JOURNAL OF APPLIED PHYSICS VOLUME 95, NUMBER 9 1 MAY 2004

Electromagnetic absorption in transparent conducting films

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(Received 29 December 2003; accepted 2 February 2004)

In this article we calculate the skin depth of indium tin oxide (ITO) films (and related materials) as a result of free electron absorption within the visible spectrum using the simple Drude model. We also discuss the consequences of finite skin depth for the transparency of current spreading layers for light emitting diode (LED) applications. Low sheet resistances are highly desirable for these layers, but the free electron density n in ITO cannot be increased much beyond $2\times10^{21}\ \mathrm{cm}^{-3}$ without pulling the plasma frequency into the red end of the visible spectrum (thus making it highly reflective); furthermore, any increases in the film thickness cause reduced transparency due to the finite skin depth δ. However, above the plasma frequency for $n\approx10^{21}\ \mathrm{cm}^{-3}$ we find that increases in the electron mobility μ_e cause increases in δ, since then approximately $\delta\propto\mu_e$. Therefore, if μ_e in ITO can be increased above present state-of-the-art values around $50\ \mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$ to intrinsic limiting values around $100\ \mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$ by improved film processing, then substantial increases in transparency are possible whilst not sacrificing the high conductivity. The output optical power of a LED using an ITO current spreading layer with high n is also approximately proportional to μ_e, so mobility increases also have a direct impact on the external power efficiency of these devices.
 [DOI: 10.1063/1.1689735]

I. INTRODUCTION

The transparent conductor indium tin oxide [In_2O_3:Sn (ITO)] and related materials are used in a variety of applications,[1,2] not least for transparent electrodes in opto-electronic devices. An important example is transparent current spreading layers in light emitting diodes (LEDs), where a low sheet resistance $R_\square$ enhances the optical output power of the device.[3] The sheet resistance $R_\square=1/ne\mu_e t$ can be reduced by increasing the free electron density n, the electron mobility μ_e, and the film thickness t. However, increases in n and μ_e also affect the electromagnetic skin depth δ associated with free electron absorption, which determines ultimately the film transparency since the power transmission coefficient T is approximately proportional to the factor $\exp(-2t/\delta)$ (as we will see later). In this article we calculate δ and T and assess the effects of increases in n and μ_e up to their limiting values in ITO films. We ignore photon absorption due to in-band states under the assumption that this can be suppressed by appropriate materials processing.

II. DRUDE MODELING OF THE OPTICAL PROPERTIES

The basic physics of ITO and related materials are now well-understood. The "optical window" for ITO is set at short wavelengths by its energy gap (around 3.8 eV)[4] and at longer wavelengths by its plasma edge, which is in the near infrared provided that the free electron density n does not exceed $2\times10^{21}\ \mathrm{cm}^{-3}$. For ITO the replacement of In^{3+} by Sn^{4+} within the host In_2O_3 lattice results in n-type doping. For doping levels above that set by the Mott criterion[5] (i.e., $n>n_c\approx10^{19}\ \mathrm{cm}^{-3}$ for ITO), one can consider ITO to have a full valence band and a parabolic conduction band partially filled by a degenerate free electron gas.[4,6]

Within this free electron framework the optical properties are described quite well by the simple Drude model but with a strongly frequency-dependent electron scattering time τ for Fermi surface electrons.[4] Electron scattering in ITO films occurs from a number of different scattering centers such as neutral impurities, phonons, and defects, though the most dominant process is likely to be from the impurity ions responsible for the doping.[6] Considering for the moment only frequencies within the visible spectrum (for which we can assume that τ is approximately constant), the Drude forms for real and imaginary parts of the dielectric function $\varepsilon=\varepsilon_1-i\varepsilon_2$ are[7]

$$\varepsilon_1=\varepsilon_\infty-\frac{\omega_N^2\tau^2}{1+\omega^2\tau^2},\quad \varepsilon_2=\frac{1}{\omega}\,\frac{\omega_N^2\tau}{1+\omega^2\tau^2},$$

