Sidebands in Strong-Field Resonance Fluorescence

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It is shown that consideration of cooperative atomic behavior in the analysis of resonance fluorescence leads to the prediction of additional sidebands.

Resonance fluorescence has become the subject of considerable interest in recent years. Theoretically,[1-6] it is an excellent vehicle for modern developments in quantum optics; experimentally,[7,8] it allows the utilization of the high spectral purity of strong laser fields. Almost all the recent theoretical work refers to the fluorescence of independent "two-level" atoms, and predicts the interesting phenomenon of reemission, for a sufficiently strong irradiation field, of three frequencies: the incident frequency ω and two sidebands at $\omega \pm \Omega$, Ω being the Rabi frequency. The effect of cooperative atomic behavior—a subject of great interest in connection with other phenomena associated with a collection of two-level systems,[9] such as superradiance and superfluorescence—has received little attention in the treatment of resonance fluorescence. This deficiency is aggravated by the fact that the only two discussions of the subject that apparently exist in the literature[3,6] make contradictory predictions concerning the effect of cooperative behavior on the number of sidebands: In one[3] I find the statement that such behavior produces additional sidebands, while in the other[6] it is claimed that the number of sidebands remains unaffected. Experiment, so far, has verified the existence of the first pair of sidebands,[7] but no effort has been made to look for additional, further sidebands. It is the purpose of this Letter to discuss the appearance of sidebands in a strong field, and to present arguments for expecting additional sidebands to result from cooperative atomic behavior. These arguments would make experimental detection of the additional sidebands an effective demonstration of the type of cooperative atomic behavior widely assumed in the treatment of collections of two-level systems.[9]

Consider a number N of identical two-level systems, all coupled identically to the electromagnetic field. They can be described as a single angular-momentum oscillator (AMO) of total angular-momentum quantum number L_0, which may be regarded as the cooperation number[3,9]; maximum cooperation is indicated by $L_0 = \frac{1}{2}N$. With use of the rotating-wave approximation, the total Hamiltonian is given by

$$H = \hbar\omega l_3 + \sum_k \hbar\omega_k (a_k^\dagger a_k + \tfrac{1}{2}) + \tfrac{1}{2}\hbar \sum_k (\gamma_k a_k^\dagger l_- + \gamma_k^* a_k l_+) + \tfrac{1}{2}\hbar(\gamma a^* l_- + \gamma^* a l_+).$$

Both the model and the notation are the same as that of Ref. 3. The AMO variables are the (dimensionless) angular-momentum components l_1, l_2, and l_3, satisfying commutation relation $[l_1, l_2] = i\hbar l_3$, etc., with $l_\pm = 2^{-1/2}(l_1 \pm i l_2)$; the variables of the kth electromagnetic-field mode, of (angular) frequency ω_k, are a_k and $a_k^\dagger$, satisfying $[a_k, a_k^\dagger] = 1$; a and a^* describe the prescribed irradiation field, which, for simplicity, I take to be exactly on resonance and of constant amplitude; the γ_k's and γ's are the appropriate coupling constants. Introducing the "reduced" variables A_k, $L_\pm$, and L_3, defined by $a_k \equiv A_k \times \exp(-i\omega_k t)$, $l_\pm \equiv L_\pm \exp(\pm i\omega t)$, $l_3 = L_3$, respectively, and utilizing the definitions

$$\mathfrak{A} \equiv \tfrac{1}{2} i \sum_k \gamma_k^* A_k \exp(-i\nu_k t), \quad A \equiv \tfrac{1}{2} i \gamma^* a \exp i\omega t,$$

where $\nu_k \equiv \omega_k - \omega$, I obtain, for the Heisenberg equations of motion,

$$\dot{L}_+ = (A^* + \mathfrak{A}^\dagger) L_3, \quad \dot{A}_k = -\tfrac{1}{2} i \gamma_k L_- \exp(i\nu_k t), \quad \dot{L}_3 = -[L_+(A + \mathfrak{A}) + (A^* + \mathfrak{A}^\dagger) L_-].$$

For sufficiently weak coupling between the AMO and the radiation field, it has been shown[3] that the field equations yield, approximately, $\mathfrak{A}(t) \simeq \mathfrak{A}_0(t) + \alpha L_-(t)$, with

$$\mathfrak{A}_0 \equiv \tfrac{1}{2} i \sum_k \gamma_k^* A_k(0) \exp(-i\nu_k t), \quad \alpha \equiv \tfrac{1}{4}\pi |\gamma(\omega)|^2 \rho(\omega),$$

where $|\gamma(\omega)|^2$ is the average of $|\gamma_k|^2$ over all k's for ω_k near ω, and $\rho(\omega)$ is the mode density at ω. The atomic equations of motion then become

$$\dot{L}_+ = (A^* + \mathfrak{A}_0^\dagger) L_3 + \alpha L_+ L_3, \quad \dot{L}_- = (\dot{L}_+)^\dagger, \quad \dot{L}_3 = -[L_+(A + \mathfrak{A}_0) + (A^* + \mathfrak{A}_0^\dagger) L_-] - 2\alpha L_+ L_-.$$

