# Reflection-Induced Source Correlation in Spontaneous Emission

Karel E. Drabe, Gerard Cnossen, and Douwe A. Wiersma
*Ultrafast Laser and Spectroscopy Laboratory, Department of Chemistry, University of Groningen, Nijenborgh 16, 9747 AG, Groningen, The Netherlands*

H. A. Ferwerda and B. J. Hoenders
*Department of Applied Physics, University of Groningen, Nijenborgh 18, 9747 AG, Groningen, The Netherlands*

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We present fluorescence spectra showing that the spontaneous-emission spectrum of a molecule in front of a mirror is not invariant on propagation. We also calculate the cross-spectral density of this light source and show that it does not obey Wolf's scaling law [Phys. Rev. Lett. **56**, 1370 (1986)].

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Wolf[1] recently showed that the spectrum of a light source cannot generally be expected to be invariant on propagation through free space. Wolf also showed that for a planar, quasihomogeneous light source the normalized spectrum in the far-field zone is only identical to the source spectrum if the spectral coherence of the light source obeys a certain scaling law. The light source satisfies the scaling law if the complex degree of coherence $\mu^0(\omega,\mathbf{r}_2-\mathbf{r}_1)$ is only a function of the variable $k(\mathbf{r}_2-\mathbf{r}_1)$, where $\omega$ is the angular frequency, $\mathbf{r}_2-\mathbf{r}_1$ is the vectorial distance between two points in the source plane, and $k$ is the absolute value of the wave vector.

It is obvious that it is quite important to investigate which light sources do or do not obey the scaling law. For instance, the question may be raised whether light sources of great cosmological interest such as stars and quasars obey the scaling law.[2] If not, then, as Wolf[2] showed, the interpretation of spectral redshifts as solely arising from the Doppler effect in an expanding universe must be reexamined. A deviation from the invariance on propagation can also be a complicating factor in obtaining frequency standards.

Bocko, Douglass, and Knox[3] recently confirmed Wolf's prediction that source correlations can lead to marked frequency shifts of spectral lines. Their experiments were performed using two acoustic sources that were partly correlated. Morris and Faklis[4,5] recently demonstrated that by using a Fourier achromat one can generate a light source that also violates Wolf's scaling law. Wolf[2] suggested that superradiance and superfluorescence may lead to source correlations and hence to light sources that are variant on propagation. An entirely different physical mechanism for source correlations might arise from correlations between the refractive index of pairs of points in a spatially random medium.[2]

In this Letter we show that spontaneous emission in front of a mirror presents a light source that violates Wolf's scaling law. Figure 2 shows two fluorescence spectra obtained at two different points in space. It is clear that these spectra are rather different. The physical origin of this effect is easily understood as arising from interference between the direct and reflected light into a certain direction $\nu$ (see also the inset of Fig. 1). If a molecule is situated at a distance $z$ from the mirror, the path difference between the interfering rays in the direction $\nu$ is $2z\cos\nu$, leading to constructive or destructive interference depending on the wavelength of the emitted light. This interference effect is basically a wide-angle interference effect as studied by Schrödinger,[6] by Halpern and Doermann,[7] and more recently by Drexhage[8] and De Martini.[9]

In order to understand this propagation effect in greater detail we have to examine the cross-spectral density function of our light source. We proceed in two steps: First we calculate the cross-spectral density of a single emitter; in the second step we account for the mode of excitation and spatial extent of our light source and sum over all emitters in the sample.

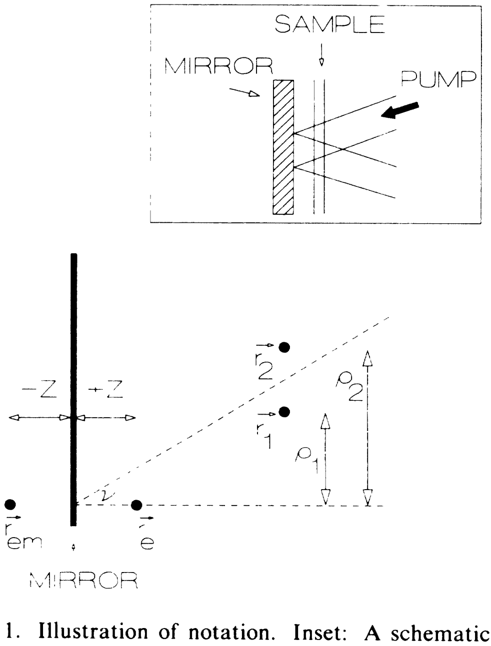

FIG. 1. Illustration of notation. Inset: A schematical outline of the geometry of the experiment.

