

# Supplementary Material – Extracting electron densities in $n$ -type GaAs from Raman spectra: Comparisons with Hall measurements.

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## I. APPROXIMATE FORMS FOR $\chi_e^0$

When the temperature  $T$  is zero, the Fermi function is the unit step function. Then, the integral in Eq. (6) in the main text has an analytic evaluation. For  $T$  finite, analytic evaluations are not possible. From the Lindhard expression for the susceptibility  $\chi_e^0$ , Eq. (6), we obtain approximate forms as expansions in terms of  $Q$  and  $R = Q^2/\Omega^2$ . We will show that up to fourth order these expansions lead to identical expressions, and will show that we can obtain the coefficients to all orders for the series in terms of powers of  $R$ . We introduce the auxiliary functions

$$g(Q, \Omega) = \frac{1}{Q^2 + 2QK\mu - \Omega^2} \quad (\text{S1})$$

$$h(Q, \Omega) = \frac{1}{Q^2 - 2QK\mu + \Omega^2}, \quad (\text{S2})$$

and note that the integrand in Eq. (6) equals  $K^2 f(R_\infty K^2/m_C)(g(Q, \Omega) + h(Q, \Omega))$ .

**1. Expansion for small  $Q$**  We write the functions  $g$  and  $h$  as Taylor expansions near  $Q = 0$ , that is

$$g(Q, \Omega) = \sum_{n=0}^{\infty} \frac{Q^n}{n!} g^{(n)}(0, \Omega) \quad (\text{S3})$$

$$h(Q, \Omega) = \sum_{n=0}^{\infty} \frac{Q^n}{n!} h^{(n)}(0, \Omega). \quad (\text{S4})$$

Explicitly and up to 4th order

$$g(Q, \Omega) \approx -\frac{1}{\Omega^2} - Q \frac{2K\mu}{\Omega^4} - Q^2 \frac{(2K\mu)^2 + \Omega^2}{\Omega^6} - Q^3 \frac{(4K\mu\Omega^2 + 8K^3\mu^3)}{\Omega^8} - Q^4 \frac{\Omega^4 + 3(2K\mu)^2\Omega^2 + (2K\mu)^4}{\Omega^{10}} \quad (\text{S5})$$

$$h(Q, \Omega) \approx \frac{1}{\Omega^2} + Q \frac{2K\mu}{\Omega^4} + Q^2 \frac{(2K\mu)^2 - \Omega^2}{\Omega^6} - Q^3 \frac{(4K\mu\Omega^2 + 8K^3\mu^3)}{\Omega^8} + Q^4 \frac{\Omega^4 - 3(2K\mu)^2\Omega^2 + (2K\mu)^4}{\Omega^{10}}. \quad (\text{S6})$$

Substituting Eqs. (S5) and (S6) into Eq. (6) in the main text, and integrating with respect to  $\mu$  we find that terms containing odd powers in  $\mu$  vanish. The resulting form for the susceptibility  $\chi_e^0$ , up to fourth order in  $Q$ , is

$$4\pi\chi_e^0(\mathbf{q}, \omega + i\gamma) = \frac{4m_C}{Q^2\pi} \int_0^\infty dK K^2 f(R_\infty K^2/m_C) \left( -Q^2 \frac{4}{\Omega^4} - Q^4 \frac{16K^2}{\Omega^8} \right). \quad (\text{S7})$$

Next, we observe that  $K = \sqrt{(2m_o m_C a_B^2/\hbar^2)E} = \sqrt{(m_C/R_\infty)E}$ , such that  $dK = \frac{m_C}{2KR_\infty} dE$ , and where the zero in energy coincides with the maximum of the valence band at the  $\Gamma$  point. Thus, the first term in Eq. (S7) is proportional to

$$\int_0^\infty dK K^2 f(R_\infty K^2/m_C) = \frac{m_C}{2R_\infty} \int_0^\infty dE \sqrt{(m_C/R_\infty)E} f(E) = \frac{m_C N_e}{2R_\infty}. \quad (\text{S8})$$

In the same fashion, the second term in Eq. (S7) is

$$\int_0^\infty dK K^4 f(R_\infty K^2/m_C) = \frac{m_C}{2R_\infty} \int_0^\infty dE ((m_C/R_\infty)E)^{3/2} f(E), \quad (\text{S9})$$

