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Resonant scattering or absorption followed by emission*

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Several papers have appeared recently indicating theoretical and experimental differences between resonant scattering and absorption followed by emission. We show that they are the same phenomenon and demonstrate that a widely used analytical treatment of radiative processes does not describe this phenomenon accurately.

I. INTRODUCTION

Recently, Klein[1] proposed that "hot luminescence" (HL), which he defined as the absorption of a photon followed by emission (AE), and resonant Raman scattering (RRS) were equivalent phenomena. This point of view was disputed by Shen[2] who used density-matrix formalism in order to argue that RRS and HL are not equivalent but are two distinct physical processes that are always simultaneously present, can interfere with each other, and differ in line shape and transient response. In particular, Ref. 2 indicated that his results can be used to account for transient phenomena, such as free induction decay (designated as coherent Raman beats by Ref. 2), which occur in the collisionless case, that is, the case where interactions of the radiation field and the scattering system with other systems (such as a thermal bath) can be neglected. In addition, there has been recent experimental work[3,4] which also contradicts Klein's contention. We, however, maintain that Klein was not in error and that RRS and AE (or HL[5]) are unequivocally the same phenomenon.

Reference 2 derived an expression for the scattering cross section for light near a resonance and identified one term as RRS and the other as HL. RRS was described as a direct and essentially instantaneous two-photon process and HL as a physically different and much slower two-step process. References 3 and 4 took a similar view and claimed to distinguish between the two processes experimentally. AE was described as a "true" absorption followed by emission while RRS was described as a nearly instantaneous, "virtual" absorption followed by emission and, therefore, when observed in a gas would not exhibit collision broadening, depolarization, or quenching.

In the past, there has been considerable lack of clarity regarding the relation of RRS to AE. Some[6] have made distinctions based on classical arguments, while others[7–9] have regarded RRS and AE as the same phenomenon and derived expressions for the RRS cross section by the intuitive use of Einstein A and B coefficients and absorption and emission line shapes.

In Sec. II, we develop an intuitive theory of RRS for the collisionless case which is shown to give the same RRS cross section as modern quantum theory and which clearly demonstrates the equivalence of RRS and AE. In Sec. III, we consider some of the special problems encountered in the quantum-mechanical theory of RRS.

II. INTUITIVE THEORY OF RRS

Consider the system whose pertinent energy levels are illustrated in Fig. 1. Initially it is in level i; it then absorbs a photon from an incident monochromatic beam of frequency ν and is raised to level k, from which it drops to level l emitting a photon of frequency ν'. To be consistent with the use of A and B coefficients, it is assumed that the g-fold degeneracy of each level is rotational, and that initially all the g_i states of level i are equally populated. It is also assumed that all broadening of the levels is natural broadening, and that the separation of each of the three levels from all other levels of the system is sufficient for interference effects to be ignored.

In this treatment each level is considered to have a width and is described by a normalized natural line-shape factor; in addition, energy is rigidly conserved. If RRS is considered to be the same as AE, then the cross section $\sigma(\nu, \nu')$ for a photon of frequency ν to be scattered as a photon of frequency ν' and the system to shift from level i to level l, is given intuitively by

$$\sigma(\nu, \nu') = \left(\frac{1}{I(\nu)/h\nu}\right) \int_0^\infty dE \left[\frac{1}{h} \frac{\Gamma_i/\pi}{[(E-E_i)/h]^2 + \Gamma_i^2}\right] \left[I(\nu) B_{ik} \frac{\Gamma_k/\pi}{[(E_k - E)/h - \nu]^2 + \Gamma_k^2}\right] \left[A_{kl}\tau_k \frac{\Gamma_l/\pi}{[(E_l - E)/h - \nu + \nu']^2 + \Gamma_l^2}\right] . \tag{1}$$

Here $I(\nu)$ is the intensity of the incident photon beam, E_a is the line center energy of level a, and $\Gamma_a = 1/4\pi\tau_a$, where τ_a is the lifetime of level a and $a = i, k, l$; A and B are the Einstein coefficients which

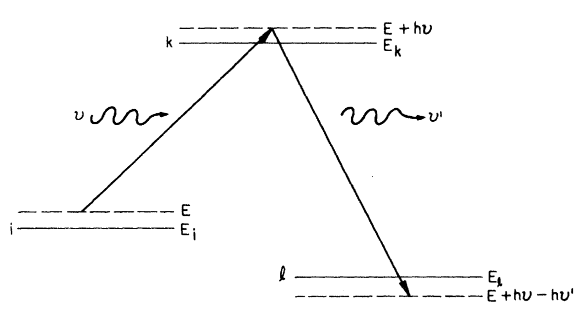

FIG. 1. System interacting with an exciting field of frequency ν and a scattered field of frequency ν'.

