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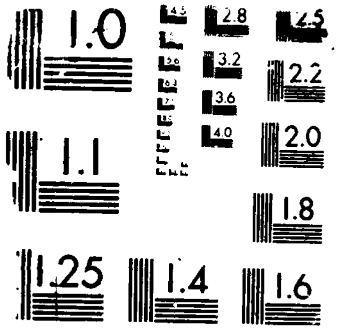

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Correlations Between Photons in Resonance Fluorescence Emitted by an Atom Near a Metal Surface

by

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in

Physical Review A

Departments of Chemistry and Physics
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CORRELATIONS BETWEEN PHOTONS IN RESONANCE FLUORESCENCE EMITTED BY AN ATOM NEAR A METAL SURFACE

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ABSTRACT

The photon correlations of resonance fluorescence emitted by a degenerate two-level atom near a metal surface are studied. An intense non-monochromatic optical laser drives a low-lying atomic transition, and the emitted fluorescence is detected by a photon counter. An expression for the spontaneous-decay operator is derived, and it appears that the various substates of the upper level have a different lifetime due to the presence of the surface, or equivalently, due to the loss of spherical symmetry. The temporal photon correlation function is then evaluated in terms of a photon-emission operator and the time-evolution operator for the atomic density matrix. The general theory is exemplified with a detailed elaboration of the $j_g = 0 \rightarrow j_e = 1$ transition, for which the two-photon correlation is obtained explicitly as a function of the lifetime A(h), depending on the atom-surface distance h, the laser linewidth, the detuning from resonance, and the dipole-coupling strength with the external field.

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I. INTRODUCTION

Irradiation of an atom by an optical laser with a frequency ω_L in close resonance with an electronic transition frequency (level separation) will cause the atom to absorb photons from the incident field. Subsequent spontaneous decay is accompanied by the emission of fluorescence radiation, which is amenable to observation in an experiment. Detailed properties of the scattered radiation, like its intensity, polarization, spectral profile and temporal photon correlations reflect the dynamical evolution of the atom in its environment. A generic method to obtain information about interatomic potentials, for instance, is by measuring the line shape of the fluorescence, which is emitted during a collision. In a similar way it should be feasible to study the behavior of an atom in the vicinity of a surface, or the alterations in its optical properties due to the presence of an optically-active substrate.

Resonance fluorescence of a free atom (gas phase or atomic beam) in a strong external field has been studied extensively during the last two decades. Theory[1-3] predicted a three-line spectrum for a two-state atom, which was confirmed by experiments.[4-6] The non-classical nature of fluorescence radiation was established by the observation of antibunching[7-9] and sub-poissonian statistics[10] in the temporal distribution of the photon emissions. Also combined time- and frequency-resolved photon correlations were found to be in excellent agreement with theory.[11-16] As stated in the first paragraph, the characteristics of the fluorescence are affected by the atom's surroundings. Most notably is the change in lifetime of an excited atomic state due to confining boundaries for the radiation, like in a cavity,[17] near a surface,[18] or in a waveguide.[19] In a series of experiments[20-24] with molecular dye films on a dielectric substrate, the theoretically-predicted[25] dependence of the decay constants (inverse lifetimes) on the distance of the molecule to the surface was found indeed. In this paper we consider an atom near a metal surface (with perfect conductivity), which is illuminated by a strong non-monochromatic laser beam, and we study the correlations between the emitted fluorescent photons. Recently, the spectral distribution of the fluorescence was calculated[26,27] for the same configuration, in the case where the atom could be accounted for by a two-state model, and where a semiclassical description of the emission process was adopted.

II. DRIVEN ATOM

An atom is situated at $\mathbf{r} = h\mathbf{e}_z$, $h > 0$, above the xy-plane, which separates the vacuum $z > 0$ from a perfectly-conducting substrate $z < 0$. This system is irradiated by a laser, with an electric-field component

$$\mathbf{E}_i(\mathbf{r},t) = E_o \mathrm{Re}\, \boldsymbol{\varepsilon}\, e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_L t-\phi(t))} \quad , \qquad (2.1)$$

where E_o is the amplitude, $\boldsymbol{\varepsilon}$ the normalized polarization vector ($\boldsymbol{\varepsilon}\cdot\boldsymbol{\varepsilon}^* = 1$), $\mathbf{k}$ the wave vector, perpendicular to $\boldsymbol{\varepsilon}$ ($\mathbf{k}\cdot\boldsymbol{\varepsilon} = 0$), and $\phi(t)$ a stochastically fluctuating phase, which gives rise to the laser linewidth. At the surface this incident field is reflected, and the sum field, evaluated at the position of the atom becomes

$$\mathbf{E}_L(t) = 2E_o \mathrm{Re}\, e^{-i(\omega_L t+\phi(t))} [\boldsymbol{\varepsilon}_\perp \cos(hk_z) + i\boldsymbol{\varepsilon}_\parallel \sin(hk_z)] \quad , \qquad (2.2)$$

where the polarization vector is divided into perpendicular and parallel components with respect to the xy-plane, and $k_z = \mathbf{k}\cdot\mathbf{e}_z$ is the z-component of the wave vector. For solids with a finite penetrability (dielectrics), the factor in square brackets is different, but the time dependence of this external field remains the same.

The laser frequency ω_L is assumed to be tuned in close resonance with the level separation $\omega_o = \omega_e - \omega_g > 0$ between two (possibly degenerate) levels with energy $\hbar\omega_e$ (excited) and $\hbar\omega_g$ (ground). If we suppress irrelevant quantum numbers, we can write the atomic wave functions as $|j_e m_e>$ and $|j_g m_g>$, with j and m the angular-momentum quantum numbers. In terms of the projectors

$$P_e = \sum_{m_e} |j_e m_e><j_e m_e| \qquad (2.3)$$

$$P_g = \sum_{m_g} |j_g m_g><j_g m_g| \qquad (2.4)$$

onto the multiplets, the atomic Hamiltonian can be represented by

$$H_a = \hbar\omega_e P_e + \hbar\omega_g P_g \quad . \qquad (2.5)$$

Coupling of the atomic dipole $\underset{\sim}{\mu}$ to the external field is governed by the interaction Hamiltonian

$$-\underset{\sim}{\mu}\cdot\underset{\sim}{E}_L(t) = -\tfrac{1}{2}\hbar\Omega(h)e^{-i(\omega_L t+\phi(t))} + \text{H.c.} \quad , \qquad (2.6)$$

where we made the rotating-wave approximation and introduced a 'Rabi operator'

$$\Omega(h) = 2(E_o/\hbar)P_e[\underset{\sim}{\mu}\cdot\underset{\sim}{\varepsilon}_\perp\cos(hk_z) + i\underset{\sim}{\mu}\cdot\underset{\sim}{\varepsilon}_\parallel\sin(hk_z)]P_g \quad , \qquad (2.7)$$

which generalizes the Rabi frequency (a real number) for non-degenerate atoms in free space.[28] Notice that $\Omega(h)$ is not Hermitian and that it depends explicitly on the normal distance h of the atom to the surface. A convenient concept will turn out to be the effective (non-Hermitian) surface-dipole moment $\underset{\sim}{\mu}_s(x)$, defined as

$$\underset{\sim}{\mu}_s(x) = 2(\cos(x)\underset{\sim}{\mu}_\perp + i\sin(x)\underset{\sim}{\mu}_\parallel) \quad , \qquad (2.8)$$

where x is a dimensionless normal-distance parameter. Then the Rabi operator becomes

$$\Omega(h) = (E_o/\hbar)\, P_e(\underset{\sim}{\mu}_s(hk_z)\cdot\underset{\sim}{\varepsilon})P_g \quad . \tag{2.9}$$

The equation of motion for the density operator $\rho(t)$ of the atom in the laser field and near the surface is

$$i\hbar\frac{d\rho}{dt} = [H_a - \underset{\sim}{\mu}\cdot\underset{\sim}{E}_L(t),\rho] - i\hbar\Gamma(h)\rho \quad , \tag{2.10}$$

where the Liouville operator $\Gamma(h)$ accounts for the spontaneous decay. Its definition will be elaborated in the next section. We mention that $\Gamma(h)$ depends on h, which embodies the second geometry dependence of the equation of motion for $\rho(t)$.

