Correlation properties of resonantly scattering light

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The space-time properties of the intensity correlator for light scattered by resonant atoms are studied. It is shown that in the measurement of such correlators by the optical mixing technique an important contribution arises from the anomalous scattered-field amplitude correlator. The simplest way of observing this correlator is in the field of a standing wave by measuring the correlation of oppositely scattered beams. The spectral and polarization properties of such a correlator are studied. Its investigation is of interest for high-resolution spectroscopy since it may yield information on the natural line width under conditions of strong Doppler broadening. It should be emphasized that this is possible in an approximation linear in the intensity of the external field, in contrast to the laser-spectroscopy techniques, which are based on nonlinear effects.

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§1. INTRODUCTION

We consider in this paper the space-time correlation properties of scattered radiation. In experiments on light scattering one usually investigates the spectral function of the scattered radiation (the normal correlator of the field amplitudes). Yet a contribution to the correlator of the intensities of the external and scattered fields [see Eq. (1)] is made also by the anomalous correlator of the complex field amplitudes. There is no such correlator in thermal radiation sources, and the correlator is not equal to zero only in an external coherent field. We discuss below the conditions under which it would be possible to observe this correlator. Its investigation is of interest for high-resolution spectroscopy, since it can yield information on the natural linewidth under conditions of strong Doppler broadening. It is important to emphasize that this is possible even in an approximation linear in the external-field intensity.

The spatial structure of the scattered light is determined by the configuration of the pump field, variation of which makes it possible to separate and measure the ordinary or the anomalous correlator. These correlators are calculated below in an approximation linear in the density of the atoms of the medium; the medium is taken to be a gas of resonant atoms.

A nonzero anomalous correlator of the dipole moments in an external field was noted by Klysho.[1] Such a correlator appears also in the theory of resonant fluorescence of an atom, in which the method of correlation functions is used.[2,3] Certain properties of the anomalous correlator were discussed by us briefly earlier.[4]

§2. FORMULATION OF THE PROBLEM

The second-order correlator, which determines the counting rate of the pair coincidences, is given by[5]:

$$G(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)=\langle \hat{E}^{(-)}(\mathbf{r}_1, t_1)\hat{E}^{(-)}(\mathbf{r}_2, t_2) \times \hat{E}^{(+)}(\mathbf{r}_2, t_2)\hat{E}^{(+)}(\mathbf{r}_1, t_1)\rangle, \quad t_2>t_1. \tag{1}$$

The angle brackets denote averaging over the vacuum of the photon field and over the states of the medium; $\hat{E}^{(+)}(\mathbf{r}, t)$ is the positive-frequency part of the Heisenberg operator of the electromagnetic field $\hat{E}^{(-)}(\mathbf{r}, t)$ is the Hermitian adjoint of $\hat{E}^{(+)}(\mathbf{r}, t)$. We note that the sequence of the field operators in G is essential; it is determined by the sequence in which the photons are registered by detectors 1 and 2.

If the scattered field $\hat{E}^{(+)}(\mathbf{r}, t)$ is mixed at the detectors with the external field $E(\mathbf{r}, t)$, then the field operator consists in this case of three parts:

$$\hat{E}^{(+)}(\mathbf{r}, t)=E(\mathbf{r}, t)+\hat{E}_s^{(+)}(\mathbf{r}, t)+\hat{E}_0^{(+)}(\mathbf{r}, t),$$

$$E_0^{(+)}(\mathbf{r}, t)=\left(\frac{2\pi\hbar\omega_0}{v}\right)^{1/2}\sum_{\mathbf{k}} a_{\mathbf{k}}\exp(-i\omega_k t+i\mathbf{kr}),$$

where $E_0^{(+)}(\mathbf{r}, t)$ is the operator of the vacuum field and $a_{\mathbf{k}}$ is the photon annihilation operator.

In the lowest approximation in the scattered field we have the following expression for G (we leave out the constant component $\sim|E|^4$):

$$G(1, 2)=[g_+(2, 1)E^*(1)E(2)+(g_-(2, 1)+g_0(2, 1))E^*(1)E^*(2)+\text{c.c.}] +g_+(2, 2)|E(1)|^2+g_+(1, 1)|E(2)|^2. \tag{2}$$

We have introduced here the following correlation functions of the scattered and vacuum fields:

$$g_\pm(2,1)=\langle\hat{E}_s^{(\mp)}(2)\hat{E}_s^{(+)}(1)\rangle, \quad g_0(2,1)=\langle\hat{E}_0^{(+)}(2)\hat{E}_s^{(+)}(1)\rangle. \tag{3}$$

The correlator G contains thus three correlation functions. The autocorrelation function g_+ determines the radiation spectrum; it satisfies the condition $g_+^*(2,1) =g_+(1,2)$. There is no such condition for the functions g_0 and g_-, and therefore their spectral functions are complex. From the physical point of view the function g_+ describes the correlation between the acts of absorption and emission of field quanta. Accordingly, the function g_- describes the correlation of two absorption acts that are separated in time. It will be shown below that the only observable quantity is the sum of the functions $g=g_0+g_-$.

