INTRODUCTION TO PRECISION LASER SPECTROSCOPY

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OUTLINE

Introduction to Precision Laser Spectroscopy

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The advent of the laser in 1960 revolutionized spectroscopy. The laser has now become a powerful tool of spectroscopy over a range extending from submillimeter wave to the vacuum ultraviolet region. Laser spectroscopy has achieved improvement in resolution, accuracy, and sensitivity many orders of magnitude compared with the traditional methods of spectroscopy. The awarding of Noble prize to Authur L. Schawlow[1] and Nicolaas Bloembergen[2] in 1981 proves the importance of this subfield of Physics.

In this talk, I will briefly review the ideas and methods of saturation and two-photon Doppler-free spectroscopy with application to atomic and molecular gases. Prof. Wing[3] will discuss these and other high resolution spectroscopic methods in more details and their applications in precision measurements and Prof. Leung[4] will talk about picosecond spectroscopy and its applications. For a complete overview of the field of precision laser spectroscopy, reader is referred to textbooks[5-7] and review articles[8,9].

I. General Features of the Laser

Principles, characteristics and properties of a variety of lasers are outside the scope of this talk, interested reader is referred to general textbooks[10-12] of laser. We will consider fundamental properties of the laser which are pertinent to laser spectroscopy.

A. Temporal Coherence and Monochromaticity

It is well known that the laser is an extremely monochromatic source of light, and its temporal coherence is very good. The laser is an optical analogy of microwave oscillator. A good laser shows small fluctuations in both intensity and frequency. Even a relatively noisy laser has a narrower spectral width than a sharp resonance of a low pressure gas. The resolving power of laser spectroscopy is therefore determined by the width of the spectral line of the sample under study rather than the linewidth of the laser output.

The resolution of a conventional spectrometer is, on the contrary, instrumentally limited. The resolving power of a grating spectrometer with its narrowest possible slit is ordinary expressed by the product of the order

of interference m and the number of grooves N. The resolution in wave number (cm^{-1}) $\delta\nu = \nu/mN$ can be approximated to $\delta\nu$ (in cm^{-1}) $\sim 1/L$, where L is the size of the grating. The resolution of Fourier transform spectroscopy may likewise be given by $\delta\nu \sim 1/D$, where D is the distance of translation of the mirror. Hence the high resolution in conventional spectroscopy is of the order of .01 cm^{-1}, corresponding to the practical limit in size of about 1 m. The resolving power of a good conventional infrared spectrometer is 10^4 - 10^5.

The Doppler broadening of the spectral line of atoms and molecules in a gas is about 10^{-6} times the frequency. Thus laser spectroscopy can easily attain a resolving of 10^6 and a resolved linewidth of the order of 100 MHz in the infrared region. Various techniques of laser spectroscopy having higher resolution than the Doppler width have been developed. Because the linewidth obtained in these methods is 10^{-3} -10^{-4} times the Doppler width, the laser must be stable to better than ±(10-100) kHz.

The quantum fluctuation $\delta\nu_{las}$ in frequency ν of laser oscillator is given by[13]

$$\delta\nu_{las} = [2\pi h\nu(\Delta\nu)^2/P]\, N_2/(N_2-N_1) \qquad (1)$$

where $\Delta\nu$ is approximated by the half resonance width of the optical resonator $\Delta\nu_{cav}$, N_1 is the lower state population and N_2 is the upper state population of the active medium. The power of stimulated emission in the optical resonantor is denoted by P, the output power being a fraction of P.

As a typical example of a gas laser at λ = 1 μm, or $\nu = 3\times10^{14}$ Hz, take $\Delta\nu = 3\times10^6$ Hz, P = 1 mW, and $N_2 = 2N_1$; one finds that the quantum noise of (1) gives a small value of $\delta\nu_{las}$ = 0.02 Hz.

A practical laser shows much larger frequency variation, because of instability of its optical resonator. The thermal expansion of the optical resonator by 10^{-8}, for example, gives rise to a decrease in laser frequency by 10^{-8}. Acoustic noises and mechanical disturbances are very often major sources of laser frequency fluctuations. It is this reason that good vibration isolation as well as good temperature control are required in lasers for high resolution spectroscopy.

