In accordance with (8), the main laws of spatial correlation also hold for temporal correlation. This is true, in particular, for the two-scale character of the function $S_t(\tau)$. And the first temporal correlation scale τ_ρ, determined at the level at which the temporal coefficient of correlation $s_t = S_t(\tau)/S(0)$ declines by e times $(s_t = e^{-1})$, is characterized by the time of transfer of turbulent inhomogeneities with a velocity v through the smaller of the radii a_t and a_{eff} (more precisely, through a_0). The expression for τ_ρ follows from (6) and (8):

$$\tau_\rho = \frac{a_0}{v} \left(\frac{1}{S_\rho} \int_0^1 d\xi \, l^2(\xi x) \right)^3. \tag{9}$$

With $\beta_0 \gg 1$ the second temporal correlation scale τ_r, determined at the level $s_t = 0$, is characterized by the time of transfer of inhomogeneities with a velocity v through the external scale of turbulence. The expression for τ_r coincides with Eq. (9) for τ_ρ if S_ρ is replaced by S_r in (9).

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A PROBLEM IN COHERENT SPONTANEOUS MOLECULAR RADIATION

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We consider controlling the direction of coherent spontaneous molecular radiation and the application of this effect in spectroscopy.

Coherent spontaneous molecular radiation [1] (or "molecular ringing" radiation, "free decay" radiation) is employed in beam masers [2], and in microwave [3, 4] and optical [5, 6] spectrometers. Until recently, it was taken for granted that coherent spontaneous radiation has the same direction and frequency as the radiation exciting the molecules [1-5]. Recent research [7-9] has indicated that coherent radiation may also be observed in a direction opposite that of the exciting traveling wave, when the polarization induced by the traveling wave is spatially modulated in amplitude by a sufficiently strong standing wave of that same radiation. The polarized radiation thus induced is at a frequency different from that of the exciting field. These articles also discuss the mechanism of this phenomenon.

The goal of this note is to indicate several other methods of spatially modulating the polarization amplitude, as well as the possibility of controlling the direction of the coherent radiation of molecules excited by a field over a wider range by varying the polarization phase through, e.g., Stark impulses. The phenomena discussed in this paper may be employed in improving the performance of spectrometers which utilize coherent radiation from molecules excited by a field.

Let us first consider the mechanism for "backwards" polarized radiation in the presence of amplitude modulation, the means for producing such modulation, and also ways in which this phenomenon may be applied in spectrometers. It is well known (see [10], for example) that the occurrence of spatial harmonics of a polarization leads to radiation of that polarization in corresponding modes of the electromagnetic field. A specific case in our discussion is that in which the excitation of a polarization is selective according to molecular speed, being produced by a sub-Doppler line width in a gas, while the relaxation of that polarization proceeds fairly slowly. In this case, for example, excitation of a gas by radiation of one frequency can lead to occurrence of a response at several frequencies (corresponding to several spectral line components within the

Doppler profile), and it is possible to transport a polarization by means of moving molecules over a distance much larger than a wavelength, permitting control by a polarization of the excitation and radiation of molecules between spatially separated regions, and so forth.

Let us consider a homogeneous gas of two-level molecules with a transition frequency ω_0, irradiated by a plane wave

$$\mathscr{E} = \mathscr{E}_0 \cos(\omega_c t - kz). \tag{1}$$

The gas pressure is assumed to be low enough that the uniform line width $\Delta\omega_0$ is less than the Doppler width. It is then well known that the radiation field will effectively interact with molecules having a speed v_z lying in the interval $\delta v_z = \Delta\omega_0/k$ about a mean speed defined by the condition $kv_z = \omega_c - \omega_0$. Changing to a coordinate system which moves with the molecules $z' = z - v_z t$, and bearing in mind that in this coordinate system the frequency of the exciting field is $\omega'_c = \omega_c - kv_z$, we obtain in the moving coordinate system an expression for the polarization, applicable over a time $\tau \ll 2\pi/\Delta\omega_0$*:

$$P' = P'_1 \sin(\omega'_c t - kz') + P'_2 \cos(\omega'_c t - kz'), \tag{2}$$

where $P'_1 = (d^2\mathscr{E}_0/\hbar\Omega')N(v_z)\sin\Omega' t$, $P'_2 = -(2d^2\mathscr{E}_0/\hbar\,\Omega')(\Delta'/\Omega')N(v_z)\sin^2(\Omega' t/2)$, d is the dipole matrix element for the transition, $N(v_z)$ is the number of molecules effectively interacting with the field (taking into account the Maxwellian velocity distribution), $\Delta' = \omega'_c - \omega_0$, and $(\Omega')^2 = (\Delta')^2 + (d\mathscr{E}_0/\hbar)^2$. The field E radiated with polarization (2) is found, as usual, by solving the Maxwell equations and, obviously, polarization (2) excites a radiated wave traveling in the same direction:

$$E' = -2\pi kL[P'_1 \cos(\omega'_c t - kz') - P'_2 \sin(\omega'_c t - kz')] \tag{3}$$