The external field is assumed to be classical and monochromatic with a frequency close to the frequency of the atomic transition ω_0:

$$E(\mathbf{r}, t)=E(\mathbf{r})e^{-i\omega t}, \quad \omega=\omega_0+\Delta, \quad |\Delta|\ll\omega_0.$$

The medium is assumed to be optically transparent, i.e.,

$$2\pi k_0 l \operatorname{Im}\chi(\Delta)\ll 1. \tag{4}$$

Here $\chi(\Delta)$ is the susceptibility of the medium to the pump field, l is the dimension of the interaction regime, and $k_0 = \omega_0/c$. It is implied that the condition (4) is satisfied also for the scattered field, so that it can be determined by perturbation theory with respect to the atom density n.

§3. CALCULATION OF THE CORRELATORS

We choose the origin inside the scattering medium. We then have in the Fraunhofer zone

$$g_\pm(\mathbf{r}_2, t_2; \mathbf{r}_1, t_1) = \frac{k_0^4}{r_1 r_2} \sum_i \left\langle \hat{p}_i^{(\mp)}\left(t_2' + \frac{\mathbf{n}_2\mathbf{r}_i(t_2')}{c}\right) \hat{p}_i^{(+)}\left(t_1' + \frac{\mathbf{n}_1\mathbf{r}_i(t_1')}{c}\right)\right\rangle,$$

$$g_0(\mathbf{r}_2, t_2; \mathbf{r}_1, t_1) = \frac{k_0^2}{r_1} \sum_i \left\langle \hat{E}_0^{(+)}(\mathbf{r}_2, t_2) \hat{p}_i^{(+)}\left(t_1' + \frac{\mathbf{n}_1\mathbf{r}_i(t_1')}{c}\right)\right\rangle; \tag{5}$$

$$t_{1,2}' = t_{1,2} - r_{1,2}/c.$$

Here $\hat{p}_i^{(+)}(t)$ is the positive-frequency part of the dipole-moment operator of an atom located at the point $\mathbf{r}_i(t) = \mathbf{r}_i(0) + \mathbf{v}_i t$ in the Heisenberg representation. By $\mathbf{n}_{1,2} = \mathbf{r}_{1,2}/r_{1,2}$ we denote the directions of the scattered rays. The summation is over the particles of the medium.

Going to the continuous limit, we introduce the operator of the dipole moment per unit phase space of the medium

$$\hat{p}^{(+)}(\mathbf{r}, \mathbf{v}, t) = \hat{p}(\mathbf{r}, \mathbf{v}, t) e^{-i\omega t}$$

and separate in it the factor that oscillates at the frequency of the external field:

$$\langle \hat{p}^{(\mp)}(\mathbf{r}, \mathbf{v}, t_2') \hat{p}^{(+)}(\mathbf{r}, \mathbf{v}, t_1') \rangle = d^2 \exp[-i\omega(t_1' \mp t_2')] F_{\pm-}(\mathbf{r}, \mathbf{v}, \tau), \qquad \tau = t_2' - t_1', \tag{6}$$

where d is the dipole moment of the transition. The correlation functions $F_{\pm-}$ depend slowly (compared with an exponential) on the difference between the times τ and satisfy the generalized Bloch equation (9).

In the case of narrow atomic resonances and small detunings, the following inequality holds

$$c/l \gg \gamma, \Delta, \tag{7}$$

and allows us to neglect the retardation in the slow oscillations of the dipole moment of the atom. In this approximation, the correlators (3) are represented as sums of products of spatial and temporal functions, with g_0 and g_- having equal spatial parts. This allows us to express the correlators of the scattered light in terms of the correlators of the medium $F_{\pm-}(\mathrm{r}, \mathrm{v}, \tau)$ in the following manner[1)]:

$$\begin{pmatrix} g_+ \\ g \end{pmatrix} = \frac{nd^2k_0^4}{r_1 r_2} \exp[-i\omega(t_1' \mp t_2')] \times \int d\mathbf{r}\, d\mathbf{v}\, f(\mathbf{v}) \exp\{-ik(\mathbf{n}_1 \mp \mathbf{n}_2)\mathbf{r} + ik\tau\mathbf{n}\mathbf{v}\} \begin{pmatrix} F_{+-}(\mathbf{r}, \mathbf{v}, \tau) \\ F_{--}(\mathbf{r}, \mathbf{v}, |\tau|) \end{pmatrix}; \tag{8}$$

$\mathrm{n} = \mathrm{n}_1$ at $\tau > 0$ and $\mathrm{n} = \mathrm{n}_2$ at $\tau < 0$. $f(\mathrm{v})$ denotes the atom-velocity distribution function, which is hereafter assumed to be Maxwellian. The fact that the sum of the correlators g_0 and g_- admits of such a representation is not obvious, and the next section will be devoted to a derivation of Eq. (8).

