IC/90/251
INTERNAL REPORT
(Limited Distribution)

International Atomic Energy Agency
and
United Nations Educational Scientific and Cultural Organization

INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS

RADIATION INDUCED LUMINESCENCE FROM A DIPOLE IMMERSED IN A THIN FILM *

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ABSTRACT

Luminescence is modelled as electromagnetic radiation from a dipole immersed in a thin film. Maxwell's equations are solved for the cases when the dipole in the thin film is oriented normal and parallel to the interfaces. Expressions for emitted electric fields *outside* the thin film are derived and are found to have a resonant denominator that vanishes at the surface polariton excitation frequencies for a thin film. Luminescent spectra are plotted and peaks are found that are identified to be associated with both surface response and bulk response. Numerical results are presented to illustrate the model by considering a vacuum-GaP-sapphire system.

MIRAMARE – TRIESTE
August 1990

* To be submitted for publication.

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1 Introduction

Radiation induced luminescence from solids was first observed by Mooradian [1] who analysed the luminescence spectra from Cu and Au. Amongst others, Boyd, Yu and Shen [2] and Crawford and Ritchie [3] reported calculations of luminescence spectra for *smooth* samples. Kash *et. al* [4] have studied the luminescent spectrum of quantum ribbons and quantum disks grown from GaAs-AlGaAs quantum wells. In our previous work [5] we investigated luminescence spectra for *rough* samples and reported calculations of zero order and first order emitted electric fields. In the present work we extend the investigation of radiation induced luminescence to the case of a dipole immersed in a *thin* film. This geometry is important both from the point of view of device applications such as opto-electronic devices, and from the standpoint of the basic physics that is exhibited.

The plan of this paper is as follows. In section 2, we solve Maxwell's equations for a system consisting of a dipole immersed in a thin film. We shall consider two cases of orientations of the dipole: *first* when the dipole is normal to the interfaces, and *secondly* when it is parallel. The results that are derived should enable one to predict the behaviour of thin film optical devices as far as emitted electric fields are concerned. Section 3 is devoted to numerical results and discussions, and our results are applied to a thin film of GaP bounded by vacuum and a substrate. Concluding remarks are made in section 4.

2 Luminescence fields from a thin film

Consider the geometry in which in region I ($z > 0$) there is a medium with a positive dielectric constant ϵ_1, and in region II ($-d < z < 0$) there is a luminescent thin film with a frequency dependent dielectric function $\epsilon(\omega)$, while region III ($z < -d$) is occupied by a substrate with a positive dielectric constant ϵ_3, as illustrated in Figure 1. The frequency dependent dielectric function, $\epsilon(\omega)$, is given by

$$\epsilon(\omega) = \epsilon(\infty) + \frac{S\omega_T^2}{\omega_T^2 - \omega^2 - i\omega\Gamma} \tag{1}$$

where $\epsilon(\infty)$ is the high frequency dielectric constant, S measures the strength of the resonance, ω_T is the TO phonon frequency, and Γ is a damping parameter. We solve Maxwell's equations for a system when there is a dipole in region II. We obtain the electric fields for all the three regions, but we shall concentrate on the fields *outside* the thin film since that is where measurements are performed. It is convenient, as Crawford and Ritchie [3] did for luminescence due to a localised dipole immersed in a solid, to apply the results to two cases: in section 2.1 we shall consider a dipole oriented normal to the thin film boundaries, and in section 2.2 we shall consider a dipole oriented parallel to the interfaces. In our model we shall consider TM modes only and the equations to be solved are; for region I

$$\nabla \wedge \nabla \wedge \vec{E}_1 - \epsilon_1 \frac{\omega^2}{c^2} \vec{E}_1 = 0 \tag{2}$$

and in region II, we have an inhomogeneous equation due to the polarisation $\vec{P}$ as a result of the immersed dipole.

$$\nabla \wedge \nabla \wedge \vec{E}_2 - \epsilon_2(\omega) \frac{\omega^2}{c^2} \vec{E}_2 = \frac{\omega^2}{\epsilon_0 c^2} \vec{P} \tag{3}$$

where for the polarisation due to the dipole, we take its dependence to be of the form

$$P_i\{e^{iQ_z z} + e^{-iQ_z z}\} e^{i(Q_x x - \omega t)} \tag{4}$$

where for the two dipole orientations $i = x$ or z and for region III, we have

$$\nabla \wedge \nabla \wedge \vec{E}_3 - \epsilon_3 \frac{\omega^2}{c^2} \vec{E}_3 = 0 \tag{5}$$

We discuss the results of the two orientations of the dipole in the next two sections.

