Electro-Optic Functions for Interpretation of Experimental Data*

Joel Grover, Stephen Koeppen, and Paul Handler
Physics Department and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801
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We reproduce here a number of line shapes which we believe will be of value to experimentalists in the field of electroreflectance. We have found them to be very useful in our work on the higher-energy critical points of germanium and gallium arsenide.

I. INTRODUCTION

Much work has been done in the field of electroreflectance since the one-electron line shapes for the field-induced change in the dielectric functions at a three-dimensional critical point were calculated by Aspnes in 1966.[1] Since then, it has been shown that excitons, thermal broadening, and nonuniform electric fields may drastically modify these line shapes.[2-6] Furthermore, it now appears that the two- and one-dimensional critical-point theories may be applicable to certain materials.[7-9] Although Cardona has cataloged the one-, two-, and three-dimensional one-electron line shapes for the various types of critical points,[2] we feel it would be very useful to the experimentalist to show the results in graphic form, including the effects of thermal broadening and electric field inhomogeneity over the entire range of their effect. We have therefore calculated the effect of broadening on the one-, two-, and three-dimensional one-electron line shapes and, in addition, the effect of field inhomogeneity on the three-dimensional line shapes. We present the results in this comment.

II. ELECTRO-OPTIC FUNCTIONS

The electric-field-induced change in the dielectric function $\Delta\epsilon$ can be expressed in terms of the one-, two-, and three-dimensional electro-optic F and G functions defined as follows[10]:

$$
\begin{aligned}
F_1(x) &= 2\pi\,\mathrm{Ai}^2(x) - H(-x)(-x)^{-1/2}\,,\\
G_1(x) &= 2\pi\,\mathrm{Ai}(x)\,\mathrm{Bi}(x) - H(x)x^{-1/2}\,,\\
F_2(x) &= \mathrm{Ai}_1(\kappa x) - H(-x)\,,\\
G_2(x) &= \mathrm{Gi}_1(\kappa x) + \pi^{-1}\ln|x| + C_0\,,\\
F_3(x) &= \pi\left[\mathrm{Ai}'^2(x) - x\,\mathrm{Ai}^2(x)\right] - (-x)^{1/2}H(-x)\,,\\
G_3(x) &= \pi\left[\mathrm{Ai}'(x)\,\mathrm{Bi}'(x) - x\,\mathrm{Ai}(x)\,\mathrm{Bi}(x)\right] + x^{1/2}H(x)\,,
\end{aligned}
\tag{1}
$$

where $\kappa = 2^{2/3}$, $H(x)$ is the unit-step function, $\mathrm{Ai}(x)$ and $\mathrm{Bi}(x)$ are the regular and irregular Airy functions,

$$\mathrm{Ai}_1(x) = \int_x^\infty \mathrm{Ai}(t)\,dt\,,$$

$$\mathrm{Gi}_1(x) = -\int_0^x \mathrm{Gi}(t)\,dt\,,$$

$$C_0 = \int \mathrm{Gi}(t)\,dt\big|_0 - \pi^{-1}\ln\kappa\,,$$

and

$$\mathrm{Gi}(x) = \pi^{-1}\,\mathrm{Im}\int_0^\infty ds\, e^{i(s^3/3+sx)}$$

is the Kramers-Kronig transform of $\mathrm{Ai}(x)$. The expressions for $\Delta\epsilon$ in terms of the F and G functions are summarized in Table I.[2] The critical points are labeled according to dimensionality and signs of the reduced effective masses μ_i, minima having all masses positive, maxima having all masses negative, and saddle points having one (M_1) or two (M_2) masses negative. In the case of two- and three-dimensional saddle points one more parameter is required, the sign of the reduced effective mass in the direction of the field, defined as

$$\frac{1}{\mu_F} = \frac{1}{\mathcal{E}^2}\sum_{i=1}^{\dim}\frac{\mathcal{E}_i^2}{\mu_i}\,,$$

where dim = dimension.