where $\omega_N^2=ne^2/\varepsilon_0 m^*$, m^* is the free electron effective mass, and ε_∞ is the high frequency relative permittivity of the ITO lattice. Collected ITO data from the literature[4,8] suggest that $\varepsilon_\infty\approx4.0$ and $m^*\approx0.35m_e$. Electron mobilities reported for high quality ITO films are around $\mu_e\approx50\ \mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$,[4,6] giving a dc scattering time $\tau_0\approx m^*\mu_e/e\approx10^{-14}$ s. Drude fits to the optical data of these films suggest that[4] $\tau\approx3.3\times10^{-15}$ s, somewhat lower than the value of τ_0 inferred from μ_e, so in our calculations we assume that $\tau\approx\tau_0/3$. The important assumption here is that $\mu_e\propto\tau_0\propto\tau$; the scaling factor relating τ and τ_0 does not affect our general conclusions.

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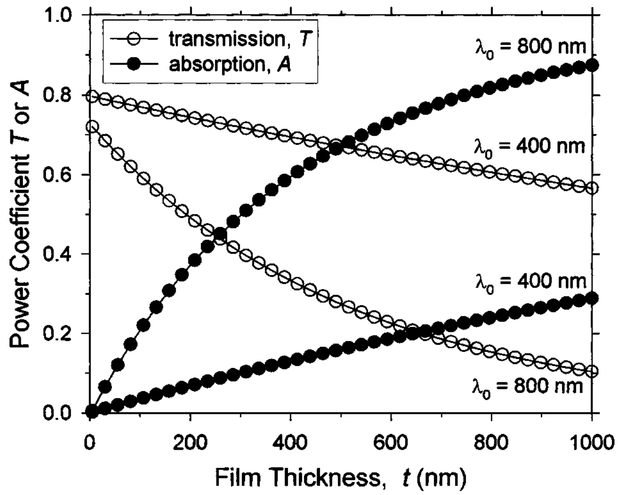

FIG. 1. The power transmission coefficient T and power absorption coefficient A as a function of film thickness t calculated for an ITO film of electron density $n=1.5\times10^{21}\ \mathrm{cm}^{-3}$ and mobility $\mu_e=50\ \mathrm{cm}^2\ \mathrm{V}^{-1}\ \mathrm{s}^{-1}$ for normally incident, incoherent light of free space wavelengths of 400 and 800 nm (we assume that the ITO film is deposited on a lossless, thick substrate of refractive index 4.0). To a very good approximation $T(t)/T(0)\approx\exp(-2t/\delta)$, whilst to a less good approximation $A(t)\approx1-\exp(-2t/\delta)$.

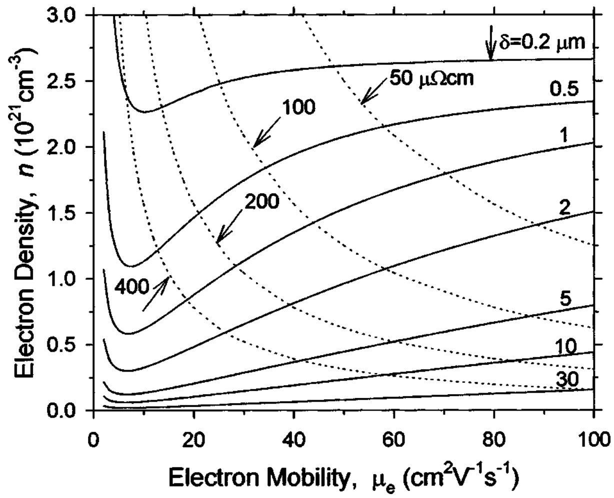

FIG. 2. Contours of the skin depth δ (in units of microns) as a function of electron density n and mobility μ_e for an ITO film at an incident wavelength $\lambda_0=800$ nm. Also shown are contours of constant resistivity $\rho=1/ne\mu_e$ in units of $\mu\Omega$ cm (dotted lines).