2.1 Dipole oriented normal to the boundaries ($P_z \neq 0, P_x = 0$)

In this section, we consider the luminescent spectra due to a dipole immersed in a thin film and oriented normal to the boundaries of the thin film. The solutions in the three regions are given in terms $E_{\alpha i}$ (with $\alpha = 1, 2, 3$ and $i = x, z$) components. In region I we have

$$E_{1x} = A e^{i(q_{1x} x + q_{1z} z)} \tag{6}$$

$$E_{1z} = -\frac{q_{1x}}{q_{1z}} A e^{i(q_{1x} x + q_{1z} z)} \tag{7}$$

In region II the general solution is the sum of a complementary function plus a particular integral, and the electric field components are given by

$$E_{2x} = [B e^{iq_{2z} z} + C e^{-iq_{2z} z}] e^{iq_{2x} x} - \frac{Q_x Q_z}{\epsilon_0 \epsilon(\omega)[Q^2 - \epsilon(\omega)\frac{\omega^2}{c^2}]} P_z \{e^{iQ_z z} - e^{-iQ_z z}\} e^{iQ_x x} \tag{8}$$

$$E_{2z} = -\frac{q_{2x}}{q_{2z}} [B e^{iq_{2z} z} - C e^{-iq_{2z} z}] e^{iq_{2x} x} - \frac{[Q_x^2 - \epsilon(\omega)\frac{\omega^2}{c^2}]}{\epsilon_0 \epsilon(\omega)[Q^2 - \epsilon(\omega)\frac{\omega^2}{c^2}]} P_z \{e^{iQ_z z} + e^{-iQ_z z}\} e^{iQ_x x} \tag{9}$$

where the inhomogeneous term is analogous to the form for a bulk response discussed by Abrikosov *et.al* [6] and Barker and Loudon [7]. In region III we have,

$$E_{3x} = D e^{i(q_{3x} x + q_{3z} z)} \tag{10}$$

$$E_{3z} = -\frac{q_{3x}}{q_{3z}} D e^{i(q_{3x} x + q_{3z} z)} \tag{11}$$

where in equations (6) to (11), the wave vectors satisfy

$$q_{\alpha x}^2 + q_{\alpha z}^2 = \epsilon_\alpha \frac{\omega^2}{c^2} \tag{12}$$

where $\alpha = 1, 2, 3$ and noting that $\epsilon_2 = \epsilon(\omega)$, and

$$Q_x^2 + Q_z^2 = Q^2 \tag{13}$$

$$q_{1x} = q_{2x} = q_{3x} = Q_x \tag{14}$$

$$q_{1z} \neq q_{2z} \neq q_{3z} \neq Q_z \tag{15}$$

Equation (14) expresses conservation of the tangential comonents of the wavevectors and equation (15) means that the normal components are not conserved. In order to satisfy the usual boundary conditions of the continuity of the tangential electric field and normal displacement vector at $z = 0$ and $z = -d$, we put $Q_x = 0$, and we obtain, after some algebra, the electric fields *outside* the thin film. In region I, we have

$$E_{1n} = F_{1n} \frac{P_z}{\epsilon_0} e^{i(\vec{q}_1 . \vec{r} - \omega t)} \tag{16}$$

where

$$F_{1n} = \left| \frac{2\sqrt{\epsilon_1} \omega q_2^2 \{\epsilon_3 q_{2z}[1 - e^{-2iq_{2z} d}] + \epsilon(\omega) q_{3z}[1 - e^{-iq_{2z} d}]^2\}}{c q_{1z} q_{2z} D_s} \right| \tag{17}$$

