Indian J. Phys. 80 (4), 361-366 (2006)

Calculation of photocurrent in Al and Be by using the free electron potential model for the crystal

R K Thapa*, S R Gurung[1], G Das[2] and R Bhattacharjee[3]

Department of Physics, Mizoram University, Tanhril 796 009, Mizoram, India
[1]Department of Physics, Pachhunga University College, Aizawl-796 001, Mizoram, India
[2]Department of Physics, Govt Kolasib College, Kolasib-796 081, Mizoram, India
[3]Department of Physics, Assam University, Silchar, Assam, India

E-mail : rkt@sancharnet.in

Received 7 October 2005, accepted 23February 2006

Abstract . We have used free electron model potential in the calculation of photocurrent from metals like aluminium and beryllium by using a spatially dependent vector potential. The results had been compared with the one calculated by using the dielectric model as proposed by Bagchi and Kar [*Phys. Rev.* **B18**, 5240 (1978)].

Keywords Photoemission, photocurrent, free electron model potential

PACS No. : 79.60.Bm

1. Introduction

Photoemission from a metal, in which an electron is excited by an incident photon and escapes to the vacuum by overcoming the surface potential barrier is well established as a technique for studying the electronic states below the Fermi level [1,2]. Many methods for photoemission calculation [3,4] have been developed where wavefunctions for the semi-infinite solids are constructed accurately.

However, in these calculations, the spatial variation of the photon field is generally neglected. An accurate calculation of the electromagnetic field in the surface region is a complex problem and first principle calculations are available [5,6] only for jellium model. On the other hand, empirical calculations of fields near the surface with 'local' dielectric function have been used to explain certain qualitative features of photoemission data for tungsten [7], aluminium [8], palladium [9] *etc.* This method has been reasonably successful even with simple wavefunctions for calculating matrix element for evaluation of photocurrent.

*Corresponding Author

In this report, we present a different type of 'local' dielectric function, which is a logarithmic function of z, and use it in deriving the vector potential in the bulk, surface and vacuum regions. The vector potential so derived will be used to calculate photocurrent from metals like Al and Be. We will compare the present results with earlier calculations [8,10-12].

2. Dielectric model and calculations of electromagnetic fields

We consider the metallic surface to be extended in the negative z-direction with surface parallel to the x-y plane such that the metal occupies all space to the left of $z = 0$ plane. The model

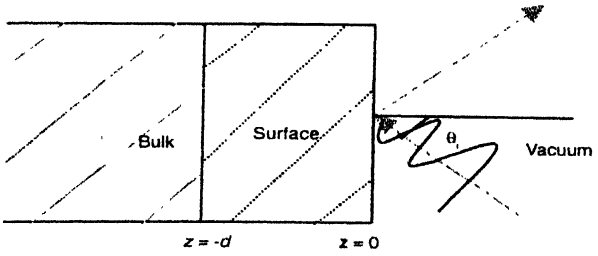

Figure 1. Schematic diagram of model for dielectric function.

dielectric function (Figure 1) for the bulk ($z \le -d$), surface ($-d \le z \le 0$) and vacuum ($z \ge 0$) is given by

$$\varepsilon(\omega,z) = \begin{cases} \varepsilon_b(\omega), & z \le -d; \\ 1+\dfrac{(\varepsilon_b-1)}{\log_e 2}\log_e\left(1-\dfrac{z}{d}\right), & -d \le z \le 0; \\ 1, & z \ge 0; \end{cases} \quad (1)$$

where $\varepsilon_b(\omega) \equiv \varepsilon_1(\omega)+i\varepsilon_2(\omega)$ is bulk dielectric function. For p-polarized light, $B(z) = B(K,\omega,z)$ (where $K = \dfrac{\omega}{c}\sin\theta_i$ is small) and it obeys the following equation [13]

$$\frac{\partial}{\partial z}\left(\frac{1}{\varepsilon}\frac{\partial B}{\partial z}\right)+\left(\frac{\omega^2}{c^2}-\frac{K^2}{\varepsilon}\right)B = 0. \quad (2)$$