The initial state of the radiation field is taken to be the ground state, so that $\langle | \mathfrak{A}_0^\dagger = 0$ and $\mathfrak{A}_0 | \rangle = 0$. An

important feature of the present formalism is the fact that the equations of motion are valid both quantum mechanically and classically, the initial classical field conditions being $\mathcal{A}_0 = \mathcal{A}_0^\dagger = 0$. The classical analysis may be applied only for L_0 sufficiently large, of course, with some additional restriction which is of no present concern.[3]

The expectation value P of the power radiated by the AMO, the fluorescence, is given by $\langle \sum_k \hbar\omega_k \dot{A}_k^\dagger A_k \rangle$ + c.c. From the equations of motion one gets

$$P(t) = \sum_k \tfrac{1}{4}|\gamma_k|^2 \hbar\omega_k \int_0^t dt_1 \langle L_+(t) L_-(t_1) \rangle \exp[-i\nu_k(t-t_1)] + \text{c.c.}$$

Utilizing the fact that the main contribution to the summation comes from the region of ω_k near ω, and approximating the summation by an integration, I obtain

$$P(t) = \int_0^\infty d\omega_k \mathcal{P}(\omega_k, t), \quad \mathcal{P}(\omega_k, t) \equiv (\alpha/\pi)\hbar\omega \int_0^t d\tau \langle L_+(t) L_-(t-\tau) \rangle \exp[-i(\omega_k - \omega)\tau] + \text{c.c.}$$

If, for large t, $P(t)$ becomes independent of the time, a steady-state situation exists. We can also regard the fluorescence as a steady-state phenomenon if the average of $P(t)$ over a reasonably short time (say, a few periods of the Rabi frequency) is constant. I anticipate this to be indeed the case for sufficiently large t, and take $\mathcal{P}(\omega_k)$, given by

$$\mathcal{P}(\omega_k) \equiv (\alpha/\pi)\hbar\omega \int_0^\infty d\tau \langle L_+(t) L_-(t-\tau) \rangle_{\text{av}} \exp[-i(\omega_k - \omega)\tau] + \text{c.c.},$$

where the subscript "av" indicates an average over t, to be the power spectrum of the fluorescence. This expression is valid both classically and quantum mechanically.

The above theory will be applied first to a two-level system (TLS). The atomic equations simplify in this case to

$$\dot{L}_+ = (A^* + \mathcal{A}_0^\dagger) L_3 - \tfrac{1}{2}\alpha L_+, \quad \dot{L}_- = (\dot{L}_+)^\dagger, \quad \dot{L}_3 = -[L_+(A + \mathcal{A}_0) + (A^* + \mathcal{A}_0^\dagger) L_-] - \alpha(L_3 + \tfrac{1}{2}).$$

These equations have been solved explicitly[3] for $\langle L_\pm \rangle$ and $\langle L_3 \rangle$. In the absence of an irradiation field ($A = 0$), $\langle L_3(t) \rangle$ decays as $\exp(-\alpha t)$ to its ground-state value—behavior that exhibits α as the Weiskopf-Wigner decay constant. In the presence of a strong irradiation field ($|A| \gg \alpha$), $\langle L_3 \rangle \simeq \langle L_3^{(0)}(t) \rangle \exp(-\frac{3}{4}\alpha t)$, where $\langle L_3^{(0)}(t) \rangle$ is a sinusoidal oscillation of (angular) frequency Ω, the Rabi frequency, defined by $\Omega^2 \equiv 2|A|^2$, and is the solution of the atomic equations of motion for $\langle L_3(t) \rangle$ when radiation is ignored. For $t \gg \alpha^{-1}$, the TLS equations for $L_\pm$ yield

$$\langle L_+(t) L_-(t-\tau) \rangle = \tfrac{1}{2}\Omega^2 \int_0^t dt_1 \int_0^{t-\tau} dt_2 \langle L_3(t_1) L_3(t_2) \rangle \exp[-\tfrac{1}{2}\alpha(2t - \tau - t_1 - t_2)].$$