§4. EQUATION FOR THE CORRELATION FUNCTIONS

The correlation functions $F_{\pm-}(\mathbf{r}, \mathbf{v}, \tau)$ are determined from the generalized Bloch equations, which take for $\tau > 0$ the form

$$\left(\frac{d}{d\tau} + \nu_-\right) F_{--} = -iV(\mathbf{r}) F_{0-}, \quad \left(\frac{d}{d\tau} + \nu_+\right) F_{+-} = iV^*(\mathbf{r}) F_{0-},$$

$$\left(\frac{d}{d\tau} + \gamma\right) F_{0-} + \gamma p(\mathbf{r} - \mathbf{v}\tau) = 2i[V(\mathbf{r}) F_{+-} - V^*(\mathbf{r}) F_{--}];$$

$$\frac{d}{d\tau} = \frac{\partial}{\partial\tau} + \mathbf{v}\nabla, \quad p(\mathbf{r}, \mathbf{v}) = e^{i\omega t}\langle \hat{p}^{(+)}(\mathbf{r}, \mathbf{v}, t)\rangle. \tag{9}$$

Here $p(\mathbf{r}, \mathbf{v})$ is the average induced dipole moment of the atom located at the point $\mathbf{r}$ and having a velocity $\mathbf{v}$; $V(\mathbf{r}) = dE(\mathbf{r})/\hbar$ is the Rabi frequency; $\nu_\pm = \gamma/2 \pm i\Delta$. The width of the upper level is γ, and the lower is assumed to be the ground level.

Equations (9) should be solved with the following initial conditions (at $\tau = 0$):

$$F_{--}(\mathbf{r}, \mathbf{v}, 0) = 0; \quad F_{+-}(\mathbf{r}, \mathbf{v}, 0) = \tfrac{1}{2}(1 + q(\mathbf{r}, \mathbf{v})); \quad F_{0-}(\mathbf{r}, \mathbf{v}, 0) = -p(\mathbf{r}, \mathbf{v}), \tag{10}$$

where q is the level-population difference. Equations (9) and (10) are written for an arbitrary inhomogeneous field with allowance for the motion of the atoms. For immobile atoms, they coincide with the equations considered in Refs. 2 and 3. For $\tau < 0$ the equations for $F_{\pm-}$ are given in the Appendix.

To obtain equations for the correlator $\langle \hat{E}_0^{(+)}(2) \hat{p}^{(+)}(1)\rangle$, we write down the Bloch equation for the atomic operators of the dipole moment $\hat{p}^{(+)}(\mathbf{r}, \mathbf{v}, t)$ and of the population-difference $\hat{q}(\mathbf{r}, \mathbf{v}, t)$ (Refs. 6 and 7):

$$\left(\frac{d}{dt} + \nu_-\right) \hat{p} = -i\hat{q}(V(\mathbf{r}) + \hat{V}_0(\mathbf{r}, t)),$$

$$\left(\frac{d}{dt} + \gamma\right) \hat{q} + \gamma = 2i[\hat{p}^+(V(\mathbf{r}) + \hat{V}_0(\mathbf{r}, t)) - \mathrm{H.c.}]. \tag{11}$$

Here

$$\hat{V}_0(\mathbf{r}, t) = e^{i\omega t} d\hat{E}_0(\mathbf{r}, t)/\hbar$$

is the "slow" part of the zero-point oscillation operator. Equation (11) is obtained by substituting in the Bloch operator equation the electromagnetic-field operator expressed from Maxwell's equations in terms of the dipole-moment operator and the operator of the zero-point field oscillations. The sequence of the operators is important here: when (11) is averaged over the photon vacuum we obtain the usual Bloch equation.

Multiplying (11) from the left by $V_0(2)$ and introducing the correlation functions

$$P(2,1) = \langle \hat{V}_0(2) \hat{p}(1)\rangle, \quad R(2,1) = \langle \hat{V}_0(2) \hat{p}^{(+)}(1)\rangle, \quad Q(2,1) = \langle \hat{V}_0(2) \hat{q}(1)\rangle,$$

we obtain the following equations:

$$\left(\frac{d}{dt_1} + \nu_-\right) P = -iV(\mathbf{r}_1) Q,$$

$$\left(\frac{d}{dt_1} + \nu_+\right) R = iV^*(\mathbf{r}_1) Q + i\langle \hat{V}_0(2) \hat{V}_0^+(1)\rangle q(1), \tag{12}$$

$$\left(\frac{d}{dt_1} + \gamma\right) Q = 2i[RV(\mathbf{r}_1) - PV^*(\mathbf{r}_1) - \langle \hat{V}_0(2) \hat{V}_0^+(1)\rangle p(1)].$$

In the resonance approximation, the correlator of the zero-point oscillations is of the form

$$\langle \hat{V}_0(2) \hat{V}_0^+(1)\rangle = A\delta(t_2 - t_1), \quad A = \frac{\gamma \exp(ik|\mathbf{r}_2 - \mathbf{r}_1|)}{2ik|\mathbf{r}_2 - \mathbf{r}_1|}. \tag{13}$$

In the derivation of (12) we have carried out a simplest separation of the mean value of the three operators

$\langle \hat{V}_0(2)\hat{V}_0^{*}(1)\hat{q}(1)\rangle$. Such a separation is rigorous in the "white noise" approximation, to which the correlator (13) corresponds.

At $t_2 > t_1$, by virtue of the causality principle, P, R, and Q are equal to zero. We note that Eqs. (9) and (12) are of like structure and differ only in the right-hand sides and in the initial conditions.

