam waist ( $Z = 0$ ),  $\phi(Z) = \exp\left[i\left(kZ - \tan^{-1}(Z/Z_0)\right)\right]$  is the phase of the Gaussian beam (with pulse duration  $\tau_p$  and wave vector  $k$ ) and the function  $F(t) = \exp\left[(-2\ln 2)t^2/\tau_p^2\right]$  gives the temporal variation of the pulse.

The spot size at a distance  $Z$  is  $w^2(Z) = w_0^2 \left[1 + (Z^2/Z_0^2)\right]$ , where  $Z_0$  is the Rayleigh range. For simplicity, we assume that the Rayleigh range is large than the sample thickness (thin sample approximation). We ignore the slight difference in the spot sizes for the CW and CCW beams at the lenses that arises because the sample is offset from the center of the ARINS. The intensity ratios in CW and CCW directions are  $(1/2 - \delta)$  and  $(1/2 + \delta)$  respectively, where, as said earlier,  $\delta$  represents the small deviation from the ideal splitting ratio of the beam splitter.

The electric field at the incident face for the two counter propagating beams can then be expressed as

$$E_{cw} = -\sqrt{1/2 - \delta} E_0 \exp\left(-r^2/w_0^2\right) F(t) \sqrt{R} \quad (27.2)$$

$$E_{cw} = \sqrt{1/2 + \delta} E_0 \exp(-r^2/w_0^2) F(t) \quad (27.3)$$

where  $R$  is the reflectivity of the uncoated flat. For the weaker CW beam,  $\alpha(I) = \alpha$ , the coefficient of linear absorption and  $n(I) = n_0$ , the linear refractive index. However, for the stronger CCW beam,  $\alpha(I) = \alpha + \beta I$  and  $n(I) = n_0 + n_2 I$ , where  $\beta$  is the effective nonlinear absorption coefficient and  $n_2$  is the nonlinear index of refraction. The electric field of a pulsed Gaussian beam at the exit face of the sample (thickness =  $L$ ) with a nonlinear absorption and nonlinear refractive index is

$$E_{exit}(r, t) = \frac{E_0(r, t)}{\sqrt{1+q}} \exp(-\alpha L/2) \exp(-ik_0 L) \exp(-ik_2 \ln(1+q)/\beta) \quad (27.4)$$

where  $E_0(r, t)$  is the incident electric field,  $q = \beta I_{in} L_{eff}$ ,  $L_{eff} = [1 - \exp(-\alpha L)]/\alpha$  is the effective length of the sample,  $I_0$  is the intensity incident on the beam splitter,  $I_{in} = (1/2 + \delta) K' I_0 = K I_0$  is the intensity incident on the sample,  $K'$  is a constant ( $< 1$ ) accounting for the reflection losses at the sample and lens surfaces while  $K = (1/2 + \delta) K'$  is another constant.

Using equations (27.1), (27.2) and (27.3), the electric field of the two counter propagating beams at the corresponding exit faces of the sample can be written as

$$E_{cw}^{exit} = -\sqrt{(1/2 - \delta)} E_0 \exp\left(\frac{-r^2}{w_0^2}\right) \exp(-\alpha L/2) \exp(-ik_0 L) F(t) \sqrt{R} \quad (27.5)$$

$$E_{ccw}^{exit} = \sqrt{(1/2 + \delta)} \frac{E_0}{\sqrt{1+q}} \exp\left(\frac{-r^2}{w_0^2}\right) \exp(-\alpha L/2) \exp(-ik_0 L) \exp(-ik_2 I_{in} L_{eff}) F(t) \quad (27.6)$$

where  $q \ll 1$  and  $\ln(1+q) \approx q$ . When the two beams arrive again at the beam splitter after one trip round the ring, the electric fields in the transmission branch are given by multiplying equations (27.5) and (27.6) by the respective splitting ratios

$$E_{cw}^t = -(1/2 - \delta) E_0 \frac{w_0}{w(Z)} \exp\left(\frac{-r^2}{w^2(Z)}\right) \exp(-\alpha L/2) \exp(-ik_0 L) \exp[i\phi(Z)] F(t) \sqrt{R} \quad (27.7)$$

