neighboring atoms. Further consideration of change in ω_1/ω_0 for F compounds with different chemical environments may improve ω_2/ω_0 here obtained. Study of the dependence of ω_1/ω_0 and ω_2/ω_0 on the covalency is in progress.

The chemical effect reflected in the intensity distributions of the F $K\alpha$ x-ray satellites produced by N^{4+} bombardment was observed and first explained quantitatively by introducing the vacancy rearrangement process in the L shell or the valence band of F^-. Γ_L estimated from the L-vacancy rearrangement probability f is directly proportional to the covalency. The technique offers promise for determining the ionicity or covalency of the chemical compounds with valence L-shell electrons.

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Intensity Correlations of a Cooperative System

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We suggest that the intensity correlation function could be an important tool in the study of atomic cooperative behavior because it contains predominantly components at $0, \pm 4\Omega$ in the limit of large Rabi frequency (2Ω) and large cooperation number. This is in contrast with the single-atom prediction where the intensity correlation function contains only frequency components at $0, \pm 2\Omega$. The master equation for the collective system is solved analytically in the secular approximation.

Many theoretical studies[1-7] of the cooperative effects in the interaction of atoms and molecules with a laser field and the vacuum of radiation have been carried out since the early work on superradiance by Dicke.[8] Several manifestations of cooperative behavior have also been observed experimentally.[9] In this Letter we discuss a commonly used model for the description of the collective behavior of two-level systems interacting with radiation in the context of resonance fluorescence. We show that the intensity correlation function[10,11] of the scattered light contains predominantly frequency components $0, \pm 4\Omega$ in the limit of large Rabi frequency (2Ω) and large cooperation number. This result is in contrast with the single-atom prediction which shows modulation only at frequency $\pm 2\Omega$ in the large-field limit. Thus the cooperative effects cause the intensity correlation function to have a very different temporal behavior which should be manifested by experiments of the type carried out by Kimble, Dagenais, and Mandel.[12] In this Letter, we also report a new approach for the solution of the master equation describing the resonance fluorescence model from a collective system and present analytical results.

The master equation describing the collective decay of an atomic system (two-level atom) in the presence of a driving laser field is given by[2a]

$$\partial W/\partial t = i\Omega[S^+ + S^-, W] - \gamma(S^+S^-W - 2S^-WS^+ + WS^+S^-) \equiv LW, \tag{1}$$

where 2Ω is the Rabi frequency and 2γ is the Einstein A coefficient. The operators S^+ and S^- are the usual polarization operators and W is the atomic density matrix. The same master equation has been adopted as a model for the description of collective effects in resonance fluorescence in the limit of strong cooperative behavior.[4] The exact solution of (1) for arbitrary number of atoms is not known. Numerical studies have been carried out for a limited number of atoms.[4,6] If the laser field is sufficiently intense, $\Omega \gg \gamma$, it is possible to develop an approximation scheme that enables us to obtain analytic results. After performing the transformation to the collective operators defined by

$$R^x = S^y, \quad R^y = S^z, \quad R^z = S^x, \tag{2}$$

we find that the Liouville operator L appearing in Eq. (1) splits into two components L_0 and L_1. The component L_0 is slowly varying in time, whereas L_1 contains rapidly oscillating terms at frequencies 2Ω and 4Ω. For intense fields, it is reasonable to make the secular approximation, i.e., to retain only the slowly varying part. Corrections to the results obtained in this fashion will be of the order $(\gamma S/\Omega)^2$, where S is the cooperation number. The new master equation has the structure

$$\partial W/\partial t = -2i\Omega[R^z, W] - \gamma\{[R^zR^z + \tfrac{1}{4}(R^+R^- + R^-R^+)]W - R^zWR^z - \tfrac{1}{4}(R^+WR^- + R^-WR^+) + \text{H.c.}\}. \tag{3}$$

Equation (3) can be solved exactly with the following results.

(A) *Steady-state fluctuations.*—The stationary solution of (3) is given by

$$W = \frac{1}{2S+1}\sum_{m=-S}^{S} |m\rangle\langle m|, \tag{4}$$

where $|m\rangle$ is an eigenstate of S^2 and S^z. The steady-state density operator (4) leads to the remarkable result that the fluctuations are of order unity rather than $1/S$:

$$(\langle S^zS^z\rangle - \langle S^z\rangle\langle S^z\rangle)/N^2 = \tfrac{1}{6}(\tfrac{1}{2} + N^{-1}), \quad (\langle S^+S^-\rangle - \langle S^+\rangle\langle S^-\rangle)/N^2 = \tfrac{1}{3}(\tfrac{1}{2} + N^{-1}). \tag{5}$$