The expression for $\langle L_3(t_1) L_3(t_2) \rangle$ can be calculated from the TLS equations of motion using an approximation which is based on neglect of commutators of field variables and atomic variables in terms that contain a product of the factors γ (or γ^*) and γ_k (or γ_k^*).[10] For simplicity, consider only the result to the lowest order in α/Ω, valid for strong fields. One obtains, for sufficiently large t,

$$\langle L_3(t) L_3(t+\eta) \rangle = \tfrac{1}{4}\exp(-\tfrac{3}{4}\alpha|\eta|)[\cos\Omega\eta - \tfrac{1}{4}(\alpha/\Omega)\sin\Omega|\eta|],$$

$$\langle L_+(t) L_-(t-\tau) \rangle = \tfrac{1}{8}[\exp(-\tfrac{1}{2}\alpha\tau) + \exp(-\tfrac{3}{4}\alpha\tau)\cos\Omega\tau],$$

$$\mathcal{P}(\omega_k) = (\alpha^2/4\pi)\hbar\omega\{\tfrac{1}{2}[(\tfrac{1}{2}a)^2 + (\omega_k - \omega)^2]^{-1} + \tfrac{3}{8}[(\tfrac{3}{4}\alpha)^2 + (\omega_k - \omega - \Omega)^2]^{-1} + \tfrac{3}{8}[(\tfrac{3}{4}\alpha)^2 + (\omega_k - \omega + \Omega)^2]^{-1}\}.$$

This result is consistent with previous work,[2,6] and displays the two sidebands displaced from the main peak by the Rabi frequency Ω. As the field becomes weaker, the closed-form result for $\mathcal{P}(\omega_k)$ (not presented here) shows that the two sidebands coalesce with the main peak.[2] A possible intuitive explanation of the presence of three peaks, even though the oscillation of $\langle L_3 \rangle$ (or $\langle L_+ L_- \rangle$) is damped completely, lies in the existence of a continuing[2] partial sinusoidal modulation of the atomic radiation (the modulation must be only partial if there exists a central peak) together with a spread[3]—due to quantum-mechanical uncertainties—in both the modulation frequency and the radiation rate.

I turn next to the case of cooperative behavior. In order to concentrate on the essential aspects of the present argument and to simplify the mathematical disucssion, I consider large cooperation, that is $L_0 \gg \frac{1}{2}$, and the field sufficiently strong so that $\Omega \gg \alpha L_0$. In this limit, the AMO may be regarded, essentially, as a classical system.[3] Interpreting the equations of motion classically, one can convert

them into

$$L_+(t) = [L_+(0) + (A^*/\alpha)] \exp[\alpha L_0 \int_0^t dt_1\, x(t_1)] - (A^*/\alpha), \quad \ddot{x} - 3\alpha L_0 x\dot{x} + [\Omega^2 - \alpha^2 L_0{}^2(1-x^2)]x = 0,$$

where $x \equiv L_3/L_0$. Neglect of the $\alpha^2 L_0{}^2$ term compared to Ω^2 yields

$$\ddot{x} - 3\alpha L_0 x\dot{x} + \Omega^2 x = 0.$$

If $x(t) = 0$, a possible solution, the power radiated is unmodulated. All other solutions of the last equation can be shown to be periodic.[3] They are not, however, sinusoidal, and thus contain harmonics. Physically, the effect of the α term in producing harmonics is explained by the fact that the ascent of L_3 is slowed by radiation while the descent of L_3 is hastened by radiation. An equivalent equation for $x(t)$ is

$$x(t) = x^{(0)}(t) + 3(\alpha L_0/\Omega) \int_0^t dt_1\, x(t_1)\dot{x}(t_1) \sin\Omega(t - t_1),$$

where $x^{(0)}(t)$ is a solution of $\ddot{x} + \Omega^2 x = 0$, andy may be regarded as the zeroth-order solution with respect to the parameter $\alpha L_0/\Omega$. Setting $x^{(0)}(t) = x_0 \cos\Omega t$ and neglecting a first-order correction to the fundamental, one obtains, up to first order,

$$x(t) \approx x_0 \cos\Omega t + \tfrac{1}{2}(\alpha L_0/\Omega) x_0{}^2 \sin 2\Omega t.$$

Now, $L_+(t)$ is the complex amplitude of oscillation of the AMO. It is clear, from the exponential form of $L_+(t)$, that this oscillation will be modulated not only at frequency Ω but also at multiples of Ω. Since the spectral resolution of modulated oscillation contains sidebands at *all* the modulation frequencies, one should expect sidebands at multiples of Ω.[11] Substituting for x into L_+ and its complex conjugate, we obtain, from $\langle L_+(t) L_-(t-\tau)\rangle_{\rm av}$, the power spectrum of the radiated field with sufficient accuracy to include the sidebands at $\omega \pm 2\Omega$, the result being

$$\mathcal{P}(\omega_k) \approx \alpha \hbar \omega L_0{}^2 \{(1 - x_0{}^2)\delta(\omega_k - \omega) + \tfrac{1}{4} x_0{}^2 [\delta(\omega_k - \omega + \Omega) + \delta(\omega_k - \omega - \Omega)] + \tfrac{1}{16}(\alpha L_0/\Omega)^2 x_0{}^4 [\delta(\omega_k - \omega + 2\Omega) + \delta(\omega_k - \omega - 2\Omega)]\}.$$

