Possible use of an absorbing cell to analyze the spectrum of scattered light

V. A. Alekseev and T. L. Andreeva

P. N. Lebedev Physics Institute, USSR Academy of Sciences
(Submitted July 12, 1974)
ZhETF Pis. Red. **20**, No. 5, 316–318 (September 5, 1974)

The spectral composition of scattered light is presently being investigated by two methods. In the first method the spectral instrument is a Fabry-Perot etalon, and in the second the spectrum is registered by optical mixing methods. We consider in this paper still another possible method of investigating the spectral composition of scattered light.

The gist of the method is to use as the spectral instrument a cell containing absorbing gas saturated by a coherent laser field. In this case a narrow transmission peak, with a width on the order of the homogeneous linewidth γ, is produced against the background of the broad Doppler absorption contour $\Delta\omega_D$. [1] Resonances of this type have been recently used in nonlinear spectroscopy to resolve a fine structure that is masked by a broad Doppler line contour (see, e. g., [2]). The absorbing cell can be used for spectroscopy of scattered light in the following manner. Simultaneously with scattering the cell with the laser field of frequency ω_1, a scattered-light signal $I_p(\omega)$ is applied to the cell. The scattered light is excited by another laser of the same type, with frequency ω_0. The frequency ω_1 of the laser radiation passing through the cell is tuned away from the frequency of the laser that causes the scattering, so that $|\omega_0 - \omega_1| \leqslant \Delta\omega_D$. The scattered-light frequency region $|\omega - \omega_1| \leqslant \gamma$ passes easier through the cell than the remaining part of the spectrum. Therefore by tuning the frequency ω_1 away from the frequency of the laser that excites the scattering, within the limits of width $\Delta\omega_p$ of the scatter-light spectrum, it is possible to plot the entire scattering spectrum.

The total intensity of the scattered light passing

through the cell with the absorbing gas is

$$J(\omega_1) = \int_{-\infty}^{\infty} I_p(\omega) e^{-\kappa(\omega) L} \, d\omega, \tag{1}$$

where L is the thickness of the cell and $\kappa(\omega)$ is determined by the usual expression for the nonlinear absorption coefficient[1)]

$$\kappa(\omega) = \alpha(\omega) \left[1 - \beta^2 \frac{(2\gamma)^2}{(2\gamma)^2 + (\omega - \omega_1)^2} \right]. \tag{2}$$

Here $\alpha(\omega)$ is the Doppler contour of the nonlinear absorption, $\beta^2 = (dE/\hbar)^2(1/\gamma\Gamma)$ is the saturation parameter, d is the dipole moment of the transition of the absorbing gas, E is the intensity of the laser-wave field, and Γ is the longitudinal relaxation. Putting $\kappa(\omega)L \ll 1$ and $\gamma \ll \Delta\omega_p$, we obtain from (1) and (2)

$$J(\omega_1) = J_b + J_p(\omega_1) = \int_{-\infty}^{\infty} I_p(\omega)[1 - \alpha(\omega)L] d\omega + I_p(\omega_1)\alpha(\omega_1)\beta^2 4\pi\gamma L. \tag{3}$$

Thus, the signal of the photodetector that registers the scattered light passing through the cell contains two components, the first of which J_b determines the constant background, and the second $J_p(\omega_1)$ determines the useful signal. As seen from (3), the ratio of the useful signal to the background is $\alpha \approx 2(\gamma/\Delta\alpha_p)\alpha(\omega_1)L \cdot 2\pi\beta^2$. For example, at $\alpha(\omega_1)L \approx 0.3$ and $\beta^2 \approx 0.6$ we have $\alpha \approx 2(\gamma/\Delta\omega_p) = 0.1$. The value $2(\gamma/\Delta\omega_p) = 0.1$ assumed here corresponds to the possibility of recording ten points on the $I_p(\omega)$ curve.