(B) *Transient behavior and the spectrum of resonance fluorescence.*—One finds the following exact result for the time evolution of the dipole-moment and inversion operators (in the frame rotating with the laser frequency ω_L):

$$\begin{aligned}\langle S^+(\tau)\rangle = {}& \tfrac{1}{2}e^{-\gamma(t-\tau)}\langle S^+(\tau) + S^-(\tau)\rangle + \tfrac{1}{4}\exp[(2i\Omega - \tfrac{3}{2}\gamma)(t-\tau)]\langle S^+(\tau) - S^-(\tau) - 2S^z(\tau)\rangle \\ & - \tfrac{1}{4}\exp[(-2i\Omega - \tfrac{3}{2}\gamma)(t-\tau)]\langle S^-(\tau) - S^+(\tau) - 2S^z(\tau)\rangle,\end{aligned} \tag{6}$$

$$\langle S^z(t)\rangle = \tfrac{1}{4}\exp[(2i\Omega - \tfrac{3}{2}\gamma)(t-\tau)]\langle 2S^z(\tau) + S^-(\tau) - S^+(\tau)\rangle + \text{c.c.} \tag{7}$$

The first-order atomic correlation function (which is proportional to the field-amplitude correlation function) is found to be

$$\lim_{t\to\infty}\langle S^+(t+\tau)S^-(\tau)\rangle = \tfrac{1}{6}[N(N+2)]\{\tfrac{1}{2}e^{-\gamma\tau} + \tfrac{1}{4}\exp[(2i\Omega - \tfrac{3}{2}\gamma)\tau] + \tfrac{1}{4}\exp[(-2i\Omega - \tfrac{3}{2}\gamma)]\tau\} \tag{8}$$

in steady state. This result yields the usual triplet structure in the resonance fluorescence spectrum because, as well known, the incoherent part of the scattered spectrum is proportional to the Fourier transform of Eq. (8). The cooperation number appears only in the form of a scale factor as previously suggested by Amin and Cordes.[13] As a result of our expansion procedure, one expects additional correction terms to Eq. (8); these have been discussed by Senitzky[5,14] using a different approach.

(C) *Intensity correlations of the radiation emitted by a collective system.*—The recent measurement[12] of the intensity correlation function in the resonance fluorescence has provided new insight into the behavior of atoms in strong laser fields. The normalized intensity correlation function in steady state is proportional to the atomic correlation function

$$\gamma^{(2)}(\tau) = \lim_{t\to 0}\langle S^+(t)S^+(t+\tau)S^-(t+\tau)S^-(t)\rangle / [\lim_{t\to 0}\langle S^+(t)S^-(t)\rangle]^2 . \tag{9}$$

The time-dependent solution of Eq. (3) leads to the following time evolution:

$$\langle S^+(t)S^-(t)\rangle = \tfrac{2}{3}S(S+1) + \tfrac{1}{6}\langle 3R^zR^z - S(S+1)\rangle e^{-(3\gamma/2)\tau} + \tfrac{1}{4}e^{-5\gamma\tau}\{\langle R^+R^+\rangle e^{4i\Omega\tau} + \langle R^-R^-\rangle e^{-4i\Omega\tau}\} + (2i)^{-1}e^{-(3\gamma/2)\tau}\{\langle R^+\rangle e^{2i\Omega\tau} - \langle R^-\rangle e^{-2i\Omega\tau}\} . \tag{10}$$

After use of the quantum regression theorem and the stationary solution (4), Eq. (10) leads to the following result for the normalized intensity correlation function:

$$\gamma^{(2)}(\tau) = 1 + \tfrac{1}{20}\{1 + 3/[4S(S+1)]\}e^{-(3\gamma/2)t} + \tfrac{3}{5}\{1 - 3/[4S(S+1)]\}e^{-5\gamma\tau}\cos(4\Omega\tau) - \tfrac{3}{4}[S(S+1)]^{-1}e^{-(3\gamma/2)\tau}\cos(2\Omega\tau) . \tag{11}$$

The corresponding result for the single-atom case $(S=\frac{1}{2})$ ${\gamma_{\rm sa}}^{(2)}$ is $(\Omega \gg \gamma)$

$${\gamma_{\rm sa}}^{(2)}(\tau) = 1 - e^{-(3\gamma/2)\tau}\cos(2\Omega\tau) . \tag{12}$$

Upon comparison of Eqs. (11) and (12) it becomes clear that the cooperative correlation function ${\gamma_{(\tau)}}^{(2)}$ has a rather different structure from its single-atom counterpart ${\gamma_{\rm sa}}^{(2)}$.

In fact, the term modulated at frequency 2Ω disappears in the limit of large cooperation numbers and new contributions, instead, become important at zero frequency [i.e., the second term of Eq. (11)] and at frequency 4Ω.[15] What is important here is that this effect is predicted even in zeroth order in the parameter $\gamma S/\Omega$. Thus, ${\gamma_{(\tau)}}^{(2)}$ provides a new tool for the study of cooperative effects.