Separating from P the purely spatial factor $A(P = AP_0)$, we can represent the correlator g_0 in the form $(\tau < 0)$

$$g_0 = \frac{nd^2k^4}{r_1 r_2}\exp[-i\omega(t_1' + t_2')]$$
$$\times \int d\mathbf{r}\,d\mathbf{v}\,f(\mathbf{v})\exp\{-ik(\mathbf{n}_1 + \mathbf{n}_2)\mathbf{r} + ik\mathbf{n}_2\mathbf{v}\tau\}P_0(\mathbf{r}, \mathbf{v}, \tau).$$

The correlator g_- is expressed in similar fashion in terms of $F_{--}(\mathbf{r}, \mathbf{v}, \tau)$. Using the equations for F_{--} at $\tau < 0$ [see the Appendix, Eq. (A.2)] and Eqs. (12), we readily see that the function $F_{--} + P_0$ satisfies at $\tau > 0$ the same equations and initial conditions as the function F_{--} at $\tau > 0$. Thus, the combination $F_{--} + P_0$ is an even function of τ and this allows us to represent the correlator g in the form (8).

§5. SPATIAL STRUCTURE OF THE CORRELATION FUNCTIONS

In an inhomogeneous pump field (for example, in a standing light wave) the functions $F_{+-}(\mathbf{r}, \mathbf{v}, \tau)$ constitute a set of spatial harmonics

$$F_{\pm -}(\mathbf{r}, \mathbf{v}, \tau) = \sum_{\mathbf{q}} e^{2i\mathbf{q}\mathbf{r}} F_{+-}(\mathbf{q}, \mathbf{v}, \tau). \tag{14}$$

Since the functions F_{+-} depend quadratically on the external field, the Fourier series contains only even harmonics. Using this expansion, we obtain the following equations for the correlators:

$$\begin{pmatrix} g_+(\mathbf{r}_2 t_2; \mathbf{r}_1 t_1) \\ g(\mathbf{r}_2 t_2; \mathbf{r}_1 t_1) \end{pmatrix} = \frac{(2\pi)^3 nd^2k_0^4}{r_1 r_2}\exp[-i\omega(t_1' \mp t_2')]$$
$$\times \sum_{\mathbf{q}} \delta(2\mathbf{q} \pm \mathbf{n}_2 k_0 - \mathbf{n}_1 k_0)\int d\mathbf{v}\,f(\mathbf{v})\,e^{i\mathbf{n}\mathbf{v}\tau k}\begin{pmatrix} F_{+-}(\mathbf{q}, \mathbf{v}, \tau) \\ F_{--}(\mathbf{q}, \mathbf{v}, |\tau|) \end{pmatrix}. \tag{15}$$

If we substitute the correlation functions (15) in the equation for $G(1,2)$, then the rapid oscillations at the optical frequency vanish, and only the dependence on the slow time τ remains.

The spatial structure of the correlator is determined by δ functions that express the conditions of the spatial locking. Since we assume that the resonance approximation is satisfied with a high degree of accuracy [condition (7)], the absolute value of the external-field vector is assumed wherever possible to be equal to k_0. We present next estimates for the correlators in the most typical cases of a traveling and a standing light wave.

Traveling wave

In the field of a plane traveling wave $E(\mathbf{r}) = Ee^{i\mathbf{k}_0\cdot\mathbf{r}}$ only $F_{+-}(\mathbf{q} = 0)$ and $F_{--}(\mathbf{q} = \mathbf{k}_0)$ differ from zero in the expansion (14). The correlator g_+ then reaches its maximum when the scattered rays have the same direction, $\mathbf{n}_2 = \mathbf{n}_1$. The correlator g differs from zero only for forward scattering, when $\mathbf{n}_1$ and $\mathbf{n}_2$ are close in direction to that of the propagation of the external field $\mathbf{k}_0$ (Fig. 1). Near this direction, in a small solid angle $\sim(k_0 l)^{-2}$, we find that $g \sim g_+$.

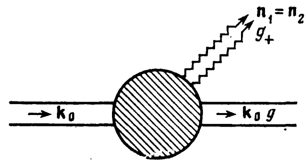

FIG. 1. Pattern of scattering in a traveling-wave field. The correlator g_+ differs from zero for arbitrary orientation of the scattered field, while the correlator g is nonzero only for forward scattering. The scattering region is shown shaded.

The forward-scattered field can be distinguished from the external monochromatic field either by its spectral composition (the spectral functions are calculated in §6), or by its polarization (§7). The anomalous correlator g can be observed in simpler fashion, however, in a standing light wave.

Standing wave

In the case of a standing light wave $E(\mathbf{r}) = E\cos\mathbf{k}_0\mathbf{r}$ the expansion (14) contains all the harmonics with $\mathbf{q}$ that are multiples of $\mathbf{k}_0$.

However, the condition of spatial locking is satisfied only by harmonics with $\mathbf{q} = 0$, $\pm\mathbf{k}_0$. At $\mathbf{q} = \pm\mathbf{k}_0$ the correlator g is anisotropic and is concentrated in a small solid angle near the directions $\pm\mathbf{k}_0$; this case is analogous to scattering in a field of a traveling wave. More interesting is a case $q = 0$. In this case a contribution to the correlator g_+ is made by rays having the same direction $(\mathbf{n}_1 = \mathbf{n}_2)$, while the correlator g receives contributions from oppositely scattered rays $(\mathbf{n}_1 = -\mathbf{n}_2)$. For the anomalous correlator, this case corresponds to absorption of two photons from oppositely traveling waves, so that the momentum transferred to the atom is zero. Then the emission of two photons should take place in opposite directions, to keep the momentum of the atom unchanged. This is shown schematically in Fig. 2. In a standing wave, the correlators g_+ and g are of the same order of magnitude for an arbitrary scattering angle.