III. SPONTANEOUS DECAY

Due to the coupling of the atomic dipole $\underset{\sim}{\mu}$ to the vacuum of the electromagnetic field, the atom decays from its excited state to its ground state, together with the emission of a fluorescent photon. In a previous paper[29] we evaluated the atomic relaxation operator $\Gamma(h)$ for an arbitrary atom with degenerate levels and near a perfect conductor. For optical transitions we can make the secular approximation[30] (sometimes called 'elimination of fast-oscillating terms'[31]) and neglect the imaginary parts of the relaxation constants. If we subsequently restrict the number of atomic levels to two, which are coupled by the laser, then $\Gamma(h)$ acquires the explicit form

$$\Gamma(h)\sigma = \tfrac{1}{2}\sum_{m_e m_g m_e'} \Big(c_{m_e m_g m_g m_e'}(h)|j_e m_e\rangle\langle j_e m_e'|\sigma + c^{*}_{m_e' m_g m_g m_e}(h)\sigma|j_e m_e\rangle\langle j_e m_e'|\Big)$$

$$- \frac{1}{2} \sum_{m_e m_g m'_e m'_g} \left(c_{m'_e m'_g m_g m_e}(h) + c^{*}_{m_e m_g m'_g m'_e}(h)\right) |j_g m_g\rangle\langle j_g m'_g| \langle j_e m_e|\sigma|j_e m'_e\rangle \quad , \tag{3.1}$$

which defines its action on an arbitrary density operator σ. Through the functions

$$b_{\perp}(x) = 1-3\left[\frac{\cos(2x)}{(2x)^2} - \frac{\sin(2x)}{(2x)^3}\right] \quad , \tag{3.2}$$

$$b_{\parallel}(x) = 1 - \frac{3}{2}\left[\frac{\sin(2x)}{2x} + \frac{\cos(2x)}{(2x)^2} - \frac{\sin(2x)}{(2x)^3}\right] \quad , \tag{3.3}$$

the relaxation constants can be expressed in terms of the matrix elements of the perpendicular and parallel components of the dipole moment, according to

$$\begin{aligned} c_{m_e m_g m'_g m'_e}(h) &= \frac{\omega_o^3}{3\pi\hbar\varepsilon_o c^3} \\ &\times [b_{\perp}(\omega_o h/c)\langle j_e m_e|\underset{\sim}{\mu}_{\perp}|j_g m_g\rangle \cdot \langle j_g m'_g|\underset{\sim}{\mu}_{\perp}|j_e m'_e\rangle \\ &+ b_{\parallel}(\omega_o h/c)\langle j_e m_e|\underset{\sim}{\mu}_{\parallel}|j_g m_g\rangle \cdot \langle j_g m'_g|\underset{\sim}{\mu}_{\parallel}|j_e m'_e\rangle] \quad . \end{aligned} \tag{3.4}$$

The presence of the surface is fully incorporated in the functions $b_{\perp}$ and $b_{\parallel}$, which approach unity for $h \to \infty$.

Next we take advantage of the rotational properties of the atomic wave functions and the fact that $\underset{\sim}{\mu}$ is a vector operator. With respect to the spherical unit vectors

$$\underset{\sim}{e}_0 = \underset{\sim}{e}_z \quad , \quad \underset{\sim}{e}_{\pm 1} = \mp(\underset{\sim}{e}_x \pm i\underset{\sim}{e}_y)/\sqrt{2} \quad , \tag{3.5}$$

we can decompose $\underset{\sim}{\mu} = \underset{\sim}{\mu}_{\perp} + \underset{\sim}{\mu}_{\parallel}$ as

$$\underset{\sim}{\mu}_{\perp} = \mu_0 \underset{\sim}{e}_z \quad , \quad \underset{\sim}{\mu}_{\parallel} = \sum_{\tau=\pm 1} \mu_{\tau} \underset{\sim}{e}^{*}_{\tau} \quad , \tag{3.6}$$

and from $\underset{\sim}{\mu}^{\dagger} = \underset{\sim}{\mu}$ it follows that

$$\mu_{\tau}^{\dagger} = (-1)^{\tau}\ \mu_{-\tau} \quad . \tag{3.7}$$

Then we write $b_0 = b_{\perp}$ and $b_{\pm 1} = b_{\parallel}$, which enables us to cast Eq. (3.4) in the form

$$c_{m_e m_g m_g' m_e'}(h) = \frac{\omega_o^3}{3\pi\hbar\varepsilon_o c^3} \times \sum_{\tau} b_{\tau}(\omega_o h/c)\langle j_e m_e|\mu_{\tau}|j_g m_g\rangle\langle j_g m_g'|\mu_{\tau}^{\dagger}|j_e m_e'\rangle \quad . \tag{3.8}$$

Matrix elements of μ_{τ} can be simplified with the Wigner-Eckart theorem, which states that[32]

$$\langle j_e m_e|\mu_{\tau}|j_g m_g\rangle = (j_g m_g 1\tau|j_e m_e)\langle j_e||\mu||j_g\rangle/\sqrt{2j_e+1} \quad , \tag{3.9}$$

and the matrix elements of $\mu_{\tau}^{\dagger}$ then follow from Eq. (3.7). Combining Eqs. (3.8) and (3.9) then yields

$$c_{m_e m_g m_g' m_e'}(h) = A_f \sum_{\tau} b_{\tau}(\omega_o h/c)(j_g m_g 1\tau|j_e m_e)(j_g m_g' 1\tau|j_e m_e') \quad , \tag{3.10}$$

where we introduced the Einstein coefficient A_f for a free atom (no surface)

$$A_f = \frac{\omega_o^3}{3\pi\hbar\varepsilon_o c^3}\ \frac{|\langle j_e||\mu||j_g\rangle|^2}{2j_e+1} \quad . \tag{3.11}$$

Since the Clebsch-Gordan coefficients are real, all relaxation constants $c_{m_e m_g m_g' m_e'}(h)$ are real. Expression (3.10) reveals the surface effect, incorporated in the parameter functions $b_{\tau}(\omega_o h/c)$ on the decay constants.

Insertion of Eq. (3.10) into Eq. (3.1) gives $\Gamma(h)$ in terms of A_f,h and geometrical Clebsch-Gordan coefficients. The coefficients $c_{m_e m_g m'_g m'_e}(h)$ are only non-zero if $m_g - m_e = m'_g - m'_e$, as follows from Eq. (3.10), so for $m_g = m'_g$ we have $m_e = m'_e$. Therefore the summation over m'_e in the first two terms of Eq. (3.1) is trivial, and furthermore we notice that the summation over m_g involves only the decay constants. With these observations the expression for $\Gamma(h)$ reduces to

$$\Gamma(h)\sigma = \tfrac{1}{2}\sum_{m_e} A_{m_e}(h)(|j_e m_e\rangle\langle j_e m_e|\sigma + \sigma|j_e m_e\rangle\langle j_e m_e|) - A_f \sum_{\tau} b_\tau(\omega_o h/c)$$

$$\sum_{\substack{m_e m_g \\ m'_e m'_g}} (j_g m_g 1\tau|j_e m_e)|j_g m_g\rangle\langle j_e m_e|\sigma|j_e m'_e\rangle\langle j_g m'_g|(j_g m'_g 1\tau|j_e m'_e) \quad , \qquad (3.12)$$

where $A_{m_e}(h)$ is defined by

$$A_{m_e}(h) = A_f \sum_{\tau} b_\tau(\omega_o h/c) \sum_{m_g} (j_g m_g 1\tau|j_e m_e)^2 \quad . \qquad (3.13)$$

From the equation of motion (2.8) we find that the population of level $|j_e m_e\rangle$ obeys

$$\frac{d}{dt}\langle j_e m_e|\rho(t)|j_e m_e\rangle = -\langle j_e m_e|(\Gamma(h)\rho(t))|j_e m_e\rangle \qquad (3.14)$$

in the absence of a laser field, and from Eq. (3.12) we obtain

$$\langle j_e m_e|(\Gamma(h)\rho(t))|j_e m_e\rangle = A_{m_e}(h)\langle j_e m_e|\rho(t)|j_e m_e\rangle \quad . \qquad (3.15)$$

Hence the population of $|j_e m_e\rangle$ decays exponentially, with an inverse lifetime equal to $A_{m_e}(h)$, which elucidates the significance of this parameter. Without a surface we have $b_\tau \equiv 1$, and the summations in Eq. (3.13) can be performed to give the result

$$A_{m_e}(h) = A_f \quad . \qquad (3.16)$$

Therefore, the population of an atomic level $|j_e m_e\rangle$ decays in free space with A_f, which is independent of m_e. This in turn expresses the rotational invariance of spontaneous decay for a free atom. Conversely, a preferred direction in space (the z-axis), which is imposed by the presence of the surface, destroys the rotational symmetry, which implies that the relaxation depends on the orientation of the atom (the m_e-value of the substate). From $b_\tau = b_{-\tau}$ we readily derive

$$A_{m_e}(h) = A_{-m_e}(h) \quad , \tag{3.17}$$

which expresses the remaining rotational symmetry about the z-axis.