satisfy the relationships

$$B_{ki} = [c^2/8\pi h(\nu_{ki})^3]A_{ki} = (g_i/g_k)B_{ik} ,$$

where $\nu_{ki} = (E_k - E_i)/h$ and g_i and g_k are the degeneracy factors for levels i and k. The first large parentheses in Eq. (1) is the inverse of the incident photon flux. This factor, multiplied by the rate at which photons of frequency ν' are produced per unit frequency, gives, by definition, the scattering cross section $\sigma(\nu, \nu')$; the integral in Eq. (1) gives this rate. The first large square brackets in the integral, multiplied by dE, is the normalized probability that the system initially had an energy E in the interval dE, $E - E_i$ from level center. It applies when level i is populated by a broadband process where collisional broadening can be neglected (flashlamp pumping, for example). The second large square brackets in the integral is the rate at which photons of frequency ν are absorbed, thus exciting the system with the given initial energy E from level i to level k and to a consequent energy $E_k - E - h\nu$ from the center of level k. The third large square brackets is the probability per unit frequency that a photon of frequency ν' is emitted, relaxing the system, now with an initial energy $E + h\nu$, from level k to level l and to a corresponding energy which is $E_l - E - h\nu + h\nu'$ from the center of level l. Note that $A_{kl}\tau_k$ is the probability of a transition to level l given that the atom was initially in level k.

By replacing the lower limit of the integral by $E = -\infty$, the integral can be evaluated by the method of residues,[10]

$$\sigma(\nu, \nu') = h\nu B_{ik} A_{kl} \tau_k F(\nu, \nu') \simeq h\nu_{ki} B_{ik} A_{kl} \tau_k F(\nu, \nu') \ , \tag{2}$$

where

$$\begin{aligned} F(\nu, \nu') = {} & \frac{4\Gamma_i \Gamma_k \Gamma_l (\Gamma_i + \Gamma_l + \Gamma_k)}{\pi^2} \frac{1}{[\Gamma_{ik}^2 + (\nu - \nu_{ki})^2][\Gamma_{kl}^2 + (\nu_{kl} - \nu')^2][\Gamma_{li}^2 + (\nu_{li} + \nu' - \nu)^2]} \\ & + \frac{\Gamma_k \Gamma_l}{\pi^2} \frac{1}{[\Gamma_{ki}^2 + (\nu - \nu_{ki})^2][\Gamma_{li}^2 + (\nu_{li} + \nu' - \nu)^2]} + \frac{\Gamma_i \Gamma_k}{\pi^2} \frac{1}{[\Gamma_{kl}^2 + (\nu_{kl} - \nu')^2][\Gamma_{li}^2 + (\nu_{li} + \nu' - \nu)^2]} \\ & + \frac{\Gamma_l \Gamma_i}{\pi^2} \frac{1}{[\Gamma_{kl}^2 + (\nu_{kl} - \nu')^2][\Gamma_{ik}^2 + (\nu - \nu_{ki})^2]} \end{aligned} \tag{3}$$

and $\Gamma_{ab} = \Gamma_a + \Gamma_b$, where $ab = ik, kl, li$.

In quantum mechanics, the cross section for the scattering of monochromatic light near a resonance is derived from the same formalism which describes ordinary Raman or Rayleigh scattering[11]; indeed, the cross section is merely the resonant term in the expression for ordinary Raman scattering. For weak fields, the differential scattering cross section is given by[12–14]

$$\left.\frac{d\sigma(\nu, \nu')}{d\Omega}\right|_{i, J_i, m_i \to l, J_l, m_l} = \frac{\pi(2\pi)^4(\nu_{kl})^3 \nu_{ki}}{h^2 \Gamma_k c^4} \left| \sum_{m_k} \langle iJ_i m_i | \vec{\mathrm{P}} \circ \vec{\lambda} | kJ_k m_k \rangle \langle kJ_k m_k | \vec{\mathrm{P}} \circ \vec{\lambda}' | lJ_l m_l \rangle \right|^2 F(\nu, \nu') \ , \tag{4}$$

where J is the total angular momentum quantum number of the appropriate level, m is the z-component quantum number characterizing each state of the level, $\vec{\mathrm{P}}$ is the dipole moment operator, $\vec{\lambda}$ is the polarization of the incident beam, $\vec{\lambda}'$ is the polarization of the scattered beam, and $F(\nu, \nu')$ is the same as in Eq. (3).