We note that in the field of a standing light wave the wave front of the weak signal is inverted because of the four-photon interaction.[8,9] This effect was observed experimentally in Ref. 10, and the coefficients of reflection of a weak signal from a standing wave were calculated by us in the resonance approximation earlier.[11]

In the case considered by us here, the role of the weak signal is played by the scattered field. Because of

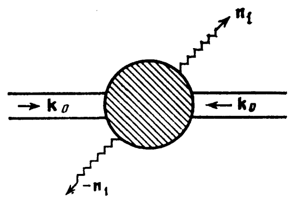

FIG. 2. Pattern of scattering in a standing wave. In this case both correlators are nonzero for any direction. In the correlators g, oppositely scattered rays correlate.

the reversal of the wave front, oppositely directed rays correlate in the anomalous mean values. We must emphasize the substantial difference between the classical picture of reflection of a weak signal from a continuous medium situated in the field of a standing wave, and the quantum picture of the scattering, which takes into account the fluctuations of the scattering medium. In the former case the intensity of the reflected signal is proportional to the square of the density of the particles and to the square of the intensity of the pump field. In the latter case we have correlators g_+ and g that are linear in the density and in the intensity of the external field. Obviously, greatest practical interest attaches to the investigation of these correlators in the case of low density of the medium and low intensity of the external field (correlators that are quadratic in the atom density were considered earlier in Ref. 12).

If we measure in experiment the correlation of oppositely scattered photons, then we can determine only the function g, since g_+ makes no contribution to $G(1,2)$ in this case. This circumstance can be important when it comes to observing a weak signal against the background of strong thermal radiation, inasmuch as there are no anomalous correlators for thermal radiation. In the case of light rays that intersect at an arbitrary angle, strongly anisotropic correlators arise and are concentrated near the surfaces of the synchronism cones.[12] Thus, by choosing a definite configuration of the external field we can observe various correlation functions of the scattered field.

§6. TEMPORAL CORRELATORS

a) Homogeneous broadening

In the case $\gamma \gg k_0 v_0$ (v_0 is the thermal velocity of the atoms) the temporal correlators $F_{+-}(\tau)$ do not depend on the concrete geometry of the external field. We denote the Fourier transforms of the functions $F_{+-}(\tau)$ and $F_{--}(|\tau|)$ by $f_{+-}(\Omega)$ and $f_{--}(\Omega)$. The function $f_{+-}(\Omega)$ is a spectral function of the scattered radiation. It is real and is independent of the phase of the external field. This function was calculated earlier in the resonance-fluorescence problem.[13] The functions $F_{--}(|\tau|)$ and $f_{--}(\Omega)$ are proportional to the square of the complex amplitude of the external field and are complex. We shall assume that the phase of the external field inside the scattering medium is zero. Then the contribution of the anomalous correlator to the counting rate of the paired coincidences can be represented in the form

$$\text{const}\cdot\{\exp[i(\varphi_1+\varphi_2)]F_{--}(|\tau|)+\text{c.c.}\},$$

where $\varphi_{1,2}$ is the optical path difference between the scattered and reference signals incident on the detectors 1 and 2. By varying the phase of the reference signal we can separate the real or imaginary part of the anomalous correlator.

We present now the expression obtained for $F_{--}(|\tau|)$ for the solution (9) and compare it with $F_{+-}(\tau)$. In the approximation linear in the intensity of the external field we have

$$F_{+-}(\tau)=\left|\frac{dE}{\hbar\nu_-}\right|^2,\quad F_{--}(|\tau|)=\left(\frac{dE}{\hbar\nu_-}\right)^2(-1+e^{-\nu_-|\tau|}). \tag{16}$$

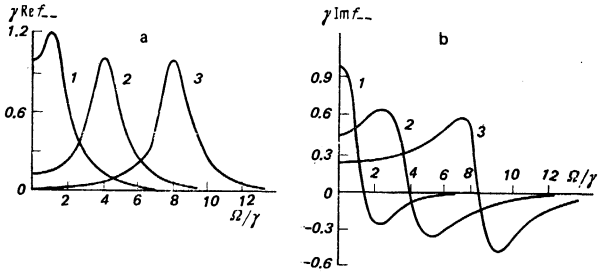

FIG. 3. Plots of the functions Re (γf_{--}) and Im (γf_{--}). The frequency unit is taken to be γ. Curves 1, 2, and 3 corresponds to $\Delta = 1$, 4, and 8, respectively. The function $\delta(\Omega)$ is omitted; $f_{--}(\Omega)$ is an even function of the frequency Ω.

In this approximation, the dipole-moment correlator $e^{i\omega\tau}F_{+-}(\tau)$ breaks up into a product of the average dipole moments $\langle p^{(-)}(t_2')\rangle\langle p^{(+)}(t_1')\rangle$. It describes the undisplaced component of the scattered radiation. The correlator

$$\exp[-i\omega(t_1'+t_2')]F_{--}(|\tau|)$$

breaks up into the product $\langle p^{(+)}(t_2')\rangle\langle p^{(+)}(t_1')\rangle$ only at $\gamma\tau \gg 1$. At $\tau = 0$ the correlator g vanishes. The reason is that the atom cannot absorb two photons simultaneously. An effect of this kind (anti-grouping of the photons) was observed experimentally in Ref. 14 in the study of the correlator of the intensities of scattered light.