To solve the above equation, we follow the prescription of Landau and Lifsitz [14] where $B = u(z)\sqrt{\varepsilon(z)}$, so that

$$\frac{d^2u}{dz^2}+k^2\left(\varepsilon-\sin^2\theta_i\right)u+\left\{\frac{1}{2\varepsilon}\frac{d^2\varepsilon}{dz^2}-\frac{3}{4}\frac{1}{\varepsilon^2}\left(\frac{d\varepsilon}{dz}\right)^2\right\}u = 0. \quad (3)$$

In vacuum, $\varepsilon = 1$ and $d\varepsilon/dz = 0$ hence eq. (3) reduces to

$$\frac{d^2u}{dz^2}+k^2\cos^2\theta_i u = 0$$

and its solution is

$$u = A_0e^{-ik_z z}+B_0e^{ik_z z}, \quad (4)$$

where $k_z = \dfrac{\omega}{c}\cos\theta_i$.

In the bulk region, $\varepsilon = \varepsilon_b$ and $d\varepsilon/dz = 0$ hence eq. (3) reduces to

$$\frac{d^2u}{dz^2}+k^2\left(\varepsilon_b-\sin^2\theta_i\right)u = 0$$

and its solution is given by

$$u = A_1e^{-ik\sqrt{\varepsilon_b-\sin^2\theta_i}\,z} \quad (5)$$

In the surface region,

$$\frac{d\varepsilon}{dz} = \frac{\varepsilon_b-1}{(z-d)}\frac{1}{\log_e 2+(\varepsilon_b-1)\log_e\left(1-\dfrac{z}{d}\right)}$$

and

$$\frac{d^2\varepsilon}{dz^2} = \frac{-(\varepsilon_b-1)}{\log_e 2}\frac{1}{(z-d)^2}.$$

As $\left[\dfrac{1}{2\varepsilon}\dfrac{d^2\varepsilon}{dz^2}-\dfrac{3}{4}\left(\dfrac{1}{\varepsilon}\dfrac{d\varepsilon}{dz}\right)^2\right] << k^2\left(\varepsilon-\sin^2\theta_i\right)$ in the surface region, hence neglecting it, eq. (3) for the surface region can be written as

$$\frac{d^2u}{dz^2}+\frac{k^2}{2\log_e 2}\left[\log_e 2+2(\varepsilon_b-1)\log_e\left|1-\frac{z}{d}\right|\right]u = 0$$

As $\left|\dfrac{z}{d}\right| < 1$ in the surface region, since z lies between 0 and $-d$, we can write

$\log_e\left(1-\dfrac{z}{d}\right) = -\dfrac{z}{d}-\dfrac{z^2}{2d^2}$ Neglecting second and higher order terms, we get

$\dfrac{d^2u}{dz^2}+\dfrac{k^2}{d\log_e 2}(1-\varepsilon_b)z+\dfrac{k^2}{2}u = 0$. which can be better expressed as

$$\frac{d^2u}{dz^2}+(az+b)u = 0$$

with $a = \dfrac{k^2}{d\log_e 2}(1-\varepsilon_b)$ and $b = \dfrac{k^2}{2}$.

Thus, the solution of eq. (6) is $u(z) = A_i(\xi)+B_i(\xi)$.

where $\xi = \dfrac{-(az+b)}{a^{\frac{2}{3}}}$ and $A_i(\xi)$ and $B_i(\xi)$ are Airy functions [15] given by

$$A_i(\xi) = 3^{\frac{2}{3}}\Gamma\left(\frac{2}{3}\right)^{-1}\left(1+\sqrt{3}\right)\left[1+\frac{1}{3!}\xi^4+\ldots.\right]$$

$$B_i(\xi) = 3^{\frac{1}{3}}\Gamma\left(\frac{1}{3}\right)^{-1}\left(\sqrt{3}-1\right)\left[\varepsilon+\frac{2}{4!}\xi^4+\ldots.\right]$$

