COMMENTS ON THE PULSE WIDTH MEASUREMENT WITH TWO-PHOTON EXCITATION OF FLUORESCENCE

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The evaluation of the pulse duration by this method requires caution, because a very similar fluorescence structure is obtained from the radiation of an ideally mode locked laser and of a free-running laser with the same oscillating bandwith.

Giordmaine et al. [1] have shown that the duration of laser pulses in the picosecond range can be obtained from photographic records of fluorescence by two-photon excitation. The purpose of the present note is to show that the evaluation of pulse duration by this method requires caution. In particular the proper interpretation of the data depends strongly on the contrast ratio in the photographic record.

In qunatum theory the rate of two-photon absorption is described by the transition matrix element A, symbolically written as

$$A = C\langle a^+ a^+ a\, a\rangle , \qquad (1)$$

where C is a collection of constants, and a^+ and a are the creation and annihilation operators of the light field. Under the experimental conditions of ref. (1),

$$a^+ = \tfrac{1}{2}(a^+(t) + a^+(t+\tau)) \qquad (2a)$$

$$a = \tfrac{1}{2}(a(t) + a(t+\tau)) , \qquad (2b)$$

where t is time and $\tau = 2nx/c$ the relative delay between parts of the light wave propagating in opposite directions, x is the distance of a point under consideration from the point of equal delay (which may be a mirror), n is the index of refraction for the laser frequency, and c the velocity of light. The photographic exposure essentially records the value of A which is time averaged through the photographic process and spatially averaged over approximately the optical wavelength due to the limited resolution of optical imaging.

Thus it is apparent that in the mentioned experiment by the two-photon absorption and fluorescence process one measures a correlation related to the class of second order correlation functions $G^{(2)}$ which were introduced by Glauber [2,3]. Similarly, the three-photon absorption and fluorescence experiment by Renzepis and Duguay [4] measures also a correlation related to Glaubers correlation functions. A well known measurement of a second order correlation function is the classical experiment by Hanbury Brown and Twiss [5]. A significant result of that experiment is that the correlation function exhibits a well-defined structure even though the incoming light is without apparent structure, i.e. it is thermal or "chaotic" light. For the same reasons the fluorescence experiment by two-photon absorption shows a structure even if the incoming light is without orderly structure. In particular, regions of higher contrast are found near $\tau = 0$ even with the light of free-running multimode lasers. In the evaluation of experimental exposures one then has the difficulty to decide whether the structure found in the photographic record is caused by free-running laser light, or by ideally mode locked laser light, or by an intermediate situation. A unique assignment is possible only if the contrast ratio is known with great accuracy.

As an illustration of this last point the results of two simple Fourier-type calculations are given. In the first case ideal mode locking is considered. N modes of equal amplitude are assumed and the frequency difference between adjacent modes is Ω. The computed fluorescence record shows a maximum relative contrast of 3 near $\tau = 0$ over a width of the order $\Delta\tau \approx (N\Omega)^{-1}$ whereas everywhere else it is. 1. In the second case a free-running laser is considered. Again N modes of equal amplitude are assumed with difference frequency Ω, however with random

phase relations between the modes. The fluorescence record in this case shows a relative contrast of 1.5 near $\tau = 0$ over the same width of $\Delta\tau \approx (N\Omega)^{-1}$ compared to unity everywhere else.

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ON THE "THREE PARAMETER" FORM OF PHENOMENOLOGICAL THEORY OF SUPERFLUIDITY

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The third coefficient in the expansion of the free energy of He II is shown to be undistinguishable from zero using experimental data present just to day.

The phenomenological theory of superfluidity [1] is based on the expansion of the free energy in terms of ρ_s and the subsequent eqation of equilibrium:

$$\frac{\hbar^2}{2m}\Delta f - \tfrac{1}{2}m(\mathbf{v}_s - \mathbf{v}_n)^2 f = \\ = -\alpha f + \beta f^3 + \gamma f^5 + \ldots, \quad (f^2 = \rho_s/m)\ . \tag{1}$$

Usually only the first two terms on the right-hand side of eq. (1) are used, $\alpha = \alpha_0(T_\lambda - T)^{4/3}$ and $\beta = \beta_0(T_\lambda - T)^{2/3}$ being empirically determined parameters [1,2]. Such a determination of the third parameter γ seems especially necessary because α and β both vanish at the λ-point.

There are two groups of experimental data from which α, β and γ can be determined. The first group concerns the bulk liquid at rest and involves precise measurements of ρ_s and the specific heat near T_λ: $\rho_s/\rho = A(T_\lambda - T)^{2/3}$ with $A = 1.44\,^{\circ}\mathrm{K}^{-2/3}$ [3], and $\Delta C = 5.20 \times 10^7$ erg/g °K [4]. These data determine the critical exponents of α, β and γ ($\frac{4}{3}, \frac{2}{3}$ and 0). In the "two parameter" form of the theory they determine completely α and β* [2]:

$$\alpha_0 = \frac{m\Delta C}{AT_\lambda} = 1.11 \times 10^{-16}\ \frac{\mathrm{erg}}{^{\circ}\mathrm{K}^{4/3}}, \\ \beta_0 = \frac{m\alpha_0}{A\rho} = 3.52 \times 10^{-39}\ \frac{\mathrm{erg\ cm^3}}{^{\circ}\mathrm{K}^{2/3}} \tag{2}$$

But in the "three parameter" form these data are sufficient only for expressing β_0 and γ in terms of α_0:

$$\beta_0 = \frac{4m\alpha_0}{A\rho}\left(1 - \frac{3m\Delta C}{4AT_\lambda\alpha_0}\right), \quad \gamma = \frac{3m^2\alpha_0}{A^2\rho^2}\left(\frac{m\Delta C}{AT_\lambda\alpha_0} - 1\right) \tag{3}$$

The second group of experimental data concerns restricted volumes of He II ($\Delta f \neq 0$) or the moving liquid and involves more rough measurements: the λ-point shift ΔT_λ in films and pores [e.g. 5] and the critical velocity v_{c2} for breakdown of superfluidity [6,7]. In the "two parameter" theory these data have to be used for verification of numerical values given in eqs. (2). It must be mentioned here that when $f \to 0$ (second order phase transition) the terms of eq. (1) involving β

* Eqs. (2) and (3) follow from the expression of ρ_s in terms of the solution f_0 of eq. (1) ($\Delta f_0 = |\mathbf{v}_s - \mathbf{v}_n| = 0$) and from the formula $\Delta c = (T_\lambda/\rho)\mathrm{d}^2/\mathrm{d}T^2(\alpha f_0^2 - \frac{1}{2}\beta f_0^4 - \frac{1}{3}\gamma f_0^6)$.