The condition $\varepsilon_1(\omega_p)=0$ defines the plasma frequency ω_p of a conducting material. When $\omega<\omega_p$ a conductor is highly reflective since then $\varepsilon_1<0$. For ITO in the limit of high electron density (i.e., $n>10^{21}\ \mathrm{cm}^{-3}$) and mobility (i.e., $\mu_e>20\ \mathrm{cm}^2\ \mathrm{V}^{-1}\ \mathrm{s}^{-1}$) then $\omega_N^2\tau^2/\varepsilon_\infty\gg1$ and $\omega_p\approx\omega_N/\sqrt{\varepsilon_\infty}\propto\sqrt{n}$, so that in this limit ω_p is determined almost exclusively by n but not by μ_e (i.e., τ). Hence, for a film to be nonreflective to light of free space wavelength λ_0 the electron density n must satisfy $n(\mathrm{cm}^{-3})<1.6\times10^{27}/\lambda_0^2(\mathrm{nm}^2)$. For efficient transmission of the whole visible spectrum (including red light of wavelength up to 780 nm), the free electron density should therefore not exceed $2.6\times10^{21}\ \mathrm{cm}^{-3}$. To allow a suitable "safety margin," we consider a plasma wavelength just over 1 μm (i.e., in the near infrared), in which case the maximum tolerable free electron density is about $n_{\max}\approx1.5\times10^{21}\ \mathrm{cm}^{-3}$. This value has already been exceeded in various ITO films (for example, Ref. 9), but clearly values of $n>n_{\max}$ are detrimental to the transparency of the films owing to the plasma edge creeping into the red part of the visible spectrum, resulting in a greatly reduced skin depth.

III. CALCULATIONS OF SKIN DEPTH AND FILM TRANSPARENCY

The importance of free electron absorption and the limitations imposed by finite skin depth on the transparency of high quality ITO films is illustrated in Fig. 1; here we calculate the power transmission coefficient T as a function of thickness t from the wave number $k=\omega\sqrt{\varepsilon}/c$ and plane wave impedance $Z=\omega\mu_0/k$ for a film of free electron density $n=1.5\times10^{21}\ \mathrm{cm}^{-3}$ and electron mobility $\mu_e=50\ \mathrm{cm}^2\ \mathrm{V}^{-1}\ \mathrm{s}^{-1}$ for incoherent light of free space wavelengths of 400 and 800 nm. We assume normal incidence and also that the film is deposited on a lossless, thick substrate of refractive index of 4.0 (appropriate for AlGaInP LEDs); this value determines $T(0)\approx0.80$, but otherwise does not influence our main conclusions. The corresponding power absorption coefficients A are also shown in Fig. 1. It is clear from these plots that the transparency of such a high quality ITO film (with resistivity about 100 $\mu\Omega$ cm) is severely limited by electromagnetic absorption within the skin depth at film thicknesses of 500 nm or more, which is more acute at the red end of the visible spectrum.

The skin depth is defined in terms of wave number k by $\delta=-1/\mathrm{Im}(k)$, so that

$$\delta=\frac{c}{\omega}\left(\frac{2}{\sqrt{\varepsilon_1^2+\varepsilon_2^2}-\varepsilon_1}\right)^{1/2}, \tag{1}$$

which can be calculated from the Drude forms for ε_1 and ε_2. Results of these calculations are shown in Fig. 2, where we plot contours of constant δ as a function of the main variable material parameters n and μ_e for an incident wavelength of 800 nm. The skin depth becomes very small for large values of n, since then ω approaches the plasma frequency ω_p. Films with high n therefore need to be very thin (typically $<\delta/5$) to preserve a high transparency. Similar contours are obtained for shorter wavelengths (e.g., 400 nm), but with larger values of δ for the same values of n and μ_e since then ω is further above ω_p. The increased values of δ when $\mu_e<10\ \mathrm{cm}^2\ \mathrm{V}^{-1}\ \mathrm{s}^{-1}$ are consequences of ω_p increasing with decreasing μ_e in this low mobility limit. For the film of Fig. 1 we calculate $\delta\approx5.9\ \mu$m and $\delta\approx1.0\ \mu$m for incident wavelengths of 400 and 800 nm, respectively. The power transmission coefficient $T(t)$ for incoherent light very closely follows the function $T(0)\exp(-2t/\delta)$, as one might have expected, with $T(0)\approx0.80$ (approximately constant for frequencies well above ω_p); for the film of Fig. 1, this gives $T(500\ \mathrm{nm})/T(0)\approx0.84$ and 0.37 for wavelengths of 400 and 800 nm, respectively,