By definition

$$\sigma(\nu, \nu') = \frac{1}{2J_i + 1} \sum_{m_i, m_l, \vec{\lambda}'} \int d\Omega \left.\frac{d\sigma(\nu, \nu')}{d\Omega}\right|_{i, J_i, m_i \to l, J_l, m_l} \ . \tag{5}$$

Let us choose the polarization of the incident beam such that it is along the z axis ($\vec{\lambda} = \vec{u}_z$). Then, summing over m_k in Eq. (4), only the term with $m_k = m_i$ is nonzero.[15] By definition[15]

$$\sum_{\vec{\lambda}', m_l} \int d\Omega \, |\langle kJ_k m_k | \vec{\mathrm{P}} \cdot \vec{\lambda}' | lJ_l m_l \rangle|^2 = \frac{c^3 h}{\nu_{kl}^3 8\pi^3} A_{kl} \tag{6}$$

and

$$\sum_{m_i} |\langle iJ_i m_i | \vec{\mathrm{P}} \circ \vec{u}_z | kJ_k m_i \rangle|^2 = \sum_{m_i, m_k} |\langle iJ_i m_i | \vec{\mathrm{P}} \circ \vec{u}_z | kJ_k m_k \rangle|^2 = \frac{(2J_i + 1)h^2 c}{8\pi^3} B_{ik} \quad . \tag{7}$$

Thus Eq. (5) reduces to

$$\sigma(\nu, \nu') = h\nu_{ki} B_{ik} A_{ki} \tau_k F(\nu, \nu') \quad . \tag{8}$$

Equation (8) is derived in a purely mathematical way. Formally, all one knows is that it describes the scattering of light near a resonance. The question as to whether it describes RRS, or AE, is answered by the fact that Eq. (8) is identical to the intuitive AE result of Eq. (2). Clearly, the physical process involved is absorption followed by emission. Since Eq. (4) is the resonant term in the quantum-mechanical expression for Raman scattering, RRS is AE, there is only one physical phenomenon involved, and there is no distinction between RRS, AE, and HL, or between "real," "virtual," direct, and two-step transitions. In addition these conclusions are supported by the historical definition of AE.[16]

III. SPECIAL CONSIDERATIONS IN THE QUANTUM-MECHANICAL THEORY OF RRS

A. Damping approximations

We have shown that Ref. 1 was not incorrect in his conception of RRS and HL (or AE). Apart from conceptual considerations, however, we note that the cross section derived by Ref. 2 is different from Eq. (4) and predicts an incorrect phenomenological picture of resonant scattering. The analysis of Ref. 2 is based on the damping approximation form of the Schrödinger equation. This approximation also has been used in a number of other papers[17,18] to solve for the resonant scattering cross section. In those papers as well, the solutions do not resemble Eq. (4) and lead to the same controversial interpretation of resonant scattering as that given by Ref. 2. The question arises, then, as to the accuracy of the damping approximation. As we point out below, this question can be simply resolved.

Writing the state vector $|\psi\rangle$ of an atom-field system as $|\psi\rangle = \sum_{s,\alpha} s_\alpha |s; \alpha\rangle$, where $|s\rangle$ is an eigenstate of energy $\hbar\omega_s$ of the free atom and $|\alpha\rangle$ is an eigenstate of energy ϵ_α of the free field, the Schrödinger equation becomes

$$\dot{s}_\alpha = -i(\omega_s + \epsilon_\alpha/\hbar)s_\alpha - \frac{i}{\hbar} \sum_{s', \alpha'} \langle s; \alpha | H | s'; \alpha' \rangle s'_{\alpha'} \,. \tag{9}$$

Here, H is the interaction Hamiltonian between the atom and the field.