Absorbing the numerical coefficients and neglecting higher terms, we write the solution in the surface region as

$$u(z) = A_2\left(1+\frac{1}{3!}\xi^3\right)+B_2\left(\varepsilon+\frac{1}{4!}\xi^4\right).$$

As $B(z) = u(z)\sqrt{\varepsilon(z)}$ and using the boundary conditions that B and $\partial B/\partial z$ are continuous at $z=0$ and $z=-d$, we obtain

$$B(z) = \begin{cases} \left(e^{-ik\cos\theta_i z}+ye^{ik\cos\theta_i z}\right), & (vacuum) \\ \left[\left(1+\dfrac{\xi^3}{6}\right)+x\left(\xi+\dfrac{\xi^2}{12}\right)\right](\varepsilon)^{\frac{1}{2}}, & (surface) \\ pq\sqrt{\varepsilon_b}\,e^{-ik\sqrt{\varepsilon_b-\sin^2\theta_i}\,z}. & (bulk) \end{cases} \quad (8)$$

The electric field components can be obtained from the ignetic field by using the relation

$$E^{\perp}(\boldsymbol{K}, \omega, z) = \frac{c}{i\omega\varepsilon}\frac{dB}{dz},$$

$$E^{\parallel}(\boldsymbol{K}, \omega, z) = -\frac{\sin\theta_t}{\varepsilon} B.$$

Thus, we can write the vector potential in three regions as

$$\tilde{A}_{\omega}(z) = \frac{A^{z}_{\omega}(z)}{A_0} = \frac{E^{z}(\boldsymbol{K} \to 0, \omega, z)}{E_0}$$

$$\begin{cases} -\sin\theta_t\left(e^{-ik\cos\theta_t z} + ye^{ik\cos\theta_t z}\right), & (vacuum) \\ \dfrac{-\sin\theta_t}{\sqrt{\varepsilon}} \quad 1+\dfrac{\xi^3}{\zeta} \;|+x|\; \xi+\dfrac{\xi^2}{12} & (surface) \\ \dfrac{-\sin\theta_t}{} \; pqe^{-ik\sqrt{\varepsilon_b-\sin^2\theta_t}\, z}. & (bulk) \end{cases} \tag{9}$$

th $p = \dfrac{y+1}{1+\dfrac{\eta^3}{6}+x\left(\eta+\dfrac{\eta^4}{12}\right.} \qquad \dfrac{1+\dfrac{\gamma^3}{6}+x\left(\gamma+\dfrac{\gamma^4}{12}\right.}{\sqrt{\varepsilon_b}\,\epsilon^{\lambda d}}$

$$x = \cdot \frac{a^{\frac{2}{3}}\sqrt{\varepsilon_b}\gamma^2+\left(\beta-\sqrt{\varepsilon_b}\lambda\right)\left|1+\dfrac{\gamma^3}{6}\right.}{\left(\sqrt{\varepsilon_b}\lambda-\beta\right)\left|\gamma+\dfrac{\gamma^4}{12}\right.-\sqrt{\varepsilon_b}a^{\frac{1}{3}}\left(1+\dfrac{\gamma^3}{3}\right.}$$

$$\frac{ik\cos\theta_t(\Gamma+x\Delta)-\left(\dfrac{a^{\frac{1}{3}}}{2}\eta^2+\rho\Gamma\right)+x\left(a^{\frac{1}{3}}+a^{\frac{1}{3}}\dfrac{\eta^3}{3}+\rho\Delta\right)}{\dfrac{a^{\frac{1}{3}}}{2}\eta^2+\rho\Gamma+x\left(a^{\frac{1}{3}}+a^{\frac{1}{3}}\dfrac{\eta^3}{3}+\rho\Delta\right)+ik\cos\theta_t(\Gamma+x\Delta)}$$

$$\lambda = ik\sqrt{\varepsilon_b-\sin^2\theta_t}, \quad \beta = \frac{\varepsilon_b-1}{4d\sqrt{\varepsilon_b}\log_e 2}, \quad \gamma = \cdot \frac{ad-b}{}$$

$$= \frac{-b}{a^{\frac{1}{3}}}, \quad \rho = \frac{\varepsilon_b-1}{2d\log_e 2}, \quad \Gamma = 1+\frac{\eta^3}{6} \text{ and } \Delta = \eta+\frac{\eta^4}{12}. \tag{10}$$