B. Spatial Coherence

The laser exhibits good spatial coherence: light waves at different parts of a laser beam have definite phase relations with each other. The spatial coherence is perfect in the single-mode laser. The very small divergence of a laser beam manifests this property of spatial coherence.

The beam divergence of the laser output is close to the theoretical

diffraction limit of coherent waves passing through a finite aperture. The beam divergence $\delta\theta$ is approximately given by

$$\delta\theta = \lambda/D \tag{2}$$

where λ is the wavelength and D is the diameter of the beam. Diffraction of light having a constant amplitude over a circular aperture is not applicable to the laser beam. The amplitude of the laser light shows a Guassing distribution about the beam axis when the laser is operated on the lowest transverse mode. When the optical field across the beam is given by

$$E(r) = E_o \exp(-r^2/w^2) \tag{3}$$

with a constant phase (plane wave front) on the surface at the origin (i.e. at beam waist), the angular distribution at a distance becomes

$$E(\theta) = A \exp\left[-(\theta/\delta\theta)^2\right], \quad \delta\theta = \lambda/\pi w \tag{4}$$

The parameter ω is called the beam size, and $\delta\theta$ is the beam divergence.

The laser beam can be focused to a small spot. The spot size is given by $f\delta\theta$, where f is the focal length. Since laser has typical divergence of 10^{-3}-10^{-4} radian, the laser beam can be focused to a size of a few times the wavelength, depending on the F-number (f/D) of focusing.

This feature allows laser spectroscopy of high spatial resolution and concentration of light energy. Metallurgical, biological and other applications of laser spectroscopy with a spatial resolution of a few micrometers have been achieved. Concentration of the laser beam gives rise to the strong optical field, which exhibits a large variety of nonlinear optical effect.

C. Tuning and Modulation

The laser is now available at thousands of different wavelengths from the vacuum ultraviolet to the submillimeter. The frequency of each laser can be tuned within the rather narrow fluorescence linewidth arround the proper frequency by adjusting the resonant frequency of its optical resonator. Wider tuning may be obtained by tuning the frequency of the laser transition with an appropriate method or by widening the bandwidth of the active medium, while suppressing the multimode-multifrequency operation.

The former method of tuning has been materialized in Zeeman tuning of gas lasers[14], pressure tuning of semiconducto diode lasers[15], tempera-

ture tuning of solid-state lasers[16], etc.. The latter method is restricted to the high gain laser, because widening of the bandwidth results in the reduction in gain. An example of this method is the high-pressure molecular gas laser[17], in which overlapping of closely spaced vibrational-rotational lines at high pressure partly compensates the gain reduction with pressure. The dye laser has such a high gain and wide bandwidth that can be tunable over a frequency range of 1000-2000 cm^{-1} in the visible region[18].

Frequency scanning within the tuning range of the laser is useful in spectroscopy. One has to notice, however, that a wider frequency tuning generally results in a larger instability of frequency. If a laser is tuned 100 GHz with a voltage of 1000V, a ripple of $\pm$ 10 mV will produce a frequency jitter of $\pm$1 MHz. Besides, the rate of frequency scan must be several times lower than the value given by the resolution in frequency divided by the time constant of observation. The frequency modulation of the laser is sometimes utilized in spectroscopy, but the modulation frequency and the maximum frequency deviation must be lower than the linewidth of the sample.

The amplitude of the laser can also be modulated. The frequency of amplitude modulation by an internal modulator is limited by the laser bandwidth, and it is still very fast compared with the speed of electronic devices. Nanosecond pulses can be produced by most gas lasers and semiconductor lasers, and picosecond and even subpicosecond pulses have been produced by glass lasers and dye lasers. Time-resolved spectroscopy by using those fast pulses has opened a the field of picosecond spectroscopy that is useful for investigating fast relaxation, energy transfer, and chemical reactions[4].

D. Brightness

The efficiencies of most lasers are fairly low. The net total power emitted from a laser is not higher than that of many other source of comparable size. Considering the narrow beam divergence and the narrow linewidth of the laser, however, we find that the laser is incomparable in brightness. The energy density per unit frequency interval and per unit solid angle is many orders of magnitude higher than any thermal source. For instance, the effective brighness temperature of a small 1 mW gas laser with a spectral width of 1 Hz is 10^{20}°K! A rather small energy of 1 mJ in a laser pulse of 1 psec duration gives a peak power of 1 GW. When this power is focused to a spot of a few micrometers in diameter, the power density will be 10^{16} W/cm^2, which is high enough to decompose any matter into ions with its optical field of 2×10^9 V/cm.