If the atom is initially in the nondegenerate level a and interacts with a monochromatic field of frequency ω, wave vector $\vec{k}$, and polarization $\vec{\lambda}$ (corresponding to the state $|a; n_{k\lambda}\rangle$ at $t = 0$), and there exists one other nondegenerate level b such that $\omega \simeq \omega_b - \omega_a$, then, ignoring, as do Refs. 2, 17, and 18, the nonresonant interactions of the applied field (the rotating-wave approximation) and the two-photon transition component of H, Eq. (9) becomes

$$\dot{a}_n = -i(\omega_a + n\omega)a_n - \frac{i}{\hbar} H^{\omega}_{ab} b_{n-1} - \frac{i}{\hbar} \sum_{c', \sigma'} H^{\sigma'}_{ac'} c'_{n, 1\sigma'} , \tag{10a}$$

$$\dot{b}_{n-1} = -i[\omega_b + (n-1)\omega] b_{n-1} - \frac{i}{\hbar} H^{\omega *}_{ab} a_n - \frac{i}{\hbar} \sum_{c, \sigma} H^{\sigma}_{bc} c_{n-1, 1\sigma} , \tag{10b}$$

$$\dot{c}_{n-1, 1\sigma} = -i[\omega_c + (n-1)\omega - \omega_\sigma] c_{n-1, 1\sigma} - \frac{i}{\hbar} H^{\sigma *}_{bc} b_{n-1} - \frac{i}{\hbar} \sum_{c'', \beta} H^{\beta}_{cc''} c''_{n-1, 1\sigma, 1\beta} \,. \tag{10c}$$

Here a_n is the amplitude of the state $|a; n_{k\lambda}\rangle$, b_{n-1} is the amplitude of state $|b; (n-1)_{k\lambda}\rangle$, and $c_{n-1,1\sigma}$ is the amplitude of the state $|c; (n-1)_{k\lambda}, 1_{k_\sigma \lambda_\sigma}\rangle$; the three states are the initial, intermediate, and final states, respectively, in a resonant scattering process.

In the damping approximation[2,17–19] Eqs. (10) are written

$$\mathring{a}_n = -i(\omega_a + n\omega)a_n - \frac{i}{\hbar} H^{\omega}_{ab} b_{n-1} - \tfrac{1}{2}\Gamma_a a_n , \tag{11a}$$

$$\dot{b}_{n-1} = -i[\omega_b + (n-1)\omega] b_{n-1} - \frac{i}{\hbar} H^{\omega *}_{ab} a_n - \tfrac{1}{2}\Gamma_b b_{n-1} , \tag{11b}$$

$$\dot{c}_{n-1, 1\sigma} = -i[\omega_c + (n-1)\omega - \omega_\sigma] c_{n-1, 1\sigma} - \frac{i}{\hbar} H^{\sigma *}_{bc} b_{n-1} - \tfrac{1}{2}\Gamma_c c_{n-1, 1\sigma} , \tag{11c}$$

where Γ_i is chosen to be the inverse of the natural lifetime of level i.

Equations (11) are the same as those used by Refs. 17 and 18 and can be readily converted in the collisionless case, the case considered here, to the equations for the matrix elements of the density operator ρ used in Ref. 2. However, the formula for calculating P_σ, the probability that a photon of frequency ω_σ is produced in the resonant scattering process $a \rightarrow b \rightarrow c$, used by Ref. 2 is not the same as that used by Refs. 17 and 18. It can be shown, however, that the two formulas yield identical results.

The probability that a photon of frequency ω_σ, wave vector $\vec{k}_\sigma$, and polarization $\vec{\lambda}_\sigma$ is created in the scattering process is, according to one view,[2,20] equal to the increase in energy of the corresponding field mode divided by $\hbar\omega_\sigma$, the energy of one photon in that mode. The increase in energy is equated to the time integral of minus the expectation value of the rate at which the vacuum field corresponding to that mode does work on the material system. The rate of work R is given by $\vec{E}_\sigma \cdot \sum_i q_i \vec{p}_i / m_i$ where $\vec{E}_\sigma$ is the field operator of the σ mode and q_i, $\vec{p}_i$, and m_i are, respectively, the charge, momentum operator, and mass of the ith constituent particle of the system. Since[21] $[\vec{r}_i, H_0] = i\hbar\vec{p}_i/m_i$ where $\vec{r}_i$ is the position of the ith particle and H_0 is the Hamiltonian of the free atom, the rate can be written as $\vec{E}_\sigma \cdot [\vec{D}, H_0]/i\hbar$ where the dipole $\vec{D} = \sum_i q_i \vec{r}_i$. Using Eqs. (11) and making the rotating-wave approximation and the dipole approximation,[21] $H = -\vec{D}\cdot\vec{E} = -\vec{D}\cdot\sum_\mu \vec{E}_\mu$ where $\vec{E}$ is the operator of the electric field and μ denotes the different modes of the field,