IV. TRANSFORMATION AND AVERAGE

In the equation of motion (2.10) the Hamiltonian is explicitly time dependent through the external field $\underset{\sim}{E}_L(t)$. With the standard transformation[33] of the density operator

$$\rho'(t) = e^{-i(\omega_L t+\phi(t))L_g}\rho(t) \quad , \tag{4.1}$$

where L_g is the commutator with the projector on the ground state, e.g.,

$$L_g\sigma = [P_g,\sigma] \quad , \tag{4.2}$$

the time dependence is greatly simplified. We find the equation for $\rho'(t)$ to be

$$i\frac{d\rho'}{dt} = (L_d(h) + L_g\frac{d\phi}{dt} - i\Gamma(h))\rho' \tag{4.3}$$

in the compact Liouville notation. Oscillations with the optical frequency ω_L have disappeared, and the phase fluctuations only enter as a multiplicative factor $d\phi/dt$ to $\rho'(t)$, which is the standard form of a stochastic differential equation. Here the Liouvillian $L_d(h)$ is defined as

$$L_d(h)\sigma = \hbar^{-1}[H_d(h),\sigma] \tag{4.4}$$

in terms of the dressed-atom Hamiltonian

$$H_d(h) = H_a + \hbar\omega_L P_g - \tfrac{1}{2}\hbar(\Omega(h) + \Omega(h)^{\dagger}) \quad , \tag{4.5}$$

which has the significance of the atom, dressed with the photons of the laser and including the dipole interaction.[30]

Due to the term $L_g d\phi/dt$ in Eq. (4.3), the density operator $\rho'(t)$ is a stochastic process. Only its average

$$\Pi(t) = \langle\rho'(t)\rangle \tag{4.6}$$

has relevance, and the equation for $\Pi(t)$ reads[34]

$$i\frac{d\Pi}{dt} = (L_d(h) - iW - i\Gamma(h))\Pi \quad , \tag{4.7}$$

with

$$W = \lambda L_g^2 \tag{4.8}$$

in terms of the half-width at half maximum λ of the Lorentzian laser profile.

Both the laser linewidth (W) and spontaneous decay ($\Gamma(h)$) give rise to damping of the free evolution ($L_d(h)$) of the dressed atom. After a transient time of the order of $(\lambda + A_f)^{-1}$, following the switch-on of the laser, the atom will have reached a steady state. If we write

$$\overline{\Pi} = \lim_{t\to\infty} \Pi(t) \quad , \tag{4.9}$$

then this $\overline{\Pi}$ is the solution of

$$(L_d(h) - iW - i\Gamma(h))\overline{\Pi} = 0 \quad . \tag{4.10}$$

Since the external field is assumed to be a CW laser, we will always consider the atom to be in the stationary state $\overline{\Pi}$.

V. FLUORESCENCE

Continuous excitation of the upper states $|j_e m_e\rangle$ by the laser and subsequent spontaneous decay to the ground states $|j_g m_g\rangle$ goes together with the emission of a stationary (fluorescence) radiation field. This signal is detected by a photomultiplier tube (PM) at a distance r from the atom. In a previous paper[29] we derived the general expression for the fluorescence in the far field, which reads

$$\underset{\sim}{E}_f(\underset{\sim}{r},t) = \frac{L^2}{4\pi\varepsilon_o c^2 r}\,[\hat{\underset{\sim}{r}}\times(\underset{\sim}{\mu}(t+\tau-r/c) + \underset{\sim}{\mu}'(t-\tau-r/c))]\times\hat{\underset{\sim}{r}} \quad . \tag{5.1}$$

Here, $\hat{\underset{\sim}{r}} = \underset{\sim}{r}/r$ is the direction of propagation, and $\tau = \hat{\underset{\sim}{r}}\cdot\underset{\sim}{e}_z h/c$ equals half the delay time between the directly-emitted wave and the wave which is first reflected by the surface. The time dependence of the operator $\underset{\sim}{\mu}$ represents the Heisenberg picture according to

$$\underset{\sim}{\mu}(t) = e^{iL(t-t_o)}\,\underset{\sim}{\mu}(t_o) \tag{5.2}$$

for any time interval $t - t_o$ and in terms of $L\sigma = \hbar^{-1}[H,\sigma]$, with H the Hamiltonian of the entire system. If we divide the operator $\underset{\sim}{\mu}$ as $\underset{\sim}{\mu} = \underset{\sim}{\mu}_\perp + \underset{\sim}{\mu}_\parallel$, then $\underset{\sim}{\mu}'$ is defined as

$$\underset{\sim}{\mu}' = \underset{\sim}{\mu}_\perp - \underset{\sim}{\mu}_\parallel \quad . \tag{5.3}$$

VI. DETECTION

From Eq. (5.1) it follows that the polarization of the fluorescence is determined by the vector operators $\boldsymbol{\mu}$ and $\boldsymbol{\mu}'$, which have a different direction in space and a different retardation time. In order to keep track of the polarization properties of the radiation, we suppose that the radiation passes a polarizer, before entering the PM, which transmits radiation with polarization $\boldsymbol{\varepsilon}_p$. From the theory of photon detection by a PM,[35,36] we then know that the detector responds to the positive frequency part of the scalar quantity

$$\mathcal{E}(t) = \mathbf{E}_f(\mathbf{r},t)\cdot\boldsymbol{\varepsilon}_p^* \quad , \tag{6.1}$$

which is the $\boldsymbol{\varepsilon}_p$ component of the fluorescence. Then we position the polarizer in such a way that the angle of incidence is zero (e.g., $\hat{\mathbf{r}}\cdot\boldsymbol{\varepsilon}_p = 0$), and we suppress the overall retardation with r/c. We then find

$$\mathcal{E}(t) = \frac{L^2 e^{iLt}}{4\pi\varepsilon_o c^2 r}\,[e^{iL\tau}\,\boldsymbol{\mu} + e^{-iL\tau}\boldsymbol{\mu}']\cdot\boldsymbol{\varepsilon}_p^* \quad , \tag{6.2}$$

where $\boldsymbol{\mu}$ and $\boldsymbol{\mu}'$ are time independent operators (Schrödinger picture). Then we notice that the surface can only have a significant effect if the distance between the atom and the xy-plane is a few optical wavelengths. Hence the retardation τ in Eq. (6.2) is of the order of a few optical cycles ($\tau \sim \omega_o^{-1}$), which implies that we can neglect the interaction term with the laser in L in the exponentials. In physical terms this means that the laser cannot cause a transition between the upper and lower states on a time scale τ, which is completely justified for optical lasers. Furthermore, the L^2 is merely an overall factor, which hardly affects the time evolution of $\mathcal{E}(t)$. Therefore, we can safely make the substitution $L\sigma \rightarrow [H_a,\sigma]/\hbar$ in $\exp(\pm iL\tau)$ and L^2. Combining everything then yields for the positive frequency part of $\mathcal{E}(t)$

$$\mathcal{E}^{(+)}(t) = \frac{\omega_o^2}{4\pi\epsilon_o c^2 r} e^{iLt}\{P_g(\underset{\sim}{\mu}_s(\omega_o\tau)^\dagger\cdot\underset{\sim}{\epsilon}_p^*)P_e\} \quad . \tag{6.3}$$

VII. PHOTON CORRELATIONS

If the active part of the PM is exposed to electromagnetic radiation it will absorb photons from this incident field. Time correlations between various photons are most conveniently expressed in the function $I_2(t_1,t_2)dt_1dt_2$ which has, by definition, the significance of the probability for the detection of a photon in $[t_1,t_1+dt_1]$ together with a detection in $[t_2,t_2+dt_2]$, and irrespective of detections at other times. The basic relation between I_2 and the incident radiation is given by[35,36]

$$I_2(t_1,t_2) = \zeta^2\langle\mathcal{E}^{(-)}(t_1)\mathcal{E}^{(-)}(t_2)\mathcal{E}^{(+)}(t_2)\mathcal{E}^{(+)}(t_1)\rangle \quad , \tag{7.1}$$

for $t_2 \geq t_1$. Here, ζ is a detector parameter (depending on efficiency, aperture, etc.) and the angle brackets indicate a quantum average. For our case, where the laser field is taken as a classical wave with a stochastic phase, the brackets also include an average over the process $\phi(t)$. The quantity $\langle\ldots\rangle$ in Eq. (7.1) is usually referred to as the intensity correlation of the field. For quantum radiation, like fluorescence, the different factors in brackets do not commute, which gives rise to non-trivial correlations between the detections of photons.

It is a standard procedure to work out the intensity correlation and to express the time regression from t_1 to t_2 in the time-evolution operator for the density operator

$$U(t) = e^{-i(L_d(h)-iW-i\Gamma(h))t} \quad , \tag{7.2}$$

and the photon-emission operator R, defined by[37]

$$R\sigma = \mathcal{E}^{(+)}(0)\sigma\,\mathcal{E}^{(-)}(0) \quad . \tag{7.3}$$

If we assume that the atom has reached its steady state Π, then the intensity correlation function is found to be

$$I_2(t_1,t_1+\tau) = \zeta^2 \mathrm{Tr}RU(\tau)R\Pi = f(\tau)I \quad , \quad \tau \geq 0 \quad , \tag{7.4}$$

which defines the function $f(\tau)$. Here, I equals the uncorrelated intensity of the radiation, which can be expressed as

$$I = \zeta \mathrm{Tr}R\Pi \quad . \tag{7.5}$$

For a zero delay time τ we have $U(0) = 1$ and $f(0) = \zeta^2 \mathrm{Tr}R^2\Pi/I$. With Eq. (6.3) we obtain

$$R^2 = 0 \quad , \tag{7.6}$$

and therefore

$$f(0) = 0 \quad . \tag{7.7}$$

This famous relation for the detection of two fluorescent photons is termed antibunching.[38] In the case that the delay time τ is very long, we find

$$f(\infty) = I \quad , \tag{7.8}$$

which shows that the second detection is independent of the first one, e.g., the detections are uncorrelated. Equation (7.8) follows from the identity

$$\lim_{t\to\infty} U(t)\sigma = \Pi \mathrm{Tr}\sigma \tag{7.9}$$

for any σ, which states that every σ evolves to a unique steady state Π and that the trace of σ is conserved in the time evolution with $U(t)$.