and in region III, we obtain

$$E_{3n} = F_{3n} \frac{P_z}{\epsilon_0} e^{i(\vec{q}_3 . \vec{r} - \omega t)} \tag{18}$$

where

$$F_{3n} = \left| \frac{2\sqrt{\epsilon_3} \omega q_2^2 e^{iq_{3z} d} \{\epsilon_1 q_{2z}[1 - e^{-2iq_{2z} d}] - \epsilon(\omega) q_{1z}[1 - e^{-iq_{2z} d}]^2\}}{c q_{1z} q_{2z} D_s} \right| \tag{19}$$

where in equations (17) and (19), D_s is given by

$$D_s = [\epsilon(\omega) q_{3z} + \epsilon_3 q_{2z}][\epsilon(\omega) q_{1z} - \epsilon_1 q_{2z}] - e^{-2iq_{2z} d}[\epsilon(\omega) q_{3z} - \epsilon_3 q_{2z}][\epsilon(\omega) q_{1z} + \epsilon_1 q_{2z}] \tag{20}$$

It is noted that the denominator D_s given in equation (20) and appearing in equations (17) and (19) occurs at the excitation frequencies for a thin film (See, for example, Cottam and Tilley [8]). Numerical applications of the results discussed in this section are given in section 3.

2.2 Dipole oriented parallel to the boundaries ($P_x \neq 0, P_z = 0$)

Consider the luminescent spectra due to a dipole immersed in a thin film and oriented parallel to the boundaries of the thin film. The fields in regions I and III are of a similar form to those given in equations (6), (7), (10) and (11), but the expressions for A and D are different. The electric fields for region II are given by

$$E_{2xp} = [B_p e^{iq_{2z}z} + C_p e^{-iq_{2z}z}]e^{iq_{1x}x} - \frac{[Q_z^2 - \epsilon(\omega)\frac{\omega^2}{c^2}]}{\epsilon_0\epsilon(\omega)[Q^2 - \epsilon(\omega)\frac{\omega^2}{c^2}]}P_x\{e^{iQ_z z} + e^{-iQ_z z}\}e^{iQ_x x} \quad (21)$$

$$E_{2zp} = -\frac{q_{1x}}{q_{2z}}[B_p e^{iq_{2z}z} - C_p e^{-iq_{2z}z}]e^{iq_{1x}x} - \frac{Q_x Q_z}{\epsilon_0\epsilon(\omega)[Q^2 - \epsilon(\omega)\frac{\omega^2}{c^2}]}P_x\{e^{iQ_z z} - e^{-iQ_z z}\}e^{iQ_x x} \quad (22)$$

where it should be noted that $B_p \neq B, C_p \neq C$ and we shall proceed by putting $Q_z = 0$, for the same reasons stated in section 2.1. The field in region I is given by

$$E_{1p} = F_{1p}\frac{P_x}{\epsilon_0}e^{i(\vec{q}_1.\vec{r}-\omega t)} \quad (23)$$

where

$$F_{1p} = |\frac{2\sqrt{\epsilon_1}\omega\{\epsilon(\omega)q_{3z}[1 - e^{-2iq_{2z}d}] + \epsilon_3 q_{2z}[1 - e^{-iq_{2z}d}]^2\}}{cD_s}| \quad (24)$$

and in region III, we obtain

$$E_{3p} = F_{3p}\frac{P_x}{\epsilon_0}e^{i(\vec{q}_3.\vec{r}-\omega t)} \quad (25)$$

where

$$F_{3p} = |\frac{2\sqrt{\epsilon_3}\omega e^{iq_{3z}d}\{\epsilon(\omega)q_{1z}[1 - e^{-2iq_{2z}d}] - \epsilon_1 q_{2z}[1 - e^{-iq_{2z}d}]^2\}}{cD_s}| \quad (26)$$

The results derived in this section are applied to a system of GaP bounded by vacuum and sapphire in the next section.