$$P_\sigma = -\frac{1}{\hbar\omega_\sigma}\int_0^\infty dt\,\langle R\rangle = \frac{i}{\hbar^2\omega_\sigma}\int_0^\infty dt\,\langle\Psi|\vec{E}_\sigma\cdot[\vec{D}, H_0]|\Psi\rangle = \frac{i(\omega_b-\omega_c)}{\hbar\omega_\sigma}\int_0^\infty dt\,(H^\sigma_{bc}b^*_{n-1}c_{n-1,1\sigma} - H^{\sigma*}_{bc}b_{n-1}c^*_{n-1,1\sigma})$$

$$= \frac{2(\omega_b-\omega_c)}{\hbar\,\omega_\sigma}\int_0^\infty dt\,\mathrm{Im}(H^{\sigma*}_{bc}b_{n-1}c^*_{n-1,1\sigma})\,, \tag{12a}$$

$$P_\sigma = \frac{2(\omega_b-\omega_c)}{\hbar\omega_\sigma}\int_0^\infty dt\,\mathrm{Im}\{[\dot{c}_{n-1,1\sigma} + (i[\omega_c + (n-1)\omega - \omega_\sigma] + \tfrac{1}{2}\Gamma_c)c_{n-1,1\sigma}]i\hbar c^*_{n-1,1\sigma}\}$$

$$= \frac{\omega_b-\omega_c}{\omega_\sigma}\Gamma_c\int_0^\infty dt\,|c_{n-1,1\sigma}|^2 + \frac{\omega_b-\omega_c}{\omega_\sigma}\int_0^\infty dt\frac{d}{dt}|c_{n-1,1\sigma}|^2 = \frac{\omega_b-\omega_c}{\omega_\sigma}\Gamma_c\int_0^\infty dt\,|c_{n-1,1\sigma}|^2 \simeq \Gamma_c\int_0^\infty dt\,|c_{n-1,1\sigma}|^2\,. \tag{12b}$$

Equation (12a) is essentially the expression for P_σ used by Ref. 2, and Eq. (12b) is the expression proffered by Refs. 17 and 18. Equation (12b) is easily recognized as the intuitive expression for the integrated rate at which the system leaves the state $|c; (n-1)_{k\lambda}, 1_{k_\sigma\lambda_\sigma}\rangle$. This obviously is also the probability the system was ever in that state and thus equals P_σ.

Equations (11) and (12b) can be easily solved for a_n, b_{n-1}, $c_{n-1,1\sigma}$, and P_σ with the result that b_{n-1}, for example, is given by[17,18]

$$b_{n-1} = \frac{-(i/\hbar)H^{\omega*}_{ab}}{s_1 - s_2}(e^{s_1 t} - e^{s_2 t})\,e^{-i(n\omega+\omega_a)t}\,, \tag{13a}$$

$$s_{1,2} = -\frac{1}{2}\left(\frac{\Gamma_a+\Gamma_b}{2} - i(\omega-\omega_{ba})\right) \pm \left[\frac{1}{4}\left(\frac{\Gamma_a-\Gamma_b}{2} + i(\omega-\omega_{ba})\right)^2 - |H^\omega_{ab}|^2/\hbar^2\right]^{1/2} \tag{13b}$$

and P_σ, for the case of a weak applied field, by[17]

$$P_\sigma = \frac{|H^\omega_{ab}|^2|H^\sigma_{bc}|^2/\hbar^4}{(\frac{1}{2}\Gamma_a - \frac{1}{2}\Gamma_b)^2 + (\omega-\omega_{ba})^2}\left\{\frac{\Gamma_b+\Gamma_c}{\Gamma_b}\,\frac{1}{(\omega_\sigma-\omega_{bc})^2 + [\frac{1}{2}(\Gamma_b+\Gamma_c)]^2} + \frac{\Gamma_c+\Gamma_a}{\Gamma_a}\,\frac{1}{(\omega_\sigma-\omega-\omega_{ac})^2 + [\frac{1}{2}(\Gamma_a+\Gamma_c)]^2}\right.$$

$$\left. - 2\mathrm{Re}\left(\frac{1}{\frac{1}{2}(\Gamma_a+\Gamma_b) - i(\omega-\omega_{ba})}\,\frac{1}{\frac{1}{2}(\Gamma_b+\Gamma_c) - i(\omega_\sigma-\omega_{bc})} + \frac{1}{\frac{1}{2}(\Gamma_a+\Gamma_c) + i(\omega_\sigma-\omega-\omega_{ac})}\right)\right\}, \tag{14}$$