With Eqs. (7.3), (6.3), (2.3) and (2.4) we find for the emission operator the general expression

$$R\sigma = \sum_{\substack{m_e m_g \\ m_e' m_g'}} |j_g m_g\rangle\langle j_g m_g'| \, \langle j_e m_e|\sigma|j_e m_e'\rangle$$

$$\times \langle j_e m_e|\underline{\mu}_s(\omega_o\tau)\cdot\underline{\varepsilon}_p|j_g m_g\rangle^* \, \langle j_e m_e'|\underline{\mu}_s(\omega_o\tau)\cdot\underline{\varepsilon}_p|j_g m_g'\rangle \quad , \tag{7.10}$$

where an overall factor $(\omega_o^2/4\pi\varepsilon_o c^2 r)^2$ is omitted, since it can be absorbed in the detector parameter ζ. Matrix elements of $\underline{\mu}_s(\omega_o\tau)$ can be evaluated explicitly with Eq. (3.9).

VIII. $\underline{j_g = 0 \rightarrow j_e = 1}$ TRANSITION

In order to study the effect of the presence of the metal surface on the photon correlations, we have to prescribe more details of the system. In the remainder of this paper we consider the important example $j_g = 0 \rightarrow j_e = 1$. For $j_g = 0$ we have $m_g = 0$ only, and in view of Eq. (3.9) this non-degenerate state couples only with $j_e = 1$, which is three-fold degenerate. We introduce the abbreviations

$$|j_g m_g\rangle = |0\ 0\rangle \rightarrow |g\rangle \quad , \tag{8.1}$$

$$|j_e m_e\rangle = |1\ m_e\rangle \rightarrow |\eta\rangle \quad , \quad \eta = m_e = -1,0,1 \quad . \tag{8.2}$$

Spontaneous decay of this atom is described by the Liouvillian $\Gamma(h)$ from Eq. (3.12), and the inverse lifetimes of the states $|\eta\rangle$ are given by Eq. (3.13). The occurring Clebsch-Gordan coefficients all have the form

$$(0\ 0\ 1\ \tau|1\ \eta) = \delta_{\eta\tau} \quad , \tag{8.3}$$

which yields for the relaxation constants

$$A_\eta(h) = A_f b_\eta(\omega_o h/c) \quad . \tag{8.4}$$

We observe that the substates $|\eta = \pm 1\rangle$ decay with $A_f b_\parallel(\omega_o h/c)$, which equals the inverse lifetime of a dipole, oriented parallel to the xy-plane. The substate $|\eta = 0\rangle$ has the Einstein coefficient $A_f b_\perp(\omega_o h/c)$, and therefore it decays as a perpendicular dipole. Notice that the differences in decay constants of the various substates are merely determined by the geometry of the system. A two-state model-atom treatment of fluorescence near a surface leaves the dipole direction, and thereby the lifetime of the excited state, undetermined. One can then choose the direction of the matrix element $\langle e|\underline{\mu}|g\rangle$ arbitrarily, and subsequently average results over a randomization of this direction. It should be obvious now that this is not a correct procedure.

The spontaneous-decay operator for this system attains the simple form

$$\Gamma(h)\sigma = \frac{1}{2}\sum_\eta A_\eta(h)(|\eta\rangle\langle\eta|\sigma + \sigma|\eta\rangle\langle\eta| - 2|g\rangle\langle g|\langle\eta|\sigma|\eta\rangle) \quad , \tag{8.5}$$

and it involves only the Einstein coefficients $A_\eta(h)$ for the three substates.

IX. ATOMIC STATE

In this section we expand the equation of motion (4.7) for the atomic state $\Pi(t)$ in matrix elements. The external field enters through the Rabi operator $\Omega(h)$, Eq. (2.9), and we shall abbreviate its non-zero matrix elements as

$$\Omega_\eta(h) = \langle\eta|\Omega(h)|g\rangle \quad , \tag{9.1}$$

which are explicitly

$$\Omega_0(h) = \frac{2}{\sqrt{3}}\frac{E_o}{\hbar}\langle 1||\mu||0\rangle \cos(hk_z)\underline{\epsilon}\cdot\underline{e}_0 \quad , \tag{9.2}$$

$$\Omega_\pm(h) = \frac{2}{\sqrt{3}}\frac{E_o}{\hbar}\langle 1||\mu||0\rangle i \sin(hk_z)\underline{\epsilon}\cdot\underline{e}^*_{\pm 1} \quad . \tag{9.3}$$

With the definitions of $L_d(h)$, W and $\Gamma(h)$ we then obtain

$$\frac{d}{dt}\langle\eta|\Pi|\eta'\rangle = -\tfrac{1}{2}(A_\eta + A_{\eta'})\langle\eta|\Pi|\eta'\rangle - \tfrac{1}{2}i(\Omega^*_{\eta'}\langle\eta|\Pi|g\rangle - \Omega_\eta\langle g|\Pi|\eta'\rangle) \quad , \tag{9.4}$$

$$\frac{d}{dt}\langle g|\Pi|g\rangle = \sum_\eta A_\eta\langle\eta|\Pi|\eta\rangle - \tfrac{1}{2}i\sum_\eta(\Omega_\eta\langle g|\Pi|\eta\rangle - \Omega^*_\eta\langle\eta|\Pi|g\rangle) \quad , \tag{9.5}$$

$$\frac{d}{dt}\langle\eta|\Pi|g\rangle = -(\tfrac{1}{2}A_\eta + \lambda - i\Delta)\langle\eta|\Pi|g\rangle + \tfrac{1}{2}i(\Omega_\eta\langle g|\Pi|g\rangle - \sum_{\eta'}\Omega_{\eta'}\langle\eta|\Pi|\eta'\rangle) \quad , \tag{9.6}$$

$$\frac{d}{dt}\langle g|\Pi|\eta\rangle = -(\tfrac{1}{2}A_\eta + \lambda + i\Delta)\langle g|\Pi|\eta\rangle - \tfrac{1}{2}i(\Omega^*_\eta\langle g|\Pi|g\rangle - \sum_{\eta'}\Omega^*_{\eta'}\langle\eta'|\Pi|\eta\rangle) \quad , \tag{9.7}$$

where the dependence of A_η and Ω_η on h is suppressed, and we introduced the detuning of the laser from resonance by

$$\Delta = \omega_L - \omega_o \quad . \tag{9.8}$$

If we make all the combinations with η and η', then Eqs. (9.4)-(9.7) constitute a set of 16 coupled first-order linear differential equations.

X. INTENSITY CORRELATIONS

For the $j_g = 0 \rightarrow j_e = 1$ transition, the photon-emission operator R, Eq. (7.10), reduces to

$$R\sigma = P_g \sum_{\eta\eta'} \langle\eta|\sigma|\eta'\rangle\langle\eta|\underset{\sim}{\mu}_s(\omega_o\tau)\cdot\underset{\sim}{\varepsilon}_p|g\rangle^*\langle\eta'|\underset{\sim}{\mu}_s(\omega_o\tau)\cdot\underset{\sim}{\varepsilon}_p|g\rangle \quad , \tag{10.1}$$

and the matrix elements of $\underset{\sim}{\mu}_s(\omega_o\tau)$ are

$$\langle\eta|\underset{\sim}{\mu}_s(\omega_o\tau)|g\rangle = [\cos(\omega_o\tau)\underset{\sim}{e}_0\delta_{\eta0} + i\,\sin(\omega_o\tau)\sum_{\eta'=\pm1} \underset{\sim}{e}^*_{-\eta'}\delta_{\eta\eta'}]\,\langle1||\mu||0\rangle\; 2/\sqrt{3} \quad . \tag{10.2}$$

We notice that the summation over η and η' in Eq. (10.1) is just a number (not an operator), and therefore $R\sigma$ is proportional to the projector P_g on the ground state for every σ. This reflects the fact that after every emission the atom is necessarily in its ground state $|g\rangle$. Since for $j_g = 0$ this state is non-degenerate, the final state after a photon emission is unique, or in other words, independent of the history of the atom. If the ground state were degenerate, then we observe from Eq. (7.10) that the density operator of the atom after emission is a superposition of (Liouville) substates $|j_gm_g\rangle\langle j_gm_g'|$, where the distribution over states depends on the matrix elements of σ, which equals the density operator just before the emission.