III. BROADENING

The effect of broadening is usually calculated by convoluting the unbroadened line shape with a Lorentzian:

$$
\begin{aligned}
\Delta\epsilon^B(\omega) &= \frac{\gamma}{\pi}\int_{-\infty}^{\infty} d\omega'\,\frac{\Delta\epsilon(\omega')}{(\omega'-\omega)^2+\gamma^2}\\
&= \Delta\epsilon(\omega+i\gamma)\,.
\end{aligned}
\tag{2}
$$

Here $\Delta\epsilon(\omega')$ is the unbroadened differential dielectric function $\Delta\epsilon_1(\omega') + i\Delta\epsilon_2(\omega')$; $\Delta\epsilon_{1,2}(\omega')$ may be found from Table I. $\Delta\epsilon^B(\omega)$ is the broadened differential dielectric function, calculated from Eq. (3) below. Although the prefactor of the F and G functions contains $1/\omega^2$, which tends to $1/(\omega+i\gamma)^2$ when broadened, this is usually neglected and the broadened $\Delta\epsilon$ is then expressed in terms of broadened F and G functions, denoted by F^B and G^B. In Fig. 1, we display $F^B(\eta)$ and $G^B(\eta)$ calculated from complex Airy functions as follows:

$$F_1^B(\eta) + iG_1^B(\eta) = 2\pi\,\mathrm{Ai}(z)[\mathrm{Ai}(z) + i\,\mathrm{Bi}(z)] - iz^{-1/2}\,,$$

TABLE I. Electric-field-induced change in the dielectric function near the different types of one-, two-, and three-dimensional critical points.

Dimension	Type of critical point		Sign of μ_F	$\Delta\epsilon_1$	$\Delta\epsilon_2$
1	minimum		+	$A_1G_1(-\eta)$	$A_1F_1(-\eta)$
1	maximum		−	$-A_1G_1(\eta)$	$A_1F_1(\eta)$
2	minimum		+	$A_2G_2(-\eta)$	$A_2F_2(-\eta)$
2	saddle points		−	$-A_2F_2(\eta)$	$A_2G_2(\eta)$
2	saddle points		+	$A_2F_2(-\eta)$	$A_2G_2(-\eta)$
2	maximum		−	$-A_2G_2(\eta)$	$A_2F_2(\eta)$
3	minimum	(M_0)	+	$A_3G_3(-\eta)$	$A_3F_3(-\eta)$
3	saddle points	(M_1)	−	$A_3G_3(\eta)$	$-A_3F_3(\eta)$
3	saddle points	(M_1)	+	$-A_3F_3(-\eta)$	$A_3G_3(-\eta)$
3	saddle points	(M_2)	+	$-A_3G_3(-\eta)$	$-A_3F_3(-\eta)$
3	saddle points	(M_2)	−	$A_3F_3(\eta)$	$A_3G_3(\eta)$
3	maximum	(M_3)	−	$-A_3G_3(\eta)$	$A_3F_3(\eta)$

$$\eta = (\omega-\omega_g)/\theta$$
$$\theta = (e^2\mathcal{E}^2/2\mu_F\hbar)^{1/3}$$
$$\frac{1}{\mu_F} = \sum_{i=1}^{\dim} \frac{1}{|\mathcal{E}|^2}\,\frac{\mathcal{E}_i^{\,2}}{\mu_i}$$

$C_0 =$ Dipole matrix element

$$A_1 = \frac{4\pi e^2C_0^{\,2}}{m^2\omega^2}\left(\frac{2\mu}{\hbar^2}\right)^{1/2}\frac{1}{(\hbar\theta)^{1/2}}$$
$$A_2 = \frac{2\pi e^2C_0^{\,2}}{m^2\omega^2}\left(\frac{4\mu_1\mu_2}{\hbar^4}\right)^{1/2}$$
$$A_3 = \frac{2e^2C_0^{\,2}}{m^2\omega^2}\left(\frac{8\mu_1\mu_2\mu_3}{\hbar^6}\right)^{1/2}(\hbar\theta)^{1/2}$$

$$F_2^B(\eta)+iG_2^B(\eta)=\mathrm{Ai}_1(\kappa z)+i\,\mathrm{Gi}_1(\kappa z)+i\pi^{-1}\ln z\,, \qquad (3)$$

$$F_3^B(\eta)+iG_3^B(\eta)=\pi\,\mathrm{Ai}'(z)[\mathrm{Ai}'(z)+i\,\mathrm{Bi}'(z)] - \pi z\,\mathrm{Ai}(z)[\mathrm{Ai}(z)+i\,\mathrm{Bi}(z)]+iz^{1/2} \times(0\le\arg z<2\pi)\,,$$

where $z=\eta+i\Gamma$ and $\Gamma=\gamma/\theta$ is a dimensionless broadening parameter. The range of broadening parameters $\Gamma=0$ to 3 covers the gamut of shapes of the broadened F and G functions. That is, for $\Gamma>3$, the line shape is similar to that shown for $\Gamma=3$ but expanded in the horizontal direction by a factor $\frac{1}{3}\Gamma$ and diminished in amplitude by $(3/\Gamma)^{4-\dim/2}$. This can be seen from the explicit asymptotic form for $\Gamma>3$: $G^B(\eta)-iF^B(\eta)\sim C_{\dim}/z^{4-\dim/2}$, where $C_{\dim}$ is a real constant depending on the dimension.