Calculation of photocurrent

Golden rule expression for the current density [16] may be written as

$$\frac{dj(E)}{d\Omega} = \frac{2\pi}{\hbar}\sum\left|\langle\psi_f|H'|\psi_i\rangle\right|^2\delta(E-E_f)\delta(E_f-E_i-\hbar\omega) \times f_0(E-\hbar\omega)\left[1-f_0(E)\right], \tag{11}$$

where H' is the perturbation responsible for photoemission by a radiation of frequency ω, $|\psi_i\rangle$, and E_i refer to the initial state wavefunction and energy and $|\psi_f\rangle$ and E_f to the final state wavefunction and energy and $f_0(E)$ denotes the Fermi occupation function. We are considering the photoemission to take place along z-axis, which is normal to the surface plane $z = 0$. We may therefore write H' in one dimension as

$$H' = \frac{e}{mc}\left[\tilde{A}_{\omega}(z)\frac{d}{dz}+\frac{1}{2}\frac{d}{dz}\tilde{A}_{\omega}(z)\right|, \tag{12}$$

where $\tilde{A}_{\omega}(z) = \left(A^{z}_{\omega}(z)\right)/A_0$ with $A^{z}_{\omega}(z)$ as the component of vector potential along z-axis and A_0 is the amplitude of incident beam. The formula for photoemission cross section can be written as

$$\frac{d\sigma}{d\Omega} \approx \frac{k^2}{\omega}\left\langle\psi_f\left|\tilde{A}_{\omega}(z)\frac{d}{dz}+\frac{1}{2}\frac{d}{dz}\tilde{A}_{\omega}(z)\right|\psi_i\right\rangle \tag{13}$$

We will use here the formula deduced in eq. (9) for $\tilde{A}_{\omega}(z)$. To evaluate the photoemission crosssection (eq. (13)), we also need the initial and final state wavefunctions.

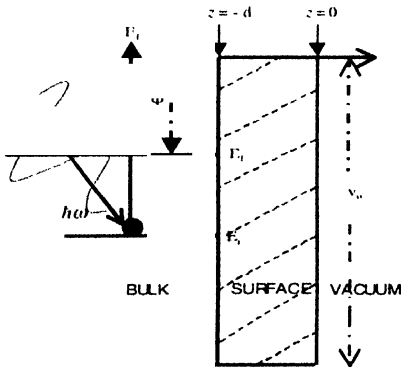

Figure 2. Schematic diagram of model potential for calculating initial state wavefunction in free electron model.

By matching the wavefunction at the surface plane $z = 0$, we may write the initial state wavefunction [17] as

$$\psi_i(z) = \begin{cases} \dfrac{m}{2\pi\hbar^2 k_i}\left\lfloor\exp(ik_i z)+\dfrac{ik_i+\chi}{ik_i-\chi}\exp(-ik_i z)\right|, & (z<0) \\ \dfrac{m}{2\pi\hbar^2 k_i}\dfrac{2ik_i}{ik_i-\chi}\exp(-\chi z), & (z>0) \end{cases} \tag{14}$$

where $\chi^2 = \frac{2m}{\hbar^2}(V_0 - E_i)$, $k_i^2 = \frac{2m}{\hbar^2}E_i$.