The cross-spectral density is defined as[1]

$$W(\mathbf{r}_1,\mathbf{r}_2,\omega)=\int_{-\infty}^{+\infty} d\tau e^{i\omega\tau}\langle E^*(\mathbf{r}_1,t)E(\mathbf{r}_2,t+\tau)\rangle , \quad (1)$$

where $E(\mathbf{r},t)$ represents the complex analytical signal at the space-time point $(\mathbf{r},t)$, the angular brackets indicate an ensemble average, $\mathbf{r}_1$ and $\mathbf{r}_2$ denote two positions, and $\omega$ denotes the angular frequency ($\omega=kc$, where $k$ is the absolute value of the wave vector, and $c$ the speed of light). In the case of spontaneous emission, it is appropriate to replace the classical fields in Eq. (1) by field operators. Particularly convenient is the Heisenberg representation of the source field given by[10]

$$E^+(\mathbf{r},t)=(A/|\mathbf{r}-\mathbf{r}_e|)\Pi(t-|\mathbf{r}-\mathbf{r}_e|/c) , \quad (2)$$

where $\mathbf{r}_e$ is the position of the emitter, $\mathbf{r}$ the observation point (which should be sufficiently far away), and $\Pi$ a deexcitation operator of the emitter. In the above expression we have contracted several constants into $A$ and have also omitted the orientational dependence of the field. Equation (1) then becomes

$$W_{\text{mol}}(\mathbf{r}_1,\mathbf{r}_2,\omega)=\int_{-\infty}^{+\infty} d\tau e^{i\omega\tau}\langle E(\mathbf{r}_1,t)E^+(\mathbf{r}_2,t+\tau)\rangle . \quad (3)$$

We may write for the electric-field operator at position $\mathbf{r}_1$ (Ref. 10),

$$E^+(\mathbf{r}_1,t)=E^+_{\text{direct}}(\mathbf{r}_1,t)-RE^+_{\text{reflected}}(\mathbf{r}_1,t) , \quad (4)$$

where $R$ is the reflectivity of the mirror, and the minus sign arises from a phase jump of $\pi$ on reflection. The subscripts "direct" and "reflected" indicate the different trajectories the spontaneous emission takes. From the functional dependence of the source-field expression on $\mathbf{r}_1$ and $t$ in Eq. (2), it is clear that we can relate $E_{\text{direct}}$ and $E_{\text{reflected}}$ in Eq. (4) according to

$$E^+_{\text{reflected}}(\mathbf{r}_1,t)=E^+_{\text{direct}}(\mathbf{r}_1,t-\tau_{1d}) , \quad (5)$$

where $\tau_{1d}$ is given below. In precisely the same way we can relate the field operator $E(\mathbf{r}_2,t)$ to the space point $\mathbf{r}_1$,

$$\begin{aligned} E(\mathbf{r}_2,t)&=E_{\text{direct}}(\mathbf{r}_2,t)-RE_{\text{reflected}}(\mathbf{r}_2,t)\\ &=E_{\text{direct}}(\mathbf{r}_1,t-\tau_{12})-RE_{\text{direct}}(\mathbf{r}_1,t-\tau_{2d}) , \quad (6)\end{aligned}$$

where $\tau_{12}$, $\tau_{2d}$, and $\tau_{1d}$ are related to path differences which follow from geometrical considerations (see also Fig. 1):

$$c\tau_{12}=|\mathbf{r}_2-\mathbf{r}_e|-|\mathbf{r}_1-\mathbf{r}_e|\approx(\rho_2-\rho_1)\sin v ,$$

$$c\tau_{2d}=|\mathbf{r}_2-\mathbf{r}_{em}|-|\mathbf{r}_1-\mathbf{r}_e|\approx(\rho_2-\rho_1)\sin v-2z\cos v ,$$

$$c\tau_{1d}=|\mathbf{r}_1-\mathbf{r}_{em}|-|\mathbf{r}_1-\mathbf{r}_e|\approx 2z\cos v ,$$

where $\mathbf{r}_{em}$ is the position of the mirror image of the emitter located at $\mathbf{r}_e$. It is to be noted that we have assumed that both $\mathbf{r}_1$ and $\mathbf{r}_2$ are sufficiently large such that in Eq. (2) differences in $1/|\mathbf{r}_i-\mathbf{r}_j|$ ($\mathbf{r}_i=\mathbf{r}_1,\mathbf{r}_2$ and $\mathbf{r}_j=\mathbf{r}_e,\mathbf{r}_{em}$) can be neglected.