IV. ELECTRIC FIELD SPATIAL INHOMOGENEITY

Aspnes and Frova[5] have shown that in the presence of a nonuniform electric field, the "effective" $\Delta\epsilon$, denoted by $\langle\Delta\epsilon\rangle$, which one calculates by a Kramers-Kronig analysis of the measured quantity $\Delta R/R$, can be related to the uniform-field line shapes for $\Delta\epsilon$ by the expression

$$\langle\Delta\epsilon\rangle=\langle\Delta\epsilon_1\rangle+i\langle\Delta\epsilon_2\rangle = -2i\kappa\int_{-\infty}^{0} e^{-2iKz}\,\Delta\epsilon(\mathcal{E}(z))\,dz\,, \qquad (4)$$

where $\mathcal{E}(z)$ is the electric field, z is the depth into the surface, $K=2\pi(n+ik)/\lambda$, n and k are the optical constants, and $\Delta\epsilon(\mathcal{E}(z))$ is the uniform-field line shape at the field $\mathcal{E}(z)$. Aspnes and Frova have shown the predictions of Eq. (4) for intrinsic germanium at several fields. For the purpose of more general applicability, it is useful to express Eq. (4) in terms of dimensionless variables as is done with the broadened line shapes of Eq. (2). We refer to the line shapes of Eq. (4) as "mixed" line shapes since the phase factor in the integrand causes mixing of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ with one another.

If we let $K=|K|e^{i\varphi}$, where $\varphi=\tan^{-1}(k/n)$, assume a linear field profile $\mathcal{E}=\mathcal{E}_s(1+z/L)$, and change the integration variable to $y=\mathcal{E}_s/\mathcal{E}$, we get

$$\langle\Delta\epsilon\rangle=\frac{-ie^{i\varphi}}{\beta}\int_1^\infty dy\,\frac{\Delta\epsilon(y)}{y^2}\,e^{-(i/\beta)(1/y-1)e^{i\varphi}}\,, \qquad (5)$$

where $\beta=\frac{1}{2}|K|L=R_1/2|K|$, where $R_1=d\ln\mathcal{E}/dz|_{z=0}$.[11] β is the ratio of the "effective" penetration depth of the light, $\frac{1}{2}|K|$,[11] to the width of the space-charge region, L. $\langle\Delta\epsilon\rangle$ in Eq. (5) depends only on the dimensionless quantities β, φ, η, and Γ. The quantities β and φ are a convenient means of describing the field inhomogeneity since, for a given semiconductor, φ depends only on the photon energy while β, the ratio of penetration depth to space-charge width, can be varied by the experimentalist by changing the electric field and sample doping.[11] We have computed curves for $\langle\Delta\epsilon_1\rangle$ and $\langle\Delta\epsilon_2\rangle$ vs η from Eq. (5) at zero broadening ($\Gamma=0$) for three values of β and for the cases $k=0$ and $k=n$. These are shown in Fig. 2. Although we have assumed a linear field profile in order to get the