3 Numerical results and discussion

The intensity of the emitted light in region I is proportional to the square of the functions derived in sections 2.1 and 2.2, and hence to $|F_{1n}|^2$, $|F_{3n}|^2$, $|F_{1p}|^2$ and $|F_{3p}|^2$. We study the frequency dependence of these quantities for a system of GaP bounded by vacuum in region I and sapphire in region III. The following constants are used: $\epsilon_1 = 1.0, \epsilon_3 = 3.1$ and for GaP, we use values from Barker [9] as $\epsilon(\infty) = 9.09, S = 2.01, \omega_T = 366.0$ cm^{-1} and Γ is taken to be $0.005\omega_T$. The luminescent spectra are given in figures 2 to 5. These figures are just part of a wide range of spectra that can be obtained, and we noted that the expressions are very sensitive to any change of wavenumber or film thickness, for the frequency range considered. The luminescent spectra are obtained as follows. Since

any elementary excitation has a defined wavevector and frequency, the figures have been plotted for two tangential wavevectors, $cq_{1x}/\omega_T = 2.5$ and 3.5, and then sweeping through the frequency range. It can be noted that the response between $\omega/\omega_T = 1.0$ to 1.105 is due to the surface modes [8], and above 1.105 of the reduced frequency there is a bulk response, recalling that this is the LO frequency given by the well known LST relation

$$\frac{\omega_{LO}}{\omega_T} = \sqrt{\frac{s + \epsilon(\infty)}{\epsilon(\infty)}} \quad (27)$$

Figures 2 and 3 illustrate the frequency dependence of $|F_{1n}|^2$ and $|F_{3n}|^2$, corresponding to the case of a dipole oriented normal to thin film as discussed in section 2.1. Figures 4 and 5 illustrate the frequency dependence of $|F_{1p}|^2$ and $|F_{3p}|^2$, corresponding to the case of a dipole oriented parallel to the thin film boundaries as discussed in section 2.1.

4 Conclusions

In this paper, we have calculated expressions related to the luminescent spectra due to a thin film. This has been done by solving Maxwell's equations in presence of a dipole immersed inside a thin film, and we considered two dipole orientations: *first*, when the dipole is normal to the thin film and *secondly*, when the dipole is parallel to the thin film boundaries. The emitted electric fields are found to have a resonant denominator that vanishes at the excitation frequencies of the thin film. The luminescent spectra have been shown to have a surface response and a bulk response. The results derived in this paper supplement some of the theoretical and experimental studies on luminescence [1 – 5], and these results have applications in thin film optical devices.

Acknowledgments

The author would like to thank Professor Abdus Salam, the International Atomic Energy Agency and UNESCO for hospitality at the International Centre for Theoretical Physics, Trieste. He would also like to thank the Swedish Agency for Research Cooperation with Developing Countries (SAREC) for financial support during his visit at the ICTP under the Associateship scheme. A discussion with Professor Persico is also acknowledged. Thanks are also due to TWAS for support at the University of Botswana, where some of this work was done.

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Figure Captions

Figure 1: The geometry studied in this paper, in which region I ($z > 0$), region II ($-d < z < 0$) and region III ($z < -d$) have dielectric functions ϵ_1, $\epsilon(\omega)$ and ϵ_3 respectively.

Figure 2: Frequency dependence of $|F_{1s}|^2$ for a tangential wavevector (a) $cq_{1x}/\omega_T = 2.5$ and (b) $cq_{1x}/\omega_T = 3.5$.

Figure 3: Frequency dependence of $|F_{3s}|^2$ for a tangential wavevector (a) $cq_{1x}/\omega_T = 2.5$ and (b) $cq_{1x}/\omega_T = 3.5$.

Figure 4: Frequency dependence of $|F_{1p}|^2$ for a tangential wavevector (a) $cq_{1x}/\omega_T = 2.5$ and (b) $cq_{1x}/\omega_T = 3.5$.

Figure 5: Frequency dependence of $|F_{3p}|^2$ for a tangential wavevector (a) $cq_{1x}/\omega_T = 2.5$ and (b) $cq_{1x}/\omega_T = 3.5$.