Thus it is not surprising that laser radiation induced a wide variety of nonlinear optical effects. Spectroscopic behaviors of nonlinear effects are in many cases utilized to obtain higher resolution.

Next, I will give a brief outline of the main ideas and methods of nonlinear spectrometer used for atomic and molecular spectroscopy without Doppler broadening. The Doppler broadening effect of a spectral line in gas sample will be treated first.

II. Doppler Broadening

A moving particle —— either an atom or a molecule —— emits or absorbs radiation which is not exactly at the quantum transition frequency ω_o corresponding to the difference between the two energy levels E_1 and E_2, as determined by

$$\hbar\omega_o = E_2 - E_1 \tag{5}$$

where $\hbar$ is Planck's constant. Instead it has a slightly shifted frequency due to the Doppler effect. The spectral line shift of a single particle is dependent on the projection of the particle velocity $\vec{v}$ along the direction of observation (Fig. 1a). In a gas, the atoms and molecules move randomly giving a different Doppler shift for each particle. At thermal equilibrium, the projection of the particle velocity along any chosen direction is given by the Maxwell distribution:

$$W(v) = \frac{1}{\sqrt{\pi}\, v_o} \exp -(v/v_o)^2$$

$$v_o = (2kT/M)^{\frac{1}{2}} \tag{6}$$

which gives a symmetrical Gaussian curve (Fig. 1b). As a result the spectral line of the assembly of particles has a symmetrical profile with its center at the quantum transition frequency ω_o (Fig. 1c). The full width of half maximum (FWHM) $\Delta\omega_D$ of the spectral line due to the Doppler broadening effect is given by:

$$\Delta\omega_D = \frac{2\omega_o}{c} \left(2 \frac{kT}{M} \ell n\ 2\right)^{\frac{1}{2}} \tag{7}$$

$$= 7.163 \times 10^{-7} \left(\frac{T}{A}\right)^{\frac{1}{2}} \omega_o$$

where M and A denote the mass and atomic weight of a particle, and T is the temperature in degrees Kelvin.

If an atom or a molecule has several transitions so close that the spectral lines overlap due to Doppler broadening, conventional methods of linear optical spectroscopy can not resolve their structure. However, a number of methods have been developed to remove these limitations, for example, the atomic and molecular beam method first used in the 1930s and successfully applied subsequently. In the 1950s, microwave optical double

resonance, level crossing, quantum beats and other spectroscopic methods were developed and used mainly for measuring atomic level structure. The advent of tunable lasers has culminated in the discovery and wide application of nonlinear laser spectroscopy which is very effective for atomic and molecular transitions.

In this talk, I should limit myself to following two fundamentally different approaches used in nonlinear laser spectroscopy to eliminate Doppler broadeing:

1. Saturation spectroscopy which is based on the changes produced in the velocity distribution of atoms and molecules interacting with a beam of laser light[19].

2. Two-photon Doppler-free spectroscopy which is based on the simultaneous absorption of photons from two laser beams of similar frequency but propagating in opposite directions.[20]

Other approaches such as polarization spectroscopy, Ramsey fringe method, and trapped-particle spectroscopy will be described by Prof. Wing[3] in later lectures in this school.

III. Saturation Spectroscopy

A Doppler-broadened spectral line is, in essence, composed of a great number of much narrower absorption or emission lines due to particles having different velocities. Therefore Doppler broadening is often described as imhomogeneous broadening. Homogeneous broadening (width 2Γ) is the width of a spectral line which is not broadened by the Doppler effect. A propagating plane wave $\vec{E}\cos(\omega t-\vec{k}\cdot\vec{r})$ from a laser can interact only with particles located within the spectral range of the homogeneous width 2Γ at the resonance frequency $\omega = \omega_0 + \vec{k}\cdot\vec{v}$. In other words, the laser field interacts only with particles that have a certain velocity projection in the direction of propagating light wave:

$$|\omega - \omega_0 + \vec{k}.\vec{v}| \leq \Gamma . \qquad (8)$$

This field can therefore change the state of a small portion of particles and distinguish them clearly from the rest. When the laser intensity is sufficiently high, preferential excitation of particles with a particular velocity alters the equilibrium distribution of particle velocities at both transition levels (Fig. 2). A hole is formed in the velocity distribuion of particles in the lower level at velocities that satisfy the resonance condition Eq. (8). On the other hand, the velocity distribution of the upper level has an excess of particles with resonant velocities. The hole depth and peak height depend on the degree of absorption saturation by the laser beam. Thus, a intense laser beam changes the velocity

distributions of atoms or molecules on both levels. This brings about a distortion of Doppler-broadened absorption or emission lines.