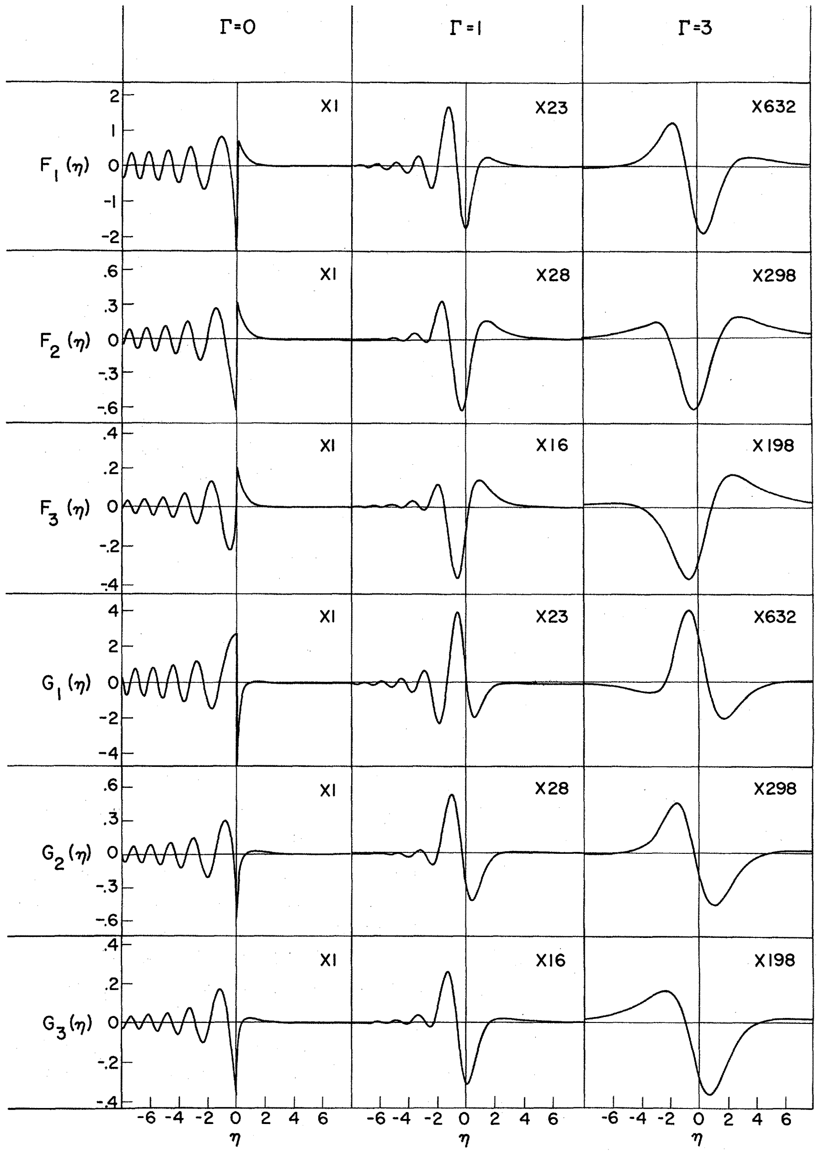

FIG. 1. One-, two-, and three-dimensional electro-optic F and G functions. $\eta=(\omega-\omega_g)/\theta$ and $\Gamma=\gamma/\theta$, where $\hbar\omega_g$ is the energy gap, $\hbar\theta$ is the electro-optic energy, and $\hbar\gamma$ is the broadening energy. The values $\Gamma=0$ to 3 cover the gamut of shapes of the broadened F and G functions. The scale factors shown are the amount by which the vertical scale has been expanded.

general expression (5), we have found that the line shapes are fairly insensitive to the detailed shape of the space-charge region. For example, we found no significant difference in the shape of $\langle\Delta\epsilon\rangle$ for a linear and an exponential field profile, with the same value for β. For a small field inhomogeneity, one is generally in the depletion region anyway,[11] in which case the field profile is in fact linear.

As previously demonstrated by Aspnes and Frova,[5,6] Fig. 2 shows that the effect of the "mixing" is to diminish the size of subsidiary oscillations and modify the relative heights of the first few peaks. For $k=0$ in the large mixing limit ($\beta>3$), $\langle\Delta\epsilon_1\rangle$ is similar to the uniform-field line shape for $\Delta\epsilon_2$, while $\langle\Delta\epsilon_2\rangle$ is similar to the negative of the $\beta=0$ curve for $\Delta\epsilon_1$, as previously noted.[5]

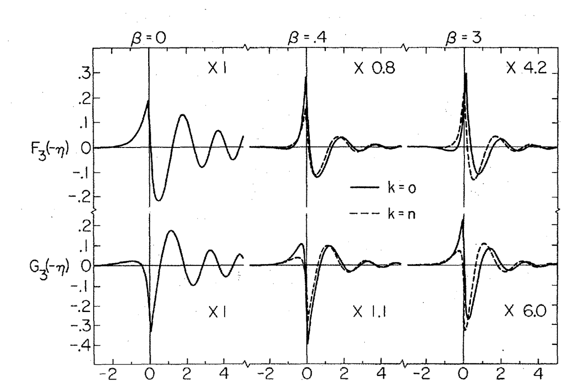

FIG. 2. Three-dimensional unbroadened ($\Gamma=0$) F and G functions for various degrees of electric field inhomogeneity β (see text). k and n are the optical constants. The values $\beta=0$ to 3 cover the gamut of shapes of the "mixed" (see text) F and G functions.