where $\omega_{ik} = \omega_i - \omega_k$. Moreover, to lowest order in H^ω_{ab}

$$s_1 = -\tfrac{1}{2}\Gamma_b + i(\omega-\omega_{ba}) - \frac{2|H^\omega_{ab}|^2/\hbar^2}{\frac{1}{2}(\Gamma_a-\Gamma_b) + i(\omega-\omega_{ba})}\,, \tag{15a}$$

$$s_2 = -\tfrac{1}{2}\Gamma_a + \frac{2|H^\omega_{ab}|^2/\hbar^2}{\frac{1}{2}(\Gamma_a-\Gamma_b) + i(\omega-\omega_{ba})}\,. \tag{15b}$$

Equation (14) is the same as the solutions of Refs. 17 and 18 and corresponds to the solution of Ref. 2, which is a low-order perturbation theory solution of Eqs. (11). The most important feature of Eq. (14) and the solution of Ref. 2 is the way they appear to divide into terms describing two distinct phenomena. The first term in Eq. (14) and the corresponding second term in Eq. (6) of Ref. 2 is said to describe a two-step[2] or stepwise[17] absorption followed by emission, and the second term in Eq. (14) and the first term in Eq. (6) of Ref. 2 an instantaneous and direct,[2] or two-quantum[17] transition. The third term in Eq. (14) is said to describe interference between the two phenomena.[17]

Pursuant to this matter, we remark that Eq. (14) can be arranged into a form identical to that of Eq. (4). Specifically, if Eq. (14) is divided by the incident photon flux and multiplied by Γ_a in order to convert the probability into a cross section, and then multiplied by the number of states in a unit interval of frequency and solid angle, it then can be converted algebraically into Eq. (4) replacing i, k, l by a, b, c, respectively. This step unambiguously resolves the question of interpretation, for it is clearly seen that the damping approximation does not lead to solutions which describe resonant scattering as the combination of two separate phenomena. Equation (14) is merely one of many possible algebraic forms; the two terms in Eq. (14) simply indicate the existence of maxima[7] in the probability of AE.

It should be pointed out, however, that there is one case for which, even in weak fields, resonant scattering should not be treated as a simple absorption followed by emission, namely, scattering to and from the atomic ground state. In this case radiative corrections are very important, in so far as obtaining an accurate solution, and it can be seen[13,22] that it is not possible to obtain an accurate expression for P_σ unless the process is treated as successive correlated absorptions and emissions of many photons. In fact, Eqs. (11) require modification[22] to take account of this special case.

Algebraic manipulation eliminates the questionable[23-25] subtracting linewidths in Eq. (14) but they still appear in Eqs. (13b) and (15) which describe the theoretical splitting or shift in frequency and change in line shape of emission from levels a and b in an applied field. However, there is no immediate reason to believe that the subtracting linewidths are incorrect and, in fact, for the case for which Γ_a and Γ_b tend to zero, Eqs. (15) give correct descriptions (insofar as nonresonant processes can be ignored[26]) of the ac Stark shift of levels a and b.[26] Moreover, for the case where $\Gamma_c = 0$, we have compared the solution of Eqs. (11) and (12b), with radiative correction terms retained, to a solution based on Heitler-Ma formalism[27] and found that the two solutions are identical, including the appearance of subtracting linewidths in the radiative correction terms.

B. Scattering time

The claims that RRS, as opposed to HL (or AE), is essentially instantaneous[2,4] and that this is the fundamental distinction between RRS and HL[2] or that RRS can be deduced[3] from the uncertainty principle to have a scattering time t_s given approximately by $1/|\nu - \nu_{ki}|$ are incorrect. The correct expression for t_s is determined by considering the scattering of a photon wave packet near a resonance.[28] The expression is similar to the expression derived[29] for the resonant scattering of a nonrelativistic wave packet. For $\nu = \nu_{ki}$, t_s is the natural lifetime of the transition, but as ν moves off resonance, t_s tends to zero. This decrease in t_s can be demonstrated experimentally[30] but a direct measurement of $t_s(\nu)$ off resonance is not possible[31] because it would require so short a light pulse that the resulting spread in its bandwidth would overlap line center, in which case a composite lifetime on the order of the natural lifetime would be observed.

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[5] Reference 2 does not specifically indicate acceptance of Klein's definition of HL as AE. However, this point is irrelevant since, as we emphasize, there is only one phenomenon involved, not two or three distinct physical phenomena (RRS and HL or RRS and AE), no matter how they are defined.

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