Denoting the emission spectrum of an emitter in free space by $S_{\text{mol}}(\mathbf{r},\omega)$ [which is the Fourier transform of the expectation value $\langle E_{\text{direct}}(\mathbf{r},t)E^+_{\text{direct}}(\mathbf{r},t)\rangle$], one obtains for the cross-spectral density of a single emitter in front of a mirror,

$$\begin{aligned} W_{\text{mol}}(\mathbf{r}_1,\mathbf{r}_2,\omega)=&S_{\text{mol}}(\mathbf{r}_1,\omega)\exp\{ik(\rho_2-\rho_1)\sin v\}\\ &\times\{(1+R^2)-2R\cos(2kz\cos v)\} . \quad (7)\end{aligned}$$

Note that the cross-spectral density is not only a function of $k(\rho_2-\rho_1)$, but also depends on the distance $z$ of the emitter to the mirror.

The above equation has been derived for the case of a single emitter. In case of an ensemble of independent emitters we may obtain the cross-spectral density by summing over all emitters:

$$W_{\text{ens}}(\mathbf{r}_1,\mathbf{r}_2,\omega)=\int dx\,dy\,dz\,\kappa(x,y,z)W_{\text{mol}}(\mathbf{r}_1,\mathbf{r}_2,\omega) , \quad (8)$$

where $\kappa(x,y,z)$ is the density of emitters as a function of coordinates $x,y,z$. The evaluation of this integral is rather cumbersome and depends on the geometry of the sample. However, sufficient insight into its functional behavior can be obtained in the following way. First, consider a monolayer of emitters parallel to the mirror. It is readily seen from Fig. 1 that the integration over $x$ and $y$ in Eq. (8) can be replaced by an integration over the angle $v$ only, and it follows that the cross-spectral density remains a function of $k(\rho_2-\rho_1)$ and $kz$. In particular, we find that for large distances the sample can be approximated by a (two-dimensional) point source, in which case an integration over $v$ is unnecessary. In addition to the spatial extent of our light source, we must also integrate over the thickness of the sample. In particular, we must account for the periodic density $\kappa(z)$ of excited molecules, which is simply proportional to the Wiener fringes formed upon excitation,[11,12]

$$\kappa(z)=1+R^2-2R\cos(2k_{pz}z) , \quad (9)$$

where $k_{pz}=k_p\cos v_p$ and the phase jump upon reflection is again taken to be equal to $\pi$. Equation (8) then becomes, for $\mathbf{r}_1,\mathbf{r}_2$ in the far field (with the *spatial* extent of the source approximated by a point),

$$W_{\text{ens}}(\mathbf{r}_1,\mathbf{r}_2,\omega)=\frac{1}{L_2-L_1}\int_{L_1}^{L_2} dz\,W_{\text{mol}}(\mathbf{r}_1,\mathbf{r}_2,\omega)\kappa(z) , \quad (10)$$

where $L_1=0$ means that the fluorescent sample is on top of the mirror and $L_2-L_1$ is the thickness of the sample. By using Eqs. (7) and (9) in Eq. (10), many terms are obtained after integration. The dominant ones are easily recognized[13] and given by

$$\begin{aligned} W_{\text{ens}}(\mathbf{r}_1,\mathbf{r}_2,\omega)=&S_{\text{mol}}(\mathbf{r}_1,\omega)\exp\{ik(\rho_2-\rho_1)\sin v\}\\ &\times\{(1+R^2)^2+2R^2\cos[(k_{pz}-k_z)(L_2+L_1)]\text{sinc}[(k_{pz}-k_z)(L_2-L_1)]\} , \quad (11)\end{aligned}$$

where $\operatorname{sinc}x = (\sin x)/x$, $k_z = k\cos v$, and $k$ is the absolute value of the wave vector of the emission.

Equation (11) is the main result of this paper. The emission spectrum is obtained by putting $\mathbf{r}_1 = \mathbf{r}_2$ (i.e., $\rho_1 = \rho_2$). Note that it is the interference term between direct and reflected light (the second term in the curly braces) that causes a deviation from the scaling law. Indeed if the reflectivity is taken to be zero, the spectrum is invariant on propagation.