$$E_{ccw}^t = (1/2 + \delta) \frac{E_0}{\sqrt{1+q}} \frac{w_0}{w(Z)} \exp\left(\frac{-r^2}{w^2(Z)}\right) \exp(-\alpha L/2) \exp(-ik_0 L) \exp[i\phi(Z)] \times \exp(-ik_2 I_{in} L_{eff}) F(t) \sqrt{R} \quad (27.8)$$

where  $w(Z)$  is the spot size at the beam splitter after one round trip. The values of  $w(Z)$  and  $w_0$  can be determined experimentally. The ARINS leakage is given by

$$|E_{out}(r, t)|^2 = |E_{cw}^t(r, t) + E_{ccw}^t(r, t)|^2 \quad (27.9)$$

Substituting equations (27.7) and (27.8) in Eq. (27.9), we get

$$|E_{out}|^2 = \left[ \frac{w_0^2}{w^2(Z)} \exp(-\alpha L) \exp\left(\frac{-2r^2}{w^2(Z)}\right) F^2(t) \right] \times \left[ \left(\frac{1}{2} - \delta\right)^2 + \frac{(1/2 + \delta)^2}{1+q} + \left(2\delta^2 - \frac{1}{2}\right) \cos(k_2 I_{in} L_{eff}) \right] |E_0|^2 R \quad (27.10)$$

For  $q \ll 1$ , equation (27.10) becomes

$$|E_{out}|^2 = \left[ \frac{w_0^2}{w^2(Z)} \exp(-\alpha L) \exp\left(\frac{-2r^2}{w^2(Z)}\right) F^2(t) \right] \times \left[ 4\delta^2 + \beta I_{in} L \delta + \left[ \left(\frac{\beta I_{in}}{4}\right)^2 + \left(\frac{kn_2 I_{in}}{2}\right)^2 \right] L^2 \right] |E_0|^2 R \quad (27.11)$$

The relevant measured quantity in this experiment is the transmitted pulse energy:

$$W = 2nc\epsilon_0 \int_{-\infty}^{\infty} \int_0^{\infty} E_{out}^2 2\pi r dr dt \quad (27.12)$$

Using equation (27.11), and carrying out the integration, we get

$$W = 2nc\epsilon_0 \left( \frac{\pi\sqrt{\pi}w^2(Z)R\tau I_0 \exp(-\alpha L)}{2\sqrt{\ln 2}} \right) \times \left[ 4\delta^2 + \frac{\beta L \delta I_{in}}{2\sqrt{2}} + \left[ \left(\frac{\beta}{4}\right)^2 + \left(\frac{kn_2}{2}\right)^2 \right] \frac{L^2 I_{in}^2}{3\sqrt{3}} \right] \quad (27.13)$$

Defining

$$I_{out} = 2(\ln 2)^{1/2} W / \pi^{3/2} \tau w^2(Z)$$

which has the dimensions of intensity, equation (27.13) can now be expressed as

$$I_{out} = 2nc\epsilon_0 R I_0 \exp(-\alpha L) \times \left[ 4\delta^2 + \frac{\beta L \delta I_{in}}{2\sqrt{2}} + \left[ \left(\frac{\beta}{4}\right)^2 + \left(\frac{kn_2}{2}\right)^2 \right] \frac{L^2 I_{in}^2}{3\sqrt{3}} \right] \quad (27.14)$$

Using  $I_{in} = KI_0$  as explained earlier, we get

$$I_{out} = 2nc\epsilon_0 R \exp(-\alpha L) \times \left[ 4\delta^2 I_0 + \frac{\beta L \delta K}{2\sqrt{2}} I_0^2 + \left[ \left(\frac{\beta}{4}\right)^2 + \left(\frac{kn_2}{2}\right)^2 \right] \frac{L^2 K^2}{3\sqrt{3}} I_0^3 \right] \quad (27.15)$$

For low intensity levels ( $q \ll 1$ ), the ARINS transmission  $I_{out}$  as a function of  $I_{in}$  is a cubic polynomial.