**2. Expansions for small  $R$**  Here we approximate the functions  $g$  and  $h$  as a series in powers of  $R$ , for  $|R| < 1$ . First we rewrite  $g$  and  $h$  as follows

$$g(Q, \Omega) = -\frac{1}{\Omega^2} \frac{1}{1 - Q^2/\Omega^2 - 2QK\mu/\Omega^2} = -\frac{1}{\Omega^2} \frac{1}{1 - R(1 + 2K\mu/Q)} \quad (\text{S10})$$

$$= -\frac{1}{\Omega^2} \sum_{n=0}^{\infty} R^n \left(1 + \frac{2K\mu}{Q}\right)^n \quad (\text{S11})$$

$$h(Q, \Omega) = \frac{1}{\Omega^2} \frac{1}{1 + Q^2/\Omega^2 - 2QK\mu/\Omega^2} = \frac{1}{\Omega^2} \frac{1}{1 + R(1 - 2K\mu/Q)} \quad (\text{S12})$$

$$= \frac{1}{\Omega^2} \sum_{n=0}^{\infty} (-1)^n R^n \left(1 - \frac{2K\mu}{Q}\right)^n \quad (\text{S13})$$

In the first step we factor either  $\Omega^2$  or  $-\Omega^2$  from the denominator, and in the second step we use the geometric series:

$$\frac{1}{1+x} = \sum_{n=0}^{\infty} (-1)^n x^n. \quad (\text{S14})$$

Note that this approach provides the coefficients for all orders explicitly, in contrast with the situation in Sec. 1, where one needs to compute the derivatives for  $g$  and  $h$  and evaluate them at  $Q = 0$ , which might be cumbersome for higher orders. In this sense, the expansion in terms of  $R$  provides a technical advantage. Up to 3th order in  $R$

$$g(Q, \Omega) \approx -\frac{1}{\Omega^2} \left(1 + R \left(1 + \frac{2K\mu}{Q}\right) + R^2 \left(1 + \frac{2K\mu}{Q}\right)^2 + R^3 \left(1 + \frac{2K\mu}{Q}\right)^3\right) \quad (\text{S15})$$

$$h(Q, \Omega) \approx \frac{1}{\Omega^2} \left(1 - R \left(1 - \frac{2K\mu}{Q}\right) + R^2 \left(1 - \frac{2K\mu}{Q}\right)^2 - R^3 \left(1 - \frac{2K\mu}{Q}\right)^3\right), \quad (\text{S16})$$

from which it follows that

$$g(Q, \Omega) + h(Q, \Omega) \approx -\frac{1}{\Omega^2} \left[2R + R^2 \left(\frac{8K\mu}{Q}\right) + R^3 \left(2 + 6 \left(\frac{2K\mu}{Q}\right)^2\right)\right]. \quad (\text{S17})$$

Substituting Eq. (S17) into Eq. (6) and integrating with respect to  $\mu$ , we obtain

$$4\pi\chi_e^0(\mathbf{q}, \omega + i\gamma) = \frac{4m_C}{\pi Q^2} \int_0^{\infty} dK K^2 f(R_{\infty} K^2/m_C) \left(-Q^2 \frac{4}{\Omega^4} - Q^4 \frac{16K^2}{\Omega^8} - \frac{4Q^6}{\Omega^8}\right). \quad (\text{S18})$$

If we keep terms up to 4th order in  $Q$  in the integrand, we obtain the approximate form for the susceptibility in Eq. (6). This observation supports our statement that a large number of approximate models for  $\chi_e^0$  can be obtained as power series of  $R$ .

In Fig. S1 we compare the exact line shape, as obtained from Eq. (7) utilizing Eq. (6), with the approximate form obtain by approximating  $\chi_e$  to zero and second order in  $Q$  to recover a Drude-like models.