The final state wavefunction is the scattering state [18] of the step potential encountered by the electron and may be written as

$$\psi_f(z) = \begin{cases} \left(\frac{m}{2\pi\hbar^2\mu}\right)^{\frac{1}{2}} \frac{2\mu}{\mu + k_f} e^{\alpha|z|} e^{ik_f z}, & (z \le 0); \\ \left(\frac{m}{2\pi\hbar^2\mu}\right)^{\frac{1}{2}} \left[e^{i\mu z} + \frac{\mu - k_f}{\mu + k_f} e^{-i\mu z} \right], & (z \ge 0); \end{cases} \quad (15)$$

where $k_f^2 = \frac{2m}{\hbar^2}E_f$ and $\mu^2 = \frac{2m}{\hbar^2}(E_f - V_0)$.

The matrix element in eq. (13) for photoemission can be evaluated by using the above expressions for the vector potential and the wavefunctions. However, to ensure convergence [4] for $z < 0$, we introduce a convergence factor $e^{\alpha|z|}$ (for $z < 0$) in the calculation of the matrix element due to lifetime effects.

Expanding the matrix element in eq. (13), we have

$$\frac{d\sigma}{d\Omega} = \int_{-\infty}^{-d} \psi_f^* \tilde{A}_\omega(z) \frac{d\psi_i}{dz} dz + \frac{1}{2}\int_{-\infty}^{-d} \psi_f^* \frac{d\tilde{A}_\omega(z)}{dz} \psi_i dz +$$

$$\int_{-d}^{0} \psi_f^* \tilde{A}_\omega(z) \frac{d\psi_i}{dz} dz + \frac{1}{2}\int_{-d}^{0} \psi_f^* \frac{d\tilde{A}_\omega(z)}{dz} \psi_i dz +$$

$$\int_{0}^{\infty} \psi_f^* \tilde{A}_\omega(z) \frac{d\psi_i}{dz} dz + \frac{1}{2}\int_{0}^{\infty} \psi_f^* \frac{d\tilde{A}_\omega(z)}{dz} \psi_i dz, \quad (16)$$

where in each region, the wave functions and the vector potentials corresponding to that region have to be used. FORTRAN program was developed to evaluate the integrals in eq. (16).

4. Results and discussion

We have used the formula of eq. (9) for vector potential in calculating $|\tilde{A}_\omega(z)|$ as a function of $\hbar\omega$ for three locations of the planes at $z = 0$ (vacuum), $z = -d/2$ (surface) and $z = -d$ (bulk). By incorporating this vector potential $\tilde{A}_\omega(z)$, photocurrent has also been calculated as a function of $\hbar\omega$ for two values of surfaces, namely, $d = 5$ a.u. and $d = 0$ (narrow surface width). We have employed the initial state wavefunction ψ_i deduced by Thapa *et al* [17] for free electron model, hence the formalism developed has been applied to the case of Al and Be which are prototype free electron metals. For each case, experimentally determined dielectric constants [19, 20] had been used and also the following parameters were used for both the cases ;

Initial state energy $E_i = 11.7$ eV,

Work function $\Phi = 4.98$ eV,

Height of the step potential $V_0 = 15.95$ eV,

Surface width $d = 2.65$ Å,

Scattering constant $\alpha = 0.2$,

Angle of incidence $\theta_i = 45^0$.