Because some of the atoms or molecule have been excited to upper state, a hole — which is often called a Bennelt hole — arises in the Doppler-broadened line profile[21]. The width of the hole is determined by the homogeneous linewidth, which can be thousands of times smaller than the Doppler width.

A related phenomenon known as "Lamb dip" forms the basis for many experiments in saturation spectroscopy. Consider the interaction of a Doppler-broadened line with a standing lignt wave. It is precisely the field that is usually present inside a laser, and it can be considered as a superposition of two counter propagating waves of the same frequency (Fig. 3). In this case, each wave burns a hole in the velocity distribution, and since the two waves are directed toward each other the holes are symmetric about the center of the Doppler profile. In essence, the laser field extracts energy from two groups of particles with different velocities. When the laser frequency is tuned to the center of the Doppler profile both holes coincide. The standing wave then interacts with only one group of particles, producing a resonance dip in the output power of the laser at the center of the transition. This effect was first investigated by Lamb in his gas laser theory, and then confirmed experimentally by two independent groups at the MIT[22] and Yale University[23].

A modification of the saturation method, which led to wide use of the method, was proposed in 1967 at three different laboratories[24,25]. The method involves the addition of a low pressure gas resonant absorption cell inside the laser cavity. The absorption saturation in the standing wave gives rise to a narrow Lamb dip at the center of the Doppler-broaden absorption line. Total saturated amplification of the component medium inside the laser cacity results in a narrow peak in the output power of the laser, which is referred to as an inverted Lamb dip. This was first successful observed by Barger and Hall for the molecular absorption cell[26]

To obtain a narrow saturation resonance at the center of an absorption line, it is not necessary to use a standing light wave. It is sufficient to have one intense travelling wave saturating the absorption and a weak wave travelling in the opposite direction acting as a probe. Molecules whose velocity v_{res}, satisfies the resonance condition $\vec{k}\cdot\vec{v}_{res} = \omega - \omega_0$, are excited by the intense travelling wave. Since the probe wave has the same frequency but is opposite in direction, it interacts only with molecules having a velocity of equal magnitude but opposite in direction to those that interact with the strong saturating wave. If the frequencies ω and ω_0 coincide, the probe beam interacts with molecules which have already been reduced in substantial number by the saturating wave. As a result, the absorption of the probe beam reveals a resonance minimum with homogeneous width and at the exact enter of the Doppler broadening

line profile. Hänsch[27] and Bordé[28] independently introduced this method in 1970. The experimental scheme is shown in Fig. 4.

IV. Two-photon Spectroscopy

This method of nonlinear laser spectroscopy without Doppler broadening was proposed by Chebotaev and Co-workers.[20] Consider a two-quantum (or photon) atomic or molecular transition in the field of a standing wave of frequency ω (Fig. 5). For a particle moving with velocity v the effective travelling wave frequency is changed by $\pm \vec{k}\cdot\vec{v}$. The only particles that can absorb two photons from one travelling wave are those for which the value of $\vec{k}\cdot\vec{v}$ satisfies the condition of two-photon resonance. However, simultaneous absorption of two photons from counter-propagating waves is possible. In this situation, the condition for two-photon resonance is only that the doubled field frequency coincides with the frequency of the two-quantum transition, i.e., with the center of the Doppler-broadened line. In this type of resonance all particles, vegardless of velocity, participate in two-photon absorption, resulting in a sharp increase of the absorption signal. The line shape of such absorption is the sum of a wide Doppler profile, representing two-photon absorption from a unidirectional wave, and a narrow resonance, corresponding to two-photon absorption by all particles for which $2\omega = \omega_0$ (Fig. 5c). The amplitude of the resonance peak at the center of the line has a high contrast equal to the ratio of Doppler width to the homogeneous width. Narrow two-photon resonances were first observed in 1974 in studies of sodium atom transitions in the visible[29] and later in studies of CH_3F molecular vibrational transitions in the infrared.[30]