Combination of Eqs. (7.5) and (10.1) gives for the intensity of the fluorescence

$$I = \zeta \sum_{\eta} n_\eta|\langle\eta|\underset{\sim}{\mu}_s(\omega_o\tau)\cdot\underset{\sim}{\varepsilon}_p|g\rangle|^2 \quad , \tag{10.3}$$

in terms of the steady-state populations $n_\eta = \langle\eta|\Pi|\eta\rangle$ of the upper levels. The two-photon correlation f(t) then becomes

$$f(t) = \zeta \mathrm{Tr} R U(t) P_g \quad , \tag{10.4}$$

which involves only a single photon-emission operator R, rather than two as in Eq. (7.4). This is again due to the fact that the ground state is non-degenerate. After the first emission the density operator equals P_g, which is the right-most factor in Eq. (10.4).

Substitution of the exponential for U(t), Eq. (7.2), into Eq. (10.4) gives an explicit expression for the correlation function f(t). The Liouvillian $L_d(h) - iW - i\Gamma(h)$, however, is a 16 × 16 matrix, which makes the evaluation of the exponential intractable, in general. In most practical cases (see the next section), the computation of f(t) is facilitated if we first transform to the Laplace domain. With

$$\tilde{f}(s) = \int_0^\infty dt\, e^{-st} f(t) \quad , \tag{10.6}$$

we find immediately

$$\tilde{f}(s) = \zeta \mathrm{Tr} R \frac{1}{s + iL_d(h) + W + \Gamma(h)} P_g \quad , \tag{10.7}$$

which only requires the inversion of the matrix. Of course, the result must be transformed back to the time domain in order to achieve an explicit expression for f(t).

XI. POLARIZATION

To fix the geometry of the experiment, there are four directions which remain to be specified. These are: (1) the laser polarization $\underset{\sim}{\varepsilon}$, (2) the laser propagation direction $\underset{\sim}{\hat{k}}$ (restricted by $\underset{\sim}{\hat{k}} \cdot \underset{\sim}{\varepsilon} = 0$), (3) the observation direction $\underset{\sim}{\hat{r}}$ and (4) the polarization unit-vector $\underset{\sim}{\varepsilon}_p$ (restricted by $\underset{\sim}{\hat{r}} \cdot \underset{\sim}{\varepsilon}_p = 0$). We shall consider two complementary situations, for which we evaluate the two-photon correlation function explicitly.

(a) Linear polarization. The laser polarization is chosen as $\mathbf{\varepsilon} = \mathbf{e}_0 = \mathbf{e}_z$, for which the direction of propagation $\hat{\mathbf{k}}$ must be in the xy-plane. Then we have $k_z = 0$, and from Eqs. (9.2) and (9.3) we find that the only non-zero matrix element of the Rabi operator is

$$\Omega_0(h) = \frac{2}{\sqrt{3}} \frac{E_o}{\hbar} \langle 1 || \mu || 0 \rangle \quad , \tag{11.1}$$

which is independent of the distance h between the atom and the surface. From Eqs. (9.4)-(9.7) it follows that this external field couples only the ground state $|g\rangle$ with the upper state $|0\rangle$,which has the decay constant $A_0(h) = A_f b_\perp(\omega_o h/c)$. The PM will be positioned in such a place that it detects radiation, which is emitted along the surface, e.g., $\hat{\mathbf{r}}$ is in the xy-plane. Then we filter the fluorescence with $\mathbf{\varepsilon}_p = \mathbf{e}_0$. For this configuration the photon-emission operator becomes

$$R_0\sigma = P_g \langle 0|\sigma|0\rangle \quad , \tag{11.2}$$

where a factor $|\langle 1||\mu||0\rangle|^2 4/3$ is absorbed in ζ.

(b) Circular polarization. In this case we take $\mathbf{\varepsilon} = \mathbf{e}_{\pm 1} = \mathbf{\varepsilon}_p$, $\hat{\mathbf{k}} = -\mathbf{e}_0 = -\hat{\mathbf{r}}$ and therefore $k_z = -k \simeq -\omega_o/c$. The interaction with the laser field now becomes

$$\Omega_{\pm 1}(h) = \frac{2}{\sqrt{3}} \frac{E_o}{\hbar} \langle 1 || \mu || 0 \rangle \; i \sin(-hk) \quad , \tag{11.3}$$

which couples only $|g\rangle$ with $|\pm 1\rangle$. Hence the relevant Einstein coefficient equals $A_{\pm 1}(h) = A_f b_\parallel(\omega_o h/c)$, and for the operator R we obtain

$$R_{\pm 1}\sigma = P_g \langle \pm 1|\sigma|\pm 1\rangle \sin^2(\omega_o h/c) \quad . \tag{11.4}$$

If we denote the wavelength of the radiation by $\lambda = 2\pi/k \simeq 2\pi c/\omega_o$, then we notice that both $\Omega_{\pm 1}(h)$ and $R_{\pm 1}$ vanish for $h = n \times \lambda/2$, with $n = 0,1,2,\ldots$. For circular polarization the fluorescence intensity is identically zero if h equals an integer multiple of λ. Furthermore, the emission operator, the Einstein coefficient and the Rabi frequency are identical for left- and right-handed polarizations, and so the photon correlations are identical (provided of course that we take $\underline{\varepsilon} = \underline{\varepsilon}_p$), which expresses again the rotational symmetry about the z-axis.

Now it is an easy matter to evaluate $\tilde{f}(s)$, Eq. (10.7), for both linear and circular polarization simultaneously. The set of equations (9.4)-(9.7) defines the matrix $s + iL_d(h) + W + \Gamma(h)$, and with some algebra we then find

$$\tilde{f}_\eta(s) = \frac{\zeta}{s} \times \frac{\frac{1}{2}(s+\frac{1}{2}A_\eta+\lambda)|\Omega_\eta|^2}{|\Omega_\eta|^2(s+\frac{1}{2}A_\eta+\lambda) + (A_\eta+s)[(s+\frac{1}{2}A_\eta+\lambda)^2 + \Delta^2]}$$

$$\times [1 - \delta_{\eta,\pm 1} \cos^2(\omega_o h/c)] \quad , \qquad (11.5)$$

for $\eta = -1,0,1$. Then the fluorescence intensity follows from

$$I_\eta = \lim_{t\to\infty} f_\eta(t) = \lim_{s\to 0} s\tilde{f}_\eta(s) \quad . \qquad (11.6)$$

XII. SPECIAL CASES

Transformation of the general result (11.5) for $\tilde{f}_\eta(s)$ to the time domain is straightforward, but the expression for $f_\eta(t)$ is very untransparent. A case of practical importance is the low-intensity limit, where the coupling strength between the dipole and the laser field

$$\Omega_R = \frac{2}{\sqrt{3}} \frac{E_0}{\hbar} |\langle 1||\mu||0\rangle| \qquad (12.1)$$

tends to zero in comparison with the relaxation parameter A_η. Normalized with the uncorrelated intensity $I_\eta = f_\eta(\infty)$, $f_\eta(t)$ is found to be

$$f_\eta(t)/I_\eta = 1 - \frac{\lambda - \frac{1}{2}A_\eta}{\lambda + \frac{1}{2}A_\eta} \; \frac{(\lambda+\frac{1}{2}A_\eta)^2 + \Delta^2}{(\lambda-\frac{1}{2}A_\eta)^2 + \Delta^2} \; e^{-A_\eta t}$$

$$+ \frac{A_\eta}{\lambda + \frac{1}{2}A_\eta} \frac{1}{(\lambda-\frac{1}{2}A_\eta)^2 + \Delta^2} e^{-(\lambda+\frac{1}{2}A_\eta)t}$$

$$\times [(\lambda^2 - \tfrac{1}{4}A_\eta^2 - \Delta^2)\cos(\Delta t) - 2\lambda\Delta\sin(\Delta t)] \quad . \qquad (12.2)$$

Figures 1 to 4 illustrate the dependence of $f_\eta(t)/I_\eta$ on the parameters Δ and λ, the laser polarization and the distance of the atom to the surface.

For higher irradiances we assume that ω_L is in very close resonance with ω_o, e.g., we take $\Delta = 0$. Then we obtain

$$f_\eta(t)/I_\eta = 1 - e^{-\alpha_\eta t}(\cos(\beta_\eta t) + \frac{\alpha_\eta}{\beta_\eta}\sin(\beta_\eta t)) \quad , \qquad (12.3)$$

with

$$\alpha_\eta = 3A_\eta/4 + \tfrac{1}{2}\lambda \quad , \qquad (12.4)$$

$$\beta_\eta = [|\Omega_\eta|^2 - \tfrac{1}{4}(\tfrac{1}{2}A_\eta - \lambda)^2]^{\frac{1}{2}} \quad , \qquad (12.5)$$

and typical behavior is illustrated in Figs. 5 and 6.