(where L is the length of the medium along the z axis), which for a transition to the laboratory coordinate system (set up in z' and ω'_c) will have the same frequency as the radiation from the polarizing molecule, which leads to the usual result [1-5], correct for a homogeneous medium. The situation changes in the case of an inhomogeneous medium. Let us consider this through the example of a pulse spectrometer in which the molecular excitation and radiation processes are separated in time (also under sub-Doppler conditions). Let the molecules be excited by a field of the form (1), which is turned on at time τ. We will assume that the amplitude of the exciting field and the pulse duration are so chosen that the molecular excitation is close to optimal ("superluminal state"). After the cessation of the excitation pulse, we now eliminate molecular polarization in layers $\lambda/4$ long and also separated from each other by $\lambda/4$; that is, we assign final conditions such that $P'_M = P'\Pi(z')$, where $\Pi(z')$ is a form factor which meanders with a period of $\lambda/2$, and which may be approximated as $\Pi(z') \approx 0.5 + 0.5\cos 2kz'$. This sort of modulation of the polarization may be achieved by dephasing a portion of the molecules by an electric field pulse applied, as was done in [6], to a periodically spaced set of stretched wire loops, shifting the molecules to another energy level by irradiating them with a shorter wavelength source, and so forth. It is important to note that once such a spatial structure is created, it will be preserved (i.e., transported by molecules moving with the same speed v_z), and it is easy to demonstrate that the polarization P'_M will have a component of the form

$$P^+ = (1/2)[P'_1 \sin(\omega'_c t + kz') + P'_2 \cos(\omega'_c t + kz')], \tag{4}$$

corresponding to the excitation of a "backwards" wave of radiation. However, for "backwards" radiation, the transition to laboratory coordinates will give a frequency which is in fact different from the frequency of the exciting radiation:

$$E^+ = -2\pi kL\{P_1 \cos[(\omega_c - 2kv_z)t + kz] - P_2 \sin[(\omega_c - 2kv_z)t + kz]\}. \tag{5}$$

Specifically, the radiation frequency ω_r will differ from the frequency of the exciting radiation by twice the Doppler shift $\omega_r(E^+) = \omega_c - 2kv_z = 2\omega_0 - \omega_c$. When, for instance, two components with resonance frequencies ω_{01} and ω_{02} are present within the Doppler line, the radiation polarizing the molecules will actually interact with two groups of molecules, the "resonance" speeds of which are defined as $kv_z^i = \omega_c - \omega_{0i}$, and upon excitation of a single frequency and production of a single final spatial modulation of the polarization, "backwards" radiation will occur at the two frequencies $2\omega_{01} - \omega_c$ and $2\omega_{02} - \omega_c$, which differ by $\Delta\Omega = 2(\omega_{01} - \omega_{02})$, and which makes possible this sort of spectroscopy within a Doppler line. Under these circumstances, the resolving power will of course be determined by the uniform line width, which in turn is bounded by the time of flight of a region with size of order L, so that $\Delta\omega_0 \gtrsim 2\pi v_T/L$. In "spatial language," the condition for resolution corresponds to the separation of two structures of length L moving with different speeds, at a distance

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*A method for calculating the polarization is described, e.g., in [11].

of the order of the spatial period $\lambda/2$. Obtaining a response from the molecules, which is the useful signal from a spectrometer, in a different direction and at a different frequency from that of the exciting radiation, can of course facilitate the successful separation of the useful signal from the latter, which is one of the main problems in microwave spectrometers [12]. Making use of the "backwards" radiation can, for example, help get rid of the shf switching (see [4]) which is applied to protect the spectrometer receiver from overloading due to the high-power pulses of exciting radiation, and to trigger the receiver at the end of a pulse for reception of radiation from the excited molecules, which in [4] has the same direction as the exciting radiation. It is well known that making such switches with sufficiently high accuracy is difficult in the short millimeter and submillimeter bands. In continuous spectrometers with spatial separation of the molecular excitation and radiation regions, of the type found in [12], reception of the "backwards" radiation also allows for the easy separation of the useful signal from the spectral lines. "Amplitude modulation" of the polarization may in this case be accomplished with diaphragms of size $\lambda/4$, positioned with a period of $\lambda/2$ between the molecular excitation and radiation regions [13].

Control of the direction of coherent molecular radiation excited by a field may also be accomplished by phase modulation of the polarization. It is obvious that the radiation direction of the polarization induced by the source field is determined by the dependence of the phase of the induced polarization on the coordinates $P = P_0 \sin[\varphi(\omega, t, x, y, z)]$, which in the particular case $\varphi = \omega t - kz$ gives coincidence between the directions of the exciting wave (1) and the polarization wave. The phase dependence on the molecular polarization coordinates may be altered, for example, by applying a corresponding Stark pulse after the radiation polarizing the gas has acted. The Stark field changes the spacing between the molecular levels under consideration, and thus changes the frequency ω_0 by an amount $\Delta\omega$ as the pulse is acting. The total polarization phase is then $\varphi = \varphi_0 + \Delta\varphi$, where the phase increment will have the form $\Delta\varphi = \Delta\omega(x, y, z)\tau$, with τ being the duration of the Stark pulse. In principle, this then permits one to control the direction of the polarized radiation. Actually, for the case $\varphi_0 = \omega t - kz$, choosing the phase increment to be of the form $\Delta\varphi = 2kz$, we find that the radiation of the polarized gas occurs laterally, opposite the direction of propagation of the exciting radiation, and a phase increment of the form $\Delta\varphi = kz - kx$ leads to "sideways" radiation along the x axis, and so forth for any given direction.

The appropriate coordinate dependence of the phase increment may be produced, for example, by a definite configuration of Stark electrodes. If we assume that the levels experience a linear Stark effect $\omega = \omega_0 + \varkappa E$, then equating the phase increment, of the form $\Delta\varphi = \varkappa E(x, y, z)\tau$, to the proper law of variation (for instance, $\Delta\varphi = 2kz$), we obtain the coordinate dependence of E which, for example, for a rectangular pulse of the E field of duration τ has the form $E = 2kz/\varkappa\tau$, which then determines the proper electrode configuration. It is clearly possible to control the polarization phase in other ways. Controlling the direction of the radiation which in the useful spectrometer signal allows, as has already been mentioned, for additional possibilities for separating the useful signal from the interference background.

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