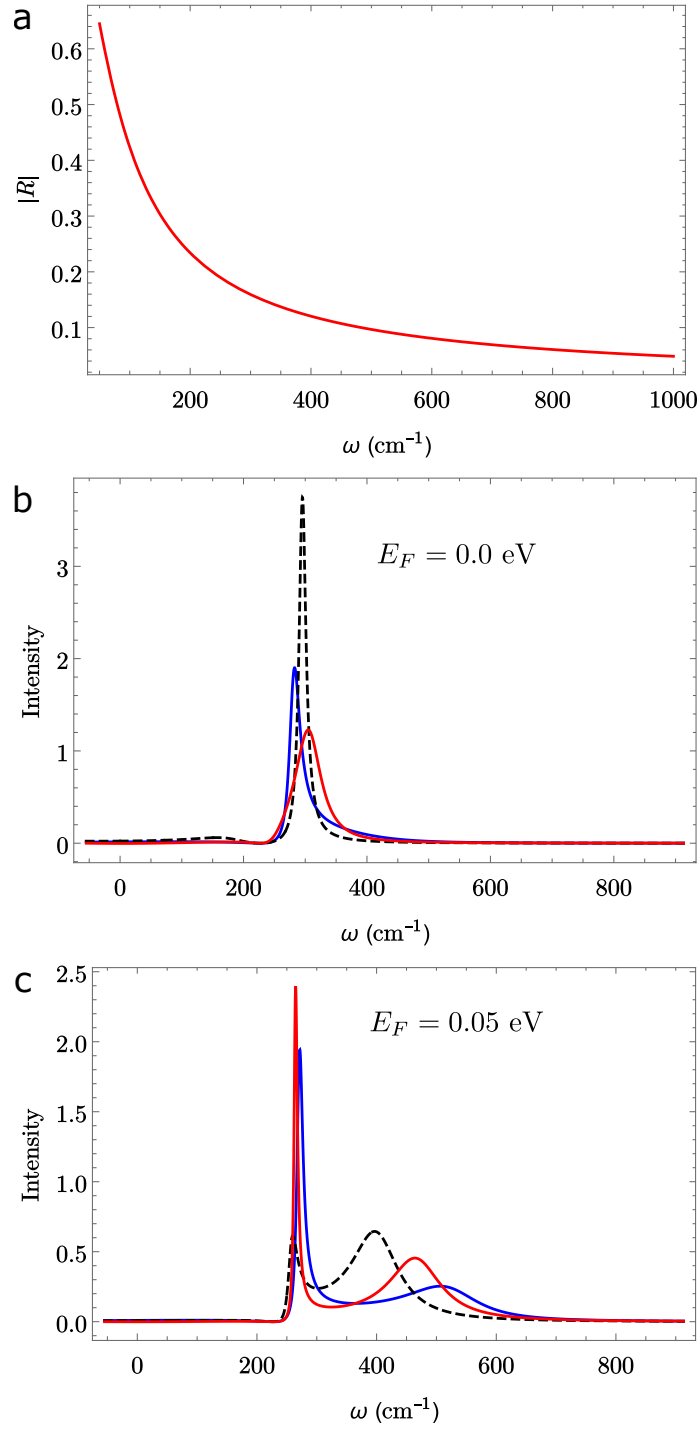


FIG. S1. A comparison between the exact (solid, blue) and zero (black, dashed) and second (red, solid) order approximate line shapes. In a) we show how  $|R|$  varies with the Raman frequency and b) and c) show the line shapes for two different values in the Fermi energy. The second order approximation in  $Q$  is needed to interpret Raman spectra. Other parameters are  $C_{FH} = -0.28$ ,  $\omega_{LO} = 284.7 \text{ cm}^{-1}$ ,  $\omega_{TO} = 267.8 \text{ cm}^{-1}$ ,  $m_{\text{eff}} = 0.067m_o$ ,  $\varepsilon_{\infty} = 10.9$ ,  $q_o = 1.03 \times 10^8 \text{ m}^{-1}$ .

## II. PARAMETERS

The following tables present the parameters used in the calculation of Raman line shape for GaAs as well as the four models for the band structure. The energies of the extrema of the conduction and valence sub-bands are referenced to the the bottom of the conduction sub-band at the  $\Gamma$  symmetry point in the Brillouin zone of the reciprocal lattice space.

Parameter	Symbol	Value
lattice constant (nm)	$a_L$	0.565
static dielectric constant	$\epsilon_0$	13.1
high frequency dielectric constant	$\epsilon_\infty$	10.9
longitudinal optical (LO) phonon energy (eV)	$\omega_{LO}$	0.0353
transverse optical (TO) phonon energy (eV)	$\omega_{TO}$	0.0332
Faust-Henry coefficient	$C_{FH}$	-0.28
Effective mass for the single equivalente conduction band $m_0$	$m_C$	0.067

TABLE S1. Dielectric response function input parameters for the intrinsic zinc blende GaAs at 300 K. The mass of the free electron is  $m_0$ . These GaAs data are from Ref. 25.

Parameter	Symbol	Value
bandgap (eV)	$E_G =   -E_{\nu\Gamma}  $	1.424
effective mass for conduction band DOS ( $m_0$ )	$m_C$	0.067
effective mass for valence band DOS ( $m_0$ )	$m_V$	0.572
number of equivalent conduction bands	$N_C$	1
number of equivalent valence bands	$N_V$	1

TABLE S2. BGN model input parameters for intrinsic zinc blende GaAs at 300 K. Data taken from Ref. 25.