XIII. CONCLUSIONS

An excited atom near a surface has a different lifetime 1/A than in free space. Standard methods to measure A and its dependence on the distance of the atom to the surface are: (1) A equals the full width of the low-intensity absorption profile and (2) 1/A equals the delay time in the emission of a fluorescent photon after excitation of the atom. In this paper we considered the situation where the atom is continuously driven by a laser field, and we evaluated the temporal correlation between photons of the emitted fluorescence. After presentation of the general theory, we focussed on the $j_g = 0 \rightarrow j_e = 1$ transition. In the most simple case, where the irradiance is monochromatic ($\lambda = 0$), exactly on resonance ($\Delta = 0$), sufficiently weak ($\Omega_R \rightarrow 0$), and has a polarization η, we found (Eq. (12.2))

$$f_\eta(t) = I_\eta(1 - e^{-\frac{1}{2}A_\eta t})^2 \quad . \tag{13.1}$$

We conclude that the two-photon correlation function f(t), which is accessible to observation by a photon counter, is essentially determined by the lifetime $1/A_\eta$ of the driven upper substate of the atom. This result will possibly provide an alternative optical technique for the investigation of atoms near a metal surface.

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FIGURE CAPTIONS

Fig. 1. Plot of the two-photon correlation function $f_\eta(t)/I_\eta$ in the low-intensity limit. Time and frequencies are given in units of $1/A_f$ and A_f, respectively, and the distance between the atom and the surface h is in units of an optical wavelength $2\pi c/\omega_o$. In this figure we take $h = 0.2$, $\lambda = 0.5$ and linear polarization. Curves a, b and c illustrate the dependence on the detuning, with $\Delta = 0$, 2 and 4, respectively. Notice that f(t) can become larger than $f(\infty)$, which implies an enhancement for the detection of a photon at time t after the detection of a photon at time zero, in comparison with the uncorrelated detection probability I.

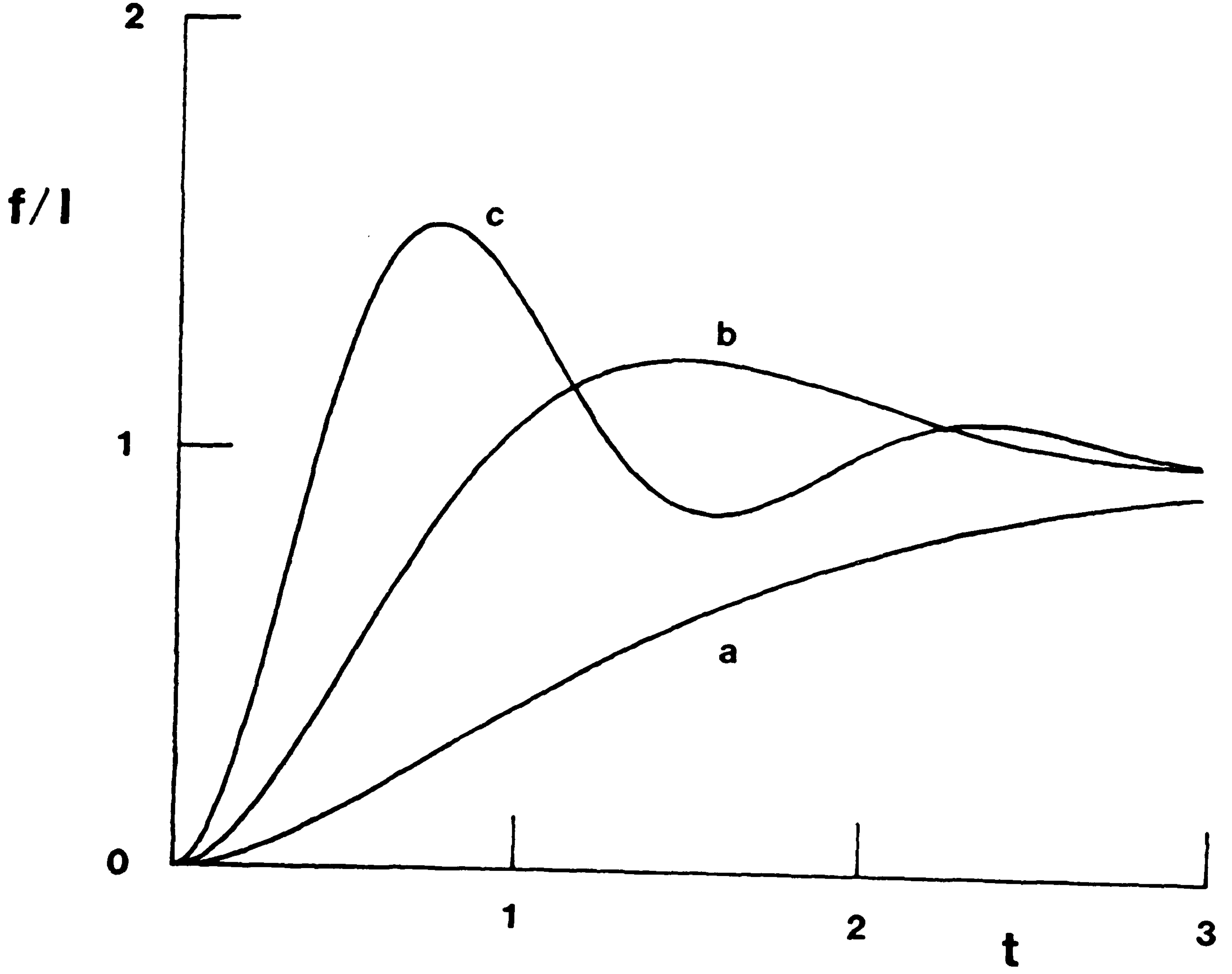
Fig. 2. Same as Fig. 1 but now we vary the laser linewidth as $\lambda = 0.2$, 0.6 and 3.0 for curves a, b and c, respectively, and the detuning is fixed at $\Delta = 4$. For a large bandwidth, curve c, the oscillatory structure is washed out entirely.

Fig. 3. Low-intensity $f_\eta(t)$ for $h = 0.2$, $\lambda = 0.1$ and $\Delta = 2$. Curves a and b correspond to linear and circular laser polarization, respectively.

Fig. 4. Two-photon correlation in the low-intensity limit with $\lambda = 0.2$, $\Delta = 2$ and circular polarization. This picture illustrates the dependence on the atom-surface distance h, with $h = 0.05$ and 0.1 for curves a and b, respectively. In curve a the approach to $f_\eta(\infty)/I_\eta = 1$ for increasing time occurs very slowly. This can be understood from the fact that for circular polarization the atom behaves effectively as a parallel dipole. Its matrix element, which determines the inverse lifetime, tends to zero for $h \rightarrow 0$, and therefore the lifetime itself becomes very large. This in turn prohibits the emission of the second photon at $t > 0$, after emission of the first photon and subsequent excitation by the laser.

Fig. 5. Correlation function for zero detuning, $h = 0.1$, $\lambda = 0.5$ and $\Omega_R = 3$. Curves a and b correspond to linear and circular polarization, respectively.