The coefficients are directly related to the coefficient of nonlinear refractive index,  $n_2$  and to the coefficient of nonlinear absorption,  $\beta$  (which may be due to saturation of absorption or two-photon absorption or excited-state absorption). As stated earlier,  $\delta$  is related to the linear leakage and limits the sensitivity. For  $\delta = 0$ ,  $I_{out} \propto I_{in}^3$ , in which case the simultaneous evaluation of  $n_2$  and  $\beta$  becomes difficult. However, a small nonzero  $\delta$  makes the analysis much simpler as each term of the polynomial can be evaluated directly. If  $\delta > 0$  and the sample exhibits two-photon absorption or excited-state absorption (i.e.  $\beta > 0$ ), then all the coefficients of the polynomial become positive and  $I_{out}$  will be a continuously increasing function of  $I_{in}$ . On the other hand, if the sample exhibits saturation of absorption (i.e.  $\beta < 0$ ) then the sign of  $\beta$  opposes the increase in nonlinear leakage. In this case, the quadratic term in the polynomial becomes negative and  $I_{out}$  shows saturation at a relatively low intensity, where the effect of  $\delta$  is cancelled by the saturable absorption coefficient,  $\beta$ . However, at higher intensities, the cubic term becomes dominant and  $I_{out}$  starts increasing continuously. For  $\delta < 0$ , the curvature of ARINS leakage as a function of  $I_{in}$  is reversed for the two processes. Thus, by carefully choosing  $\delta$ , both the origin of nonlinearity and the values of nonlinear coefficients can be conveniently obtained.

### ARINS simulations:

The calculated ARINS transmission using equation (27.15) with typical values of  $n_2 = 1.8 \times 10^{-6} \text{ cm}^2 / \text{GW}$  and  $\beta = 0.378 \text{ cm} / \text{GW}$  by choosing different values of  $\delta$  is shown in figure 27.2.