For electron densities up to around $n_{\max}\approx1.5\times10^{21}\ \mathrm{cm}^{-3}$ and mobilities $\mu_e>10\ \mathrm{cm}^2\ \mathrm{V}^{-1}\ \mathrm{s}^{-1}$ then ε_1

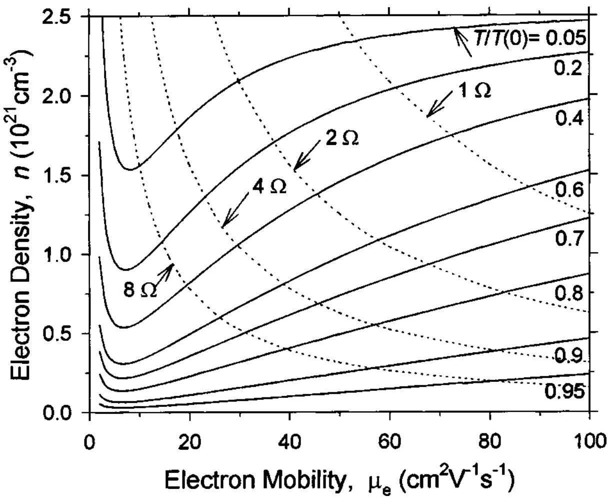

FIG. 3. Contours of the normalized power transmission coefficient $T(t)/T(0)$ as a function of electron density n and mobility μ_e for an ITO film of thickness $t=500$ nm at an incident wavelength $\lambda_0=800$ nm. Also shown are contours of constant sheet resistance $R_\square=1/ne\mu_e t$ in units of Ω (dotted lines).

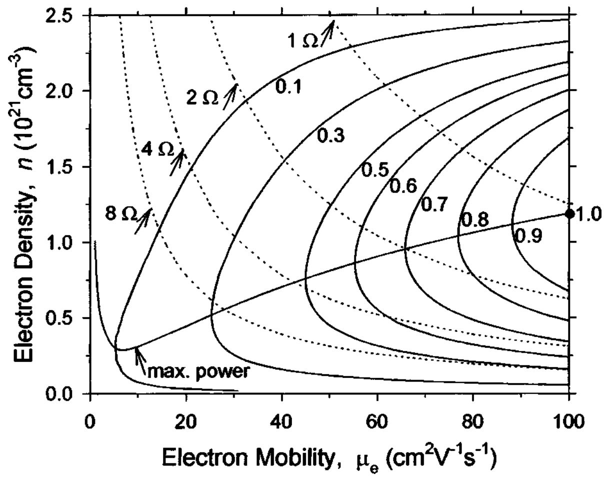

FIG. 4. Contours of maximum LED output power as a function of electron density n and mobility μ_e for an ITO spreading layer of thickness $t=500$ nm at an incident wavelength of $\lambda_0=800$ nm. The labeled values are normalized to the maximum output power when $\lambda_0=800$ nm and $\mu_e=100\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$. Also shown are contours of constant sheet resistance (dotted lines) and the locus of maximum output power.

$\approx\varepsilon_\infty$ and $\varepsilon_2\approx\omega_N^2/\omega^3\tau\ll\varepsilon_\infty$. We can now use Eq. (1) to deduce a simple approximate formula for the skin depth in this limit that illustrates clearly its dependence on the main variable material parameters n and μ_e, giving

$$\delta\approx\frac{2c}{\omega}\frac{\sqrt{\varepsilon_1}}{\varepsilon_2}\approx\frac{2m^*}{Z_\infty e^2}\frac{\omega^2\tau}{n}, \tag{2}$$

where $Z_\infty\approx 377\ \Omega/\sqrt{\varepsilon_\infty}$ is the high frequency wave impedance of ITO. Equation (2) yields a good approximation for δ towards the bottom right hand portion of Fig. 2. From Eq. (2) we observe that $\delta\propto\omega^2\mu_e/n$ (assuming that $\tau_0\propto\tau$), so that increasing n leads to the expected reduction in δ as ω_p is reduced, together with an associated reduction in the transparency of the film. However, Eq. (2) indicates that an *increase* in mobility for fixed n actually *increases* the skin depth and *increases* the film transparency. Since $\delta\propto\omega^2$, the smallest value of δ within the visible spectrum occurs for red light. Hence, if the transparency of an ITO film is deemed adequate for red light, it will be even better for blue light since then we are even further from the plasma edge and the skin depth is approximately four times larger.