We present the expressions for the spectral functions:

$$f_{+-}(\Omega)=2\pi\left|\frac{dE}{\hbar\nu_-}\right|^2\delta(\Omega),$$
$$f_{--}(\Omega)=\left(\frac{dE}{\hbar\nu_-}\right)^2\left(-2\pi\delta(\Omega)+\frac{1}{\nu_-+i\Omega}+\frac{1}{\nu_--i\Omega}\right). \tag{16'}$$

The anomalous-correlator spectrum (shown in Fig. 3) contains not only the undisplaced component, but also two Lorentz peaks that are symmetrically placed relative to the external-field frequency. Thus, in the lowest approximation in the intensity of the external field, the anomalous correlator contains more information on the scattering medium (on the line width) than the correlator g_+.

We present also an expression for $F_{\pm -}(\tau)$ in the next order in E^2

$$F_{+-}(\tau)=2\left|\frac{dE}{\hbar\nu_-}\right|^4\left(-2+e^{-\gamma|\tau|/2}\left[\cos(\Delta\tau)+\frac{\gamma}{\Delta}\sin(\Delta|\tau|)\right]\right),$$
$$F_{--}(|\tau|)=4\left(\frac{dE}{\hbar\nu_-}\right)^2\left|\frac{dE}{\hbar\nu_-}\right|^2\left[1-e^{-\nu_-|\tau|}\left(1-\frac{\nu_-|\tau|}{2}\right)\right].$$

b) Inhomogeneous broadening

So far, the atoms were assumed to be at rest. The thermal motion leads to an inhomogeneous broadening of the spectral lines. Let us see how the correlation functions change under the conditions of strong Doppler broadening $k_0 v_0 \gg |\nu_-|$. In the case of a traveling wave, the correlator averaged over the thermal motion of the atoms is of the form

$$\langle F_{+-}(\tau)\rangle_T=\int d\mathbf{v}f(\mathbf{v})F_{+-}(\mathbf{q}=0,\mathbf{v},\tau)e^{ik\tau\mathbf{n}_1\mathbf{v}}$$
$$=\frac{\pi^{1/2}}{k_0v_0\gamma}\left|\frac{dE}{\hbar}\right|^2\exp\left\{-\frac{\Delta^2}{(k_0v_0)^2}-\frac{(\tau k_0v_0\sin\theta)^2}{4}-i\Delta\tau(1-\cos\theta)\right\}. \tag{17}$$

Here θ is the angle between the propagation direction and the observation direction. The correlation time $(k_0 v_0 \sin\theta)^{-1}$ depends on the direction of the scattered ray. We see that by varying the detuning Δ or the correlation time τ we can obtain from the correlator $\langle F_{+-}(\tau)\rangle_T$ information only on the Doppler width of the line. Since the anomalous correlator for a traveling wave differs from zero only in the case of forward scattering, the thermal motion of the atoms does not change the width of the spectral function.

In the case of a standing light wave there remains in the correlator $F_{+-}(\tau)\rangle_T$ only the real part, equal to the real part of expression (17). The anomalous correlator has, for an arbitrary observation direction, the form

$$\langle F_{--}(|\tau|)\rangle_\tau = \frac{\pi^{1/2}}{k_0 v_0 \nu_-}\left(\frac{dE}{\hbar}\right)^2 \exp\left[-\frac{(k_0 v_0 \tau \sin\theta)^2}{4} - \nu_-|\tau|\right] \operatorname{sh}(\nu_-|\tau|\cos\theta). \quad (18)$$

If $\theta \sim 1$, we should consider delay times $\tau < (k_0 v_0)^{-1}$. Under this condition we have $|\nu_-|\tau \ll 1$ and the factor ν_-, which contains the natural line width, drops out of Eq. (18). Therefore at large observation angles we can obtain from the anomalous correlator information only on the Doppler-broadened line. At a small observation angle $\theta < \gamma/k_0 v_0$ and at $\gamma\tau \sim 1$, by tuning the frequency of the external field, we can determined from the anomalous correlator (18) the natural line width under the conditions of strong Doppler broadening.

For strong transitions (for example, for atoms of alkali metals), the parameter $\gamma/k_0 v_0$ is not very small, of the order of $\sim 10^{-2}$. Therefore the separation of the scattered field from the incident one entails no great difficulty. For weak transitions (γ/kv_0 is small) such a separation can be carried out by using the polarization properties of the scattered field.

§7. POLARIZATION PROPERTIES OF THE SCATTERED-FIELD CORRELATORS

In the analysis of the polarization properties of scattered radiation, account must be taken of the degeneracy of the atomic levels. We represent the field in the form of a sum of fields with definite polarization

$$\mathbf{E}(\mathbf{r}, t) = \sum_s E_s(\mathbf{r}, t)\mathbf{e}^s(\mathbf{n}),$$

where $\mathbf{n} = \mathbf{r}/r$. The anomalous correlator of the components of the polarization is of the form

$$\langle \hat{E}_{s_2}(\mathbf{r}_2 t_2)\hat{E}_{s_1}(\mathbf{r}_1 t_1)\rangle = \frac{k^4}{r_1 r_2}\exp[-i\omega(t_1'+t_2')]\int d\mathbf{r}\, d\mathbf{v}\, f(\mathbf{v})$$
$$\times \sum_{m\mu} \exp\{-ik\mathbf{r}(\mathbf{n}_1+\mathbf{n}_2) + ik\mathbf{v}\mathbf{n}_1\tau\}\langle \hat{\sigma}_{m_2\mu_2}(\mathbf{r}, \mathbf{v}, t_2')$$
$$\times \hat{\sigma}_{m_1\mu_1}(\mathbf{r}, \mathbf{v}, t_1')\rangle (\mathbf{e}^{s_2}(\mathbf{n}_2)\mathbf{d}_{\mu_2 m_2})(\mathbf{e}^{s_1}(\mathbf{n}_1)\mathbf{d}_{\mu_1 m_1}). \quad (19)$$