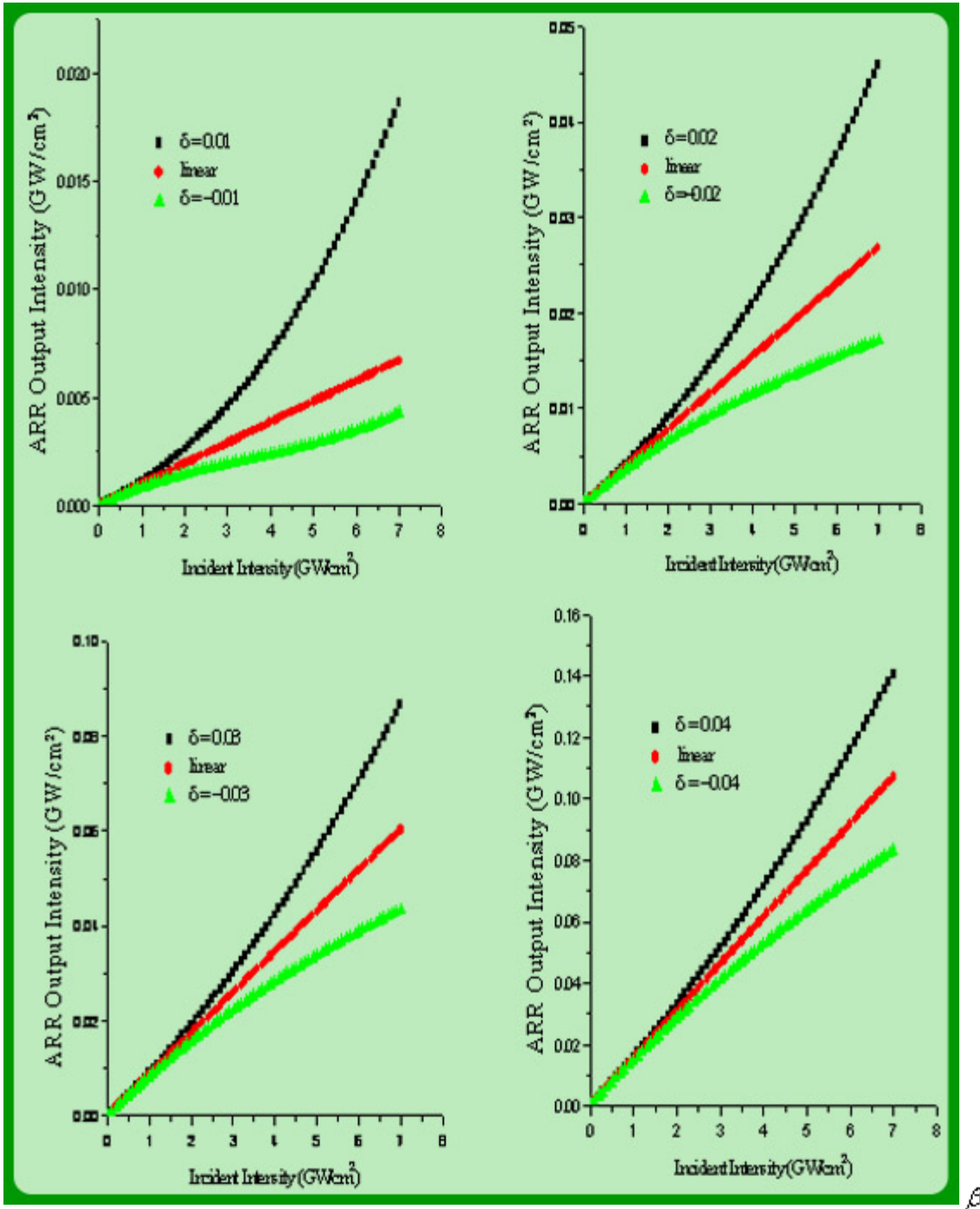


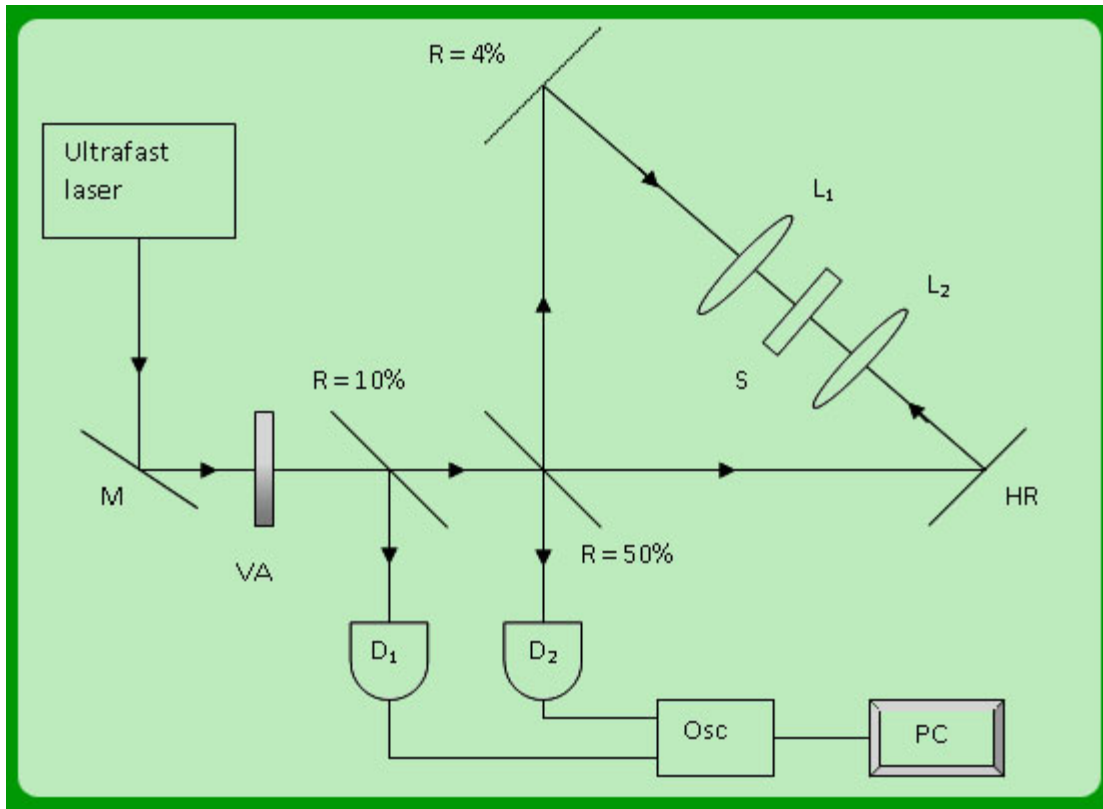
Figure 27.2: ARINS simulation for (a)  $\delta = \pm 0.01$ , (b)  $\delta = \pm 0.02$ , (c)  $\delta = \pm 0.03$  and (d)  $\delta = \pm 0.04$ .