Fig. 6. The parameters in this figure are $\lambda = 0.5$, $\Delta = 0$, $\Omega_R = 8$, and the laser is circularly polarized. Curves a and b represent the change in correlation as a function of h. We take h = 0.1 and 1 for a and b respectively. It appears that the atom-surface distance can affect the photon-correlation function dramatically, which implies that a measurement of $f_\eta(t)/I_\eta$ might appear to be a sensitive technique for the determination of h of atomic adsorbates.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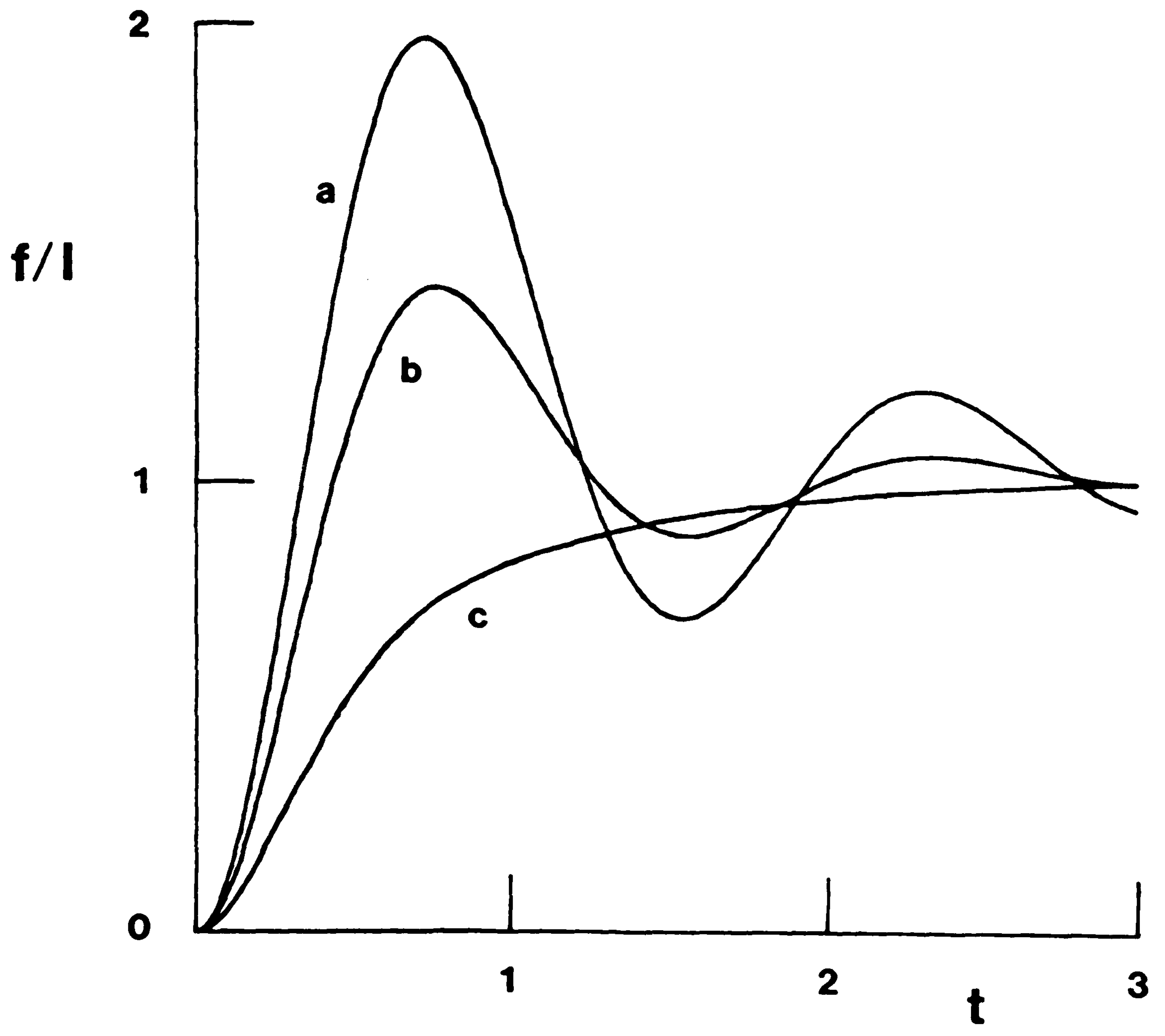
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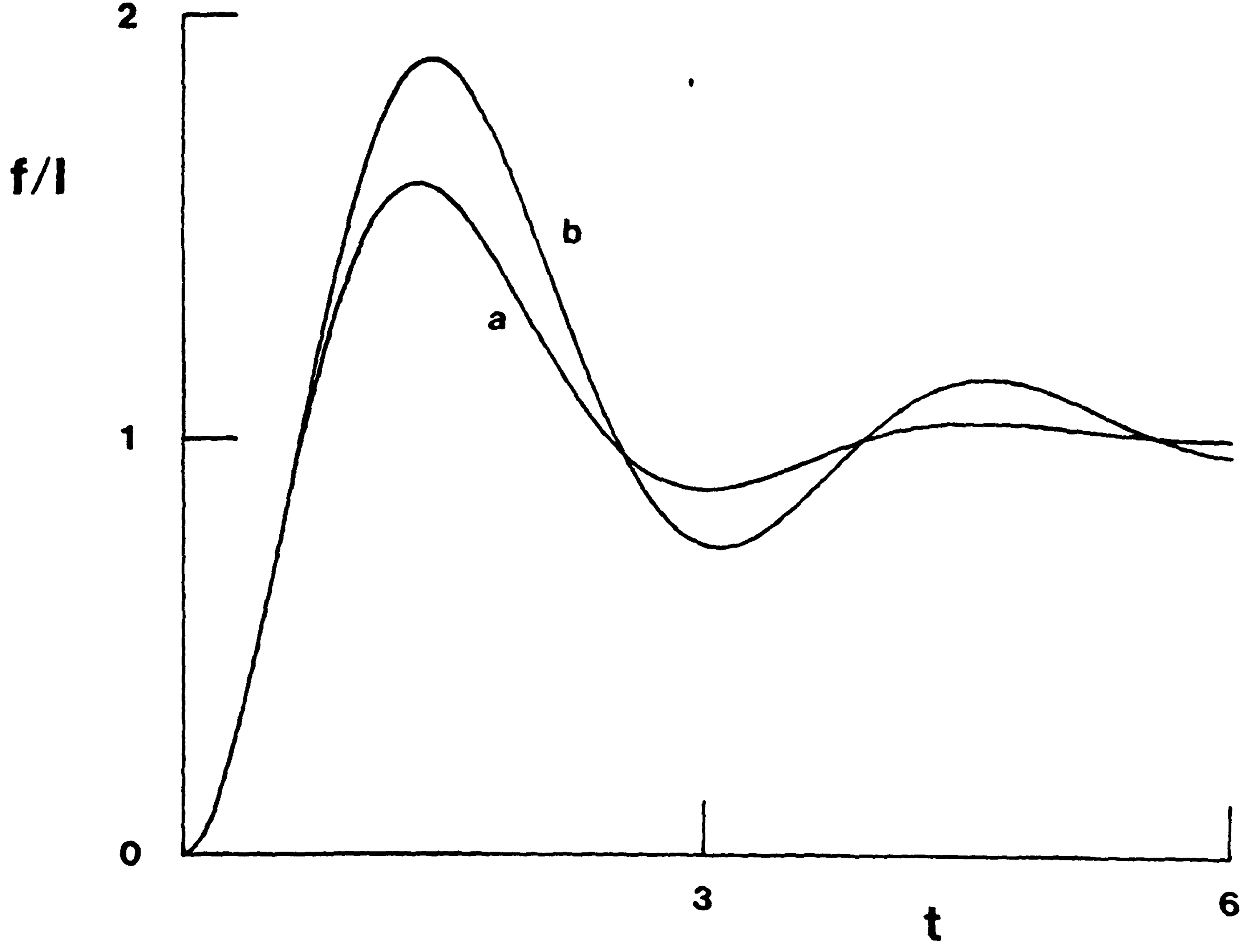
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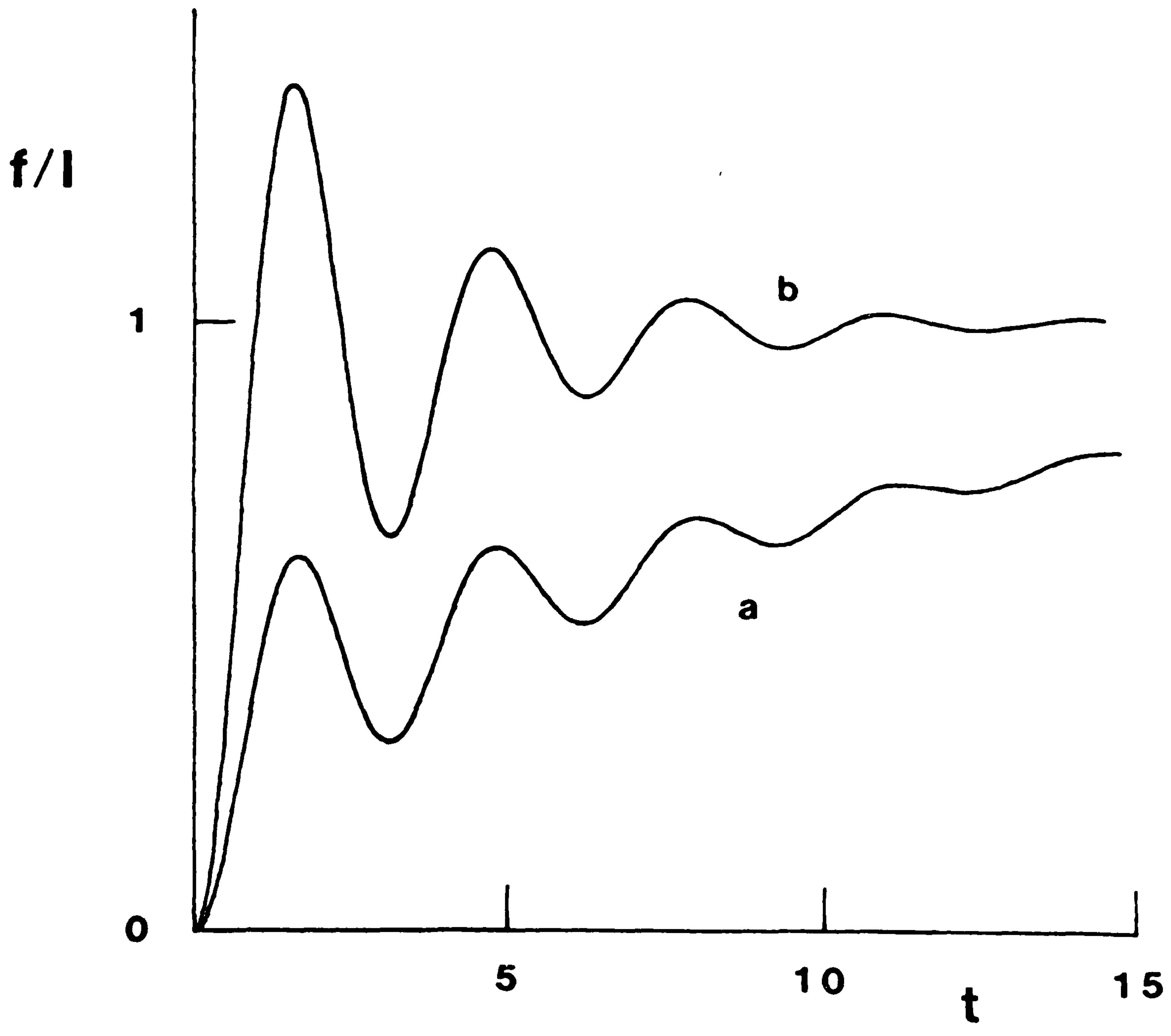
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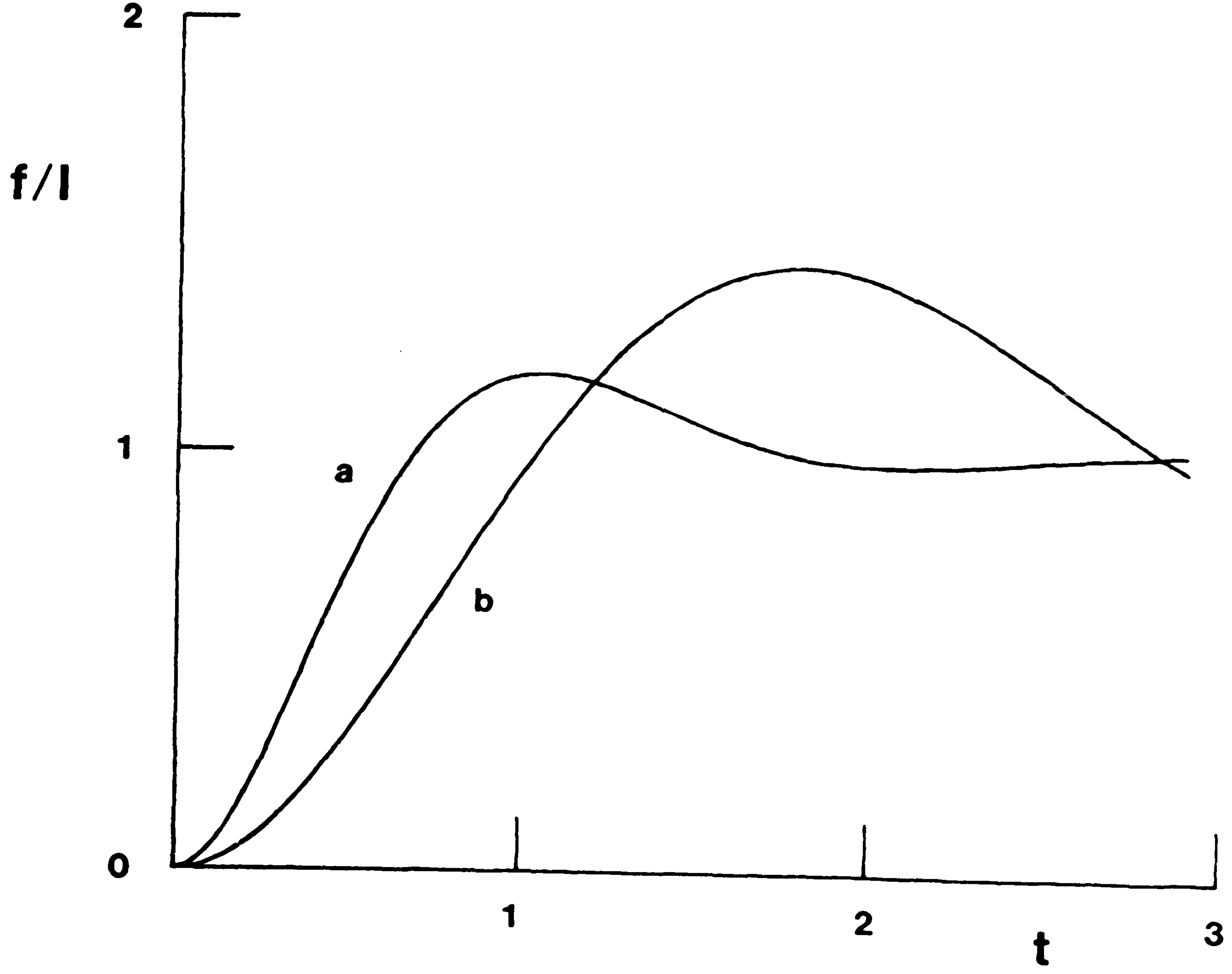
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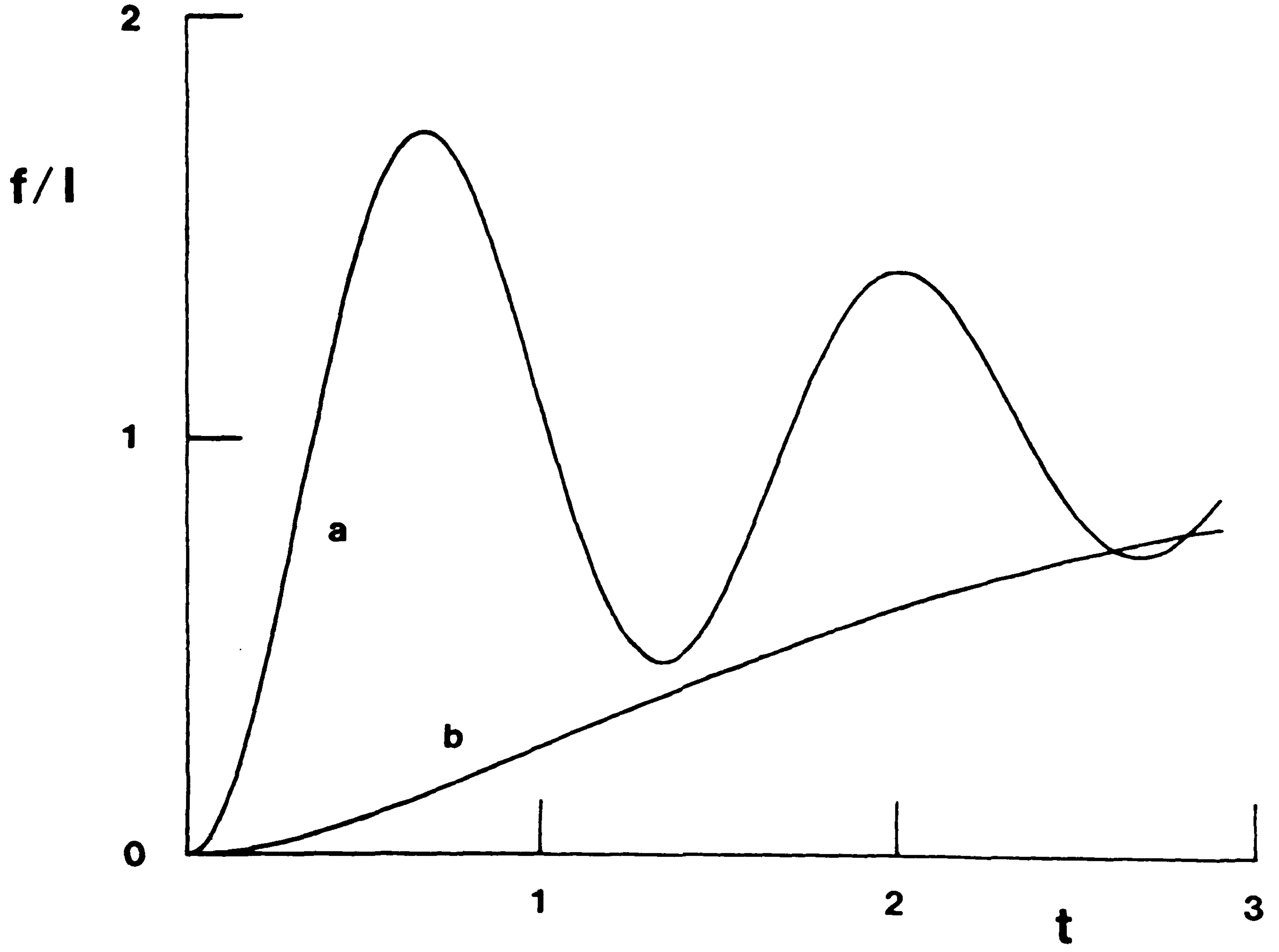
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