The lower and upper levels have angular momenta j_1 and j_2, and their sublevels are designated respectively by the indices μ and m. By $\mathbf{d}_{\mu m}$ we denote the dipole-moment matrix element. The equation for the two-dimensional atomic correlator $\langle\hat{\sigma}(2)\hat{\sigma}(1)\rangle$ can be easily calculated with the aid of the Bloch equations with account taken of the degeneracy of the atomic states,[15] using the method described in §4.

Let the pump field be a superposition of opposing waves with polarizations $E^{(\pm)}$:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{(+)}e^{ik_0 r} + \mathbf{E}^{(-)}e^{-ik_0 r}.$$

Then in the lowest order in the pump field we obtain the expression

$$\langle \hat{\sigma}_{m_2\mu_2}(\mathbf{v}, t_2')\hat{\sigma}_{m_1\mu_1}(\mathbf{v}, t_1')\rangle = [V^{(+)}_{m_1\mu_2}V^{(-)}_{m_2\mu_1}(e^{-\nu_-\tau} - e^{-ik\mathbf{v}\tau})$$
$$+ V^{(-)}_{m_1\mu_2}V^{(+)}_{m_2\mu_1}(e^{-\nu_-\tau} - e^{ik\mathbf{v}\tau})](\nu_- + ik\mathbf{v})^{-1}(\nu_- - ik\mathbf{v})^{-1}, \quad (20)$$
$$V^{(\pm)}_{m\mu} = \mathbf{E}^{(\pm)}\mathbf{d}_{m\mu}/\hbar.$$

Equation (20) contains only the interference term connected with the absorption of the photons from different light beams. Substituting (20) in (19) and summing over the sublevels, we have

$$\langle E_{s_2}(t_2)E_{s_1}(t_1)\rangle = \frac{(k_0 d)^4 n}{r_1 r_2}(2\pi)^3\delta(\mathbf{n}_1+\mathbf{n}_2)\exp[-i\omega(t_1'+t_2')]\int d\mathbf{v} f(\mathbf{v})$$
$$\times \exp(ik_0\mathbf{v}\mathbf{n}_1\tau)\sum_{\varkappa;\alpha=\pm} G_\varkappa[(\{e^{s_1}(\mathbf{n}_1)\otimes E^{(\alpha)}\}_\varkappa\{e^{s_2}(\mathbf{n}_2)\otimes E^{(-\alpha)}\}_\varkappa)$$
$$\times (e^{-\nu_-\tau} - e^{-i\alpha\tau\mathbf{k}_0\mathbf{v}})](\nu_- + i\mathbf{k}_0\mathbf{v})^{-1}(\nu_- - i\mathbf{k}_0\mathbf{v})^{-1},$$
$$G_\varkappa = (2j_1+1)^{-1}\begin{Bmatrix} 1 & 1 & \varkappa \\ j_1 & j_1 & j_2 \end{Bmatrix}^2. \quad (21)$$

The curly bracket denotes a $6j$ symbol, and $\{a\otimes b\}_\varkappa$ is the tensor product of the vectors a and b of rank $\varkappa$. Let the scattered fields and the pump fields propagate along a single line

$$\mathbf{n}_2 = -\mathbf{n}_1, \quad \mathbf{n}_1 \| \mathbf{k}_0.$$

To separate the scattered field from the incident field, we consider the field configuration shown in Fig. 4. The polarization of the opposing waves will be assumed linear and orthogonal, and the polarization of the scattered field in the directions $\mathbf{n}_1$ and $\mathbf{n}_2$ are specified by the unit vectors $\mathbf{e}(n_1) \parallel \mathbf{E}^{(+)}$ and $\mathbf{e}(n_2) \parallel E^{(-)}$. In other words, we are considering the correlation of fields that are back-scattered without change of polarization[2)] (this process is shown schematically in Fig. 4 by the dashed line). Obviously, in this situation the scattered field can be easily discriminated by means of its polarization from the pump field passing through the medium. Calculations carried out for this special case in accordance with Eq. (21) yield

$$\langle \hat{E}_y(r_2 t_2)\hat{E}_x(r_1 t_1)\rangle = \frac{(k_0 d)^4 N\pi^{1/2}E^{(+)}E^{(-)}}{2r_1 r_2 \nu_- k_0 v_0}$$
$$\times \exp[-i\omega(t_1'+t_2')](G_1+G_2)(-1+e^{-2\nu_-|\tau|}), \quad (22)$$

where N is the total number of scattering particles. The dependence of the coefficient $G_1 + G_2$ on the angular momentum of the lower level $j_1 = j$ for transitions with $\Delta j = 0, \pm 1$ is given below:

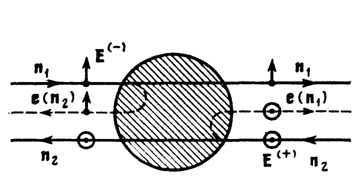

FIG. 4. Scheme of the polarizations of the pump and scattered-signal waves. The standing light wave is made up of two linearly polarized rays with orthogonal polarization vectors $\mathbf{E}^{\pm}$. The reflected waves are shown dashed. The polarizations of the reflected waves are designated by $\mathbf{e}(\mathbf{n}_1)$ and $\mathbf{e}(\mathbf{n}_2)$.

$$\frac{G_1+G_2}{G_0} \qquad \begin{matrix}\Delta j=+1\\ \dfrac{j(6j+7)}{5(j+1)(2j+3)}\end{matrix} \qquad \begin{matrix}\Delta j=-1\\ \dfrac{(j+1)(6j-1)}{5j(2j-1)}\end{matrix} \qquad \begin{matrix}\Delta j=0\\ \dfrac{2j(j+1)+1}{5j(j+1)}\end{matrix}.$$

We see that the considered effect is possible for atoms whose lowest state has a nonzero angular momentum.

At $\gamma\tau > 1$ the exponential in (22) can be neglected, and the anomalous correlator depends on the frequency of the external field only via the factor ν_-^{-1}. Thus, the anomalous correlator (22) has under the conditions of the strong Doppler broadening a Lorentz shape with a natural width $\gamma/2$.

§8. CONCLUSION

The foregoing analysis shows that the measurement of the space-time correlation function $G(1,2)$ makes it possible to determine the normal and anomalous correlators of the complex amplitudes of the scattered field. Under ordinary conditions, a plane traveling wave is incident on the scattering medium, and light scattered at a certain angle is emitted. It is possible then to measure only the spectral function of the normal correlator. The anomalous correlator makes a contribution to the forward scattering, and is therefore difficult to measure it against the background of the strong pump field. In a standing-wave field the scattering picture is greatly different. In this case, by measuring the correlation of the rays scattered in the same direction, or of oppositely scattered rays, we can determine separately the normal or the anomalous field correlators. These correlators turn out to be of the same order and the anomalous correlator carries just as complete information on the scattering medium as the ordinary spectral function. The spectral function of the anomalous correlator is complex and its phase is determined accurate to the phase difference between the pump field and the reference signal. By varying this difference we can separate the real or imaginary part of the spectral function $f_{--}(\Omega)$.

In the linear approximation in the intensity of the external field, the normal correlator contains only the undisplaced component, whereas the anomalous correlator has also displaced components [see Eq. (16)]. This circumstance can be significant in the spectroscopy of narrow atomic and molecular resonances, since it makes it possible to separate the fine structure of the resonance levels against the background of the Doppler broadening. It is important to emphasize that this is possible in an approximation linear in the intensity of the external field, in contrast to laser-spectroscopy methods, which are based on nonlinear effects.

It should be noted that the measurement of the anomalous correlators may turn out to be useful also in the study of other (nonresonant) media (dielectrics, liquid crystals, etc.). They can be used to obtain additional information not only on the spectra but also on the spatial properties of the scattering medium.

APPENDIX

The correlation function F_{--} satisfies the relation $F^*_{--}(\tau)=F_{++}(-\tau)$. Therefore to find F_{--} at $\tau<0$ it suffices to calculate at $\tau>0$ the function F_{++}, which satisfies a system of equations similar to (9),

$$\left(\frac{d}{d\tau}+\nu_+\right)F_{++}=iV^*(\mathbf{r})F_{0+},\quad \left(\frac{d}{d\tau}+\nu_-\right)F_{-+}=-iV(\mathbf{r})F_{0+},$$
$$\left(\frac{d}{d\tau}+\gamma\right)F_{0+}+\gamma p^*(\mathbf{r}-\mathbf{v}\tau)=2i[F_{++}V(\mathbf{r})-F_{-+}V^*(\mathbf{r})] \qquad \text{(A.1)}$$

with the following initial conditions at $\tau=0$:

$$F_{++}=0;\quad F_{-+}={}^1/_2(1-q);\quad F_{0+}=p^*.$$

From this we get for $\tau<0$

$$\left(-\frac{\partial}{\partial\tau}+\mathbf{v}\nabla+\nu_-\right)F_{--}=-iV(\mathbf{r})F_{-0},\quad \left(-\frac{\partial}{\partial\tau}+\mathbf{v}\nabla+\nu_+\right)F_{-+}=iV^*(\mathbf{r})F_{-0}$$
$$\left(-\frac{\partial}{\partial\tau}+\mathbf{v}\nabla+\gamma\right)F_{-0}+\gamma p(\mathbf{r}+\mathbf{v}\tau)=2i[F_{-+}V(\mathbf{r})-F_{--}V^*(\mathbf{r})] \qquad \text{(A.2)}$$

with initial conditions at $\tau=0$:

$$F_{--}=0;\quad F_{-+}={}^1/_2(1-q);\quad F_{-0}=p.$$

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1) We leave out the monochromatic (undisplaced) radiation component, which contributes only to the forward scattering.

2) We might also say that we are dealing with forward field scattering with change of polarization.

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