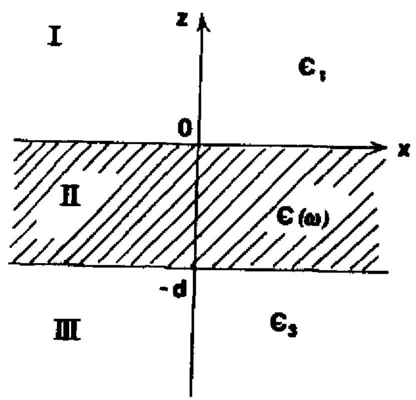
z
I
ε1
0
x
II
ε(ω)
-d
III
ε3

Fig.1

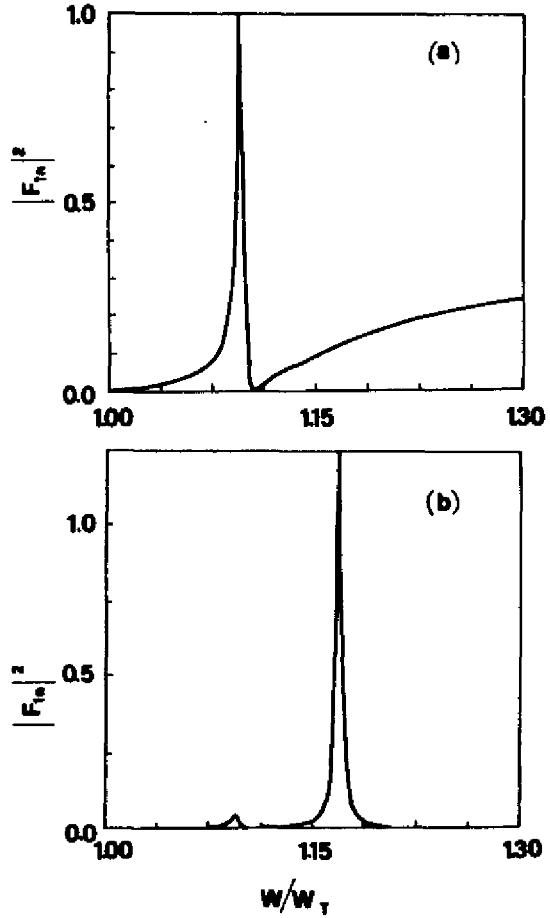
1.0
(a)
0.5
0.0
1.00
1.15
1.30
(b)
1.0
0.5
0.0
1.00
1.15
1.30
W/W T

Fig.2

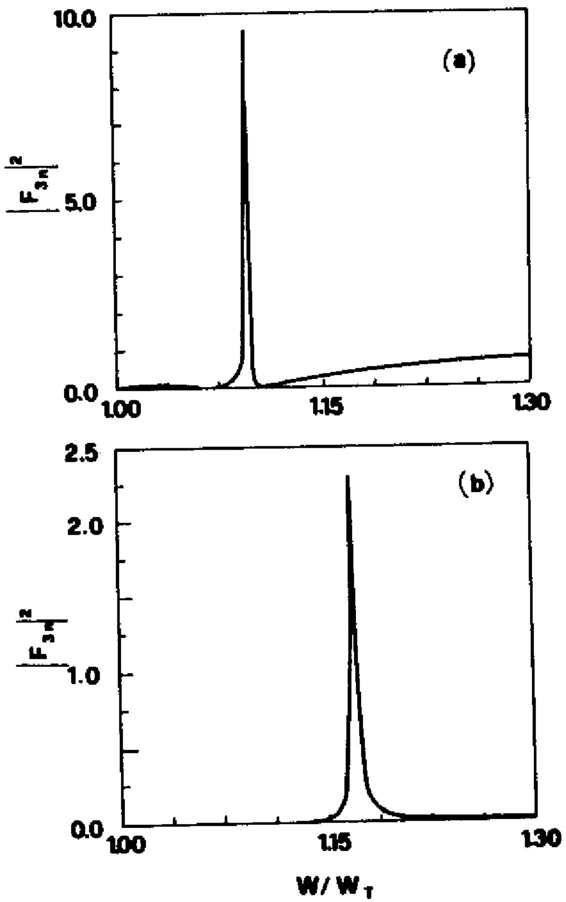
10.0
(a)
$|F_{3n}|^2$
5.0
0.0
1.00
1.15
1.30
2.5
(b)
2.0
$|F_{3n}|^2$
1.0
0.0
1.00
1.15
1.30
W/W_T