Parameters	Symbol	Value
bandgap (eV)	$E_G =   -E_{\nu\Gamma}  $	1.424
bottom of the conduction L sub-band (eV)	$E_{cL}$	0.29
bottom of the conduction X sub-band (eV)	$E_{\nu X}$	0.48
top of the degenerate valence $\Gamma$ sub-band (eV)	$-E_{\nu\Gamma}$	1.424
spin-orbit splitting (eV)	$-E_{so}$	0.34
top of split-off valence $\Gamma$ sub-band (eV)	$-E_{so\Gamma} = -E_{\nu\Gamma} - E_{so}$	1.764
effective mass of conduction $\Gamma$ sub-band ( $m_0$ )	$m_{c\Gamma}$	0.063
non-parabolicity factor for conduction band $\Gamma$ sub-band	$\zeta$	0.824
transverse $L$ sub-band mass ( $m_0$ )	$m_{tL}$	0.075
longitudinal $L$ sub-band mass ( $m_0$ )	$m_{lL}$	1.9
effective mass of conduction $L$ sub-band ( $m_0$ )	$m_{cL} = (m_{lL} m_{tL}^2)^{1/3}$	0.222
transverse $X$ sub-band mass ( $m_0$ )	$m_{tX}$	0.19
longitudinal $X$ sub-band mass ( $m_0$ )	$m_{lX}$	1.9
effective mass of conduction $X$ sub-band mass ( $m_0$ )	$m_{cX} = (m_{lX} m_{tX}^2)^{1/3}$	0.409
light hole mass of valence $\Gamma$ sub-band ( $m_0$ )	$m_{lh}$	0.082
heavy hole mass of valence $\Gamma$ sub-band ( $m_0$ )	$m_{hh}$	0.51
effective mass of valence $\Gamma$ sub-band ( $m_0$ )	$m_{\nu\Gamma}$	0.53
split-off band mass of the valence sub-band at $\Gamma$ ( $m_0$ )	$m_{so}$	0.15
number of equivalent conduction $L$ sub-bands	$N_{cL}$	4
number of equivalent conduction $X$ sub-bands	$N_{cX}$	3

TABLE S3. PDOS model input parameters for intrinsic zinc blende GaAs at 300 K. Data from Ref. 25

Parameter	Symbol	Value
$\Gamma$ sub-band coefficients	$E_{\Gamma 0}$	1.519 eV
	$A_{\Gamma}$	0.540 meV/K
	$B_{\Gamma}$	204 K
$L$ sub-band coefficients	$E_{L 0}$	1.815 eV
	$A_L$	0.605 meV/K
	$B_L$	204 K
$X$ sub-band coefficients	$E_{X 0}$	1.981 eV
	$A_X$	0.460 meV/K
	$B_X$	204 K

TABLE S4. Coefficients for the temperature dependence of the conduction band extrema. Data taken from Ref. 25

Band model	Coefficient	Value
BGN	$a_0$	17.3292
	$a_1$	13.1545 ( $\text{eV}^{-1}$ )
	$a_2$	-37.4789 ( $\text{eV}^{-2}$ )
	$a_3$	26.5678 ( $\text{eV}^{-3}$ )
	$a_4$	53.7760 ( $\text{eV}^{-4}$ )
PDOS2	$a_0$	17.5156
	$a_1$	12.8520 ( $\text{eV}^{-1}$ )
	$a_2$	-34.5783 ( $\text{eV}^{-2}$ )
	$a_3$	-27.3728 ( $\text{eV}^{-3}$ )
	$a_4$	223.720 ( $\text{eV}^{-4}$ )
PDOS4	$a_0$	17.5198
	$a_1$	12.3461 ( $\text{eV}^{-1}$ )
	$a_2$	-35.5542 ( $\text{eV}^{-2}$ )
	$a_3$	40.1630 ( $\text{eV}^{-3}$ )
	$a_4$	0
PDOSNPG	$a_0$	17.6913
	$a_1$	13.8441 ( $\text{eV}^{-1}$ )
	$a_2$	-31.5108 ( $\text{eV}^{-2}$ )
	$a_3$	-57.3738 ( $\text{eV}^{-3}$ )
	$a_4$	544.398 ( $\text{eV}^{-4}$ )

TABLE S5. Coefficients used in Eq. (18) and Fig. 4 in the main text for the four band models for GaAs.