In Fig. 2 the fluorescence spectrum (lower curve) is shown from the molecule nile blue in front of a mirror. This spectrum is obtained from a position at a large distance from the sample, in which case the spatial extent can be considered as a point. This spectrum is described by Eq. (11) and depends only on the angle of observation. The other spectrum in Fig. 2 (upper curve) is obtained from a position in space closer to the mirror, at a distance such that the spatial extent of the source has to be taken into account. Now the integration over the angle $v$ should be performed, which tends to wash out the $\cos(2kz\cos v)$ term in Eq. (7). The relevance of this observation is that in a plane parallel to the mirror and sufficiently close to the fluorescent film, the spectrum is the same at every point in this plane. Spectra taken at points closer to the film than the spectrum of Fig. 2 (upper curve) are identical to the spectrum of nile blue obtained without a mirror. In other words, it is possible to define a source plane in which the spectrum is the same at every point across it.

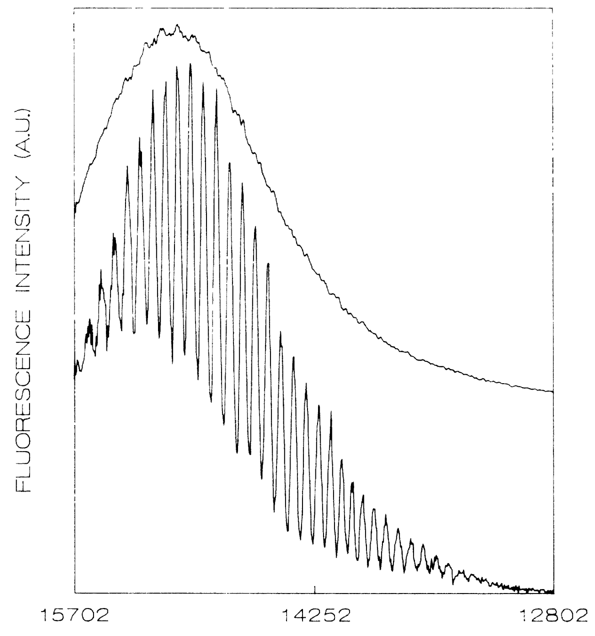

FIG. 2. Fluorescence spectra of nile blue obtained at two different points. The upper spectrum is taken at a distance of 1 mm from the film; the lower spectrum is taken at a distance of 100 mm from the film. Both spectra were obtained from the same sample. Note that the upper spectrum has been shifted for ease of comparison. The polymer film, containing nile blue, was 2.5 $\mu$m thick and at a distance of 54 $\mu$m from the mirror.

The experiments were performed using polymer layers (1–10 $\mu$m thickness) of poly(4-vinyl-piridine) doped with the fluorescent molecule nile blue. The films were spin coated on glass plates and placed parallel to an Al mirror. The nile-blue molecules were excited by a low-divergent HeNe laser having a beam diameter of 1 mm. The low power density (less than 6 mW/mm$^2$) used precludes any significant contribution from stimulated emission in the spectra. The pump beam, polarized perpendicular to the plane of incidence, is incident at an angle $v_p$ with respect to the mirror normal. The emission spectrum was detected with an $f/1.6$ lens. The focal point could be located at various points outside the sample. A second lens (210-mm focal length), placed about 500 mm from the first lens, ensures that only light near the focal point of the first lens is properly focused into a 1-m-grating spectrometer. At the exit slit of the spectrometer a photomultiplier tube was used for detection. The particular choice of the laser beam diameter inside the sample and the position of the focal point of the first lens determines the detection angle and angular resolution.

To summarize, we have shown that spontaneous emission in front of a reflecting surface presents a light source that violates Wolf's scaling law. A noteworthy property of the source is that a plane can be defined in which the spectrum is identical at every point. However, at points outside this plane the spectrum is position dependent. The position dependence basically arises from interference of light traveling directly and via the mirror to the point of observation. However, the effect is not limited by the coherence length of the source (which in our case of nile blue is only a few $\mu$m).

As radiation quite often can be expected to be contaminated with reflected light, this violation of the invariance of the spectrum on propagation can occur under rather general conditions. Of course, the *quantitative* extent to which the observed spectrum is dependent on position is determined by a number of factors, of which angular and spectral resolution are most important. As a significant amount of light (about 10%–20%) out of the Universe reaches us via reflection of particles present in interstellar dust,[14] it remains a challenge to explore the relevance of our findings.

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