The limiting structure width that can be resolved in the $I_p(\omega)$ spectrum by this method is equal to 2γ. Values $2\gamma \approx 2\times10^5$ Hz were obtained[4] in an absorbing cell containing, e.g., I_2, whose component $P(13)$ of the (43—0) band coincides with the $\lambda = 0.5145$ nm line of an argon laser. This value is approximately one tenth the limiting band width γ_{FP} of a Fabry-Perot etalon with base 150 cm.

We note that the considered method contains a harmful signal J_n due to spontaneous emission of the absorbing-gas molecules that have become excited in the course of saturation of the gas by the laser field. It is easy to show that the ratio of the useful signal J_p to the noise is

$$J_p/J_n = B_p(\lambda^2/\Delta\omega_p) = \bar{n}. \tag{4}$$

Here B_p (quanta/cm^2 sec sr) is the brightness of the scattered light, λ is the radiation wavelength, and $\bar{n}$ is the average number of photons per field oscillator in the scattered light. An approximately equal noise is produced in optical-mixing spectroscopy.[5] Typical values of n in the case of scattering in a liquid are $\bar{n} \approx 10^{-2}$. Such values of n correspond to excitation of a scattered spectrum of width $\Delta\omega_p \approx 10^6$ Hz in a liquid with an extinction coefficient $dR/dQ \approx 10^{-7}$ cm^{-1} sr^{-1} by a laser of 0.3 W power, with focusing on an area $10^4\lambda^2$. The ratio (4) is therefore small, and this gives rise to significant difficulties in the registration process. However, this noise can be eliminated in the method under consideration. To this end it suffices, e.g., to tune the frequency ω_1 to another line, which is connected with the work in transition by a common lower level. It is assumed here that the gas pressure is low enough for the collision-induced level-mixing frequency to be much lower than the rate of the radiative decay. In the case of I_2, the component $P(13)$ is located $\sim 3\times10^{10}$ Hz away from the nearest component $Q(13)$ or $R(13)$, which shares a common lower level with $P(13)$, so that this calls for a considerable change in the laser frequency. This change can be produced, e.g., by the electrooptical frequency-mixing method used in[6]. The spontaneous noise produced at the frequency of the component $P(13)$ can be filtered out in this case by an ordinary Fabry-Perot interferometer with $\gamma_{FP} \approx 10^9$ Hz. The working range of the indicated method is bounded by the values $\gamma \geq 10^5$ Hz and $\Delta\omega_D \approx 10^9$ Hz, i.e., $10^5 < \Delta\omega_p \leq 10^8$ Hz. It is important that it is precisely this range of $\Delta\omega_p$ which is the most difficult to investigate by the two other methods. The optical-mixing method and the Fabry-Perot interferometer method.

We note in conclusion that the usual methods of laser nonlinear spectroscopy call for the laser line to coincide with the line of the investigated transition, thus imposing serious limitations on the generality of this method. The method proposed by us is universal, since one absorbing cell suffices for the investigation of the spectra of the scattering of light from all substances.

We are grateful to B. Ya. Zel'dovich and I. I. Sobel'man for a discussion.

1)The resultant additional structure in $\kappa(\omega)$, which was observed in[3], is immaterial in this case.

[1]W. E. Lamb, J. Phys. Rev. 134, 1428 (1964).
[2]M. D. Levenson and A. L. Schawlow, Phys. Rev. 6, 10 (1972).
[3]E. V. Baklanov and V. P. Chebotaev, Zh. Eksp. Teor. Fiz. 61, 922 (1971) [Sov. Phys.-JETP 34, 490 (1972)].
[4]T. J. Rayan, D. G. Youmans, L. A. Hackel, and S. Ezekiel, Appl. Phys. Lett. 21, 320 (1972).
[5]G. B. Benedek, in: Matière et Rayannement, Presses Universitaires de France, Paris, 1969, p. 49.
[6]Z. Bay, G. G. Luther, and J. A. White, Phys. Rev. Lett. 29, 189 (1972).