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magnitude as the one in the absence of the cooperative effects and therefore should be more directly accessible to experimental observation.

[15] If one considers a system with large numbers of atoms which are emitting independently, then one finds (Ref. 11)

$$\gamma^{(2)}(\tau) = 1 + \tfrac{1}{4}e^{-2\gamma\tau}(1 + \tfrac{1}{2}e^{-\gamma\tau} + \tfrac{1}{8}e^{-3\gamma\tau}\cos(4\Omega\tau) + \tfrac{1}{2}e^{-(5/2)\gamma\tau}\cos(2\Omega\tau).$$

Again, there is a large difference between the structure of $\gamma_{(\tau)}{}^{(2)}$ obtained under cooperative conditions and the above expression as far as the weights and decay rates of the various contributions are concerned.

Variational Method in Turbulence Theory

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The weighted mean square of the Navier-Stokes equation is mimimized with a complete set of realizability inequalities as constraints. Expansion of moments in complete orthogonal functions leads to successive approximations without ever involving moments of order higher than 4. Alternatively, the expansions may be in Wiener-Hermite kernels, thereby automatically satisfying the realizability constraints. The approach extends to other classical and quantized systems with polynomial nonlinearity.

The nonlinearity of the Navier-Stokes (NS) equation couples velocity-field moment equations of all orders. However, the values of moments up to fourth order are sufficient to determine whether an ensemble of velocity fields satisfy the NS equation in mean square. We suggest that this can be the basis of a closed formulation, useful for computation, in which moments of order higher than 4 never appear and the missing information contained in the initial values of the higher moments shows up as freedom in the evolution of the moments retained.

The NS equation may be written as

$$L_i(t) = dy_i/dt + \textstyle\sum_j \nu_{ij} y_j + \sum_{jk} A_{ijk} y_j y_k = 0, \qquad (1)$$

where the $y_i(t)$ are the real amplitudes of linearly independent modes of the velocity field, ν_{ij} is a damping matrix with positive eigenvalues, and $A_{ijk} + A_{jki} + A_{kij} = 0$ (conservation of $\sum_i y_i{}^2$ by the nonlinearity). Consider the limit $\kappa \to 0$ of the constraint on the ensemble[1]

$$\textstyle\sum_i \int_0^T \langle [L_i(t)]^2 \rangle \rho_i(t)\,dt = \kappa^2, \qquad (2)$$

where $\langle\,\rangle$ denotes ensemble average, and $\rho_i(t) > 0$ for $0 \leqslant t \leqslant T$. Equation (2) expresses approximation to the NS equation in mean square, with $L_i(t) = O(\kappa)$ in typical realizations. It implies, via Schwarz inequalities expressing positivity of the probability density, that the hierarchy of NS many-time moment equations is satisfied with errors $O(\kappa)$ as $\kappa \to 0$, if the moments of all orders exist in the limit. Equation (2) involves only second-, third-, and fourth-order moments. It can be used variationally to determine values of these moments that solve the NS equation provided the values are constrained to be realizable (i.e., represent an ensemble with nowhere negative probability density).

For a single real stochastic variable b the necessary and sufficient conditions for realizability are[2]

$$I_{2n}(\lambda) = \langle (\sum_{r=0}^{n} \lambda_r b^r)^2 \rangle$$

$$= \sum_{r,\,s=0}^{n} \lambda_r \lambda_s \langle b^{r+s} \rangle \geqslant 0 \quad (n = 1, 2, \ldots, \infty) \qquad (3)$$

for all real λ_s such that $\sum_{s=0}^{n} \lambda_s{}^2 > 0$. Equation (3) states that the symmetric matrix $Q_{rs} = \langle b^{r+s} \rangle$ has no negative eigenvalues. The moments $\langle b^n \rangle$ $(n > 2N)$ of an ensemble with given $\langle b^n \rangle$ $(n \leqslant 2N)$ are nonunique.[3] If $I_{2N}(\lambda) > 0$ (all λ) we can take $\langle b^{2N+1} \rangle = 0$ and verify by minimizing with respect to λ_{N+1} that a sufficient condition for $I_{2N+2}(\lambda) > 0$ (all λ) is

$$\langle b^{2N+2} \rangle > \max[(\sum_{s=0}^{N} \lambda_s \langle b^{s+N+1} \rangle)^2 / I_{2N}(\lambda)], \qquad (4)$$

where the maximum is with respect to the λ_s at $\sum_{s=0}^{N} \lambda_s{}^2 = 1$. This can be continued indefinitely yielding all $I_{2n}(\lambda) > 0$, provided only that the arbitrarily prescribed $\langle b^{2n} \rangle$ $(n > N)$ grow fast enough with n. Very large values of $\langle b^{2N+2} \rangle$ in (4) corre-