In Fig. 3 we plot contours of normalized power transmission coefficient $T(t)/T(0)$ as a function of n and μ_e at 800 nm wavelength for an ITO film of thickness $t=500$ nm. Since $T(t)/T(0)\approx\exp(-2t/\delta)$, the contours of $T(t)/T(0)$ follow those of constant δ shown in Fig. 2. We obtain similar contours of $T(t)/T(0)$ for smaller values of t, except with values of $T(t)/T(0)$ closer to 1. Again, Eq. (2) yields a good approximation for δ towards the bottom right hand portion of Fig. 3. Hence, increased T at fixed n is achieved by increasing μ_e. This result has an important consequence if high transparency layers with very low sheet resistance $R_\square$ are required. For example, if μ_e could be doubled whilst preserving high n then, assuming that $\mu_e>10\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$ to start with, in principle the layer could be grown to twice the thickness whilst preserving the same value of T; hence, $R_\square$ for this layer could be reduced by a factor of 4, which is a very significant result that illustrates the importance of maximising μ_e.

From Fig. 3 we see that values of $T(t)/T(0)$ very close to 1 are obtained over a wide range of μ_e but at low n, which clearly is not compatible with the common requirement of low sheet resistance $R_\square$. This is illustrated by the contours of constant $R_\square$ in Fig. 3, where very low values of $R_\square$ (e.g., around 1 Ω for a 500-nm-thick film) are only possible if the transparency of the film is compromised.

IV. IMPLICATIONS FOR LED SPREADING LAYERS

In this context, we consider finally the application of ITO films and related materials for LED current spreading layers. The optical power transmitted through a current spreading layer is proportional to the factor $(n\mu_e t)^{1/2}\times\exp(-2t/\delta)$, where the term $(n\mu_e t)^{1/2}$ accounts for the increased spreading current as the thickness t is increased[10] and the term $\exp(-2t/\delta)$ accounts for the increased electromagnetic absorption. The output power increases monotonically with increasing mobility μ_e for all fixed values of n and t, but is maximized when varying n or t for fixed μ_e. For example, when $t=4\delta$ the output power has a maximum value proportional to $(n\mu_e\delta)^{1/2}$, which from Eq. (2) is approximately proportional to μ_e for large n and μ_e. Hence, if mobility μ_e ($>10\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$) could be doubled while maintaining high n then there will be a doubling of the optical power transmitted by the current spreading layer, another significant result.

We illustrate this further in Fig. 4, where we use Eq. (1) to plot the contours of normalized LED output power as a function of n and μ_e for a 500-nm-thick film at a wavelength of 800 nm. For each value of μ_e there is an optimized value of n for maximum output power which lies on the locus shown in Fig. 4. This optimization is not too critical, however, since the output power falls away gradually for nonoptimized values of n. We observe that a reduced sheet resistance $R_\square$ gives rise to a larger optical output power provided that $n < n_{max} \approx 1.5 \times 10^{21}$ cm^{-3} (i.e., ω_p is still well into the infrared). Therefore, $R_\square$ for a current spreading layer should be reduced by increasing μ_e to its maximum possible value while maintaining n just below n_{max}. This might be achieved by improved film processing so that μ_e approaches its intrinsic limit, which in ITO is set at around 100 cm^2 V^{-1} s^{-1} as a result of ionized impurity scattering.[6]

V. SUMMARY AND CONCLUSIONS

In summary, we have used the simple Drude model to investigate the parameters important for electromagnetic absorption in ITO films of high carrier density. A simple approximation for the skin depth above the plasma frequency shows that it is proportional to electron mobility. We have then used this result to show that an increase in mobility gives rise to an increase in the optical transparency of an ITO film and also the optical output power of a LED when using ITO as its current spreading layer. These results have bearing on the applications of alternative transparent conductor systems, where it may be possible to achieve even higher mobilities; for example, electron mobilities as high as 140 cm^2 V^{-1} s^{-1} have been reported in $InGaO_3(ZnO)$,[11] but free electron densities are presently very low in this material.

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