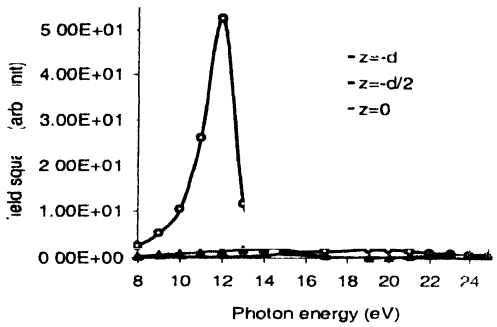

Figure 3. Plot of $|A(\omega,z)|^2$ as a function of photon energy for Al

In Figure 3, the plot of variation of $|\tilde{A}_\omega(z)|^2$ against photon energy is shown for three locations of surface planes in the case of Al. For the surface region ($z = -d/2$), we find that $|\tilde{A}_\omega(z)|^2$ increases to a maximum at photon energy $\hbar\omega$ ~ 12 eV it then decreases as $\hbar\omega$ is further increased. There is a minimum in $\tilde{A}_\omega(z)$ at $\hbar\omega = 15$ eV and with further increase in photon energy, it showed a second hump of very small magnitude $|\tilde{A}_\omega(z)|$ at $\hbar\omega \sim 18$ eV. The important point to note here is that $\tilde{A}_\omega(z)$ showed a minimum at $\hbar\omega = 15$ eV, which is the plasmon energy of Al. Similar trends had been observed by Levinson *et al* [21], Feibelman [5] and Barberan and Inglesfield [22]. The behaviour of $|\tilde{A}_\omega(z)|$ in the case of other locations of surface planes, for example at $z = -d$ (bulk) and $z = 0$ (vacuum), different than in the case of surface ($z = -d/2$).

The plot of variation of $|\tilde{A}_\omega(z)|^2$ against photon energy for Be shows same behaviour as in the case of Al and is shown in Figure 4. In the case of Be also we find that the variation of $|\tilde{A}_\omega(z)|^2$ with photon energy is interesting only for the surface region ($z = -d/2$). Here also, the plot of $|\tilde{A}_\omega(z)|^2$ against $\hbar\omega$ showed a maximum at $\hbar\omega = 13$ eV, and decreased towards minimum as $\hbar\omega$ was increased further. Most minimum in $|\tilde{A}_\omega(z)|^2$ in the case of Be occurred at $\hbar\omega = 20$ eV and showed

almost a constant minimum as $\hbar\omega$ increased further. Similar behaviour was obtained by Thapa and Kar [24] in which minimum was obtained at $\hbar\omega = 20$ eV.

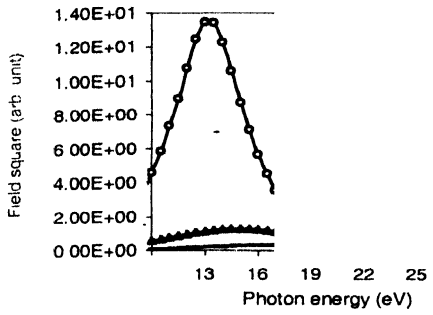

Figure 4. Plot of $|\tilde{A}_\omega(z)|^2$ as a function of photon energy for Be

In Figures 5 and 6, plots of photocurrent as a function of $\hbar\omega$ in the case of Al and Be are shown respectively, for two cases of surface widths namely $d = 2.65$ Å and $d = 0$ (narrow surface width). As it is a model calculation, we have chosen the same value of physical parameters for E_f, Φ, V_0 etc. However, the respective values of dielectric constant of each metal had been used for the calculations. In both the metals, we find that for $|\tilde{A}_\omega(z)|^2$, the behaviour of photocurrent is similar to the behaviour of $\tilde{A}_\omega(z)|^2$. The reason for this being that is involved in the matrix element of eq. (11). The results of photocurrent in the case of Al showed trends as obtained by Levinson [21], Feibelman [5], Thapa and Kar [8,11]. Similarly, in the case of Be, we find that the photocurrent results showed similar behaviour as obtained by Bartynski *et al* [23], Thapa and Kar [24].