Fig.3

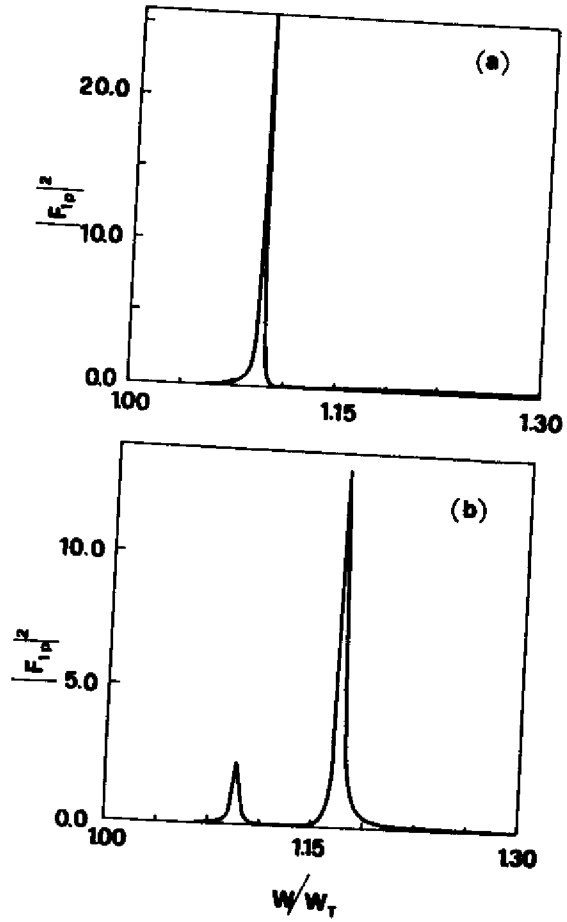
(a)
20.0
$|F_{1p}|^2$
10.0
0.0
1.00
1.15
1.30
(b)
10.0
$|F_{1p}|^2$
5.0
0.0
1.00
1.15
1.30
W/W_T

Fig.4

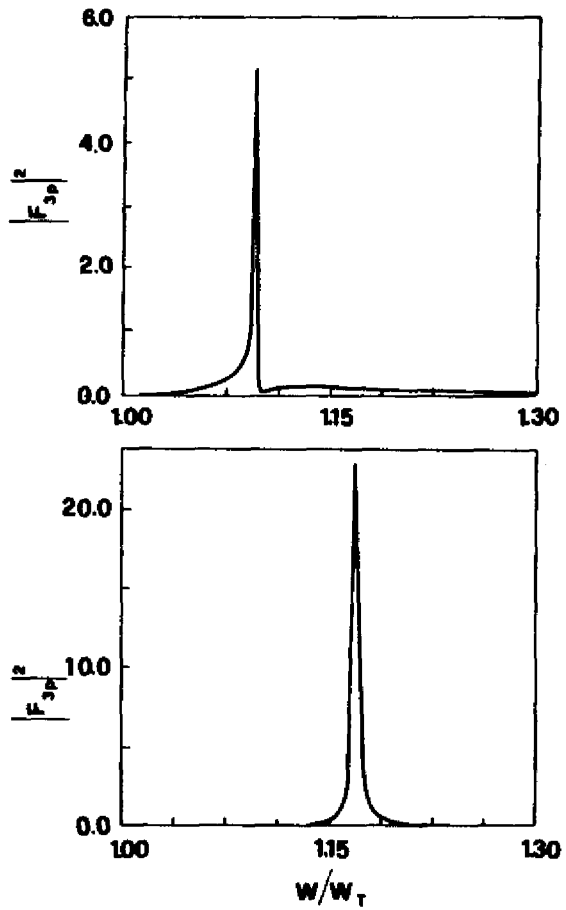

Fig.5