The above simulation shows that the ARINS leakage is very sensitive to both sign and magnitude of  $\delta$ . In fact, it is the combination of both  $\delta$  and  $\beta$  which determines the sign and magnitude of the quadratic term in equation (27.15). If the sample exhibits nonlinear absorption (not saturation) then  $\beta > 0$ . In this case, for  $\delta > 0$ , all the coefficients of the polynomial will be positive and  $I_{\text{out}}$  will be a continuously increasing function of  $I_{\text{in}}$ . However, if the sample exhibits saturation of absorption ( $\beta < 0$ ), then the quadratic term in equation (27.15) becomes negative and  $I_{\text{out}}$  shows saturation at a relatively low

intensity where the effect of  $\delta$  is cancelled by the saturable absorption. Thus the curvature experiences a point of inflexion in this case. At higher intensities, the cubic term in equation (27.15) dominates and therefore  $I_{out}$  increases continuously. For  $\delta < 0$ , the curvature of ARINS leakage as a function of  $I_{in}$  is reversed for the two processes.

### Experimental setup:

Figure 27.3 shows a typical ARINS setup. The pulsed laser beam from an ultrafast laser is first attenuated by a variable attenuator such as a neutral density filter mounted on a motorized rotational stage. A small part of the incoming attenuated beam is reflected by a thin glass plate having reflectivity  $R = 10\%$  and is measured by a calibrated reference photodiode ( $D_1$ ) to give the incident power. The transmitted signal of ARR is measured by another calibrated photodiode ( $D_2$ ). A lens is used to couple the leakage signal to the photodiode to ensure the coupling of entire transmitted beam. The signals from the two photodiodes are acquired by the data acquisition system.



**Figure 27.3: ARINS experimental setup.** M- mirror, L1, L2 – lenses, HR – high reflectivity mirror, S – sample, VA – variable attenuator D1, D2- detectors, Osc – digital oscilloscope, PC – microcomputer.

### Advantages and disadvantages of ARINS:

- Since both beams travel the same path inside the ring, it is immune to any external disturbances
- Extreme sensitivity due to  $\sim 10 \mu\text{rad}$  phase change detection capability. Hence, suited for measurement of nonlinearity of thin films ( $< 100\text{nm}$  thick film demonstrated.)
- Has unique capability of discriminating against different nonlinear optical processes based on their response times.

### disadvantages

- Does not provide the sign of nonlinearity. It has to be supplemented by other technique like Z-scan to get the sign of  $\beta$  and  $n_2$ .
- Since this technique is based on the time difference between the arrivals of two pulses at the sample, it can't be used for cw beam.

**Sources of errors and precautions:**

- a. The sensitivity of this technique depends on the value of  $\delta$ . Too large value of  $\delta$  brings down the sensitivity of this technique considerably. Since  $\delta = 0$  also makes it impossible to separate the values of  $n_2$  and  $\chi$ , care should be taken to maintain the value of  $\delta$  at least around 0.02 - 0.04.

**References:**

1. P. Vasa, B. P. Singh, P. Taneja and P. Ayyub, Opt. commun. **233**, 297(2004)

**Recap****In this lecture you have learnt the following**

- Theory of ARINS technique for the measurement of third order nonlinear susceptibility  $\chi^{(3)}$ .
- Corresponding experimental details.
- sources of errors and precautions and
- Its merits and demerits.