However, when $k=n$, the effect of the mixing is less pronounced in terms of modifying the first few peaks and one does not get the complete interchange of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ in the limit of large β. For $\beta>3$, the shape of $\langle\Delta\epsilon\rangle$ is the same as for $\beta=3$, while the magnitude decreases as $1/\beta$.

We have found the line shapes shown in Figs. 1 and 2 to be of great value in our experimental work and we hope that others will find them as useful.

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[1]D. E. Aspnes, Phys. Rev. 147, 554 (1966); 153, 972 (1967).

[2]M. Cardona, *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1969), Suppl. 11, Chap. 7.

[3]D. F. Blossey, Phys. Rev. B 2, 3976 (1970).

[4]B. O. Seraphin and N. Bottka, Phys. Rev. 145, 628 (1966).

[5]D. E. Aspnes and A. Frova, Solid State Commun. 7, 155 (1969).

[6]A. Frova and D. E. Aspnes, Phys. Rev. 182, 795 (1969).

[7]S. Jasperson, S. Koeppen, and P. Handler, in Proceedings of the Tenth International Conference on the Physics of Semiconductors, Cambridge, Mass., 1970, p. 432 (unpublished).

[8]S. Koeppen, P. Handler, and S. Jasperson, Phys. Rev. Letters (to be published).

[9]S. Pond (unpublished).

[10]Reference 2, pp. 172, 173, and 180. There seem to be some errors present in the definition of the two-dimensional electro-optic functions on p. 180. The correct expressions are given in Eqs. (1).

[11]S. Koeppen and P. Handler, Phys. Rev. 187, 1182 (1969).

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ERRATA

Raman Scattering in Gray Tin. C. J. Buchenauer, M. Cardona, and F. H. Pollak [Phys. Rev. B 3, 1243 (1971)]. Because of errors in composition, the associations of Figs. 1 and 2 with their captions were incorrect. These figures, but not their captions, should be interchanged.

Phase Transition in a Wigner Lattice, Leslie L. Foldy [Phys. Rev. B 3, 3472 (1971)]. The accuracy of the moments given in Table I should have been specificed to be "one part in 10^4 or better" rather than "one part in 10^5 or better." However, the uncovering of a small error in the program has revealed that even this claim is not correct, but that the error in the table never exceeds 0.00028. The corrected moments of maximum interest are (a) for the bcc lattice: $u_1=0.511\,389$, $u_4=0.203\,078$, $u_6=0.149\,391$, $u_8=0.117\,720$; (b) for the fcc lattice: $u_1=0.513\,194$, $u_4=0.202\,642$, $u_6=0.149\,074$, $u_8=0.117\,414$. These values are believed to be correct to about one unit in the last quoted digit. A check of the calculation of Ingham and Jones [Proc. Roy. Soc. (London) 107, 636 (1925)] of the inverse-sixth-power lattice sums reveals that their sum for the bcc lattice is not correct to the accuracy quoted but should be 12.253 662 rather than 12.2533^{70}_{66}. The check between the spectral determination of u_4 and this value is then validated to almost one part in 10^6. The quoted value of the sum S_3 should be changed to 2.0389×10^{-2}, while S_4 is unchanged. These changes cause the lower end of the transition line in Fig. 4 to lie slightly higher without any change in the conclusions. The vibrational spectra are not changed to the accuracy quoted.

The following reference was inadvertently omitted from Ref. 10: F. W. de Wette, Phys. Rev. 135, A287 (1964).

The equation for the transition line on p. 3477 should have $\Delta h(\tau)$ rather than $h(\tau)$ on the left-hand side. The fifth sentence starting on p. 3478 should end with 10^2 rather than 10^{-2}.

Anisotropic X-Ray Absorption in a Single Crystal of Gallium, A. I. Kostarev and W. M. Weber [Phys. Rev. B 3, 4124 (1971)]. In line 1 of the abstract "unpolarized" should read "unpolarized and polarized." On p. 4130, paragraph 1, "$\phi_1=84°$ in (10)..." should read "$\phi_0=84°$ in (10)....."

Energy Levels of Bloch Electrons in Magnetic Fields, H. H. Hosack and P. L. Taylor [Phys. Rev. B 3, 4091 (1971)]. In the last term of Eq. (2) the denominator

$$2\mathcal{E}_g[1-(n+\tfrac{1}{2})\hbar\omega/\mathcal{E}_g$$

should be replaced by

$$2\mathcal{E}_g[1-(n+\tfrac{1}{2})\hbar\omega/\mathcal{E}_g]^{1/2}\,.$$