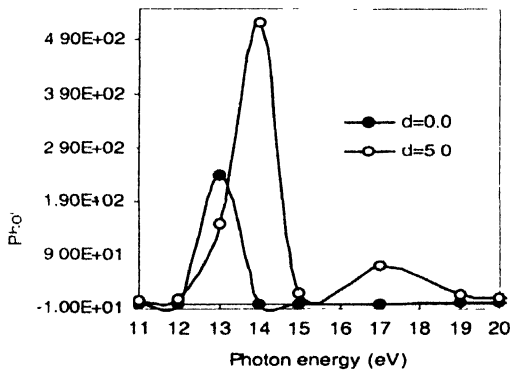

Figure 5. Photocurrent as a function of photon energy for Al with

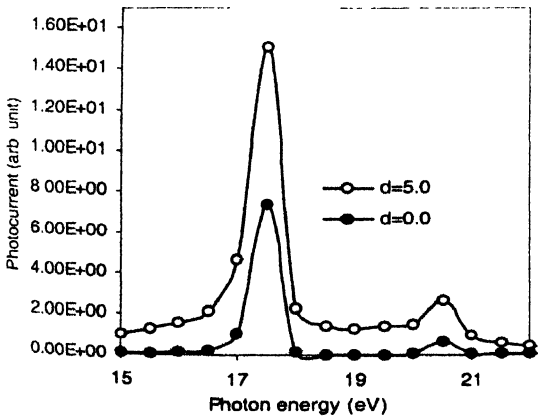

Figure 6. Photocurrent as a function of photon energy for Be with $\alpha = 0.2$.

5. Conclusion

The plots of photon field as well as photocurrent (Figures 5,6) agree qualitatively with the experimental results. We feel that proper choice of Initial and final state wavefunctions will enable us to extend the model to transition metals and semiconductors. Even though the choice of this dielectric model has no theoretical background, the dielectric model does work well specially in the case of Al.

Acknowledgment

RKT acknowledges a research grant and SRG fellowship from CSIR, New Delhi.

References

[1] B A Orlowski *Surface Sci* **200** 144 (1988)

[2] R McLean and R Haydock *J Phys.* **C10** 1929 (1977)

[3] A Liebsch *Phys. Rev. Lett* **32** 1203 (1974)

[4] J B Pendry *Surface Sci.* **57** 679 (1976)

[5] P J Feibelman *Phys Rev Lett.* **34** 1092 (1975), *Phys. Rev.* **B12** 1319 (1975)

[6] G Mukhopadhyay and S Lundquist *Solid State Commun.* **21** 629 (1977)

[7] A Bagchi and N Kar *Phys Rev.* **B18** 5240 (1978)

[8] P Das, R K Thapa and N Kar *Mod Phys Lett* **B5** 65 (1991)

[9] R K Thapa and N Kar *Phys. Stat. Sol. (b)* **179** 391 (1993)

[10] P Das and N Kar *Phys Stat. Sol. (b)* **187** 551 (1995)

[11] R K Thapa and N Kar *Indian J Phys.* **64A** 321 (1990)

[12] Zaithanzauva and R K Thapa *Indian J. Pure Appl. Phys.* **34** 843 (1996)

[13] L D Landau and E M Lifsitz *Electrodynamics of Continuous Media* (New York : Pergamon) Sec. **86** p293 (1984)

[14] L D Landau and E M Lifsitz *Electrodynamics of Continuous Media* (New York : Pergamon) Sec **88** p304 (1984)

[15] A K Ghatak, I C Goyal and S J Chua *Mathematical Physics* (Delhi : Mc Millan India) p66 and p459 (1995)

[16] D R Penn *Phys. Rev. Lett.* **28** 1041 (1972)

[17] R K Thapa, P Das and N Kar *DAE Proc Solid State Phys Symp.* (IIT Madras) **32C** 396 (1989)

[18] R K Thapa and N Kar *Indian J. Pure Appl Phys* **26** 620 (1988)

[19] J Weaver *Handbook of Chemistry and Physics of Solids* (Boca Raton, Ohio : CRC Press)

[20] N W Ashcroft and N D Mermin *Solid State Physics* (New York Holt, Reinehart and Winston) (1976)

[21] H J Levinson, E W Plummer and P J Feibelman *Phys Rev Lett* **43** 953 (1979)

[22] N Barberan and J E Inglesfield, *J. Phys.* C **14** 3114 (1981)

[23] R A Bartynski, E Jensen, T Gustaffson and E W Plummer *Phys Rev.* **B 32** 1921 (1985)

[24] R K Thapa and N Kar *Phys. Rev.* **B 51** 17980 (1995)