V. Conclusion

There is not enough room to discuss all of the precision laser spectroscopic methods. The discovery of narrow optical resonances induced by laser radiation, however, has formed the basis for superhigh-resolution nonlinear spectroscopy and precision spectroscopy. This is now one of the most promising trends in laser spectroscopy. Some laboratories are running and have already carried out experimentson measurements of fundamental constants by use of narrow nonlinear resonances. Narrow molecular resonances have made it possible to create optical oscillators with very high frequency stability; this and the direct measurement of optical frequency has already introduced a single joint quantum standard of time and length. The application of precision spectroscopy to fundamental measurements will be covered in Prof. Wing's talk.

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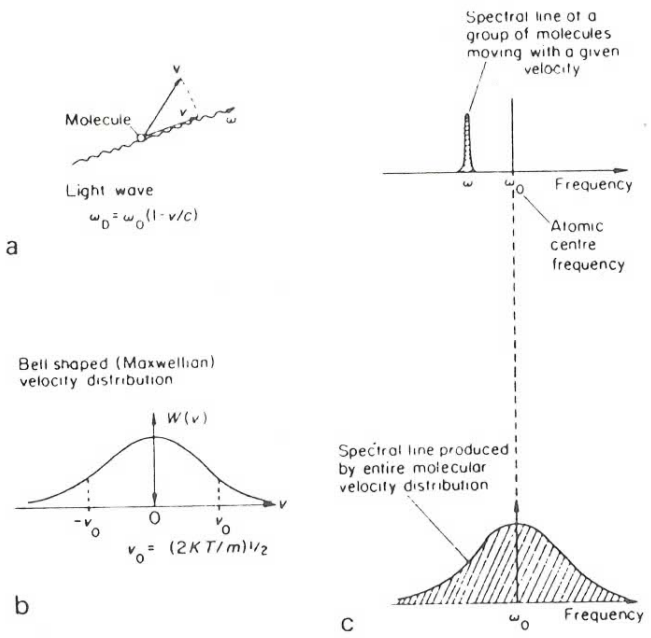

Fig. 1 Influence of the Doppler effect on the shape of a spectral line: a — Doppler shift due to a particle moving with velocity v. The Doppler effect shifts the emission or absorption frequency from v_0 to v; b — thermal distribution of particle velocities; c — corresponding spectral lines. The upper curve is the response of particles moving with velocity v as in a. The lower curve shows the responsed of particles over the entire thermal distribution.

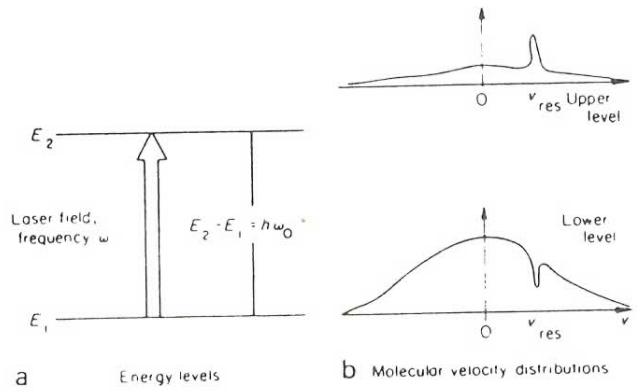

Fig. 2 Changes in the particle velocity distribution for two transition levels under the action of a laser travelling wave of frequency ω: a — Energy level diagram; b — velocity distribution projected onto the wave vector for particles in lower and upper transition levels (v_{res}: $(\omega-\omega_0)c/\omega_0$ projection of resonant velocity.