The simple classical model that has been used contains no mechanism for line broadening, so that the spectrum is infinitely sharp, of course. However, it is reasonable to expect that such a mechanism will not affect the existence of sidebands at $\omega \pm 2\Omega$. The ratio of the strength of the second pair of sidebands to that of the first pair is given by $\frac{1}{4}(\alpha L_0/\Omega)^2 x_0{}^2$. Since I have assumed the strong inequality $\alpha L_0/\Omega \ll 1$ (and noting that $0 \leq x_0{}^2 \leq 1$), the second pair of sidebands may, perhaps, appear too weak, in comparison with the first pair, for experimental detection. However, these sidebands indicate resonances that may be detectable if one is looking at a small frequency region that contains no other resonances. Furthermore, the smallness of $\alpha L_0/\Omega$ was used mainly in order to facilitate the calculation. One needs only $4\alpha L_0 \lesssim \Omega$ in order to separate the sidebands from the central peak in a TLS, for instance.[2] Observation of the additional resonances would—because of its qualitative nature—be a rather dramatic verification of the fact that a number of two-level systems interacting with the optical radiation field may be regarded under certain conditions as a single, classical, angular-momentum oscillator.

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[11]A critique of the reasons which lead the authors of Ref. 6 to believe that no additional sidebands arise due to cooperative atomic behavior will be found in Ref. 10.

Ionic Motion in α-AgI

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A molecular-dynamics study of silver diffusion in superionic conductor α-AgI is performed. Interionic potentials are constructed using Pauling's ideas of ionic radii. The diffusion constant for silver and its temperature dependence are in good agreement with experiment. Good agreement is also obtained for the silver density map with the experiment of Cava, Reidinger, and Wuensch.

α-AgI has been extensively studied[1] because of its interest as a solid fast ion conductor (often called superionic conductor). In this Letter we present a molecular-dynamics (MD) study of this material. Our calculations show that in the computer model the diffusion constant of Ag^+ and their distribution in the interstices of the thermally agitated iodine lattice are faithfully reproduced. In addition, MD provides a detailed picture of the complete process of self-diffusion. The central issue in all such calculations is the construction of potential functions which, when used in the MD calculations, produce structural and dynamical correlations which compare favorably with those observed in the laboratory. The MD trajectories then porvide a wealth of microscopically detailed information which can be very hard to obtain by conventional experimental means. A study of CaF_2 along these lines has already been reported.[2]

For CaF_2 the ionic model given by Kim and Gordon[3] is sufficiently good.[2] In α-AgI this problem is not so straightforward. We have constructed effective pair potentials which provide a simple means of describing, with reasonable accuracy, the structural and dynamical properties of α-AgI. Schommers[4] had to use quite unphysical forces to keep the iodine lattice vibrating as a stable structure; in his model each iodine was attached to bcc lattice position by harmonic springs. In our model no such unphysical elements have been introduced. For AgI, we use

$$V_{ij}=\frac{A_{ij}(\sigma_i+\sigma_j)^n}{r^n}+\frac{Z_iZ_je^2}{r}-\frac{1}{2}(\alpha_iZ_i^2+\alpha_jZ_j^2)\frac{e^2}{r^4}-\frac{W_{ij}}{r^6}, \quad (1)$$

where i, j describe the type of ions; A_{ij} the repulsive strength; σ_i, σ_j the particle radii; α_i, α_j the electronic polarizabilities. If σ's, α's, and W's are known, we need to determine five parameters (namely A_{ij}'s, n, and $|Z_i|=|Z_j|$). By assuming $A_{ij}=A$, the situation is considerably simplified. The repulsive term then implies that each ionic "contact" contributes energy A, i.e., the coefficient of r^{-n} is scaled according to the sum of particle radii. Low-temperature crystal structure, cohesive energy, and compressibility may be used to determine these three parameters.

However, AgI, and α-AgI in particular, pose special problems in this respect. It is certainly not purely ionic and the estimate of the cohesive energy from the Born-Haber cycle can be in considerable error. The γ-AgI compressibility, however, is known[5] ($\simeq 1\times10^{-11}$ cm^3/erg) and from phonon dispersion measurement[6] there is evidence of $|Z|\simeq 0.6$.

As to the σ's, Pauling's[7] concept of ionic radii is a means of expressing the bond length. Using the concept literally, we write $\sigma_{Ag}+\sigma_I$=Ag-I distance and $2\sigma_I$=I-I distance. Because of the large