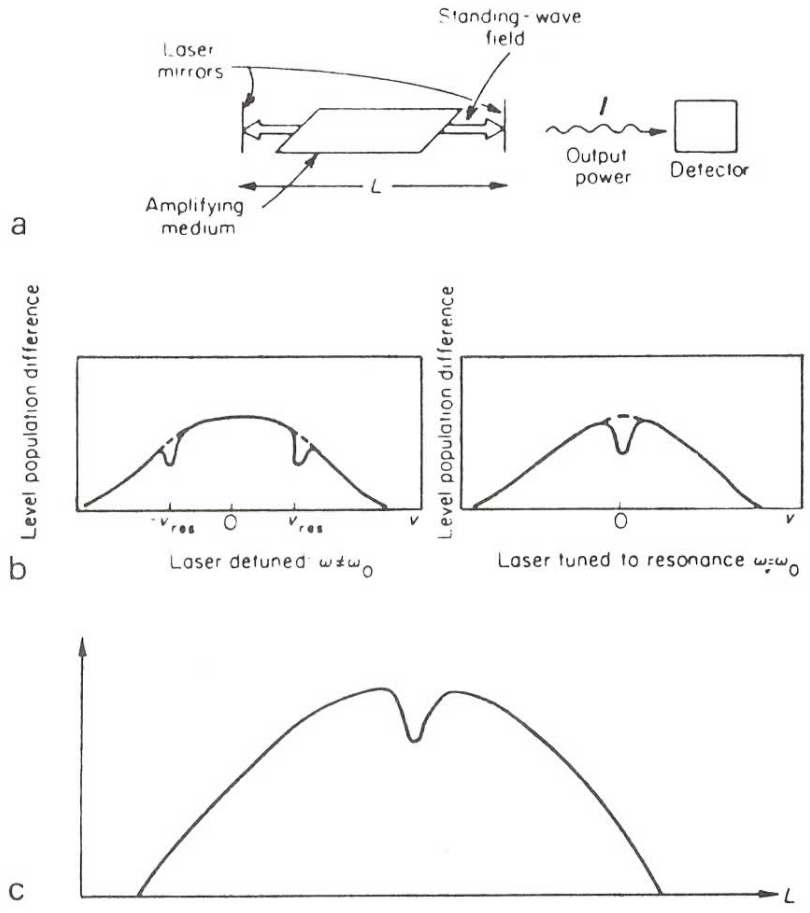

Fig. 3 Lamb-dip experiment: a — experimental arrangement: b — velocity distribution curves. Note that the saturated velocity groups can overlap only when the laser frequency is tuned to the centre of the Doppler profile: c — power output curve. Laser intensity is plotted as a function of separation between the laser mirrors. The narrow dip in the centre is the Lamb dip.

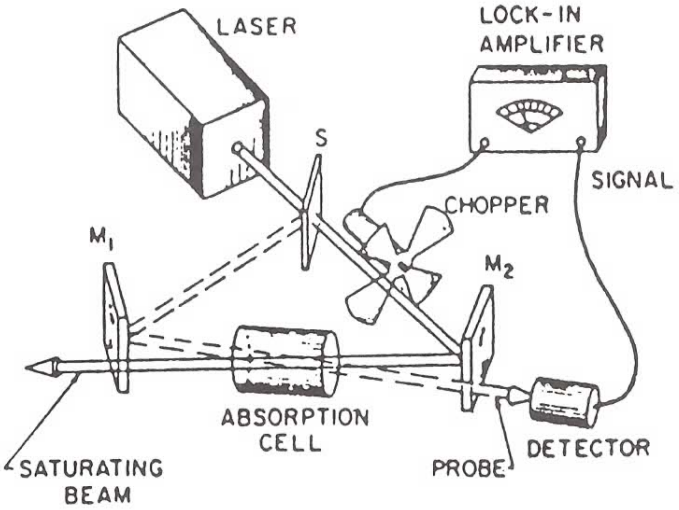

Fig. 4 Apparatus for Doppler-free spectroscopy by saturated absorption.

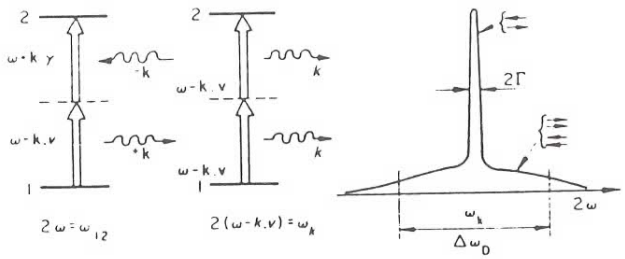

Fig. 5 Two-photon narrow resonance in a standing wave: a — compensation for the Doppler shift by simultaneous absorption of the photons from two travelling waves propagating in opposite directions: b — its absence when unidirectional photons are absorbed; c — the shape of narrow resonance in a two-photon absorption.